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Plastic pollution in Moreton Bay sediments, Southeast Queensland, Australia

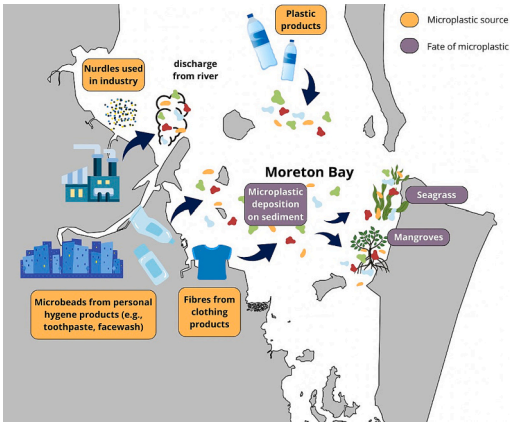
Elvis D. Okoffo [a,*], Emmeline Tan [b], Alistair Grinham [c], Sai Meghna Reddy Gaddam [b], Josie Yee Hang Yip [b], Alice J. Twomey [b], Kevin V. Thomas [a], Helen Bostock [b]

[a] Queensland Alliance for Environmental Health Sciences (QAEHS), The University of Queensland, 20 Cornwall Street, Woolloongabba, QLD 4102, Australia
[b] School of the Environment, Faculty of Science, The University of Queensland, Australia
[c] School of Civil Engineering, Faculty of Engineering, Architecture, and Information Technology, University of Queensland, Australia

HIGHLIGHTS

- Quantitative analysis of seven types of plastics in marine sediment samples by Pyr-GC/MS
- Total plastic concentrations ranged from 3.3 to 2194.2 μg/g of sediment across sites.
- Polyethylene dominates plastics in the sediment's samples.
- Estimated minimum plastic budget for Moreton Bay is around 7000 t.

GRAPHICAL ABSTRACT



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ABSTRACT

The mounting issue of plastic waste in the aquatic ecosystem is a growing source of concern. Most plastic waste originates on land and a significant proportion of this eventually finds its way into the marine environment, which is widely regarded as a major repository for plastic debris. Currently, there exists a substantial gap in our understanding of how much plastic, the main polymer types, and the distribution of plastic in the marine environment. This study aimed to provide information on mass concentrations of a range of plastics in the surface sediments in the semi-enclosed Moreton Bay, just offshore the large city of Brisbane, Southeast Queensland, Australia. Surface sediment samples were quantitatively analysed for a suite of 7 common plastic polymer types (i.e., polystyrene (PS), polycarbonate (PC), poly-(methyl methacrylate) (PMMA), polypropylene (PP), polyethylene terephthalate (PET), polyethylene (PE) and polyvinyl chloride (PVC)) using a pressurized liquid extraction (PLE) followed by double-shot microfurnace pyrolysis coupled to gas chromatography mass spectrometry (Pyr-GC/MS). The advantage of this approach is that it can measure plastics below the limit of visual detection. The study revealed that $\Sigma_7$plastics were consistently present in the samples, although the

* Corresponding author.
E-mail address: e.okoffo@uq.edu.au (E.D. Okoffo).

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concentrations displayed a wide range of concentrations from 3.3 to 2194.2 μg/g across different sites. Among the polymers analysed, PE and PVC were found at the highest concentrations, ranging from 2.3 to 1885.9 μg/g and 3.0–979.5 μg/g, respectively. Based on the average concentrations of plastics measured, the dry bulk density and volume of sediments within the top 10 cm of the bay, it was estimated that there is a minimum of 7000 t of plastics stored in the surface sediments of the bay. This study is the first to report the mass concentrations of identified plastics and identify the main polymer types in Moreton Bay. This is important information to develop management plans to reduce the plastic waste entering the coastal marine environment.

1. Introduction

Small plastic particles (i.e., microplastics, particles <5 mm and nanoplastics, particles <1 μm), formed during the degradation of larger plastic products, are found throughout the oceans (Martin et al., 2020; Gomiero et al., 2019; Mani et al., 2019; Haave et al., 2019). Most plastic pollution is washed into waterways and delivered to the coast by rivers, where it is primarily trapped nearshore in estuaries and coastal ecosystems (Haave et al., 2019; Besseling et al., 2017; Eerkes-Medrano et al., 2015; Dibke et al., 2021; Li et al., 2016; Shi et al., 2022; Hidalgo-Ruz et al., 2012), with some making it offshore into the open ocean (Martin et al., 2020; Pattiaratchi et al., 2022). While the sight of visible plastic debris in the rivers and on the beach is distressing, the presence and impact of microplastics go largely unnoticed due to their microscopic size (Hidalgo-Ruz et al., 2012). However, the consequences of plastic particles in our waterways are far-reaching and profound. These tiny plastic particles can be ingested by microscopic aquatic organisms, leading to disruptions in the food chain and subsequent consequences for marine ecosystems (Besseling et al., 2017; Eerkes-Medrano et al., 2015; Dehaut et al., 2018; Cox et al., 2019; Waring et al., 2018). Evidence suggests the environmental fate of plastics can have negative impacts on invertebrates, including crustaceans and bivalves, reducing survival rate, feeding ability, and immune system function (Li et al., 2016; Rochman et al., 2017). Moreover, the consumption of plastics by seafood species poses a threat to human health (Ribeiro et al., 2020; Ribeiro et al., 2021). Additionally, plastics can leach harmful chemicals into the environment and attract and concentrate heavy metals and organic pollutants, exacerbating the contamination of marine habitats (Rillig et al., 2021; Ribeiro et al., 2019).

According to global assessments, merely 1 % of plastic particles that enter the marine environment remains afloat on the surface waters (Law, 2017; van Sebille et al., 2015). These findings have prompted some to hypothesize that plastic particles may descend to deeper layers within the water column (Wang et al., 2016; Kooi et al., 2017; Reisser et al., 2015) and become trapped in sediment (Van Cauwenberghe et al., 2013). Hence, the sequestration of plastic particles in sediment is regarded as the ultimate repository for marine plastic pollution, offering an explanation for the unexpectedly low levels discovered in surface waters (Martin et al., 2020). The impact of plastic debris on marine organisms is influenced by various factors that determine their distribution and availability in either the water column or sediments (Gomiero et al., 2019). Typically, high-density particles tend to sink and accumulate in sediments, while low-density particles remain buoyant on the sea's surface (Gomiero et al., 2019). Nonetheless, due to the formation of biofilm on their surfaces (Oberbeckmann et al., 2015; Rummel et al., 2017), low-density plastic particles can eventually settle and amass on the seafloor, alongside non-buoyant plastic particles, posing a threat to benthic organisms and ecosystems.

Despite many recent reported studies (Gomiero et al., 2019; Brandon et al., 2019; Zhu et al., 2021; Zhang et al., 2021; Stile et al., 2021), there is still a substantial lack of knowledge and information on the mass concentration and amount of plastic pollution in many coastal environments, especially in Australia. The goal of this study was to determine the concentration, distribution and main polymer types contributing to plastic pollution within the surface sediments of the enclosed coastal embayment of Moreton Bay, just offshore the city of Brisbane, Southeast Queensland, Australia. The bay is subject to diverse sources of pollution and anthropogenic pressure such as aquaculture (West et al., 2019) and inshore fisheries (Thurstan et al., 2019; Pascoe et al., 2014) tourist activities (Ruhanen et al., 2019), shipyards, leisure and commercial vessels (Cohen et al., 2019) and household and industrial discharges (Townsend et al., 2019; Morelli and Gasparon, 2019), including sewage and urban stormwater discharges (Saeck et al., 2019). Within the catchments there is also significant agricultural runoff that results in the direct loading of pesticides, nutrients, and sediments during major flood events (Grinham et al., 2021; Lockington et al., 2017; Grinham et al., 2018). However, there is currently limited information on the extent of plastic pollution in the enclosed Moreton Bay, and how it might be impacting the range of marine ecosystems within the bay. By determining the concentration and distribution of the plastic polymers accumulating in the bay, we can start to address this pervasive problem of marine plastic pollution to preserve the natural ecosystems of the economically, socially, and culturally significant Moreton Bay, also known as Quandamooka Country to the Traditional Owners in the region. Traditional names are used in the text for the islands within the bay.

Regional setting.

Moreton Bay is a large coastal embayment that extends 115 km long and 40 km wide. With an area of 1523 km^2 and an average depth of 6.8 m, Moreton Bay is one of Australia's largest estuarine systems. The geomorphology of Moreton Bay is characterized by both oceanic and riverine processes. There are three oceanic tidal channels called North and South Passages, while the large sand islands North Stradbroke Island/Minjerrabah and Moreton Island/Mulgumpin, that form the eastern side of the bay, are dominated by wave processes. There are four large rivers that flow directly into the bay including the Brisbane, Logan, Caboolture, and Pine rivers (Gibbes et al., 2014). The Brisbane River has the largest catchment of 13,600 km^2, which starts in Great Dividing Range, then flows through native forest and rural farmland used for grazing and cropping, before flowing through the centre of Ipswich and Brisbane City and out through the Brisbane Port at the highly modified river mouth. The Logan is the second largest catchment of 3860 km^2 and flows from the rainforests on peaks of the Main Range National Park through rural farmland and through the city of Logan into the south of Moreton Bay (Neil, 1998). Over the last century, since European invasion, there has been significant intensification of land clearing for farming and urbanisation. This has led to two-fold increase in the area of mud cover in central Moreton Bay between 1970 and 2015 (Lockington et al., 2017). Most of this mud has been transported into the bay during high rainfall/flooding events associated with La Nina such as 2011[41] and recent floods in 2022.

Moreton Bay is host to a wide diversity of different marine ecosystems including internationally recognised RAMSAR wetlands protected for migratory birds, seagrass, mangroves, and saltmarshes, and coral reefs (Pandolfi et al., 2019). Many of these ecosystems have been impacted by anthropogenic activities, especially the increasing amounts of mud entering the bay (Lockington et al., 2017). The extent of seagrass in the bay is dynamic depending on recent flood events (Maxwell et al., 2019; Kovacs et al., 2019), while mangroves have recently been expanding into the saltmarsh regions in the bay due to rising sea levels (Lovelock et al., 2019). Most of these coastal marine ecosystems have been found to be very effective at trapping mud and plastic particles

(Huang et al., 2020), therefore some of our samples deliberately targeted sediment samples from the seagrass and mangrove areas or regions.

2. Materials and methods

2.1. Chemicals and materials

Information on the chemicals and materials used in the study are provided in the Supporting Information (Text 1).

2.2. Sediment sampling

Sediment samples were collected from 47 sites within Moreton Bay, from around the Brisbane River mouth area (north), and from the southern part of the bay fed by the Logan River, as well as from mangroves and seagrass beds on North Stradbroke Island/Minjerrabah (Fig. 1). Global positioning system (GPS) coordinates were recorded from each sampling site (Table S1). It should be noted that the samples were collected for other projects analysing mud, heavy metals and pollutants in the bay and we have utilised what samples were available for the region at the time of this study. A stainless-steel sediment grab sampler (Van Veen grab sampler, Kc Denmark A/S, Silkeborg, Denmark) was used to collect the samples from a small work boat. The surface area of the grab sampler was 250 cm^2 and the maximum penetration depth was 10 cm. After each retrieval, the sediments were released onto a stainless-steel tray, and samples were then scooped up directly using pre-cleaned wide-mouth clear glass jars (125 mL). Sediment samples from mangrove and seagrass areas were collected directly via scooping the sediment directly into glass jars during low tide. These glass jars were rinsed with MilliQ water, followed by acetone and dichloromethane (DCM), and then wrapped in aluminium foil prior to sampling. During the sample collection process, field blanks ($n = 5$, opened glass jars filled with pre-DCM cleaned hydromatrix (inert diatomaceous earth sorbent)) were used to estimate the degree of contamination of the samples during sampling. After the sampling, the samples were immediately placed in a cool, dark storage container and transported directly to the laboratory, where they were stored at 4 °C until they were analysed.

2.3. Sample processing

Sediment samples were processed and extracted following a previously published method developed for analysing plastics in biosolids (Okoffo et al., 2020a; Okoffo et al., 2020b). The initial aim of this study was to test this method for use on marine sediment samples. Briefly, sediment samples were mixed thoroughly using a metal spoon while wet, freeze-dried and pulverised to fine grains with a mortar and pestle. Following this, the samples were sieved through 2 mm sieves (from Endecott), to ensure homogeneity and to remove larger particles such as shell fragments and gravels. The mass concentration data obtained in this study with the combined PLE and Pyr-GC/MS methods are independent of particle size, colour, shape, or particle numbers and allows for a more accurate reporting of the total mass of plastics (including both microplastics (particles <2 mm) and nanoplastics (particles <1 μm) in each sediment sample. To minimize potential contamination of samples, sieves were removed immediately after sieving, wrapped, and stored in aluminium foil. Between samples, the mortar and pestle and the sieves used were thoroughly washed with MilliQ water and DCM to avoid cross contamination. For every sample pretreatment batch, 5 procedural blanks (consisting of pre-washed hydromatrix) were treated the same way as samples. Approximately 5 ± 0.1 g of each sediment sample obtained were weighed into a 10 mL pre-cleaned (with DCM) PLE stainless steel extraction cell and filled or topped up to the brim with pre-cleaned (with DCM) hydromatrix. After filling the cells, the samples were spiked with 20 μL of 2 mg/mL solution of deuterated polystyrene (PS-d_5) internal standard and extracted using DCM at 180 °C and 1500 psi as previously reported (Okoffo et al., 2020a; Okoffo et al., 2020b). Immediately after extraction, 240 μL of each sample extract was transferred into 80 μL pyrolysis cups (Eco-Cup LF, Frontier Labs, Japan), evaporated for 30 min in a fume hood while covered in aluminium foil to prevent contamination from airborne contaminants and loaded on an auto sampler for Pyr-GC/MS analysis. The applied PLE extraction and Pyr-GC/MS methods have been validated to resolve issues related to the thermal degradation, dissolution, solubility, and extracts stability known for some of the plastic types analysed (Okoffo et al., 2020a; Okoffo et al., 2020b). Previous work using this method has successfully quantified the amount of plastic in 1 ± 0.1 g biosolid samples from single WWTPs. Due to the lower concentration of plastic in marine sediments we initially analysed 1 g and 5 g of sediments for plastics detection. We chose 5 g to get a more representative concentration of plastics. The validation of the methods with sediments samples are explained in the method performance section below.

2.4. Plastics analysis

The sediment samples were analysed for 7 common plastic types (i.e. polystyrene (PS), polycarbonate (PC), poly-(methyl methacrylate) (PMMA), polypropylene (PP), polyethylene terephthalate (PET), polyethylene (PE) and polyvinyl chloride (PVC)) using a double-shot component of a Multi-Shot Micro-Furnace Pyrolyzer (EGA/PY-3030D)

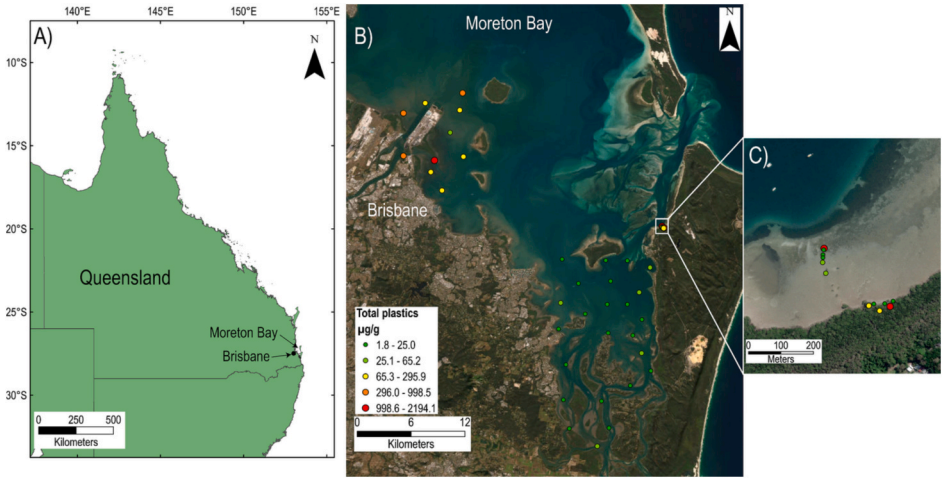

Fig. 1. An aerial view of southern Moreton Bay region in Southeast Queensland, Australia (Fig. 1A). Fig. 1B shows the 47-sediment sampling locations that were the focus of this study. Fig. 1C depicts a zoomed in sampling locations for the seagrass and mangrove sites on Minjerabah (North Stradbroke Island).

equipped with an auto-shot sampler (AS-1020E) (both Frontier Lab's, Japan) and attached to a Shimadzu GC/MS – QP2010-Plus (Shimadzu Corporation, Japan) as previously reported (Okoffo et al., 2020a; Okoffo et al., 2020b). These plastic types account for >70 % of the global plastics demand (PlasticsEurope, 2022) and are commonly consumed in Australia (represents >50 % of total plastics consumed) (Kyle O'Farrell, 2021; O'Farrell, 2018; O'Farrell, 2019). The first pyrolysis shot of the double-shot technique (ramped from 100 to 300 °C) was used as a clean-up step to thermally remove potentially interfering volatile and semi-volatile organic materials co-extracted from the sediment samples. The second pyrolysis shot (conducted at 650 °C for 12 s) was used to quantitatively measure the plastics identified in the samples. Detailed double-shot Pyr-GC/MS parameters are given in Table S2 of the Supporting Information. To improve the selectivity in the quantification of PET and PC in the samples, a second aliquot of the extracted samples were derivatized by adding 10 μL of tetramethylammonium hydroxide (TMAH, 25 % in methanol, Sigma-Aldrich) to the individual samples in pyrolysis cups prior to analysis (Okoffo et al., 2021). It should be noted however that the thermochemolysis of the samples were performed in single-shot mode at 650 °C for 0.2 min (12 s) as the indicator ions selected for PET and PC quantification were not stable using the double-shot parameters (Okoffo et al., 2021). That is, the sample extracts were run in duplicate, one using the double shot method and one using the single shot method (for the thermochemolysis). Besides the change in pyrolysis temperature, all the other parameters remained the same.

2.5. Plastics quantification

To identify and quantify the target plastic types in the sediment samples, specific indicator ions were selected for each polymer type as previously described (Okoffo et al., 2020a; Okoffo et al., 2020b; Okoffo et al., 2021) (summarised in Table S3). The applied method was optimized to resolve indicator ions selectivity against several biogenic polymers and organic materials, and matrix-related issues known for some of the plastic types (Okoffo et al., 2020a; Okoffo et al., 2020b). Calibration curves were performed for the 7 plastic types by PLE extracting standards and aliquoting into pyrolysis cups (8 points ranging from 0.08 to 33 μg/cup, having $R^2 \geq 0.95$). Calibration curves injected at the beginning and end of each batch were calculated by plotting the peak area ratio of the selected indicator ions to the internal standard ions, (i.e., PS-d_5-monomer, Table S3) versus the concentration of each target plastic type and using the integration results for quantification. PS-d_5 was used as an internal standard for all the target plastics (all values reported in this study were corrected for the recovery of the internal standard). The limit of quantification (LOQ) for each target plastic was calculated as the concentration of a peak with a signal to noise ratio of 10:1 or as 10 times the baseline noise. Method detection limits (MDLs) were calculated from concentrations in field, procedural and laboratory blanks and calculated as the mean concentration + 3 times the standard deviation of detected concentrations, Table S4. For calculating average concentrations and standard deviations in blanks, ½ LOQ was inserted where concentrations were < LOQ. For plastic types where all blanks were below LOQs then MDLs are listed as the LOQ (Table S4). All reported data were blank subtracted.

2.6. Quality assurance and quality control

Extensive quality assurance and quality control (QA/QC) measures were taken to minimize and prevent possible contamination of the samples including the use of 100 % cotton laboratory coats, avoiding the use of plastic materials whenever possible during sample collection, pre-treatment, extraction, analysis, and laboratory procedures, working in a fume hood where possible and use of field, laboratory, and procedural blanks. The field, laboratory, and procedural blanks were used to monitor processing and extraction contamination. Detailed QA/QC procedures are provided in Text S3. In all fields, laboratory and procedural blank samples, the target plastics were either not detected or close to the MDLs (Tables S4).

2.7. Plastic budget for surface sediments of Moreton Bay

To calculate an **estimated** budget of plastics for the surface sediments of Moreton Bay, we adapted an approach that is used to estimate sediment budgets in coastal and estuarine environments (Bostock et al., 2007). We used the average plastic concentration (C – μg/g) × the average dry bulk density (DBD – g/cm^3) of the sediment samples (see below; Table S5) × the area of the bay (A - km^2) × the depth (D – 10 cm; depth of grab sample - see above (USEPA, 2001)):

$$\text{Total mass of plastic} = C \times DBD \times A \times D_{10}$$

The dry bulk density (DBD) of each sediment sample is determined by taking a known volume of sediment from the grab sample using a 5 cm^3 (5 cc) syringe, which is then dried to determine the dry weight of the known volume (APHA, 1998) (Table S5). This allows for the conversion of the concentration of plastic per gram to a concentration by volume of sediment. DBD is calculated:

$$\text{DBD}\left(\text{g/cm}^3\right) = \text{Dry Weight (g)}/\text{Volume}\left(\text{cm}^3\right)$$

Due to the differences in the plastic concentration for each region, the average $\Sigma_7$plastic was determined for each region – North, South (including seagrass, as most of the seagrass is in the southern area (Maxwell et al., 2019) and mangroves (Lovelock et al., 2019):

$$\text{Total}_{\text{North}} = C_{\text{North}} \times DBD_{\text{North}} \times A_{\text{North}} \times D_{10}$$

$$\text{Total}_{\text{South}} = C_{\text{South}} \times DBD_{\text{South}} \times A_{\text{South}} \times D_{10}$$

$$\text{Total}_{\text{Mangrove}} = C_{\text{Mangrove}} \times DBD_{\text{Mangrove}} \times A_{\text{Mangrove}} \times D_{10}$$

The total mass of plastic for the entire Moreton Bay (MB) was then calculated based on the concentration for each region:

$$\text{Total plastic MB} = \text{Total}_{\text{North}} + \text{Total}_{\text{South (Seagrass)}} + \text{Total}_{\text{Mangroves}}$$

There are several limitations to this approach. First it uses the average plastic concentration in each of the regions, some of which have been poorly sampled and there is a wide range in concentrations in each of these regions, indicating the patchy nature of the plastic deposition. Second it assumes that plastic is only present down to 10 cm depth. Given that plastic was invented in the 1950s and became widely used in 1970s it is highly likely that plastic is present deep in the sediments across the bay.

3. Results and discussion

3.1. Method performance

Quantitative analysis of plastics in the sediment samples were performed by pressurized liquid extraction (PLE) followed by double-shot microfurnace pyrolysis coupled to gas chromatography mass spectrometry (Pyr-GC/MS). This method has previously been used for biological and biosolids samples, allowing for the extraction of both microplastics (particles <5 mm) and nanoplastics (particles <1 μm) particles of the seven most common plastic types (i.e., PC, PE, PET, PMMA, PP, PS and PVC) into DCM and subsequent quantification on Pyr-GC/MS. Here we validate the method for the identification and quantification of seven plastics in sediment samples – independent of particle size and capturing both microplastics and nanoplastics in the samples (Okoffo et al., 2020a; Okoffo et al., 2021). Compared with available particle-related characterization techniques such as Raman and Fourier-transform infrared (FT-IR)) spectroscopies that reports data on the number, size, shape, colour, and polymer types of particles, the method used in this study provides a mass-based concentration of the

total amount of plastics in samples that enables a mass concentration of plastics in the environment to be estimated. Despite the apparent advantages of using mass-based concentration data to quantify the pollution levels of plastics, limited studies have described the mass concentration of plastics in marine sediment samples.

To assess sample homogeneity and reproducibility of the selected method for plastics analysis within the entire sediment samples, ten duplicates of randomly selected sediment samples were analysed in different batches (i.e., 10 separate 5 g aliquots, from 5 sampling sites were individually examined). The relative standard deviation (RSDs, %) of the individual plastic concentrations in the duplicate samples were < 30 %. In brief, the average RSD, % between the samples were 7 % for PP, PC and PE, 13 % for PET, 5 % for PVC, 5 % for PMMA and 18 % PS. The RSD, % of the sum of total plastic concentrations ($\Sigma_7$plastics) in each 5 g extract ranged from 5 % to 22 % across all sites. Variability was influenced primarily by the PP, PVC, and PE concentrations.

3.2. Concentrations of plastics in Moreton Bay

Plastics were detected at all 47 sites with concentrations of the sum of the 7 plastics ($\Sigma_7$plastics) ranging 3.3–2194.2 μg/g, Table 1, Fig. 1. The plastic concentrations were highly heterogeneous in the surface sediment samples within the bay (Fig. 1). The highest concentrations of the total plastics ($\Sigma_7$plastics) were 2194.2 μg/g from site 54 A offshore the Brisbane River mouth (located in the northern region of the bay), 2124.2 μg/g from site S6 (seagrass area/region) and 1680.3 from site M5 (mangrove area/region), while several samples had very low plastic concentrations of 3–6 μg/g from the seagrass and mangrove sites on

Table 1
Concentrations of plastics (μg/g) in sediments samples. Where an analyte was not detected, <method detection limit (<MDL) is reported.

ID	Region	PP	PET	PS	PMMA	PVC	PC	PE	$\Sigma_7$plastics
56	North	4.3	<MDL	<MDL	<MDL	58.3	<MDL	126.3	189.0
55	North	2.6	2.6	<MDL	<MDL	61.7	<MDL	204.8	271.7
35	North	6.0	1.4	<MDL	<MDL	58.0	<MDL	68.4	133.8
N53	North	4.5	3.8	<MDL	<MDL	99.4	1.5	384.6	493.9
36	North	4.8	9.5	0.2	<MDL	206.6	2.8	774.7	998.5
38	North	6.5	1.8	<MDL	<MDL	47.3	<MDL	240.3	295.9
54 A	North	4.6	9.9	0.5	<MDL	292.7	0.7	1885.9	2194.2
52	North	2.1	1.5	<MDL	<MDL	33.1	<MDL	112.4	149.1
53	North	2.4	<MDL	1.4	<MDL	23.5	<MDL	25.6	53.0
N70	North	4.5	4.8	<MDL	<MDL	80.3	1.2	373.1	463.8
S5	Seagrass	<MDL	0.1	<MDL	<MDL	<MDL	<MDL	10.6	10.7
S6	Seagrass	76.1	<MDL	0.9	0.2	979.5	0.7	1066.9	2124.2
S4	Seagrass	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	3.3	3.3
S3	Seagrass	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	6.0	6.0
S1	Seagrass	<MDL	0.5	<MDL	<MDL	10.8	<MDL	53.9	65.2
S2	Seagrass	2.6	0.4	0.2	<MDL	7.0	<MDL	24.1	34.3
M1	Mangrove	2.4	<MDL	<MDL	<MDL	<MDL	<MDL	3.2	5.5
M4	Mangrove	141.7	11.9	0.7	<MDL	297.0	0.7	1228.3	1680.3
M5	Mangrove	<MDL	0.8	<MDL	0.3	18.9	1.7	114.2	135.7
M3	Mangrove	<MDL	<MDL	<MDL	<MDL	3.6	<MDL	10.1	13.7
M2	Mangrove	<MDL	0.3	<MDL	<MDL	5.5	<MDL	9.0	14.8
M6	Mangrove	0.9	0.8	<MDL	<MDL	21.4	0.5	109.2	132.9
1	South	1.1	4.4	3.2	<MDL	9.7	28.0	9.4	55.8
19	South	0.4	1.6	0.4	<MDL	9.8	21.0	13.9	47.0
123	South	<MDL	2.0	<MDL	<MDL	24.6	43.4	83.5	153.5
128	South	<MDL	1.6	<MDL	<MDL	19.8	12.0	15.1	48.5
129	South	<MDL	1.2	<MDL	<MDL	20.2	58.2	95.8	175.4
134	South	<MDL	0.8	<MDL	<MDL	9.5	17.2	23.9	51.5
136	South	<MDL	0.8	<MDL	<MDL	7.3	9.6	12.3	30.0
122	South	<MDL	0.7	<MDL	<MDL	14.6	22.0	13.7	50.9
2	South	0.5	1.2	1.1	<MDL	9.6	14.0	18.5	44.9
3	South	0.8	2.8	1.0	<MDL	11.8	24.2	25.0	65.6
117	South	<MDL	1.2	0.2	<MDL	6.0	19.6	30.7	57.7
119	South	<MDL	1.1	<MDL	<MDL	4.8	12.0	19.6	37.5
122	South	<MDL	0.7	<MDL	<MDL	14.6	22.0	13.7	50.9
163	South	<MDL	0.8	<MDL	<MDL	6.1	8.4	6.3	21.6
5	South	0.5	2.1	1.0	<MDL	13.6	32.2	11.6	61.0
6	South	0.4	2.8	0.4	<MDL	11.4	14.4	7.2	36.7
115	South	<MDL	<MDL	<MDL	<MDL	15.5	13.4	15.8	44.7
164	South	<MDL	2.4	<MDL	<MDL	8.1	31.6	9.2	51.3
177	South	<MDL	1.6	<MDL	<MDL	16.5	36.8	52.6	107.5
179	South	<MDL	11.6	<MDL	<MDL	8.4	12.0	11.8	43.8
7	South	0.4	4.2	0.5	<MDL	13.8	28.4	29.4	76.6
120	South	<MDL	0.8	<MDL	<MDL	6.7	12.0	8.6	28.2
124	South	0.8	1.8	<MDL	<MDL	4.9	6.4	6.5	20.4
126	South	<MDL	4.0	<MDL	<MDL	6.9	10.8	13.5	35.2
127	South	<MDL	0.2	<MDL	<MDL	3.0	9.4	2.3	14.8
Total		270.9	102.5	11.6	0.4	2581.7	528.7	7384.8	10,880.6
Minimum		0.4	0.1	0.2	0.2	3.0	0.5	2.3	3.3
Maximum		141.7	11.9	3.2	0.3	979.5	58.2	1885.9	2194.2
Mean		11.8	2.6	0.8	0.2	60.0	16.0	157.1	231.5
Median		2.4	1.6	0.6	0.2	13.8	12.0	19.6	51.5
SD		31.6	3.0	0.8	0.05	156.8	13.5	360.8	494.3
Sum of plastics (% contribution to total sum of plastics)		2.5	0.9	0.1	0.004	23.7	4.9	67.9	100

Polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polycarbonate (PC), polyethylene terephthalate (PET), polystyrene (PS), and poly-(methyl methacrylate) (PMMA).SD: standard deviations.

North Stradbroke Island/Minjerrabah. The sediment samples from the seagrass and mangrove areas were the most heterogeneous with some very high concentrations >1500 μg/g and some very low concentrations (3–6 μg/g). Overall, mangroves and seagrasses have been reported to be an effective trap for sediments and plastics (Martin et al., 2020) with studies demonstrating that environments with elevated sediment accumulation rates, such as vegetated coastal habitats like seagrasses, salt marshes, and mangroves, maintain persistent and substantial concentrations of plastics (Huang et al., 2020; Duarte et al., 2013; Middelburg et al., 1997; Mohamed Nor and Obbard, 2014; Jones et al., 2020; Cozzolino et al., 2020).

Therefore the samples from each of the areas/regions sampled (i.e., around Brisbane River mouth (northern region), Mangroves areas, Seagrass areas, Southern bay fed by the Logan River) were averaged to provide a more representative plastic concentration for each area (Table 2, Fig. 2). Overall, the northern samples around the Brisbane River mouth contained the highest total concentration of plastics ($\Sigma_7$plastics) with an average concentration of 524 μg/g, while the samples from the south of the bay, contained an average plastic concentration of 55 μg/g (Table 2, Fig. 2). This is likely due to the size of the catchments, with the Brisbane River that flows into the centre of the Moreton Bay draining a significantly larger catchment (~10,000 km^2) and flowing through more urban environments than the Logan river, which flows into the south of the bay (3860 km^2).

3.3. Concentrations of plastic polymer types in Moreton Bay

The concentrations of 7 different polymer types (i.e., PE, PVC, PP, PC, PET, PS and PMMA) were quantified in the samples (Table 1). PE was detected at the highest concentration and was detected at every site (2.3–1885.9 μg/g). PVC was detected at 43 sites (3.0–979.5 μg/g), PET at 39 sites (<1–11.9 μg/g), PC at 33 sites (<1–58.2 μg/g), PP at 23 sites (<1–141.7 μg/g), PS at 14 sites (<1–3.2 μg/g) and PMMA at 2 sites (<0.2–0.3 μg/g). The concentrations of each type of plastic polymer were highly variable with different concentrations present at each site, Table 1. The highest concentrations of PE were detected at site 54 A (1885.9 μg/g, in the northern region) with elevated concentrations also detected at site M4 (1228.3 μg/g, mangroves region) and at site S6 (1066.9 μg/g, seagrass region). Similarly, PVC concentrations were higher at sites S6, M4 and 54 A (979.5, 297.0 and 292.7 μg/g, respectively). The highest concentrations of PC and PET were observed at sites 129 (northeast of the bay) and M4 (mangrove region) (58.2 and 11.9 μg/g, respectively), with the highest concentrations of PP and PS detected at sites M4 (mangroves region) and 1 (northeast region) (141.7 and 3.2 μg/g, respectively).

PE represented more than half of the plastics concentration (52–80 %) for the North, Mangrove and Seagrass regions except for the southern region which had 39 % (Table 2), while PVC accounted for 17–40 % of the plastics concentration in each region. In the southern region of the bay PC was present and accounted for about 37 % of the plastic concentrations. Although the sediment samples from the other regions (i.e., north, mangrove and seagrass) recorded PC concentrations, the average PC mass concentration for the regions were far below 1 μg/g which accounted for about 0 % in each region. Similarly, the average PP concentration for the southern part of the bay was low (0.2 μg/g). However, PP was present in the other regions and accounted for about 1–7 % of the plastic concentrations. We should point out that although the percentage of PC, PP, PET, PS and PMMA in Table 2 is recorded as 0 % for some regions, it is essential to note that the reported percentage may not accurately represent the absolute absence of the polymer in the relevant area. The numerical concentration values, even if not explicitly provided in the table, is still non-zero, and the 0 % value is due to rounding or the fact that it is below the error range.

Although PP is one of the most versatile and extensively used polymers worldwide (Rogers, 2020); second only to PE in volume (PlasticsEurope, 2022; PlasticsEurope, 2019); it made up <10 % of the total plastics concentration measured in the sediment samples analysed, while there were also only trace amounts of PS and PMMA present in the samples (Table 1). PP may not be as common in the sediments due to its slightly lower density (<1 g/cm^3; Table S6), making it less dense than water and it tends to be buoyant, making it more likely to remain suspended in the water column rather than settling into the sediment. The lower density of PP allows it to float or be transported with water currents, potentially leading to lower concentrations in sediment. Similarly, while PS (1.05–1.06 g/cm^3, Table S6) and PMMA (1.16–1.20 g/cm^3, Table S6) are denser than water, they may remain suspended due to its small particle size, which can be influenced by factors like polymer morphology and the physical conditions of the water, hence, the low concentrations and detections of PS and PMMA in the sediments. PS and PMMA particles can adsorb onto sediment surfaces or aggregate with other particles, affecting their settling behaviour. For example, the particle size of PS and PMMA can influence its settling behaviour with smaller particles staying suspended for longer periods, contributing to lower concentrations in sediment. PS and PMMA particles can also adsorb onto sediment particles or organic matter in the water column, potentially reducing their availability for settling into sediment. Based on this we propose that PS and PMMA may be more prevalent in the water column. Also, biological processes, such as microbial degradation or interactions with organisms, may have influence the fate of PS and PMMA in the water column and sediment. PMMA polymers are used in optical lenses, acrylic nails, paint, laptops, smartphone display screens, interior and exterior panels, canopies, LCD screens, personal care products, and furniture among others (Ali et al., 2015) (Table S6). Hence, the mostly non-detection of PMMA in the analysed sediment samples possibly implies that PMMA leakage into the bay may not be a current issue.

Across all samples analysed in this study, PE contributed to 67.9 % of the concentrations of the total plastics quantified ($\Sigma_7$plastics) in the sediment samples, followed by PVC at 23.7 %, PC at 4.9 %, PP at 2.5 % and PET at 0.9 % (Table 1). The plastics concentrations profile follows the reported annual consumption of PE > PVC > PET > PP > PS for Australia in 2019–2020 (Kyle O'Farrell, 2021; O'Farrell, 2018; O'Farrell, 2019), suggesting a direct link between the consumption of plastics in Australia and the contribution in the coastal environment. While PE represented about 26 % of the total plastics consumed (Kyle O'Farrell, 2021; O'Farrell, 2019), it was 67.9 % of the plastic mass in the sediment samples analysed. It should be noted however, that the consumption data for PS included expanded PS and PS, PE included high-density, low-

Table 2
Mean plastic concentration and percentage of each plastic polymer type for each of the regions.

Area	PE		PVC		PP		PC		PET		PS		PMMA		Sum
	μg/g	%	μg/g	%	μg/g	%	μg/g	%	μg/g	%	μg/g	%	μg/g	%	
North	419.6	80 %	96.1	18 %	4.2	1 %	0.6	0 %	3.5	1 %	0.1	0 %	<MDL	0 %	524
South	21.3	39 %	10.6	19 %	0.2	0 %	20.1	37 %	2.2	4 %	0.3	1 %	<MDL	0 %	55
Mangrove	245.7	74 %	57.7	17 %	24.2	7 %	0.5	0 %	2.3	1 %	0.1	0 %	0.04	0 %	331
Seagrass	194.1	52 %	166.2	44 %	13.1	4 %	0.1	0 %	0.2	0 %	0.2	0 %	0.03	0 %	374

Polymers are listed in order of highest concentration/% to lowest: polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polycarbonate (PC), polyethylene terephthalate (PET), polystyrene (PS), and poly-(methyl methacrylate) (PMMA), method detection limit (<MDL).

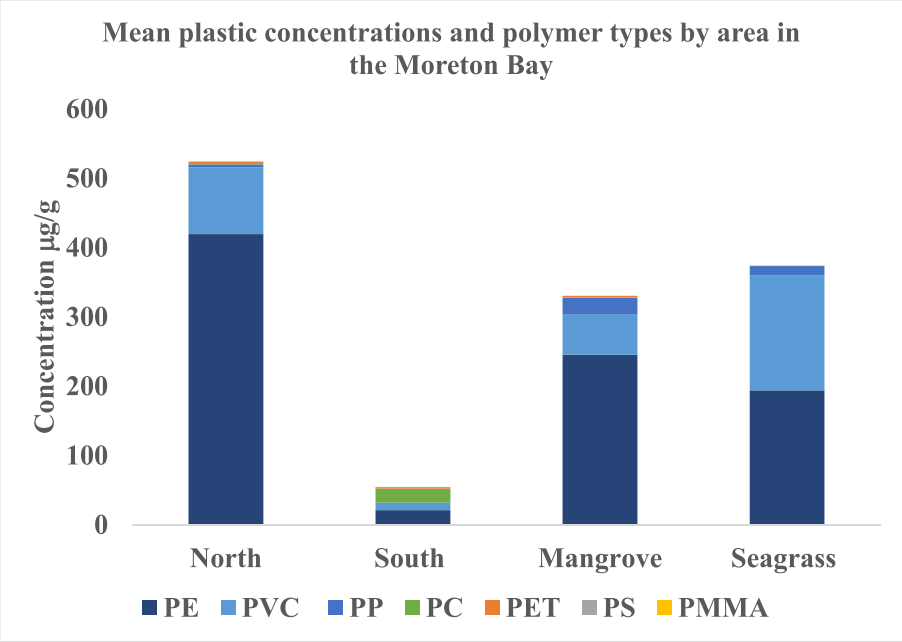

Fig. 2. Mean plastic concentration in samples from each of the regions listed in order of highest concentration. Polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polycarbonate (PC), polyethylene terephthalate (PET), polystyrene (PS), and poly-(methyl methacrylate) (PMMA).

density, and linear low-density PE, and there were no Australian consumption data for PMMA and PC. PE (both high and low density) is the largest volume polymer used globally (PlasticsEurope, 2022; PlasticsEurope, 2019) and is one of the cheapest plastic material types with applications in items including food and general packaging such as carrier bags, consumer goods, electronics, household goods, toys, plastic bottles, single-use plastic bags, films, and personal care products (Ziajahromi et al., 2017a; Ziajahromi et al., 2017b; Akarsu et al., 2019; Kazour et al., 2019) (Table S3). The wide use of this plastic, especially for single-use items is presumably leading to the increased leakage into waterways and Moreton Bay. PVC is used in wastewater treatment plant (WWTPs) pipes, containers, building materials and furniture, packaging, electronics, fibre for clothing etc., (Allsopp and Vianello, 2000; Insight, 2020) (Table S6), which may have increased its leakage into waterways reaching the bay. Interestingly, the Australian annual consumption data on plastics does not capture and provide data for PC usage in Australia, however, PC was widely detected at high concentrations (Table 1) in the sediment samples analysed from the bay. PC is commonly used for plastic lenses in eyewear, in medical devices, automotive components, protective gear, greenhouses, digital disks (CDs, DVDs, and Blu-ray), electronics/electrical applications, mobile phones and exterior lighting fixtures, among others (Table S6), with the detections possibly suggesting leakage into the bay. While the annual consumption data for plastics in Australia, as presented in the study, does not explicitly provide information on the usage of PC in the region, it is important to note that the absence of such data does not necessarily establish a direct relationship between PC consumption and recent plastic pollution issues in the bay. The lack of consumption data for PC makes it challenging to draw definitive conclusions about its contribution to plastic pollution in the studied area.

It is noted that there can be several different sources of these plastic types reaching Moreton Bay and its surrounding environment including through industrial discharges, WWTPs discharges, connecting waterways (rivers, lakes, creeks), stormwater, road runoff, fishing activities, recreational and commercial boating activities, among others. Hence, catchment population habits or socioeconomics, trade waste, and WWTP discharges could have contributed to the variability between the concentrations of plastics measured. Also, the degradation of commonly used consumer products (e.g., plastic bottles, packaging products, plastic containers, toys, electrical and electronic devices, built environment, decorating and construction materials, among others) reaching the bay into smaller plastic fragments due to physical and chemical processes or exposure to UV radiation may have contributed to the plastics pollution levels in the bay (Okoffo et al., 2019; Browne et al., 2011; Murphy et al., 2016; Carr et al., 2016). The highly variable concentrations across the bay may also be influenced by the dynamic ocean tidal currents and the flood waters flowing into the bay and out through the North and South passages.

The profile of the plastics quantified in the sediment samples is in agreement with the numerous studies that have reported plastic particles of different size ranges in sediment samples (Gomiero et al., 2019; Shi et al., 2022; Zhu et al., 2021; Birami et al., 2022; Kukkola et al., 2022; Liu et al., 2022) that included PE, PVC, PC, PP, PET, PMMA, PS, among others. However, it is difficult to directly compare the data reported in this study to these other studies due to the broad range of methods used for sampling, processing, and the particle size detection limit reported in the literature, with most studies reporting/detecting plastic particle sizes >1 μm in sediment samples, as compared to the concentrations of all particles <2 mm in this study. To the best of our knowledge, there has only been 2 other marine sediment studies that have used Pyr-GC/MS approaches that have reported mass concentrations of plastics in sediment samples (Gomiero et al., 2019; Zhang et al., 2021), where we can directly compare with our study. The first study analysed sediment samples from South Korea, Japan and USA for PET and PC and measured concentrations of up to130 and 14 μg/g, respectively (Zhang et al., 2021). These concentrations are in a similar range to the concentrations of these polymers reported for this study. Another study from Boknafjorden (Norway) (Gomiero et al., 2019) quantified the mass concentrations of the same 7 plastics in marine sediment samples as analysed in this study. Although the study used a slightly different sample pretreatment approach as compared to the PLE used in this study, they found that the most abundant polymer present in all investigated sites was PE, with values ranging from 32.3 to 139.2 μg/kg (0.0323–0.1392 μg/g) (Gomiero et al., 2019). This was followed by PVC and PET which ranged from 9.0 to 120.0 μg/kg (0.009–0.12 μg/g) and from 12.0 to 13.5 μg/kg (0.012–0.135 μg/g), respectively. PP ranged

from 10.0 to 78.4 μg/kg (0.01–0.0784 μg/g). PS and PMMA were only detected in 60 % and 40 % of the investigated sites, respectively, with PC not found in any of the analysed samples. The concentrations reported in the Norwegian study were several orders of magnitude lower than those found in the samples in our study. This is likely due to the fact that our study was conducted on coastal sediments in close proximity to a large urban population and associated plastic sources, although differences due to analytical techniques, sampling methods, sample pre-treatment cannot be ruled out.

3.4. Plastic budget for surface sediments of Moreton Bay

Based on the plastic concentrations of the different regions of the bay we developed a first estimate of the total plastic budget for the surface sediments of Moreton Bay, Australia (See Methods above; Table 3). Due to the large discrepancies in the plastic concentrations in the different regions of the bay we have used the average concentration of $\Sigma_7$plastics for each region. The average concentration of plastics in μg/g is converted to μg/cm^3 by multiplying this by the average dry bulk density of the sediment sample (DBD; measurements in Table S5) for each region. The volume of the surface sediment in the bay is calculated assuming a depth of 10 cm (the depth the grab can sample to), multiplied by the total area for the northern, southern and mangrove regions in the bay (USEPA, 2001) (Table 3). We have used the most recent assessment of the total area of mangroves (Lovelock et al., 2019) Table 3). The seagrass analyses is incorporated in the southern area measurements, as seagrass areas are highly dynamic (Kovacs et al., 2019), and seagrass is mostly found in the South and Eastern parts of the bay in the recent surveys (Maxwell et al., 2019). The plastic budget for the surface sediments of Moreton Bay is estimated to be a minimum of 7000 t (Table 3). For context, this would fill 3 Olympic size swimming pools.

We acknowledge that there are considerable limitations and assumptions with our method. However, it is necessary to convert the plastic from ug/g of sediment to a total mass to convey the scale of the plastic pollution within Moreton Bay. It is clear that the semi-enclosed embayment of Moreton Bay is effectively trapping a significant amount of plastic that is flowing into the bay from the adjacent urban catchments. However, large areas of the bay have not yet been analysed for plastics, especially in the northern part of the bay, which is the main depocenter for the mud (Lockington et al., 2017) and therefore the plastic concentration may be higher in this region. The plastic concentrations may also be greater after flood events.

It is important to quantify the plastic pollution in coastal embayment's and harbours adjacent to urban catchments around Australia and globally in order to assess the scale of the pollution and the main plastic polymers that are contributing to it. This will provide a baseline to assess future approaches to manage plastic waste and minimize the leakage into the marine environment.

4. Conclusion

The work presented here tests a new analytical method for marine sediments using the Pyr-GC/MS. Based on the analyses of 47 surface sediment samples from a range of environments within Moreton Bay, offshore the city of Brisbane, Australia, we provide the first estimate of mass-based concentration levels and polymer composition of plastics in the surface sediments of this semi-enclosed coastal embayment. We estimate a minimum plastic budget of 7000 t in Moreton Bay. Of the main polymer types analysed, PE and PVC were ubiquitous and present in the highest concentrations, although the polymer concentrations were highly variable between sites. This likely reflects the wide range of uses and sources of these plastics to marine ecosystems and the dynamic environment which may be transporting them via tidal currents and floods. This study establishes baseline data on the concentration of plastic, the main types of plastics, and the distribution of these plastics in the urban coastal environment of Moreton Bay.

Table 3
Estimated plastic budget in surface sediments of Moreton Bay.

	Sediment from South/ Seagrass area[a]	Sediment from North area	Sediment from Mangroves area	Total
Plastics concentration - μg/g ($\Sigma_7$plastics)	118[a]	524	330	
DBD of sediment - g/cm^3 (Table S5)	1.07	0.84	1.68	
Area in the bay - km^2	224	1299	184[b]	1707
Depth of sediment - cm	10	10	10	10
Volume of sediment - cm^3	2.24E+12	1.3E+13	1.84E+12	1.707E+13
TOTAL plastic budget - tonnes	283	5718	1020	7021

[a] Average concentration plastic and DBD of all the southern and seagrass samples. The total seagrass area is dynamic for the bay (Maxwell et al., 2019), but the seagrass is mostly found in the south and east of the bay and therefore the seagrass averages are included in the South area.
[b] Mangrove area based on 2019 survey (Lovelock et al., 2019).

CRediT authorship contribution statement

Elvis D. Okoffo: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Emmeline Tan:** Data curation, Formal analysis, Investigation, Visualization, Writing – review & editing, Validation. **Alistair Grinham:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Visualization, Writing – review & editing. **Sai Meghna Reddy Gaddam:** Formal analysis, Investigation, Visualization, Writing – review & editing. **Josie Yee Hang Yip:** Formal analysis, Investigation, Visualization, Writing – review & editing. **Alice J. Twomey:** Methodology, Resources, Writing – review & editing. **Kevin V. Thomas:** Methodology, Resources, Supervision, Writing – review & editing, Project administration. **Helen Bostock:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – review & editing.

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

Additional information on Pyr-GC/MS parameters, QAQC and results of plastic in blanks can be found in the Supporting Information. Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.170987.

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