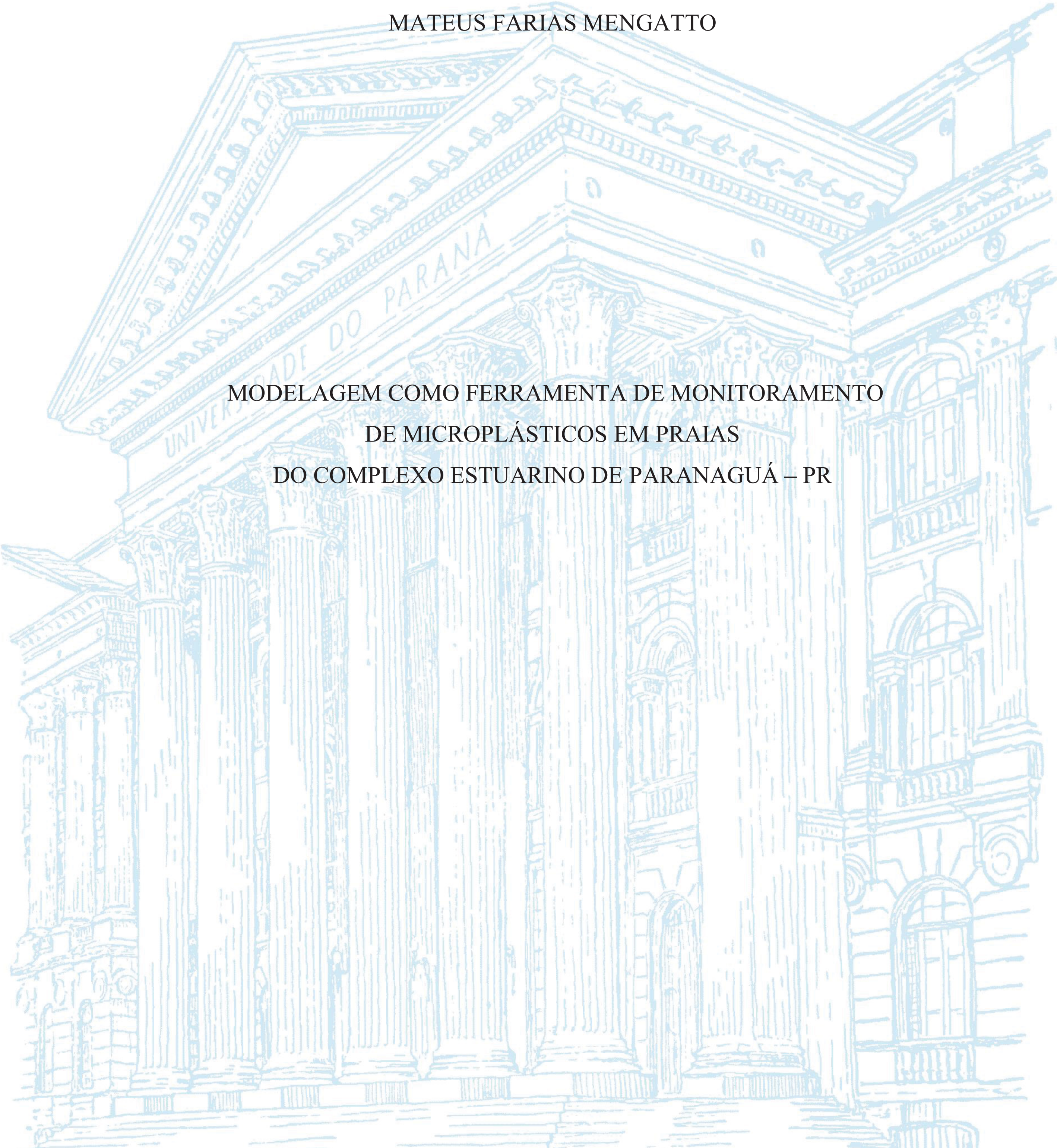

UNIVERSIDADE FEDERAL DO PARANÁ

MATEUS FARIAS MENGATTO

MODELAGEM COMO FERRAMENTA DE MONITORAMENTO DE MICROPLÁSTICOS EM PRAIAS DO COMPLEXO ESTUARINO DE PARANAGUÁ – PR

PONTAL DO PARANÁ
2021

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MODELAGEM COMO FERRAMENTA DE MONITORAMENTO DE MICROPLÁSTICOS EM PRAIAS DO COMPLEXO ESTUARINO DE PARANAGUÁ – PR

Dissertação apresentada ao curso de Pós-Graduação em Sistemas Costeiros e Oceânicos, Campus Pontal do Paraná – Centro de Estudos do Mar, Universidade Federal do Paraná, como requisito parcial à obtenção do título de Mestre em Sistemas Costeiros e Oceânicos.

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“If the sea is sick, we'll feel it. If it dies, we die.
Our future and the state of the oceans are one.”
Sylvia Earle

RESUMO

Os microplásticos (MPs) têm sido foco de preocupação ambiental, pois esses poluentes potencialmente tóxicos à saúde dos organismos aquáticos podem comprometer ecossistemas costeiros e marinhos. O Complexo Estuarino de Paranaguá (CEP), sítio RAMSAR de grande importância ecológica, compreende quatro unidades de conservação federais e abriga o segundo maior porto do Brasil e a maior cidade litorânea do Paraná (157 mil habitantes), potenciais fontes de MPs. Neste estudo, MPs na fração de tamanho de 1 a 5 mm foram investigados em 19 praias arenosas do CEP. As amostras de sedimento superficial (0-5 cm) foram coletadas na linha de deixa de marés de sizígia e, em laboratório, processadas por peneiramento e analisadas visualmente quanto ao numero e forma de MPs. Um total de 389 MPs foi encontrado em 16 dos locais de amostragem, destes 63,7% eram espumas, 13,8% fragmentos de plástico rígido, 12,8% fragmentos de tinta, 7,2% pellets de plástico, 1,8% filmes e 0,5% linhas. A maioria das praias da Área de Proteção Ambiental (APA) de Guaraqueçaba apresentou MPs. No CEP, as atividades urbanas e portuárias foram consideradas como as fontes mais prováveis de MPs. A obtenção de dados de campo são uma parte essencial no monitoramento da poluição por MP e de ações eficazes para a gestão e mitigação deste poluente. No entanto, grandes campanhas de amostragem podem ser caras, o que mostra a importância do uso de modelos de rastreamento de partículas, pois eles podem otimizar o monitoramento de MP sem grandes esforços de amostragem. Nesse contexto, também avaliamos um modelo de rastreamento de partículas - o modelo 2D TrackMPD acoplado ao modelo hidrodinâmico MOHID para investigar se essa abordagem poderia ser aplicada para auxiliar no monitoramento de MPs no CEP. Os resultados do 2D TrackMPD mostram que diferentes trajetórias e destinos das partículas são esperados para pontos de liberação de MPs distantes e destacam que os principais pontos de acúmulo de MPs, a partir dos pontos de liberação simulados, situam-se próximos as cidades de Paranaguá e Antonina. Encontramos uma correlação positiva e significativa entre a saída do modelo e os dados observacionais de MPs de praias não urbanizadas, mas correlações negativas e não significativas ao se considerar praias urbanizadas, comprometendo a precisão do modelo. Nossos resultados mostram que o modelo requer uma validação mais robusta para ser aplicado como uma ferramenta de monitoramento e apoio às ações de gestão de unidades de conservação. No entanto, as simulações destacam o movimento transfronteiriço dos MPs entre distintas áreas do CEP, principalmente os liberados na Baía de Paranaguá. A potencial exportação dos MPs para a APA de Guaraqueçaba e a plataforma oceânica adjacente aumenta a preocupação com a poluição dos MPs nesta área. Além disso, nosso trabalho traz informações sobre a dinâmica *source-to-sink* de MPs neste complexo estuarino, que é importante sob o ponto de vista da poluição por plástico local e também global.

Palavras-chave: Poluição marinha.Sedimento. Praia arenosa. Estuário. Modelo de rastreamento de partículas.

ABSTRACT

Microplastics (MPs) have been the focus of environmental concerns, as these toxic pollutants to aquatic organisms' health may compromise ecosystems. The Paranaguá Estuarine Complex (PEC), a RAMSAR site of great ecological importance, comprises four federal conservation units and harbors the second-largest port in Brazil and the largest coastal city of Paraná, both potential MPs sources. In this study, MPs (size fraction 1 to 5 mm) were investigated on 19 sandy beaches of the PEC. Surface sediment samples (0-5 cm) were collected at the high tide line and, in the laboratory, processed (sieving) and visually analyzed for its MPs content. A total of 389 MPs were found at 16 of the sampling sites. Of these 63.7% were foam, 13.8% hard plastic fragments, 12.8% paint fragments, 7.2% plastic pellets, 1.8% films, and 0.5% lines. The majority of the Guaraqueçaba Environmental Protection Area (EPA) beaches presented MPs. At the PEC, urban and harbor activities are considered the most likely MPs sources. Field data is an essential part of MP's pollution monitoring and sound management and mitigation actions. However, large sampling campaigns can be costly, which showcases the importance of MP particle-tracking models as these may optimize MP monitoring without large sampling efforts. In this context, we also assess a particle-tracking model - the 2D TrackMPD model framework coupled with MOHID Water OGCM accuracy to investigate if this approach could be applied to aid MPs monitoring at the PEC. The 2D TrackMPD outputs show that different particle trajectories and fates are expected for distant MPs release points and highlight that the Paranaguá and Antonina cities surrounding are the main MPs accumulation hotspots within the PEC. A positive and significant correlation was found between the model output and MPs observational data from non-urbanized beaches, but negative and insignificant correlations are observed when considering urbanized beaches, compromising the model accuracy. Our results show that the model requires a more robust validation to be usable as a monitoring tool and to support protected areas management actions. Nevertheless, the simulations highlight a transboundary movement of MPs between different areas on the PEC, mainly the releases on Paranaguá Bay. The export of MPs to the Guaraqueçaba EPA and the adjacent ocean shelf raises MPs pollution concerns for this area. Additionally, our work brings information regarding the source-to-sink dynamic of MPs in this estuarine complex, which has importance from a local and global perspective on plastic pollution.

Key-words: Marine pollution. Sediment. Sandy beach. Estuary. Particle-tracking model.

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LISTA DE ABREVIATURAS E SIGLAS

2D – *two dimensions;* duas dimensões
3D – *three dimensions*; três dimensões
APA – Área de Proteção Ambiental
CEP – Complexo Estuarino de Paranaguá
DNOS – *National Department Against Drought*
EPA – *Environmental Protection Area*
MP – *microplastic;* microplástico
NaCl – *sodium chloride*
nMDS – *non-metric multidimensional analysis*
OGCM – *Ocean general circulation model*
PDF – *probability density function*
PEC – *Paranaguá Estuarine Complex*
PERMANOVA - *Permutational multivariate analysis of variance*
Pv – *probability value*
WWTP - *wastewater treatment plant*

LISTA DE SÍMBOLOS

% – *percentage*
º – *degree*
cm – *centimeter*
cm^3 – *cubic centimeter*
g cm^{-3} – *grams per cubic centimeter*
Kg – *Kilograms*
Km^2 – *square Kilometers*
m – *meter*
m s^{-1} – *meter per second*
m^3 d^{-1} – *cubic meter per day*
m^3 s^{-1} – *cubic meter per second*
mm – *millimeter*; milímetro
MP m^{-2} - *microplastic per square meter*
MP m^{-3} – *microplastic per cubic meter*
MPs d.w. kg-1 – *microplastics per Kilogram dry weight*
N – *sample size*
p – *p-values*
R – *correlation coefficient*
µm – *micrometer*

SUMÁRIO

HIGHLIGHTS

- Microplásticos (1–5 mm) no Complexo Estuarino de Paranaguá foram avaliados.
- A distribuição dos microplásticos foi heterogênea e ampla ao longo do estuário.
- O modelo TrackMPD foi aplicado em um estuário subtropical brasileiro.
- Os resultados do modelo foram comparados com amostras de microplástico *in situ.*

RESUMO EM LINGUAGEM ACESSÍVEL

Microplásticos (MPs), partículas de plástico com tamanho de 1 µm a 5 mm, são poluentes prejudiciais à saúde de organismos marinhos cuja presença em ambientes costeiros e marinhos é preocupante. As principais fontes de MPs para o meio ambiente são atividades portuárias, industriais, de navegação, turismo, pesca, aquacultura e a descarga de efluentes domésticos. Nos ambientes costeiros, como praias e estuários, a poluição por MPs é particularmente problemática, pois são regiões que abrigam ecossistemas ricos em espécies com importância ambiental e econômica. O Complexo Estuarino de Paranaguá (CEP), localizado no litoral Sul do Brasil, abriga quatro áreas de conservação ambiental federais em uma de suas margens e o segundo maior porto do Brasil e a maior cidade do litoral do estado do Paraná na outra. O conhecimento da presença e distribuição de MPs em regiões estuarinas, como o CEP, é fundamental para traçar estratégias de mitigação e dar base para ações de manejo de suas áreas de preservação ambiental. Aqui, apresentamos o primeiro levantamento sobre a presença e distribuição de MPs em praias do CEP e testamos a aplicação de um modelo computacional de rastreamento de partículas para o monitoramento deste poluente. Para isso, coletamos amostras de areia de 19 praias do CEP e as analisamos quanto a seu conteúdo de MPs. Encontramos um total de 389 MPs nas praias do CEP, em sua maioria fragmentos de espuma, distribuídos espacialmente de forma variada. Em quase todas as amostras coletadas dentro do CEP foram encontrados MPs, inclusive dentro Área de Proteção Ambiental (APA) de Guaraqueçaba. Considerando as atividades humanas realizadas no CEP, identificamos as atividades portuárias e urbanas como as fontes mais prováveis dos MPs; porém, consideramos que as atividades realizadas por comunidades locais nas praias também podem contribuir com MPs para o CEP. Os resultados da simulação numérica realizada com o modelo 2D TrackMPD integrado ao modelo hidrodinâmico MOHID destacam que a liberação de MPs nas áreas mais urbanizadas da área estudada, onde ficam o porto e a cidade de Paranaguá, representa potenciais fontes desses poluentes para a APA de Guaraqueçaba e a plataforma oceânica adjacente. No entanto, a comparação entre os resultados da simulação numérica e dos dados observados no campo indicam que trabalhos futuros ainda são necessários para que o mesmo seja validado e aplicado como uma ferramenta no monitoramento da poluição por MPs no CEP.

1. INTRODUÇÃO

Microplásticos (MPs), por definição, são partículas sólidas sintéticas ou de matriz polimérica, com forma regular ou irregular, de dimensões entre 0.001 mm e 5 mm e, de origem primária ou secundária, e que são insolúveis em água (FRIAS; NASH, 2019). Essas partículas são potencialmente deletérias a saúde de organismos aquáticos, ameaçando a biodiversidade, e comprometendo funções ecossistêmicas (ANDRADY, 2017). Por isso, a poluição dos ambientes marinhos por essas partículas representa uma preocupação mundial e que é tratada como urgente (SAPEA, 2019).

Uma parte significativa dos MPs que chegam ao ambiente marinho é retida nos ambientes costeiros (p.e., praias e estuários) (ZHANG et al., 2017). O acúmulo de MPs em estuários é particularmente problemático, uma vez que esses ambientes são habitats essenciais para o desenvolvimento de espécies em todos os níveis tróficos, algumas com importância ecossistêmica e econômica (GRAY et al., 2018). Nesse sentido, recentemente houve um aumento no número de trabalhos que investigam os MPs em ambientes estuarinos, inclusive no Brasil (p.e., LIMA et al., 2015; DE CARVALHO; BAPTISTA NETO, 2016; CASTRO et al., 2016; VENDEL et al., 2017; FIGUEIREDO; VIANNA, 2018; ALVES; FIGUEIREDO, 2019; BAPTISTA NETO et al., 2019; OLIVATTO et al., 2019; GORMAN et al., 2020; OLIVEIRA NOVAES et al., 2020; LINS-SILVA et al.,2021; ZAMPROGNO et al., 2021), sendo um deles no Complexo Estuarino de Paranaguá (CEP; VIEIRA et al., 2021).

O CEP, é um estuário subtropical, que possui ampla importância ecológica. Este complexo, compreende áreas de manguezal e é cercado por um dos últimos remanescentes da Mata Atlântica (*Natural World Heritage Site* – UNESCO, 1999), protegidos por lei por meio da instauração de unidades de conservação, entre elas, quatro federais (Área de Proteção Ambiental de Guaraqueçaba, Estação Ecológica de Guaraqueçaba, Parque Nacional do Superagui, e Reserva Biológica Bom Jesus). Em contrapartida, o CEP também abriga o segundo maior porto brasileiro, o Porto de Paranaguá, e adjacente a ele, a maior cidade costeira do litoral do Paraná (município de Paranaguá, estimativa de 157 mil habitantes; IBGE, 2021). Principalmente em seu eixo Leste-Oeste, o CEP possui diversas fontes potenciais de MPs, como atividades portuárias e industriais, navegação, turismo, pesca, aquacultura, e poluição por efluentes domésticos. Todas estas fontes são amplamente aceitas pelos pesquisadores como responsáveis por quantidades significativas de plásticos para os ambientes costeios. Estudos recentes relatam a presença de partículas de MPs em hepatopâncreas de ostras no CEP (VIEIRA et al., 2021), e a presença de pellets plástico (GORMAN et al., 2019; MOREIRA et al., 2016) depositados em praias arenosas adjacentes à foz do estuário. Além disso, alguns trabalhos regionais evidenciam a presença de macroplásticos (>2,5 cm) em praias interiores e adjacentes ao CEP (KRELLING et al., 2017; KRELLING; TURRA, 2019), no conteúdo recuperado através de arrastos de fundo (POSSATTO et al., 2015) e no conteúdo estomacal de tartarugas marinhas (GUEBERT-BARTHOLO et al., 2011; NUNES et al., 2021). No entanto, até o momento, ainda há carência de dados sobre a distribuição espacial de MPs no interior do CEP.

Monitorar a presença e distribuição dos MPs nos ambientes costeiros e marinhos gera conhecimento importante para o entendimento das dinâmicas de transporte e acúmulo, o qual é essencial no âmbito de estratégia de mitigação e gestão desses poluentes (GESAMP, 2019). E estudos de monitoramento ambiental, em sua grande parte, ocorrem por meio de avaliações quantitativa das concentrações de MPs nos ambientes, realizada através de campanhas de amostragem nos mais variados compartimentos, como água, sedimentos e organismos. Contudo, campanhas de amostragem envolvem alto custo financeiro e demandam tempo (*ver* MILLER et al., 2017), principalmente para extensas áreas. Nesse contexto, as simulações numéricas de rastreamento de partículas representam uma ferramenta alternativa para gerar informaçõe a respeito das fontes, sumidouros e caminhos de MPs, melhorando nossa capacidade de mapear áreas de risco (HARDESTY et al., 2017). Além disso, uma vez que se estabeleça uma correlação positiva entre a distribuição simulada e a distribuição ambiental dos MPs, estes modelos preditivos têm o potencial de preencher lacunas de dados na ausência de observações (SOUSA et al., 2021), assim obtendo um potencial de otimizar o monitoramento desses poluentes no ambiente.

Dentro deste contexto, a conexão continente-oceano propiciada por ambientes estuarinos faz destes ambientes ideiais para melhorar nossa compreensão dos processos envolvidos na entrada de MPs no oceano. Ainda, a complexa hidrodinâmica e rede de interações ambientais, sociais e econômicas do CEP (PROCOPIAK et al., 2017; ESTADES, 2003) o tornam um ambiente ideal para a abordagem da modelagem numérica de rastreamento de partículas no mapeamento de MPs. A presença de MPs no CEP apresenta um risco iminente aos organismos que são essenciais à manutenção ecossistêmica e à subsistência de comunidades locais. O monitoramento dos MPs no CEP torna-se imprescindível para uma gestão ambiental eficaz, principalmente no âmbito das unidades de conservação. Nesse contexto, este trabalho tem como objetivos (i) identificar a presença e distribuição de MPs (1 a 5 mm) em praias arenosas do CEP, gerando uma primeira avaliação destes poluentes; e (ii) implementar um modelo de rastreamento de partículas, a fim de identificar áreas com maior potencial de acumulação de MPs no CEP e (iii) avaliar a acurácia do modelo selecionado comparando os resultados da simulação com dados observacionais, buscando uma ferramenta para otimizar o monitoramento de MPs na região.

O trabalho foi estruturado em dois capítulos principais apresentados em formato de artigo científico, seguindo a formatação das revistas pretendidas para submissão. Os capítulos são seguidos por um texto de integração e conclusão final do trabalho. O Capítulo I intitulado "*Primeira avaliação da abundância de microplásticos em sedimentos de praias arenosas do Complexo Estuarino de Paranaguá (sítio RAMSAR)*" apresenta a identificação e classificação de MPs (fração de tamanho de 1 a 5 mm) em sedimentos superficiais (0 – 5 cm) de 19 praias arenosas do CEP. O trabalho foi recentemente submetido à Revista *Marine Pollution Bulletin* (ISSN: 0025-326X; Qualis A1 na área de biodiversidade; *Impact Factor* - 4.049; *CiteScore* - 6.7) como um artigo de *Baseline*, formatado sem subdivisões. O Capítulo II intitulado "*Modelagem de rastreamento de partículas como uma ferramenta de monitoramento de microplásticos em um sistema estuarino subtropical*" apresenta a implementação de um modelo de rastreamento de partículas (TrackMPD), trazendo aspectos metodológicos e a avaliação da acurácia do modelo por meio da comparação entre os resultados do modelo e os dados observacionais apresentados no Capítulo I. O Capítulo II, será submetido à Revista *Environmental Pollution* (ISSN: 0269-7491; Qualis A1 na área de biodiversidade; *Impact Factor* - 6.792; *CiteScore* - 9.3) no formato de *Research Paper.*

2. CAPÍTULO I

A first assessment of microplastic abundance in sandy beach sediments of the Paranaguá Estuarine Complex (RAMSAR site)

Primeira avaliação da abundância de microplásticos em sedimentos de praias arenosas do Complexo Estuarino de Paranaguá (sítio RAMSAR)

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Abstract

Here we present the first assessment of microplastics (1–5 mm) abundance in drift line sediments from nineteen sandy beaches at the Paranaguá Estuarine Complex (PEC), a subtropical estuarine system from South Brazil. This estuarine system harbors the second largest grain port in Brazil and a RAMSAR site, the Guaraqueçaba Environmental Protection Area (EPA). Sediment samples were washed through a 5- and 1-mm mesh sieve and then visually inspected. We found a total of 398 microplastic particles, of which the majority (63.7%) were foams, 13.8% hard plastic fragments, 12.8% paint fragments, 7.2% pellets, 1.8% films, and 0.5% lines. The most probable microplastic sources for PEC beaches are urban and port activities. However, small communities and marine sources may also contribute to MP presence. Almost all beaches within the EPA were contaminated by microplastics, which represents a threat to marine biota and may hinder the conservation unit goal.

Keywords: Microplastic; Estuary; Spatial distribution; Conservation Unit; South Brazil.

Highlights

- The presence of microplastics (1 – 5 mm) in a subtropical estuary is evaluated.
- Small spatial scale variability is important for estuary microplastic distribution
- Foam and hard plastic fragments are the dominant morphotypes.
- The highest microplastic abundances were found in an Environmental Protected Area.

Microplastics (MPs) are 1 µm – 5 mm size synthetic solid particles or polymeric matrix, which are insoluble in water, with regular or irregular shapes derived from primary or secondary sources (Frias and Nash, 2019). Primary MPs are manufactured within these sizes, such as 5 mm plastic pellets and micro-beads in cosmetics, while secondary MPs originate from the fragmentation of larger plastic debris (GESAMP, 2015).

MPs pollution in marine environments is a worldwide concern that needs to be urgently addressed (SAPEA, 2019). Ubiquitous on all marine environments, estimations suggest that more than 6.4 k tons of microplastics are present only on Great Pacific Garbage Patch (Lebreton et al., 2018), with ninety-eight percent of primary microplastics have been from land-based origins from coastal, industrial and domestic activities (Boucher and Friot, 2017). In coastal zones, microplastics amount have often a positive and significant correlation with nearby population density (Van Cauwenberghe et al., 2015, Hitchcock and Mitrovic, 2019). Concern about MPs is around the toxicity of low molecular weight chemical species present in plastic, such as residual monomers, chemical additives (Andrady, 2017), and the absorption of persistent organic pollutants, which can cause deleterious effects on marine biota (Anbumani and Kakkar, 2018; Botterell et al., 2019).

The Paranaguá Estuarine Complex (PEC) is a subtropical estuary with vast mangrove forest belts surrounded by an Atlantic Forest Reserve (Natural World Heritage Site – UNESCO, 1999) protected by Brazilian federal law through the Guaraqueçaba Environmental Protection Area (Guaraqueçaba EPA), Guaraqueçaba Ecological Station, Superagui National Park, and Biological Reserve of Bom Jesus. The Guaraqueçaba EPA comprises a vast portion of the PEC water bodies, salt marshes, tidal flats, sandy beaches, and habitats for various marine fauna that support traditional fisheries communities' livelihood. However, in the estuary, there is also the second-largest grain port in Brazil (Paranaguá Port), and the most populated city at the Paraná state coast, Parunguá city, plus four others. Even comprising wastewater treatment plants (WWTPs), high concentrations of sewage indicators (chemical markers and fecal indicator bacteria) are found around Paranaguá (Cabral et al., 2018; Martins et al., 2010), representing a potential MPs source (Cole et al., 2011).

MPs presence has been reported on oysters retrieved within the PEC (Vieira et al.,2021) and at beaches close to the estuary mouth (Gorman et al., 2019; Moreira et al., 2016), and also macroplastics (>2.5 cm) are founded on inner beaches (Krelling et al., 2017; Krelling and Turra., 2019) and during bottom trawling in the estuary (Possato et al., 2015). This plastic contamination evidence on PEC gives important clues that MP would also be found in environmental compartments within the estuary. The PEC inner sandy beaches are compartments that have the potential to receive and accumulate MPs. In this context, this study aims to assess MPs (1 – 5 mm) on PEC sandy beaches, analyzing their spatial variability along the estuary and potential associated sources.

The PEC, located in the state of Paraná, South Brazil (25°30' S, 048°25' W) (Fig. 1), comprises 600 Km² subdivided into two main water bodies, the Paranaguá and Antonina Bay (330 Km²) and the Laranjeiras and Pinheiros Bay (200 Km²), east-west and north-south axis, respectively (Lana et al., 2001; Marone et al., 2005). It is classified as a partially mixed estuary, mainly controlled by tides (Noernberg et al., 2006), with a residence time of about 3.49 days (Marone et al., 1995). The beaches of the PEC, in general, have a narrow and steep beach face followed by a wide intertidal plain with a small slope gradient (Rosa and Borzone, 2008). Well sorted fine sands compose beach sediments near the estuarine mouth; however, towards the interior of the estuary, beach face grain size increases as the plains get muddier (Rosa and Borzone, 2008). The innermost estuarine beaches present a decrease in energy gradient due to reducing ocean waves' contribution (Rosa and Borzone, 2008).

Nineteen PEC sandy beaches were sampled in December 2020, comprising a broad spatial scale along the estuary (Fig. 1). Beaches were classified as urbanized (n=14) – nearby urban centers or communities (i.e., Itiberê River (S7) and Emboguaçu River (S9) are considered as urbanized even if there are no settlements at the beach but both beaches are in the surroundings of Paranaguá City) – and non-urbanized beaches – beaches which aren't occupied, without local anthropogenic influence (Fig. 1). Most beaches are only accessible by boat, which yields a long displacement time; therefore, sampling was performed over two subsequent spring tide cycles (Table S1 and S2). Beach length determined the number of sampling points, with three different sampling points on beaches with over 250 meters in length (n=6) and only one sampling point in those smaller than that (n=12). An exception to this was the Galemas Island (S19), with two sampling points, even though it has less than 250 m in length because it is separated into two sections by a large rock. Following Alvarez-Zeferino et al. (2020), samples were taken along the high tide line. 10-meter sections of beach define the sampling points, which were randomly picked for each beach. A 10-meter rope marked at every 1-meter was extended parallel to the high tide line to avoid sampling bias. At each sampling point, three randomized sediment sample replicates were collected with a stainless steel core (20 cm diameter and 5 cm depth), yielding approximately 1,570 cm³ and 3 Kg of dry sediments. Samples collected were placed in aluminum trays until processing in the lab.

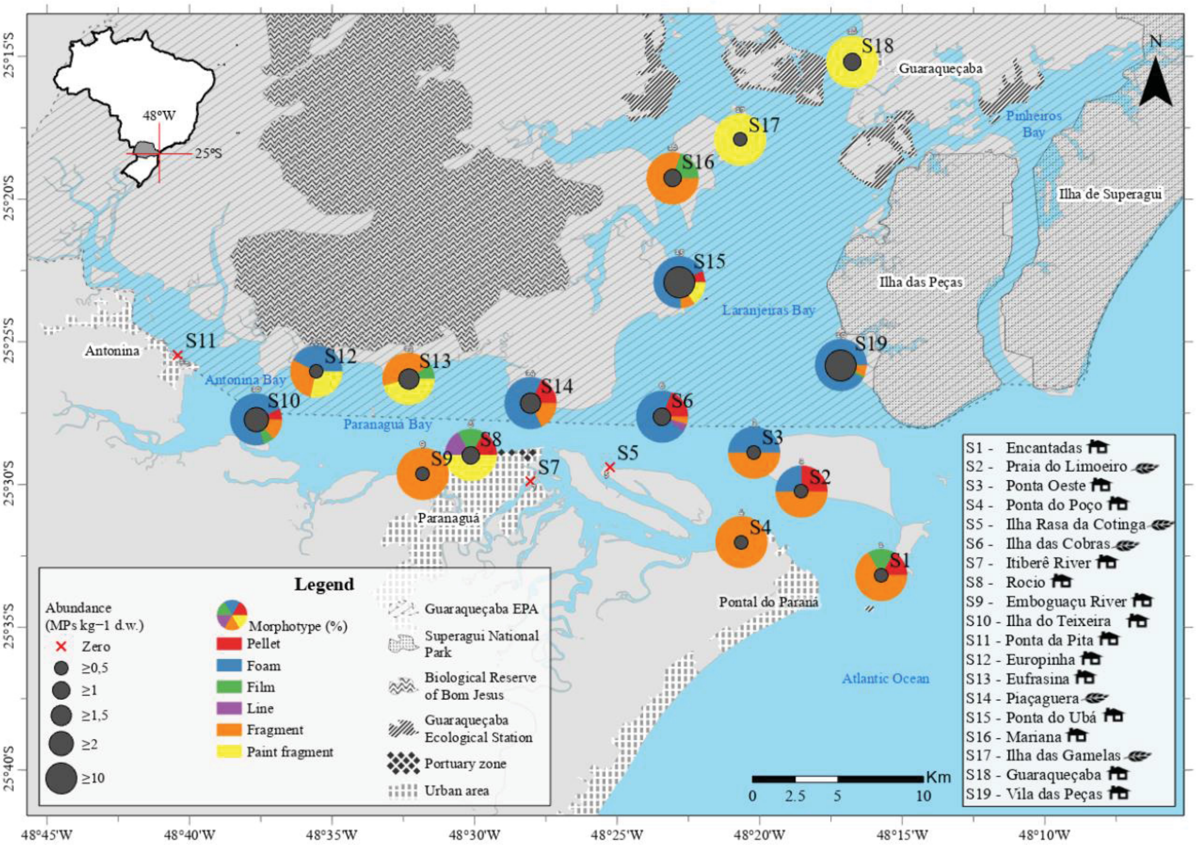

Fig. 1. Map of the Paranaguá Estuarine Complex and study sampling sites and popular names. Gray circle sizes represent MP abundance (MPs d.w. Kg^{-1}); pie charts, MP morphotype proportion; square texture polygons, urban areas; diamond texture polygons, the Paranaguá and Antonina port zones; Guaraqueçaba Environmental Protected Area - more spaced diagonal lines' polygon; Superagui National Park – straight lines' polygon; Guaraqueçaba Ecological Station – less spaced diagonal lines' polygon; Biological Reserve of Bom Jesus – zigzag lines' polygon; House and leaf figures after beach names denote their classification as urbanized or non-urbanized, respectively.

In the lab, each sample (replica number one) was oven-dried at 60 ºC for 24 hours and dried sieved in a laminar flow bench, and the <1 mm sample was stored in aluminum trays for further analysis. The other samples (two other replicas) were wet sieved with water previously filtered through a 250 µm sieve. Samples with large amounts of sediment in the 5 to 1 mm size fraction were also subjected to flotation for MP extraction. For this, a saturated sodium chloride solution (NaCl, ρ: ~1,2 g cm^{-3}) was used (Prata et al., 2019). The NaCl solution was added to the sample in a beaker glass, in a ratio of four to one volume, respectively, stirred for 2 min with an overhead mechanical stirrer, and allowed to settle for 3 minutes. The supernatant was then vacuum filtered with a Whatman® GF/C ~1 µm (47 mm) filter. During MP extraction, air contamination monitoring was carried out with an exposed Petri dish with a wet glass fiber filter.

All sieved content was placed on Petri dishes and visually inspected with a ZEISS SteREO Discovery V8 (80x) optical stereomicroscope, and MPs were separated. Following De Witte et al. (2014), a hot needle was used for MP confirmation (Fig. 2a). MPs were then classified as hard plastic fragments, foam, film, line, and pellet (Fig. 2), following the morphological descriptors of the Guidelines for the Monitoring and Assessment of Plastic Litter in the Ocean (GESAMP, 2019). In addition, we also classified MP as paint fragments separately from hard plastic fragments (Gaylarde et al., 2021). As paint fragments can be more brittle than hard plastic (Gaylarde et al., 2021), this characteristic was considered during the visual classification. To standardize the MP's 1 – 5mm interval size and avoid the data analysis overestimations, we measured MPs particles with the software ImageJ (Fiji package; Schindelin et al., 2012) considering their longest length size (Isobe et al., 2014). MPs were also classified in 1mm interval size classes (1 – 2mm, 2 – 3mm, 3 – 4mm, and 4 – 5 mm size) for further assessment comparisons. The colors were defined visually, considering the dominant color.

Microplastic data for each location is reported as the mean of three replicate analyses, expressed as the number of particles (items), and the abundance expressed as the number of particles per kilogram of dry beach sediment (MP d.w. Kg^{-1}). For comparisons with other studies, we also express MP abundance per sampled area (MP m^{-2}) and volume (MP m^{-3}) through the stainless steel core area and volume, respectively (Table S4). A Permutational multivariate analysis of variance (PERMANOVA), based on the six beaches with three sampling points (S1, S2, S6, S12, S14, and S15), was performed to analyze the sample variability between beaches ("Site" - fixed factor) and sampling points ("Point" - fixed factor nested to "Site"), through the MPs multivariate morphotype matrix. An ordination with non-metric multidimensional analysis (nMDS) was performed to support PERMANOVA results. A zero-adjusted Bray-Curtis coefficient was employed to treat the denude samples in our matrix, adding a 'dummy variable' (value = 1) to the original abundance matrix (Clarke et al., 2006). Both nMDS and PERMANOVA (the adonis2 function in the vegan library; R statistical software, 2021) analyses were performed using the Bray-Curtis distance method.

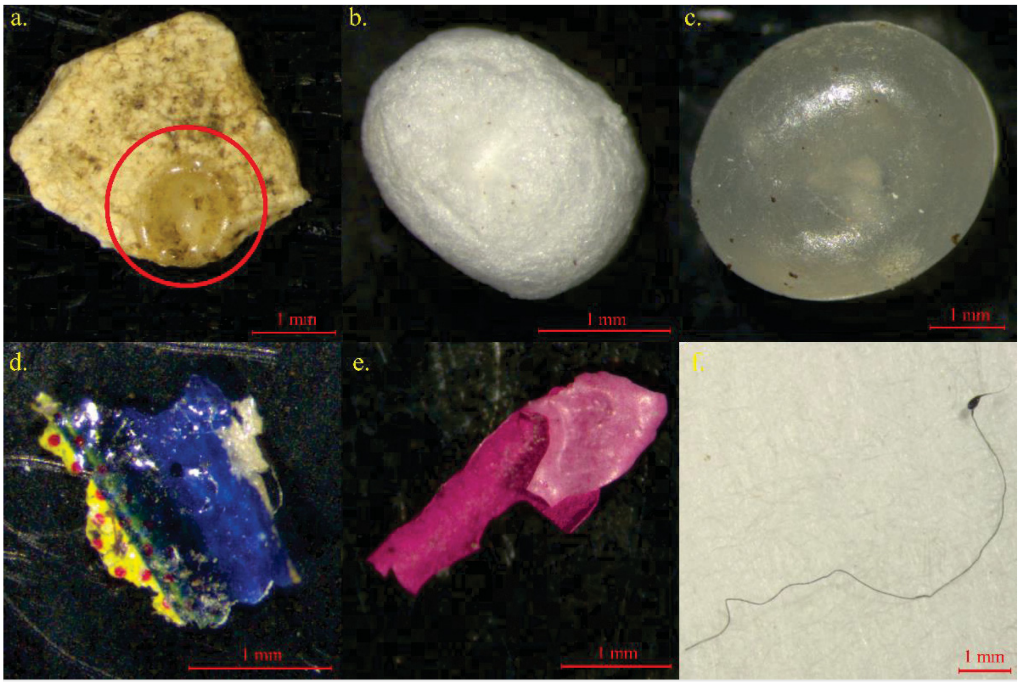

Fig. 2. MP morphologies (a) "yellowing" hard plastic fragment, (b) white foam, (c) transparent pellet, (d) blue film, (e) pink paint fragment, and (f) black fiber identified at the sandy beaches within the Paranaguá Estuarine Complex. The red circle in panel (a) shows deformation from the hot needle test.

We found MPs particles (total = 389 items) at most sampling sites, except at beaches Ilha Rasa da Cotinga (S5), Itibirê River (S7), and Pontal da Pita (S11). Ponta do Ubá (S15, = 91 MPs, 21.4 MPs replica^{-1}; Fig. 3) and Vila das Peças (S19, = 50 MPs, 23.6 MPs replica^{-1}) beaches presented the highest number of items per sample. Regarding MPs abundance per dry sediment weight, PEC sandy beaches comprised an average of 1.2 items d.w. Kg^{-1}, with the highest value found at Vila das Peças (7.8 items d.w. Kg^{-1}), although most beaches (n=7) had less than 1 item d.w. Kg^{-1} (Fig. 1). These quantities are relatively small compared with other Brazilian estuaries. In northern Brazil, at the Pedra Branca beach (Pará) fluvial-estuarine system, Oliveira Novaes et al., 2020 reported 20,166.7 particles m^{-3} (0.3 – 5 mm size fraction), against the 15,074.31 particles m^{-3} (extrapolated abundance) observed on Vila das Peças (S19) at the PEC. While in Southeastern

Brazil, at the Guanabara Bay (Rio de Janeiro), a maximum of 1300 particles m^{-2} (1 µm - 5 mm size fraction; de Carvalho and Baptista Neto, 2016), while PEC estimates a maximum of 188 particles m^{-2}, on Vila das Peças (extrapolated abundance). Worth mentioning here that these estuaries' adjacent areas are more urbanized than the PEC, in terms of demographic densities and urban areas. Nevertheless, the widespread presence of MPs within the PEC sandy beaches raises concerns about this pollutant and its impacts on the environment, especially at the environmental protected areas of the PEC

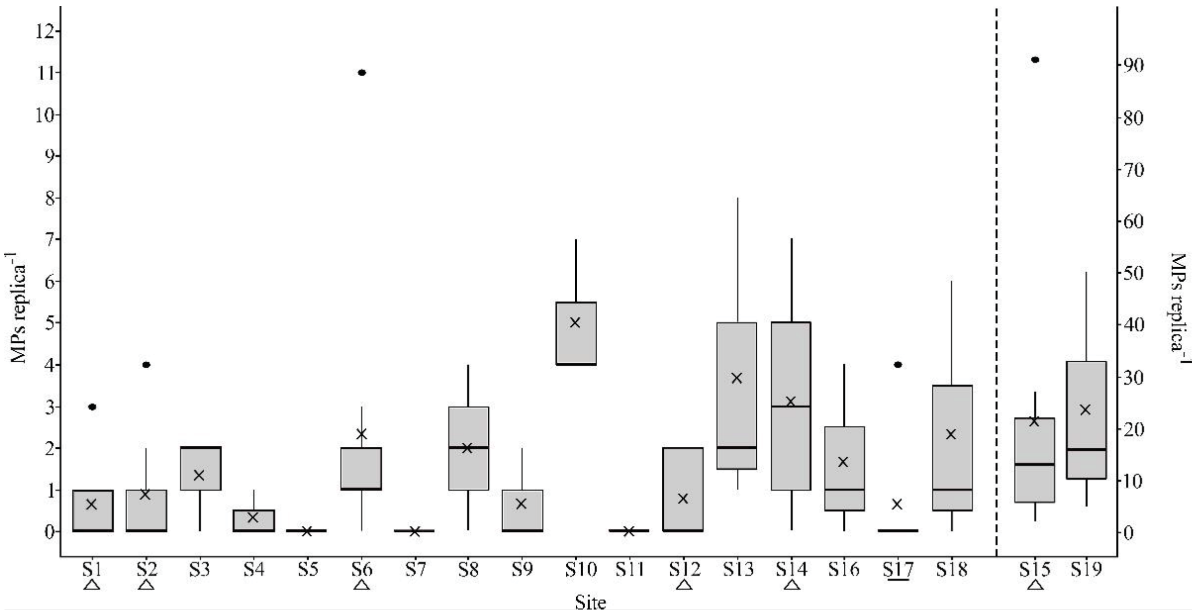

Fig. 3. Boxplot of the number of MP items per replica observed at PEC sandy beaches. Note that the dashed line separates S15 and S19 with a different scale y-axis. Crosses (X) represents mean replica values; black dots (•), outlier values (considering the interquartile range - IQR); black triangles (Δ), beaches with three sampling sites; and the black line (-) under beach S17, two sampling sites. For sampling location, please refer to Fig. 1.

MPs observed in this study are commonly reported in the literature at other sandy beaches in the world. Foam (63.7%) and hard plastic fragments (13.8%) dominated the samples, followed by paint fragments (12.8%), and pellets (7.2%), and soft plastics (film, 1.8%) and lines (0.5%) (Fig. 1). Hard plastic fragments were the most widespread MP morphotype observed at the PEC; these particles were present in thirteen beaches. In contrast, lines were the least observed MP morphotype at the PEC, in the 1 to 5 mm size fraction, only present at beach Ilha das Cobras (S6) and Rocio (S8). Secondary MPs, such as foam and hard plastic, may derive from a diverse range of larger plastic items fragmentation, while primary MPs, such as pellets, are derived from on-land and at sea commercial activities (Boucher and Friot, 2017). Even though MPs morphotype may help determine the possible sources of the contamination (GESAMP, 2015), it is virtually impossible to source-point secondary MPs. At the PEC, secondary MPs are probably related to household waste disposal from urban centers located adjacent to the estuary. Indeed, beached marine debris found within the PEC by previous studies was dominated by large plastics and large foam pieces (Possatto et al., 2015; Krelling et al., 2017; Krelling and Turra, 2019). According to Possatto et al. (2015), Krelling et al. (2017), and Krelling and Turra (2019), they were related to sewage input (domestic and ship-based).

The presence of pellets at our sampling sites, mainly at the Paranaguá Bay beaches, must be highlighted. Considering that pellets can enter the environment through maritime vessels and port activities during the transport or loadings (GESAMP, 2015; Turra et al., 2014), the Paranaguá and Antonina port activities are the most probable sources for the pellets observed in this study. Compared with Santos Estuary (São Paulo) (maximum – 377 pellets m^{-2}; Manzano, 2009), which comprises the largest port of Latin America, the pellets abundance in PEC beaches is small (maximum - 14 pellets m^{-2} in S15; Fig S4). This comparison may suggest a possible positive correlation between the abundance of pellets and the magnitude of port activity, however, this assumption needs further investigation. We cannot disregard marine sources for these particles once pellets have also been observed at the beaches near the PEC mouth (Moreira et al., 2016; Gorman et al., 2019). Nevertheless, their presence in the Guaraqueçaba EPA sandy beaches may represent a potential monitoring tool for port activity spatial influence range.

Additionally, paint fragments (12.8%) may bring important information about possible MPs sources in the PEC. Paint fragments usually are associated with boats and ships' coatings and superstructures such as piers and oil rigs and contain synthetic polymers like alkyds, epoxy resins, poly(acrylate/styrene), and polyurethane (Gaylarde et al., 2021). This type of MP was observed in 30% of the sampling sites and is probably associated with vessel navigation, piers, and moorings. However, at the Guaraqueçaba EPA, it was the only MP type observed at Ilha das Gamelas (S16) and Guaraqueçaba (S17), suggesting that these particles may also have a local origin, associated with small fishery boat coatings.

MPs observed in this study presented a mean size of 3.066 mm, with most particles' sizes ranging between 2- and 3-mm (35.7%) (Fig. 4). Foams influenced this mean once were 74.8% of the 2 to 3 mm size particles. Pellets size range between 2- and 5-mm size; however, most (64.2%) comprised between 4- and 5-mm size. Notwithstanding, the paint fragments particles sizes were mostly between 1 and 3 mm (74%). Hard plastic fragments showed almost equal size classes distribution; even with the highest abundance between 2 and 3 mm (29.6%), the other size classes reached around 20% to 25%. The MPs presented various colors, composed of white/translucent (23.9%), blue (7.97%), green (4.6%), red (3.8%), and black (2.8%) particles (Fig. 5). Other colors as yellow, grey, and pink were present but accounted for less than 1% of the observed colors. Nevertheless, most particles were discolored and presented a "yellowing" surface (55.3%) (Fig. 2a, Fig.5), most probably related to the plastic particle's advanced degradation processes (Auta et al., 2017). According to Cole et al. (2011), high oxygen availability and direct exposure to sunlight can increase the weathering process of beached marine debris, turning particles more brittle, forming cracks and ''yellowing''. As weathering processes change MPs' adsorption performance and behavior of pollutants (Sun et al.,2020), future studies should explore the relationship between MPs and other pollutants (i.e., organic compounds and heavy metals).

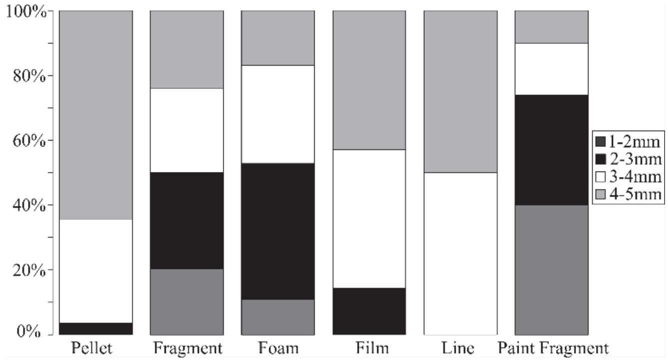

Fig. 4. Particle size classes [1 - 2mm (dark-gray), 2 – 3mm (black), 3 – 4mm (white), 4 – 5 mm (gray)] percentages for each MP morphotype.

The absence of MPs at some sites may be related to random sampling or beach depositional or particle transport dynamics, especially considering the Itiberê River and Ponta da Pita comprised within urbanized areas. Statistical analysis demonstrates that MP's spatial variability is critical in evaluating the heterogeneous distribution of this pollutant between our sampling sites. PERMANOVA indicated, significantly ($p<0.05$), that variability between beaches is higher than between sampling points (Table 1). However, the high R^2 from analysis residual indicated that replicas are responsible for explaining the most variability. High heterogeneity between the retrieved replicas ($n=3$) is an important factor, emphasizing the importance of considering small spatial scale variability (i.e., a few meters) to comprehend PEC`s MPs distribution better. The nMDS ordination supports PERMANOVA results, highlighting that MP morphologies also differ, mainly on replicas level, but also between sampling points (Fig. 6). At Ponta do Ubá (S15), one sampling point presented only paint fragments and films, while other sampling points did not present these morphologies but instead had foams, hard plastic fragments, and pellets.

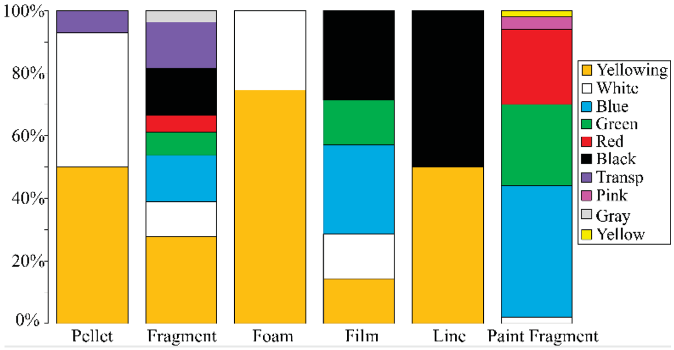

Fig. 5. Particle colors [Discolored "yellowing" (Orange), White (white), Blue (blue), Green (green), Red (red), Black (black), Transparent (purple), Pink (pink), Gray (gray), Yellow (yellow)] percentages for each MP morphotype.

Nevertheless, the fact that sampling campaigns occurred on different occasions must be considered when comparing MPs abundance between sampling sites, especially given that in-between campaigns, high precipitation occurred (Table S3). According to Krelling and Turra (2019), at the PEC, high precipitation and high riverine discharge can increase the input of marine debris. Though we observe an increase in MPs abundance at sampling sites from the second campaign, our sampling design does not allow us to correlate it to precipitation. Hence, the influence of precipitation over MPs abundance and distribution at the PEC needs to be further assessed by future studies. Another aspect is small-scale temporal variability over MPs abundance. At the PEC mouth, Moreira et al. (2016) reported that tides are a primary factor in pellet distribution variability, favoring the accumulation of particles with time, the same was seen for macroplastics (Bettim et al., 2021). The overlapping effect bias of these factors can be avoided by sampling all sites concomitantly. However, given access limitations and the distance between PEC beaches, this could only be possible to perform with multiple teams per campaign. Therefore, we recommend that future studies better constrain temporal and seasonal variability influence over MPs abundance and distribution at the PEC sandy beaches.

Table 1. Results of PERMANOVA comparing MP morphotype composition matrix from beaches (Site), and Sampling Point nested in Site; p-Value significance codes: '***' 0.001 '**' 0.01 '*'0.05.

	df	R^2	Pseudo-F	p-Value
Site	5	0.323	5.548	0.001 ***
Site: Point	12	0.255	1.824	0.006 **
Residual	36	0.42		
Total	53	1		

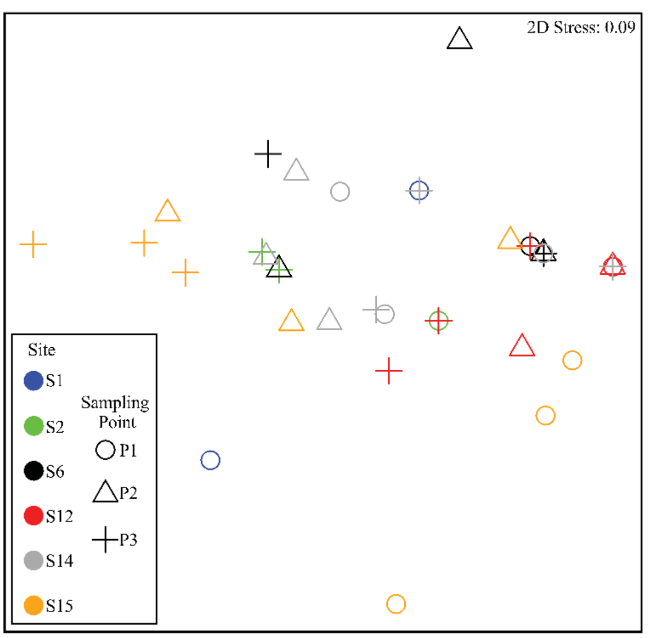

Fig. 6. nMDS grouping of MPs morphotype in samples from the six beaches [Encantadas (S1 - blue), Praia do Limoeiro (S2 - green), Ilha da Cobras (S6 - black), Europinha (S12 - red), Piaçaguera (S14 - gray), and Ponta do Ubá (S15 - orange)] with three sampling sites (P1 – circle; P2 – triangle, and P3 – cross).

In the vegetated flooded areas of the PEC, mangroves and salt marshes are common (Noernberg et al., 2006). These environments are susceptible to MPs accumulation (Lloret et al., 2021; Zamprogno et al., 2021). Mangrove vegetation structures can inhibit microplastic translocation, trapping the particles (Li et al., 2019). For macroplastics, Ivar do Sul (2014) reported that mangroves can retain the items for long periods (months-years). So, the investigation of MPs in these ecosystems could enhance our understanding of MPs distribution on our study area. Moreover, these vegetated areas can also provide information about MPs' historic contamination (Lloret et al., 2021). Additionally, mangroves and salt marshes also harbor a diversity of marine species and it is well reported that ingestion of MPs can cause adverse effects on marine fauna (Browne et al., 2008), such as reduced feeding activities, loss of energy, and decline in survival, with a result of the decrease in species abundance and richness (Pinheiro et al., 2020). Moreover, considering sandy beach marine fauna's crucial role in trophic energy flows (Costa et al., 2017), a negative impact on these organisms can lead to consequences for the whole ecosystem. Additionally, seafood consumption is also a potential route for human exposure to MPs (Van Cauwenberghe and Janssen, 2014). A recent study (Vieira et al., 2021) reported MPs in the hepatopancreas of oysters (*Crassostrea gasar*) retrieved from different sites within the PEC. These authors also report higher MPs concentrations in specimens retrieved in front of the Antonina and Paranaguá harbors. In this sense, the presence of MPs at the PEC beaches represents a threat for benthos fauna, and subsequently, for the local ecosystems. MPs may also impact socio-environmental aspects, once the traditional fisherman communities livelihood depends directly on the extraction of local natural resources.

In summary, this study is the first baseline of the presence, abundance, and morphotypes of the 1 to 5 mm size fraction microplastic particles on the Paranaguá Estuarine Complex sandy beaches. Our results highlight that even those sites located within the protected areas are subjected to MPs input. Even if MPs abundance values at the PEC are smaller than those found in other Brazilian estuaries, these pollutants threaten ecosystem health and hamper the fulfillment of the conservation units goal. The potential sources of these particles may be the Paranaguá and Antonina urban and port activities. Procopiak et al. (2017) also correlate these PEC south margin municipalities as sources of garbage and chemistry pollutants. However, to assert these assumptions it is necessary to make further assessments of the microplastic contribution of PEC urban centers. Besides, the adjacent ocean and local human activities, the last mainly for urbanized beaches, could also act as potential MPs sources for the PEC. Further assessments are required to understand MPs dynamics at this dynamic ecosystem, including studies that focus on the temporal and seasonal variability of MPs distribution, abundance, and morphotype on the beaches and other compartments such as mangrove, salt marshes, and water column.

Acknowledgments

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REFERENCES

Following the Federal University of Paraná normative, all cited references are at the end of this document.

2.1. Supplementary material

Table S1. Sampling locations and date for each Sampling Point and replicas; Beaches reference (Site), name (Description), sampling day (Day), the sampling time of day (Time), sampling point (Point), replica, and Location (Latitude and Longitude – decimal degrees); Field Observations for each Site.

Site	Description	Day (2020)	Time	Point	Replica	Latitude (°)	Longitude (°)	Field observations
S1	Encantadas	12/4	10:35	1	R1	-25.57175	-48.31573	Visually there is the presence of marine debris (large quantities); Houses and walls on the sand of the beach in the middle section (Sampling point 2); A community inhabits next to the beach; Intense tourism;
					R2	-25.57172	-48.31574	
					R3	-25.57168	-48.31569	
			11:10	2	R1	-25.56705	-48.31557	
					R2	-25.56705	-48.31557	
					R3	-25.567	-48.31558	
			11:40	3	R1	-25.56472	-48.31675	
					R2	-25.5647	-48.31679	
					R3	-25.56466	-48.31681	
S2	Praia do Limoeiro	12/16	08:35	1	R1	-25.52243	-48.35873	Visually there is the presence of marine debris; Very small waves;
					R2	-25.52246	-48.35864	
					R3	-25.52247	-48.35864	
			09:00	2	R1	-25.52319	-48.35743	
					R2	-25.52321	-48.35742	
					R3	-25.52321	-48.35739	
			09:30	3	R1	-25.52344	-48.35667	
					R2	-25.52344	-48.35664	
					R3	-25.52345	-48.3566	
S3	Ponta Oeste	12/16	10:05	1	R1	-25.50279	-48.38226	Visually there is the presence of marine debris, in less proportion compared to the others.
					R2	-25.50282	-48.38231	
					R3	-25.50282	-48.3823	
S4	Ponta do Poço	12/3	10:10	1	R1	-25.54973	-48.38898	Intense east/northeast wind during sampling; apparently, more organic matter in the sediment content;
					R2	-25.54974	-48.38898	
					R3	-25.54975	-48.38904	
S5	Ilha Rasa da Cotinga		11:13	1	R1	-25.5104	-48.45811	Salt marshes near the beach, mainly on the R3. A few pieces of marine debris (we collected to clean the beach, not analysis).
					R2	-25.51042	-48.45808	
					R3	-25.51044	-48.45804	
S6	Ilha das Cobras	12/16	10:45	1	R1	-25.48428	-48.4309	Visually there is presence of marine debris; Very small waves (smalles than S2)
					R2	-25.48425	-48.43088	
					R3	-25.48423	-48.43088	
			11:00	2	R1	-25.48395	-48.4307	
					R2	-25.48394	-48.43065	
					R3	-25.48393	-48.43066	
			11:10	3	R1	-25.4835	-48.43035	
					R2	-25.48346	-48.43032	
					R3	-25.48343	-48.4303	

Continuation – Table S1

Site	Description	Day (2020)	Time	Point	Replica	Latitude (°)	Longitude (°)	Field observations
S7	Itiberê River	12/3	13:26	1	R1	-25.5178	-48.49986	Visually there is the presence of marine debris; MPs in waterline; Marinas on the front of the beach;
					R2	-25.5178	-48.49986	
					R3	-25.51782	-48.49989	
S8	Rocio		11:57	1	R1	-25.50405	-48.53112	Visually there is the presence of marine debris (large quantities), plastic bags on the trees; Access to Paranaguá City;
					R2	-25.50404	-48.53114	
					R3	-25.50403	-48.53118	
S9	Emboguaçu River		12:39	1	R1	-25.51394	-48.55644	Visually there is the presence of marine debris (large quantities); Our mariner relates people throwing trash bags directly on the river;
					R2	-25.51395	-48.55641	
					R3	-25.51396	-48.5564	
S10	Ilha do Teixeira	12/14	15:18	1	R1	-25.48539	-48.64393	Wall limits the beach upper; A little community inhabits the beach;
					R2	-25.48537	-48.64395	
					R3	-25.48534	-48.64397	
S11	Ponta da Pita	12/14	14:15	1	R1	-25.45171	-48.6851	Smalls walls in some beach sections; Planted grass limits the upper beach;
					R2	-25.45169	-48.68512	
					R3	-25.45174	-48.68516	
S12	Europinha	12/14	12:52	1	R1	-25.45978	-48.61148	Wall limits the upper beach on sampling point 1; Visually there is the presence of marine debris; A community inhabits the beach;
					R2	-25.45979	-48.61151	
					R3	-25.45981	-48.61154	
			13:15	2	R1	-25.46018	-48.61231	
					R2	-25.46021	-48.61233	
					R3	-25.46023	-48.61235	
			13:30	3	R1	-25.46041	-48.61267	
					R2	-25.46041	-48.61267	
					R3	-25.46042	-48.61268	
S13	Eufrasina	12/14	12:08	1	R1	-25.47735	-48.4978	Wall limits the beach upper; Large quantity of biogenic material, like oyster shells; A community inhabits the beach.
					R2	-25.46404	-48.56388	
					R3	-25.46405	-48.56388	
S14	Piaçaguera		10:50	1	R1	-25.47662	-48.50106	Trees in beach upper limit; Visually there is the presence of marine debris;
					R2	-25.47659	-48.50106	
					R3	-25.47663	-48.50099	
			11:09	2	R1	-25.47681	-48.49981	
					R2	-25.47681	-48.4998	
					R3	-25.47681	-48.49979	
			11:15	3	R1	-25.47732	-48.49784	
					R2	-25.47733	-48.49784	
					R3	-25.47735	-48.49779	

Continuation – Table S1

Site	Description	Day (2020)	Time	Point	Replica	Latitude (°)	Longitude (°)	Field observations
S15	Ponta do Uba	12/15	08:03	1	R1	-25.41587	-48.42208	Visually there is the presence of marine debris; Beach Left limit with plastic bags on the trees; A community inhabits the beach; Houses on the sand of the beach.
					R2	-25.41585	-48.42208	
					R3	-25.41584	-48.42208	
			08:24	2	R1	-25.41343	-48.42152	
					R2	-25.41341	-48.42153	
					R3	-25.4134	-48.42151	
			08:40	3	R1	-25.41167	-48.42073	
					R2	-25.41165	-48.42072	
					R3	-25.41161	-48.42068	
S16	Mariana	12/15	09:20	1	R1	-25.35882	-48.42518	Visually there is the presence of marine debris; A community inhabits the beach;
					R2	-25.35884	-48.42517	
					R3	-25.35889	-48.42515	
S17	Ilha das Gamelas	12/15	10:00	1	R1	-25.33726	-48.38902	Visually there is the presence of marine debris, in less proportion compared to the others.
					R2	-25.33728	-48.38902	
					R3	-25.33728	-48.38903	
			10:20	2	R1	-25.3386	-48.38968	
					R2	-25.33858	-48.38968	
					R3	-25.33852	-48.3897	
S18	Guaraqueçaba	12/15	11:00	1	R1	-25.2978	-48.33071	The smallest beach sampled; Wall limits the beach upper and left; City boat pier on the side of the beach; Runoff from the city goes to the beach;
					R2	-25.29777	-48.33075	
					R3	-25.29777	-48.33076	
S19	Vila das Peças	12/15	12:00	1	R1	-25.45686	-48.3367	Visually there is the presence of marine debris; A community inhabits next to the beach;
					R2	-25.45684	-48.33669	
					R3	-25.45683	-48.33669	

Table S2. Elevation levels from Time of the Day; data from tide table of the Directorate of Hydrography and Marine Navigation (DHN) of Brazil (2020).

Day	Time	Level (m)
Dec 3	05:06	1.4
	09:58	0.4
	16:34	1.2
	21:58	0.2
Dec 4	05:49	1.3
	10:26	0.5
	17:13	1.1
	22:36	0.3
Dec 14	02:23	1.5
	07:11	0.6
	07:58	0.6
	10:51	0.5
	15:09	1.3
	21:13	0.2
Dec 15	03:08	1.6
	07:26	0.6
	09:21	0.7
	11:51	0.6
	15:58	1.3
	21:32	0.2
Dec 16	03:56	1.5
	08:02	0.6
	10:47	0.8
	12:54	0.7
	16:47	1.2
	22:13	0.2

Table S3. Rainfall accumulation in mm per period (2020 date) from each river; data from HIDROINFOPARANÁ (Instituto Água e Terra of Paraná State).

River (Station)	Nov 26 to Dec 03	Dec 04 to Dec10	Dec 11 to Dec 17
Nhundiaquara (Morretes)	36.6	151	61
Cachoeira (Vila Nova)	32	132.4	130
Cachoeira (Pinguela)	72.8	88	95.8
Guaraqueçaba (Colônia Rio Verde)	19	71.2	117.6
PEC Mean	160.4	442.6	404.4

Table S4. Beaches references (Site), name (Description), the number of Sampling Points (S. Points), and the sum of MPs particles found for each morphotype – Pellet; Hard Plastic Fragment; Foam; Film; Line; and Paint Fragment; MP abundances per Kg (MP Kg-1); Extrapolated data abundances – '*' symbol – to comparison with other studies – per m^2 (MP m-2) and m^3 (MP m-3).

Site	Description	S. Points	Pellet	Hard Plastic Fragment	Foam	Film	Line	Paint Fragment	MP replica^{-1}	MP Kg^{-1}	MP m^{-2} *	MP m^{-3} *
S1	Encantadas	3	1	4	0	1	0	0	0.7	0.2	5.3	424.6
S2	Praia do Limoeiro	3	2	4	2	0	0	0	0.9	0.3	7.1	566.2
S3	Ponta Oeste	1	0	2	2	0	0	0	1.3	0.4	10.6	849.3
S4	Ponta do Poço	1	0	1	0	0	0	0	0.3	0.1	2.7	212.3
S5	Ilha Rasa da Cotinga	1	0	0	0	0	0	0	0	0	0	0
S6	Ilha das Cobras	1	4	1	15	0	1	0	2.3	0.8	18.6	1486.2
S7	Itiberê River	1	0	0	0	0	0	0	0	0	0	0
S8	Rocio	1	1	0	0	1	1	3	2	0.7	15.9	1273.9
S9	Emboguaçu River	1	0	2	0	0	0	0	0.7	0.2	5.3	424.6
S10	Ilha do Teixeira	1	1	2	11	1	0	0	5	1.7	39.8	3184.7
S11	Ponta da Pita	1	0	0	0	0	0	0	0	0	0	0
S12	Europinha	3	0	2	3	0	0	2	0.8	0.3	6.2	495.4
S13	Eufrasina	1	0	5	0	1	0	5	3.7	1.2	29.2	2335.5
S14	Piaçaguera	3	5	5	18	0	0	0	3.1	1	24.8	1981.6
S15	Ponta do Ubá	3	14	17	132	1	0	29	21.4	7.1	170.7	13659
S16	Mariana	1	0	4	0	1	0	0	1.7	0.6	13.3	1061.6
S17	Ilha das Gamelas	2	0	0	0	0	0	4	0.7	0.2	5.3	424.6
S18	Guaraqueçaba	1	0	0	0	0	0	7	2.3	0.8	18.6	1486.2
S19	Vila das Peças	1	0	5	65	1	0	0	23.7	7.9	188.4	15074

3. CAPÍTULO II

Particle-tracking model as a tool for microplastic pollution monitoring in a subtropical estuarine system

Modelagem de rastreamento de partículas como uma ferramenta de monitoramento de microplásticos em um sistema estuarino subtropical

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Abstract

Microplastic (MP) pollution has been the focus of marine environmental concerns. Monitoring MPs is essential to efficient mitigation and management of this pollutant. Modeling trajectories of MPs may be a great tool to understand the behavior of this pollutant in marine environments. The Paranaguá Estuarine Complex (PEC) is a RAMSAR site and anthropized estuary, which comprises various potential sources of MPs. This study applied the 2D TrackMPD model framework coupled with MOHID Water OGCM to simulate MPs trajectories trends and accumulation hotspots given by probability density function (PDF) at the PEC. We assess the model's accuracy through a data-model comparison approach using the correlation between model probabilities from PDFs output and observational data of MPs in sandy beaches. The 2D TrackMPD outputs show that MPs from distant release points have different particle trajectories and fates, and the highest PDF probabilities highlight that the main MPs accumulation hotspots within the PEC are located next to the Paranaguá and Antonina cities. The adjacent ocean represents an insignificant MPs source to the PEC. The data-model comparison yields a positive and significative correlation with non-urbanized beaches; however, no significant and negative correlation considering urbanized beaches, compromising model accuracy. We attribute these disparities to either model parameters or field data representativeness. Although the model may require more robust validation to support protected areas management actions, its output highlights the transboundary movement of MPs between different areas of the estuary and the adjacent ocean shelf, and also the role of the Paranaguá sources to export MPs to the Guaraqueçaba Environmental Protection Area, raising plastic pollution concerns for this area and on the global perspective.

Keywords: Microplastic; Particle trajectory; Paranaguá Estuarine Complex; RAMSAR site.

Highlights

- The TrackMPD framework was implemented for a subtropical estuary.
- A microplastic particle-tracking accuracy was assessed through field samples.
- Primary fates and trajectories differences were among distant release points.
- Paranaguá and Antonina cities' nearby areas are microplastic accumulation hotspots.

3.1. Introduction

Plastic pollution has gained scientific and societal attention over the last decades, given the high rate of plastic production and inadequate waste management on land (Jambeck et al., 2015; GESAMP, 2019). The 1 µm to 5 mm size plastic particles, nominative microplastics (MPs) (Frias and Nash, 2019), presence in the marine environment needs to be urgently addressed (SAPEA, 2019). The concerns about MPs pollution in aquatic systems lie around their ubiquitousness and their potential effects on biota (Wang et al., 2019; Hale et al., 2020). In general, 80% of plastic debris on the ocean has land-based sources (Andrady, 2011). A significant part of land-based sources supplied to the marine environment is retained on the coastal areas sediments on beaches, wetlands, and estuaries (Zhang, 2017).

Estuarine areas can receive high inputs of MPs through multiple sources as inland river discharges, urban runoffs and sewage, port and industrial activities, fisheries, tourism, agriculture, and aquaculture (GESAMP, 2016; Andrady, 2017). MPs accumulation in estuaries is particularly problematic, once these environments are essential habitats for species development in all trophic levels, some with ecosystemic and economic importance (Gray et al., 2018). These transition coastal environments have been a target of recent MPs research (e.g., Alves and Figueiredo, 2019; Baptista Neto et al., 2019; Forero-López et al., 2021; Gray et al., 2018; Hitchcock and Mitrovic, 2019; Sruthy and Ramasamy, 2017; Zheng et al., 2019;

Zuo et al., 2020). These studies' research approach on quantitatively assessing MP’s pollution in different environmental compartments (i.e., water, beaches, bottom sediments, and organisms) by generating observational data.

The quantitative assessment constitutes essential information to understand key factors that influence MP’s accumulation on environmental compartments and represents a necessary step to planning mitigation and management actions regarding this pollutant (GESAMP, 2019). However, sampling large areas with numerous samples can be time and money costly (see Miller et al., 2017). Another methodology with the potential to bring important information about MPs’ sources, sinks, and pathways is the particle-tracking model approach (Hardesty et al., 2017).

Particle-tracking models have been developed and applied for MPs in different regions (Alosairi et al., 2020; Atwood et al., 2019; Ballent et al. 2013; Daily and Hoffman, 2020; Genc et al., 2020; Gorman et al., 2020; Isobe et al., 2014; Iwasaki et al., 2017; Jalón-Rojas et al., 2019b; Sousa et al., 2021; Zhang et al., 2020). The model frameworks of MPs particle-tracking generally use Lagrangean functions coupled with an ocean general circulation model (OGCM). OGCMs are widely used to improve our understanding of coastal and oceanic processes (e.g., water renewal time in semienclosed environments - Braunschweig et al., 2003; sedimentary dynamics - Franz et al., 2017; and pollutants distribution - Pierini et al., 2012). The complexity of this kind of modeling increased over the last decade, mainly adding different processes of particles behavior (Khatmullina and Chubarenko, 2019).

Although sophisticated models seem promising, accurate experimental or field data validations are required (Khatmullina and Chubarenko, 2019). Model validation can increase the model utility, confidence in results, and understanding of model output uncertainty (Hardesty et al., 2017). Few studies tested the accuracy of the numerical particle-tracking of MPs. Accuracy tests generally compare the model results with field data of MPs abundance in water (Atwood et al., 2019; Daily and Hoffman, 2020; Iwasaki et al., 2017), beach sediments (Atwood et al., 2019; Gorman et al., 2019), and in sessile organisms (Sousa et al., 2021). High accuracy on the correlation between particle tracking models prediction and field data can fill data gaps in the absence of observations (Sousa et al., 2021). Therefore, besides the potential to understand MPs dynamics, MP’s particle-tracking models allow their use to predict MPs distribution without large sampling campaigns, reducing the cost of assessing MPs pollution.

The Paranaguá Estuarine Complex (PEC; southern Brazilian southeast coast) is a promising estuarine environment for particle-tracking simulations, once has a complex hydrodynamic and encompasses a scenario that contrasts preserved areas with urbanized.

However, the Delft 3D particle-tracking model was already implemented for marine debris on PEC area (Krelling et al., 2017) without MP's approach and a valid N-S PEC axis hydrodynamic. MP's presence in PEC is reported on estuarine sandy beaches (Mengatto and Nagai, *submitted*) and oysters hepatopancreas (Vieira et al., 2021), and also on beaches located adjacent to the estuary mouth (pellets; Gorman et al., 2019; Moreira et al., 2016). Additionally, macroplastics (>2.5 cm) have also been studied at the PEC bottom (Possatto et al., 2015), beaches (Krelling et al., 2017; Krelling and Turra, 2019), and sea turtles (Guebert-bartholo et al., 2011; Nunes et al., 2021).

Hence, in this study, we applied the TrackMPD particle-tracking for the PEC coupled with a valid hydrodynamic model to the entire PEC (including the N-S axis; Franz et al., 2021), aiming to identify MP particles' trajectories and fate, and testing the model's accuracy to predict MPs accumulation hotspots in this estuarine complex.

3.2. Study area

3.2.1 PEC physical settings

The PEC is a subtropical estuary located in the southern Brazilian state of Paraná (25.5º S, 48.4º W) (Fig. 1). This estuarine complex surface area comprises 600 Km², divided at the Paranaguá and Antonina Bay (330 Km²) and the Laranjeiras and Pinheiros Bay (200 Km²), east-west and north-south axis, respectively (Lana et al., 2001; Marone et al., 2005). It is classified as a partially mixed estuary, mainly controlled by tides (Noernberg et al., 2006), with a residence time of about 3.49 days (Marone et al., 1995). The tidal regime is mainly semidiurnal, and the range average is 2.2 m (Marone et al., 1995). The maximum observed current velocity approaching 0.85 m s^{-1} in the ebb and 1.10 m s^{-1} in flood (Marone et al., 2005). River runoff varies seasonally, from approximately 7 x 10^6 m^3 d^{-1} during the winter to 28 x 10^6 m^3 d^{-1} in the summer in the east-west axis, with a general annual mean river freshwater input up to 200 m^3 s^{-1} (Marone et al., 2005).

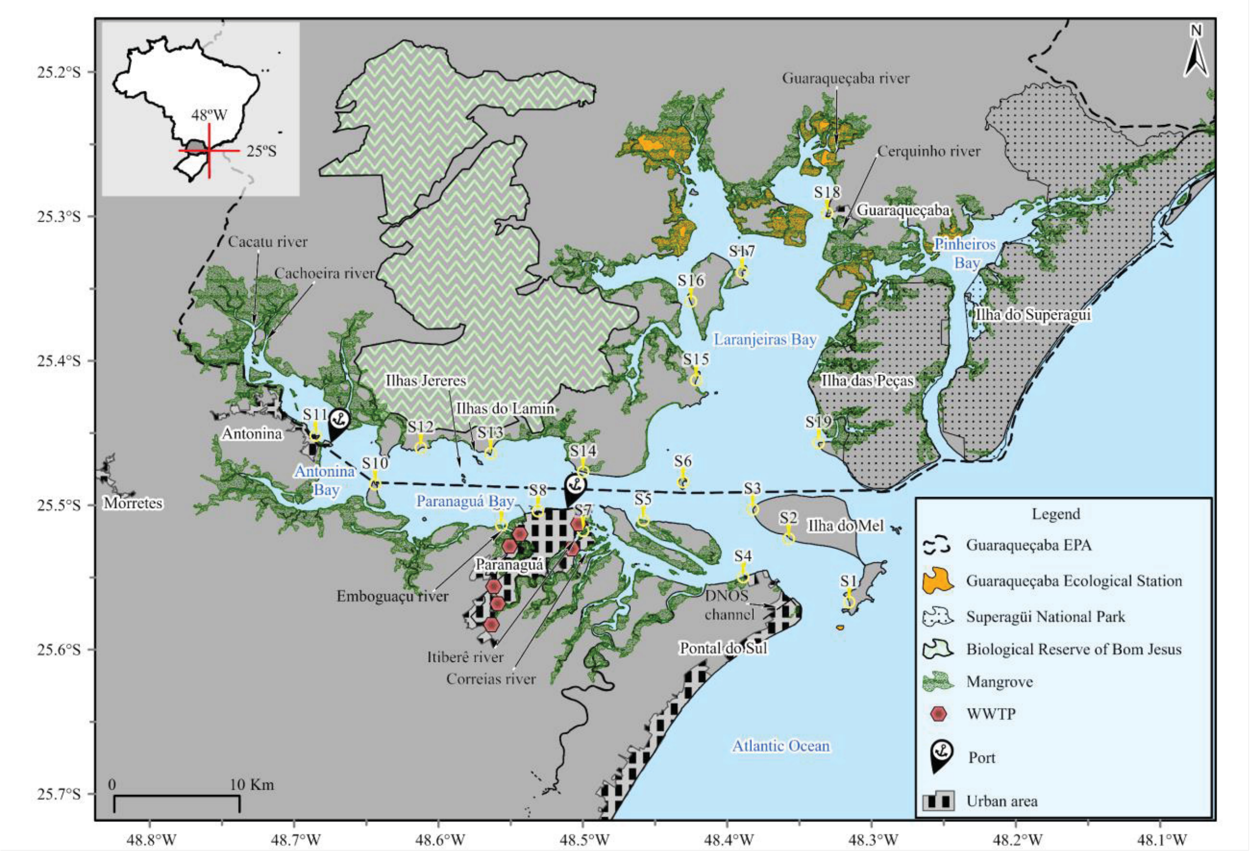

Fig. 1. The Paranaguá Estuarine Complex map and Mengatto and Nagai (*submitted*) sampling sites (yellow circles). The protected areas limits of Guaraqueçaba Environmental Protection Area (dashed outline); Guaraqueçaba Ecological Station (orange polygon); Superagui National Park (dotted texture polygons); and Biological Reserve of Bom Jesus (zigzag texture). The anchor symbols represent port locations; black rectangles texture polygons represent cities urban areas; and red hexagons represent the WWTPs. Green textures represent natural mangrove vegetation.

3.2.2. PEC's ecologic and anthropization status

The PEC is a RAMSAR site that comprises vast mangrove forest belts surrounded by a significant portion of the South-East Atlantic Forest Reserve (Natural World Heritage Site – UNESCO, 1999). Four Federal Conservation Units (FCU) ensured by Brazilian law are established within the PEC: The Environmental Protection Area (EPA) of Guaraqueçaba, Guaraqueçaba Ecological Station, Superagui National Park, and Biological Reserve of Bom Jesus (Fig. 1). Therefore, the PEC has socioecological importance, representing an essential habitat for terrestrial, estuarine, and marine species and supporting the local fisheries communities' livelihood. Nevertheless, PEC has an increasing degradation level (Estades, 2003) despite the valuable preserved areas, mainly on the east-west axis.

The Paranaguá Bay harbors one of the biggest ports of Latin America (the Paranaguá Port), and the Paranaguá City, the most populated center at the Paraná coastal zone (estimative of 154.936 inhabitants; IBGE, 2021). Waste-water treatment plants (WWTP) of Paranaguá have their discharges on Itiberê and Emboguaçu River, south and north of the city, respectively (Fig.1.), however, the treatment compass about 90% of the sewage (Instituto das Águas do Paraná, 2017). The Itiberê River is the primary domestic and industrial sewage source to the PEC (Martins et al., 2010; Cabral et al., 2018). Furthermore, the PEC comprises four population centers and a second port (the Ponta do Felix Port) at Antonina City.

As marine litter sources in the PEC, Krelling et al. (2017) associated the harbor area, a mooring area inside the estuary, the mouth of Itiberê River, and in front of the channel of the National Department Against Drought (DNOS channel), which are related with sewage, domestic and harbor inputs. Moreover, PEC encloses areas with port and industrial activities, navigation, tourism, fishing, aquaculture, landfills, and poor sanitation next to the mangrove vegetation and riverbanks (Procopiack et al., 2007; Silva et al., 2015), which can be correlated as potential MPs sources. Besides, for MPs, the WWTPs are not 100% efficient in retaining the particles, also represented as a source of this contaminant (Karbalaei et al., 2018).

3.3. Material and methods

3.3.1. The TrackMPD model

The TrackMPD is a recent tracking-particle model framework developed in Matlab (Jalón-Rojas et al., 2019b) that allows simulation of a diversity of processes as beaching, washing-off, windage, sinking, deposition, degradation, and biofouling. Besides, the advantage of this model is the versatility of using velocity data from different OGCMs, and it is also a comprehensive and user-friendly tool (Jalón-Rojas et al., 2019b). The TrackMPD toolbox used was the framework v.1 (available on: <https://github.com/IJalonRojas/TrackMPD/tree/master/TrackMPDv1_Toolbox>) ran in MATLAB R2017b version. The model was run using the 2D velocity fields of PEC from the Brazilian Sea Observatory, simulated on the MOHID Water Modelling System (Franz et al., 2021). The TrackMPD is originally a three-dimensional model but has a 2D version, ignoring the vertical term (Jalón-Rojas et al., 2019b). So, the 2D TrackMPD considers only horizontal hydrodynamic and thus is representative for floating microplastic, that is, with density lower than seawater (~1.02 g/cm^3; e.g., polyethylene – 0.917 to 0.965 g/cm^3; polypropylene – 0.85 to 0.94 g/cm^3; Hidalgo-Ruz et al., 2012).

The PEC MOHID OGCM included three downscaling levels, the third with a horizontal grid resolution of 200 m x 200 m for PEC (Fig. 2). The tidal constituents were from FES2014 (Finite Element Solution), using 31 tidal constituents (Franz et al., 2021). The bathymetries were defined based on Brazilian Navy nautical charts and from local data measured by local institutions and companies and collected by the Center for Marine Studies (Franz et al., 2021). The Global Forecast System obtained atmospheric boundary conditions with a 0.25º horizontal resolution (Franz et al., 2021). River freshwater inputs used on the model were monthly averages calculated using data from the National Water Resources Information System (https://www.snirh.gov.br/hidroweb/) or published papers and thesis (Franz et al., 2021).

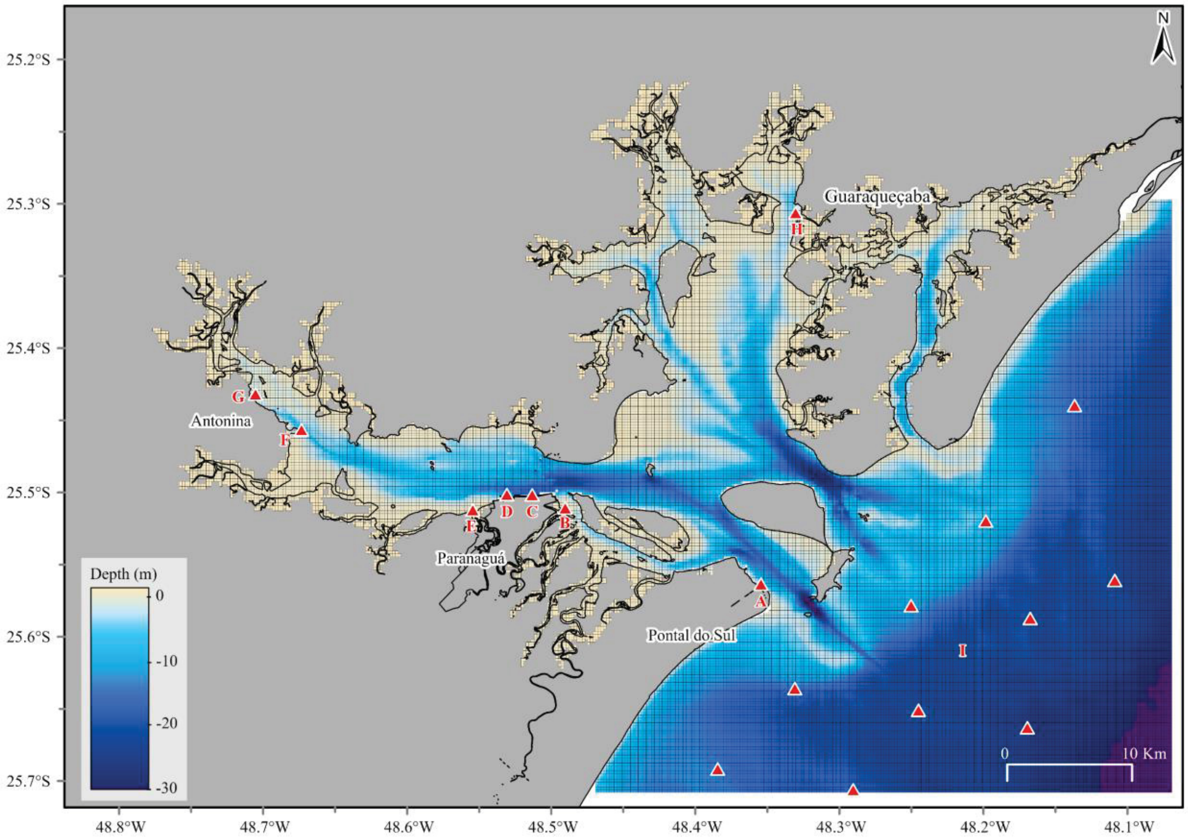

Fig. 2. CEP MOHID water model grid data and TrackMPD release points (red triangles; for letters reference, please see Table 1) – release point "I" comprises ten locations represented by the ten triangles located on the ocean basin.

Due to the lack of general wind patterns regional studies, we did not apply the windage drag on TrackMPD. Only beaching behavior was applied to the particles in our simulations; No specific size and density were defined for the particles, considering they only as positive buoyancy particles without sinking. Particle release points (N = 9) were chosen along with the PEC according to the proximity of potential MPs sources (Table 1). The simulation was run for 40 days. Due to the lack of measured data about MPs discharge in the PEC, we modeled the releases equally for all release points regarding the number of particles. Ten particles every six hours were released for 30 days from each release point (total =10440 particles). For release point I, particles were released from ten different locations as one particle per location (Fig. 1). The emission occurred under four tidal conditions, starting on October 1st, 2014, the beginning of the neap tide cycle. After this period, no more particles were released. The previously released particles stayed submitted to drifting until the last ten days, configuring approximately three times the estuary residence time.

Table 1. Model release points, site description, and MPs potential sources related.

Release point	Description	Potential sources
A	DNOS channel	Sewage discharges and urban runoff from Pontal do Sul; vessel transit and marinas activities; and local tourism;
B	Itiberê and Correias's river mouth	WWTP and sewage discharges; urban runoff from Paranaguá City (Martins et al., 2010; Cabral et al., 2018); vessel transit; and marinas activities;
C	Paranaguá Port	Accidental loss during cargo loading and transport; pellets (Pereira, 2014);
D	Paranaguá City	Industrial and urban waste;
E	Emboguaçu river mouth	WWTPs discharges, urban runoff, landfills on the river margin and mangrove areas (Silva et al., 2015);
F	Ponta do Félix Port	Accidental loss during cargo loading and transport;
G	Antonina City	Urban runoff;
H	Guaraqueçaba City	Sewage discharge and urban runoff;
I	Anchoring ship zone	Accidental loss during cargo transport; Marine source; Marine debris fragmentation;

3.3.2. Probability density functions (PDF) and model accuracy test

For better visualization and interpretation, model results of all particle trajectories and fates were analyzed with the aid of probability-density maps calculated through probability density functions (PDFs). PDFs are calculated by the probability that a particle moves from one location to another over a time interval by counting the number of particles per bin and then normalizing by the total number of particles and maps through binning particles position in histograms (Jalón-Rojas et al., 2019a). Aggregating integral curves define the PDFs for such particles (Van Sebille et al., 2018). This analysis can be used to determine the expected tracer concentrations and is widely used to predict expected dispersal patterns of materials in turbulent processes (Mitarai et al., 2009). We calculated PDFs for a 0.01° x 0.01° grid resolution (approximate 1,02 km²) to assess, quantitatively, the most probable MP accumulation areas of the MPs from each release point, represented through de highest probabilities.

To test the model output accuracy, we compare the probability values calculated through a PDF considering all release points (all particles), with MP (1 – 5 mm size) field data of nineteen PEC beaches sampled at the high tide line on spring tide conditions in December of 2020 (Mengatto and Nagai, *submitted*). The field data were compared with the probabilities in the bin that's corresponded to their locations. Pearson's and Spearman's (ranks) tests correlation was implemented with R statistical software (2021). Probabilities and MPs field data correlation were analyzed considering all samples, and after separating the urbanized beaches occupied by fisheries communities or nearest to cities (N=14) – S1 (Encatadas), S3 (Ponta Oeste), S4 (Ponta do Poço), S7 (Itiberê River), S8 (Rocio), S9 (Emboguaçu River), S10 (Ilha do Teixeira), S11 (Ponta da Pita), S12 (Europinha), S13 (Eufrasina), S15 (Ponta do Ubá), S16 (Mariana), S18 (Guaraqueçaba), and S19 (Vila das Peças) – and non-urbanized beaches, without direct human influence (N=5) – S2 (Limoeiro), S5 (Ilha Rasa da Cotinga), S6 (Ilha das Cobras), S14 (Piaçaguera), and S17 (Ilha das Gamelas) (for the beaches locations, please refer to Fig.1).

3.4. Results

3.4.1. 2D TrackMPD model outputs

The PEC 2D TrackMPD particles differ in trajectories, particle fate (Fig. S1), and probability values distribution (Fig. S2 until S10), especially among distant release points. Altogether, almost 80% of MPs were beached, and 20% remained on the water at the end of simulations. Besides, 76 particles (less than 1%) got out of the model domain; specifically, those from DNOS channel (A) and Anchoring ship zone (I) release points. MPs show a general mean of 7 (± 9.7) days of movement; this value, however, changes according to the release point location. For release point A, on estuary mouth, particles trajectories duration is 13 (± 11.8) days on average, and towards the inner estuary, this decrease (Fig. 2). For release points B, C, and D, placed on the middle of the Paranaguá Bay, trajectories duration average are 4.7 (± 8), 9 (± 9), and 9.8 (±8.6) days, respectively, while for the most inner release points, less than three days (E = 1.1 ± 3.8; F = 2.7 ± 4.9; G = 0.5 ± 1.4; H = 2.5 ± 3.5). From release point I, particles trajectory duration average was higher (19.8 ±10.4).

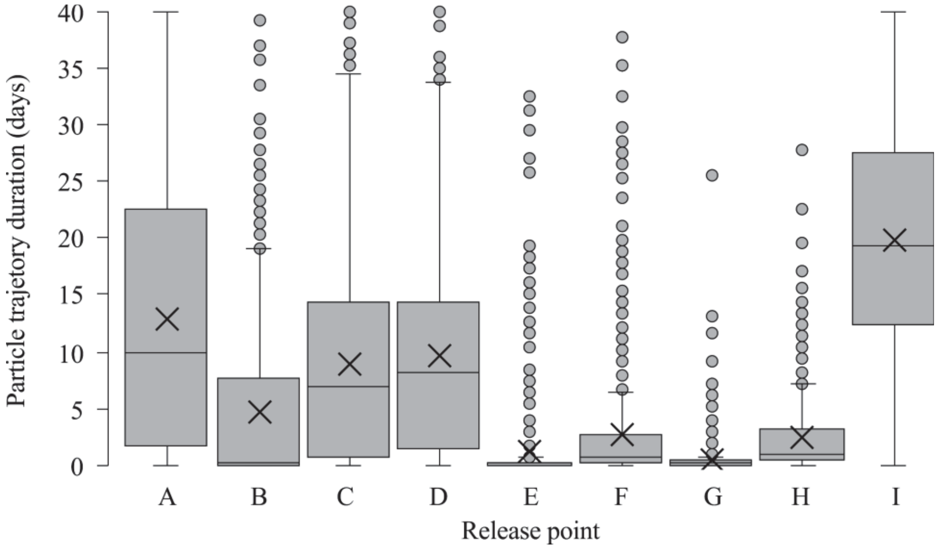

Fig. 3. Boxplot of particles trajectory duration per Release point; X – average particle trajectory duration; grey circles - outliers (considering the interquartile range - IQR).

The highest probability values (Pv) calculated through PDFs are found around areas where particles are predominantly beaching. Pv values were considered high relative to the proximity of the maximum Pv of each release point; considering all release points (for Data-model comparison, below), median and high Pv values are >0.01 and >0.03, respectively. Particles from the DNOS channel (release point A), beaching next to the estuary mouth, on the shoreline nearby the release point (Pv = 0.045), at the Ilha do Mel estuary facing the southern section (Encantadas beach; “S1” in Fig. 1; Pv = 0.034), and at the southeast margin of Ilha da Cotinga (Pv = 0.028; Fig S1 – A and S2). Around 40% of particles from A remain on the water at the end of simulations, most (38%) at the Atlantic Ocean shelf adjacent to the PEC. In contrast, the innermost release points had more than 80% of particles beached inside the estuary (Table 2). It is important to highlight that the 2D TrackMPD model considers that beaching occurs when particles' movement stops reaching inside the defined data domain polygons. However, the 2D approach beaching does not consider the presence of rigid structures (i.e., port docks), located above the tide levels, in which beaching would not be possible. These occurred almost for all release points (except H and I), however, was important for B, C, D, and E, which had more than 10 % of the particles beached at the Paranaguá Port dock (Table 2).

Table 2. Model output for each particle status at the end of the simulation (% of particles). Water – particles that remain on the water, and Beached – beached particles; and its location inside the estuary, on the oceanic shelf, and beached on port docks. The estuary limit was defined following Marone et al. (2005).

Release point	Water		Beached			
	Estuary	Oceanic shelf	Estuary	Oceanic shelf	Paranaguá Port dock	Ponta do Félix Port dock
A	2.8%	38.1%	50.9%	6.9%	0.1%	-
B	8.0%	0.6%	91.4%	-	15.9%	-
C	15.3%	1.6%	83.1%	-	29.6%	-
D	15.2%	1.1%	83.7%	-	25.2%	-
E	1.0%	-	99.0%	-	9.9%	-
F	3.5%	-	96.5%	-	1.8%	1.9%
G	-	-	100.0%	-	-	0.8%
H	0.8%	-	99.2%	-	-	-
I	0.9%	86.8%	5.0%	2.0%	-	-

The particles released next to Paranaguá City and Port show similar trajectories, limited to the inner portion of the Paranaguá Bay and spread over the Laranjeiras Bay, reaching the ocean through both the north and south estuarine mouth channels, even if their fates differ. From the Itiberê and Correia's river mouth release point (B), most particles remain next to the release point on the river's mouth and the Cotinga channel (Pv = 0.190), with some spreading on the Paranaguá and Laranjeiras Bays (Fig. S1 – B). Still, release points C and D show similar trajectories and fates, reaching farther into the Paranaguá and Laranjeiras Bays than those from release point B. From these sources, although the highest Pv occurred at the Paranaguá Port dock (Pv = 0.121 for C – Fig. S3; and Pv= 0.112 for D – Fig. S4), relatively high Pv are also found at the Rocio Beach (Fig. 1 – "S8"; Pv = 0.089 for C; Pv = 0.075 for D), on the mangrove areas at the port opposite margin (Pv = 0.046 for D), on the surroundings of Ilhas Jereres and Ilha do Lamin (Pv = 0.032 for D), and at Ilha das Cobras (Pv = 0.037 for C; Pv = 0.035 for D). While particles released from the Emboguaçu river mouth (E), the innermost release point on Paranaguá Bay, remain within the estuary, mainly inside the Paranaguá Bay, with few particles reaching the Laranjeiras Bay (Fig. S1 – E), and particles quickly beaching in the same bin of release point E (Pv = 0.352). Also, this release point influences the Rocio Beach area with relatively high Pv (= 0.164; Fig. S6).

The particles released from more internally located points on the estuary (release points Ponta do Félix Port, Antonina City, and Guaraqueçaba City; Fig. S1 – F, G, and H, respectively) remain within the PEC, and the majority of particles reach the estuary tributaries mouths, such as the Cachoeira and Cacatu river mouth for particles released from points F and G (Pv = 0.041 and 0.124, respectively), and the Guaraqueçaba river for point H particles (Pv = 0.056).

Additionally, particles from the Ponta do Félix Port release point (F) also reach the Paranaguá and Laranjeiras Bays, the latter with fewer particles. Mainly, release point F shows the highest Pv next to the Ponta do Felix port (Pv = 0.154; Fig. S7) and release points G and H (Fig S8 and S9, respectively) in the same bin where they are allocated (Pv = 0.319 and 0.213, respectively).

Few particles from release point I enter and beached inside the PEC; most remain in the water column at the adjacent ocean basin at the end of simulations (Fig S1 – I; Table 2). Pv from this release point is too small inside the PEC (maximum Pv = 0.003 on the southeast margin of Ilha da Cotinga; Fig. S10), representing a minor contribution for the estuary.

3.4.2. Data- model comparison

The PDF considering all release points is represented in Figure 3. The highest probabilities are observed on Emboguaçu River mouth (Pv = 0.039), followed by Antonina City (Pv = 0.036), and the Rocio beach at Paranaguá City (Pv = 0.033). These are the leading MPs accumulation hotspots given from the release points used on the 2D TrackMPD output. The model also shows that relatively high probability values are found at the Paranaguá ports docks (Pv = 0.031). However, as these are rigid structures, we can not consider them as MPs accumulation hotspots (explanation above). Notwithstanding, relatively high and medium probabilities values are observed at the Itiberê River mouth (Pv = 0.024), Guaraqueçaba City and Cerquinho River mouth (Pv = 0.026), Cachoeira and Cacatu river mouth (Pv = 0.017), Ilha das Cobras (Pv = 0.011), and on dense mangrove areas located at the south of Ponta do Felix port (Pv = 0.010) and on the Paranaguá port opposite margin (Pv = 0.009).

The correlation results between the 2D TrackMPD Pvs and observational MPs (1 – 5 mm size fraction) abundance data on PEC sandy beaches are shown in Table 3 and presented in Figure 5. When considering all observational data sampling points, no significant values are observed. However, when urbanized and non-urbanized beaches are considered separately, a strong positive ($R = 0.881$) and significant ($p < 0.05$) Person's correlation is observed between model output and non-urbanized beaches field data, with a positive but not significant ($p > 0.1$) Spearman's test result.

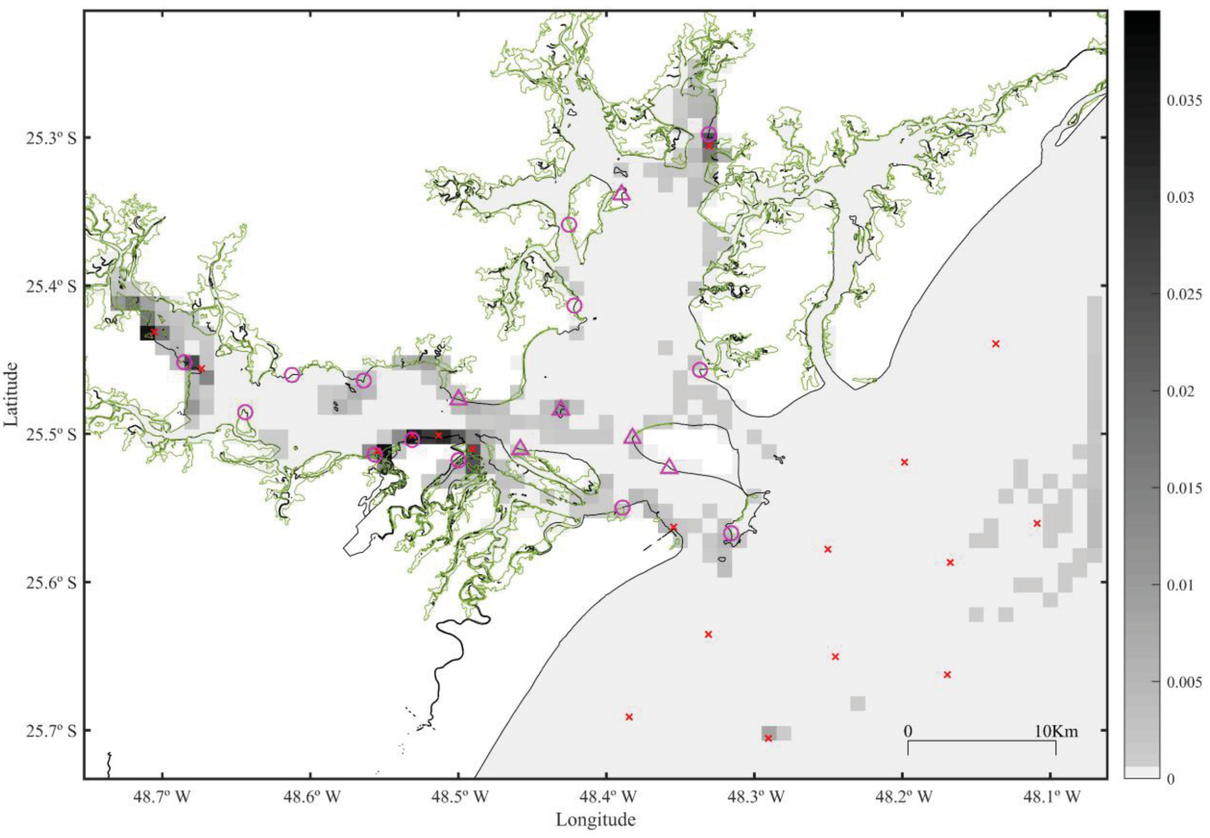

Fig. 4. Probability density distribution (Pv; grayscale bar) with all PEC 2D TrackMPD implementation release points. Red 'X' represents all release points locations; Beaches sampled by Mengatto and Nagai (submitted) are represented by magenta circles – urbanized beaches, and magenta triangles – non-urbanized beaches. Green contours represent the mangrove areas.

3.5. Discussion

3.5.1. 2D TrackMPD model outputs and MPs hotspots within the PEC

Overall, our results show that MPs distribution on PEC differs depending on release point location, though near MPs sources can influence the same areas with different MPs concentrations. Additionally, the probability-density maps obtained provide a good insight into potential MPs accumulation hotspots within the PEC inner areas (Fig. 3).

Table 3. Pearson's and Spearman's test results of correlation between model probability and MPs field data from Mengatto and Nagai (*submitted*); '*' in Pearson's p-value represent significance ($p<0.05$).

	Pearson's correlation				Spearman's correlation		
	R	t	Df	p-value	ρ	S	p-value
All samples	-0.142	-0.591	17	0.562	-0.041	1186.3	0.869
Urbanized beaches	-0.209	-0.742	12	0.473	-0.183	538.3	0.531
Non-urbanized beaches	0.881	3.225	3	0.048*	0.700	6	0.233

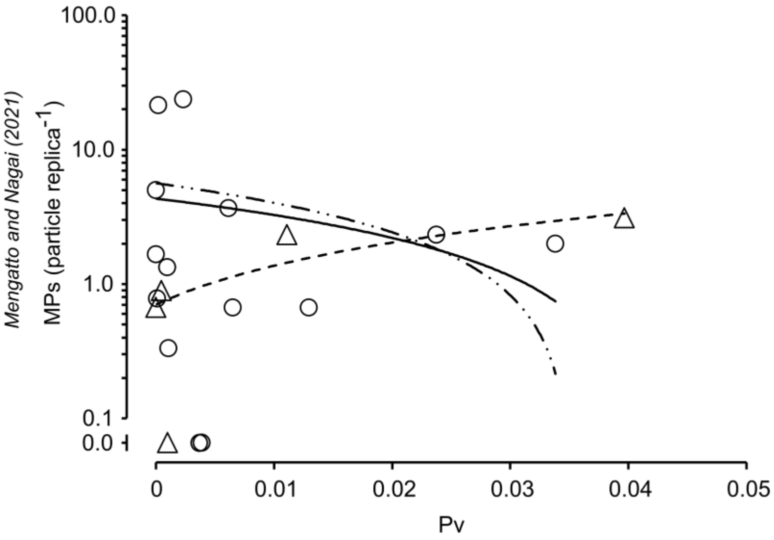

Fig. 5. Correlation between model PDF output (Pv) and MPs observational data from Mengatto and Nagai (*submitted*). Circles – Urbanized beach samples; Triangles – non-urbanized beach samples; dash-dot line – urbanized beaches samples linear regression; dashed line – non-urbanized beaches samples linear regression; black line - all samples linear regression.

The 2D TrackMPD results indicate that MPs sources are inside the PEC, with insignificant contribution (0.6 % of particles) from ocean sources (model output for release point I). The modeled trajectories and MPs fates also suggest that the PEC could function as an MP source for the adjacent continental shelf, specifically MPs from sources located in the east-west axis from the middle part of the estuary towards the mouth, which includes the Paranaguá Port area. These particles are mainly exported to the adjacent ocean shelf via the northern mouth of the estuary (Fig. S1 – B, C, D), except for the DNOS channel, following a similar pathway of the suspended sediment transport depicted by Mayerle et al. (2015). Considering that longshore currents flow to the north, the PEC may also serve as a potential MPs source for the Superagui National Park sandy coastal beaches. Our model results also highlight the importance of local sources for MPs distribution in different PEC bays. The more internal the source, the higher the number of particles that remain within the bay. For example, MPs sourced at the inner portion of the estuary tend to be dispersed and beached within these areas. In the case of Guaraqueçaba City, particles beaching around the release point (H), following the direction of tributary rivers. Besides, release point H influences MPs' presence in mangrove areas that comprise the Guaraqueçaba Ecological Station (Fig S9). Thus, considering that release points simulated here are the main for these inner areas of the estuary (F, G, and H), mitigation in MPs inputs around these sources (Antonina City and Port, and Guaraqueçaba City) could influence the MPs pollution directly in the inner areas (i.e., the Cachoeira, Cacatua, and Guaraqueçaba rivers’ mouth areas).

Our model results highlight that MPs sourced at the Paranaguá Bay (release points B, C, and D) also have the potential to be exported to the Laranjeiras Bay, reaching a large portion of the Guaraqueçaba EPA, this implies that special attention should be given to MP pollution in PEC protected areas. This TrackMPD output is corroborated by Mengatto and Nagai (*submitted*), who reported the presence of plastic pellets on beaches located in the Guaraqueçaba EPA domain, associating port activities as probable sources (needs investigation). Notably, particles released from the Paranaguá Port (release point C) are widespread along the Paranaguá Bay, reaching the ports opposite margin mangroves, the inner part of the Laranjeiras Bay, and are also exported to the adjacent continental shelf. The Paranaguá port receives loads of imported resin pellets (Pereira, 2014), and losses during unloading may be a source for this type of MP to the surrounding areas. If the port activities are a source of plastic pellets to the EPA, this finding must be further investigated, and monitoring these activities as a source of MP pollution to the PEC should be carried out.

TrackMPD MPs differ from macro litter trajectories applied by Krelling et al. (2017). These authors implemented the Delft3D model showed trackers being exported to the adjacent continental shelf and PEC mouth adjacent sandy beaches, and no particles remain in the estuary. In an overall context, MP transport is probably different from macro litter in the PEC. However, the comparison with Krelling et al. (2017) must be careful. These authors do not implement particle beaching behavior, not exploring if particles can beach inside the estuary. Besides, particles displacement on the Laranjeiras Bay is not seen, maybe due to the lack of data for the PEC north-south axis, differently from the MOHID hydrodynamic model used here, which has monthly mean river discharges from both axes in the PEC (Franz et al., 2021). Nevertheless, it is a fact that both modeling trajectories, MPs and macro litter (Krelling et al., 2017), have a transboundary movement in the PEC, and it must be considered for management actions. In addition, PEC contributes as a source of MPs for the adjacent shelf but not as a sink for MPs from this ocean area, similar to macro litter (Krelling et al., 2017), thus can be considered as a good source of these floating plastics for the marine environment.

3.5.2. Data-model comparison

The comparison between the 2D TrackMPD output and field data yields no significant correlation when considering all sampled and urbanized beaches. However, it shows a good agreement when non-urbanized beaches are considered separately, emphasizing local MPs input as potentially important, also highlighted by Mengatto and Nagai (*submitted*). Since at non-urbanized beaches, local sources are insignificant, MPs abundances in these sites are influenced by external sources, yielding a better correlation with the model results than for the urbanized beaches. Worth noting that the field data shows that MPs are present even in non-urbanized beaches where the model output indicated zero PDF (Fig. 4).

Several factors may be involved in this model-data comparison outcome, encompassing factors from the model initial run parameters choice to the field data acquisition. Beached particles on rigid structure sites (almost 10% of the total; 2D version ignoring vertical term) can occur in port docks and in other structures such as piers or seawalls, influencing the Pvs of the model output. Indeed, 3D modeling is a more robust option for the TrackMPD and can result in different particle trajectories, mainly in sites with strong vertical turbulence, affecting the horizontal displacement (Jalón-Rojas et al., 2019a). Although, according to Jalón-Rojas et al. (2019a), the 2D approach can predict the general patterns of MPs' trajectories and fates. So, our results can be satisfactory for floating MPs, which hardly to sinking either due to incrustation or biofilm, being driven only for horizontal currents. The 3D run is still recommended (Jalón-Rojas et al., 2019a) and must be further evaluated for the PEC, compared with our 2D outcomes to assess the main differences for floating MPs also to evaluate the fates and trajectory to the non-buoyancy polymers.

Another model-related factor may be contour washing-off, which might explain the modeled presence of MPs in sites where Mengatto and Nagai (*submitted*) did not observe MPs, such as the Itiberê river and the Ponta da Pita beaches. However, this behavior was not applied in our simulation since we lack data on particles' half-life on beaches. Recently Hinata et al. (2020) developed a model that accounts for backwash probabilities estimations; however, this model still needs further field data validation. Additionally, three factors should be considered regarding the field data. First, observational data refer to MPs in the 5 – 1 mm size fraction, not considering the abundance of other sizes fractions, such as the < 1 mm or even the mesoplastics (5 - 25 mm) since particle size is not specified in the simulation. Although, for some simulations, the particle movement is practically the same for buoyancy particles from different densities and sizes (Alosairi et al., 2020; Sousa et al., 2021), trajectories can vary depending on the size, polymer density, and shape of the particle released (Khatmullina and Chubarenko, 2019). Secondly, Mengatto and Nagai (2021) did not realize the chemical analysis, so the particle's polymer density was not explored. For TrackMPD 2D, it would be interesting the comparison with <1 g/cm^3 density MPs, which are hard to sink. Lastly, the chosen sampling design may also underrepresent the general MPs abundance variability on PEC beaches as samples were retrieved in different sampling campaigns in distinct weeks and subsequent tidal cycles, which may have an overlapping effect over MPs abundances (Mengatto and Nagai, *submitted*).

Other factors that may have affected our data-model comparison results include insufficient particles in the simulation, missing punctual or diffuse sources (local or external), and the balance between the number of particles per release point. These are critical points that have the potential to modify the robustness of our results. Diffuse sources such as domestic, industrial effluents, or even atmospheric inputs (Akdogan and Guven, 2019), are challenging to account for. Additionally, mesoplastics (> 5mm) and larger plastic fragments (>25 mm) weathering can also act as sources for MPs. Particularly on beaches, where high oxygen availability and intense sunlight exposure enhance plastic degradation, cracking, and consequently fragmenting larger plastic, and thus, these sites can be considered a hotspot of MP generation (Andrady, 2011).

Our model-data comparison could not validate the 2D TrackMPD for the PEC, and a more robust validation is required, considering both model and field adjustments. Mangrove sediments may be a potential environment to test our models' accuracy since our results show accumulation on these areas, mainly next to the Itiberê river, on the port opposite margin, and the inner Antonina Bay. The MOHID OGCM for the PEC considers the mangrove areas in the grid data, driving water fluxes through these, mainly on the high tides. When we include mangroves as emerged areas in the data domain map, the model can drive the MPs particles to these areas, and beaching may occur when particles stop inside the map polygon. MPs accumulation in mangrove sediments is likely to occur (Deng et al., 2020; Zamprogno et al., 2021). According to Li et al. (2019), mangrove vegetation can inhibit the process of translocation of microplastics from mangrove surface waters to seawater on flood conditions, retaining the particles.

Additional environmental variables could also be considered within our model run to improve model output. At Santos Bay (Southeastern Brazil), Gorman et al. (2020) applied a particle-tracking model to predict plastic pellets concentrations on neighboring beaches, explaining 45% of observed field data pellet concentrations. The insight was to apply a Generalized Additive Model, including the model, rainfall, and beach zone, which explained 95% of observed beach pellet concentrations rates (Gorman et al.,2020). Nevertheless, the 2D TrackMPD simulations displayed potential MP accumulation areas from multiple sources within the PEC. This highlights that considering particle-tracking models allied to environmental parameters may be an excellent approach to improve model results and represent an alternative tool for this type of pollutant monitoring.

3.6. Conclusion

The TrackMPD particle-tracking model in two dimensions showed to be a promising tool for detecting the most probable areas of MPs accumulation in the PEC. It highlights that the Paranaguá area is a potential MPs source for the Guaraqueçaba EPA and the Superagui National Park, and Guaraqueçaba City for Guaraqueçaba Ecological Station. It also serves as an alert regarding the potential of MPs pollution in these protected areas. Besides, the model shows that PEC MPs have a transboundary movement between different areas of the estuary and presents a source-to-sink dynamic, important information from a local and global perspective of plastic pollution.

The data-model comparison highlights that future studies should (i) conduct further model validation, including a higher number of observational data, such as beaches and other depositional environments, (ii) apply the TrackMPD 3D approach, and (iii) compare it to different modeling tools. Thus, we consider that increasing MP investigations and sampling efforts in diverse PEC environments is a crucial next step to provide a robust validation for particle trajectory modeling application towards the sustainable management of PEC's coastal protected areas.

Acknowledgments

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Following the Federal University of Paraná normative, all cited references are at the end of this document.

3.7. Supplementary material

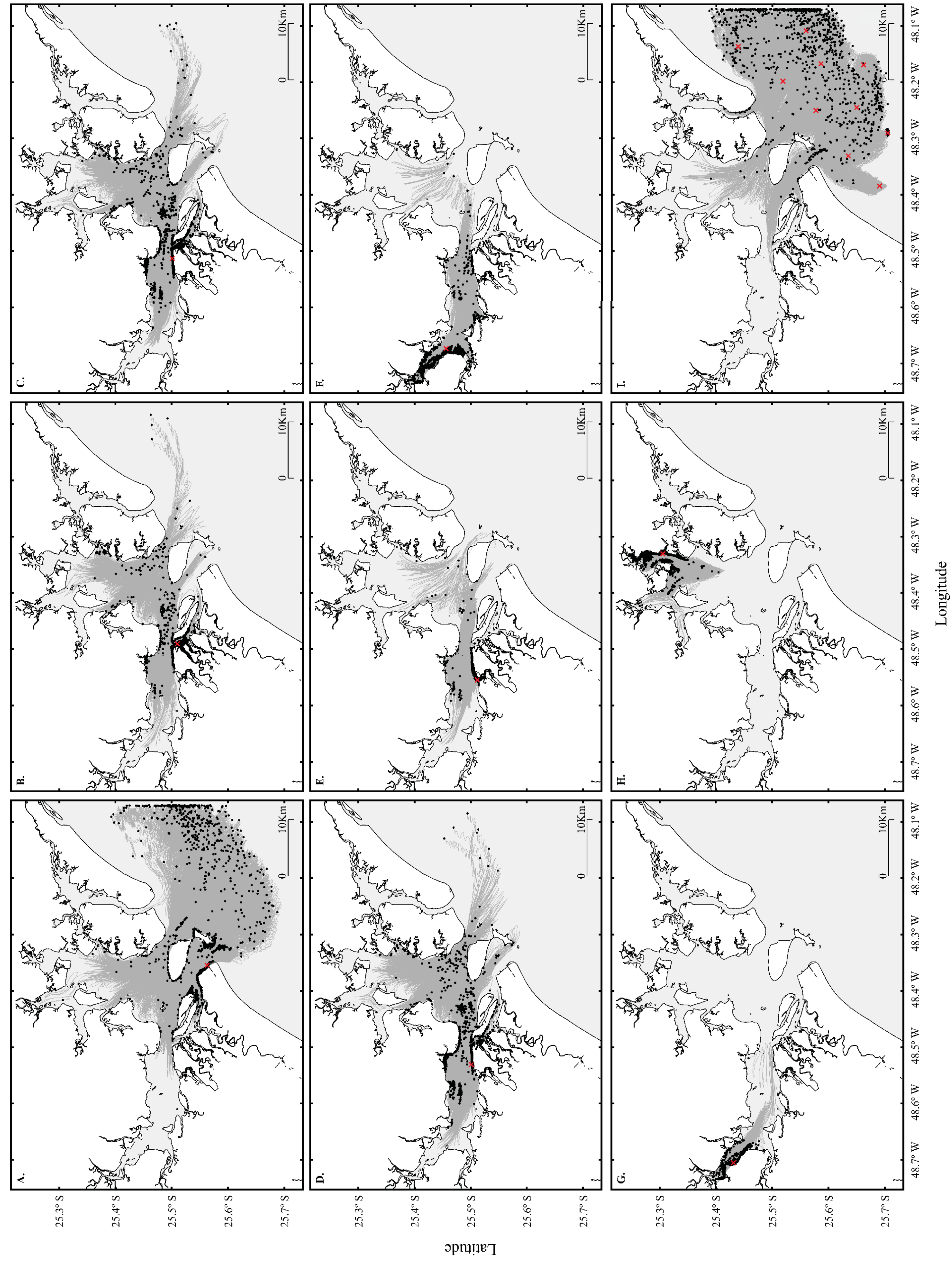

Fig. S1. PEC 2D TrackMPD trajectories (gray lines) and fates (black dots) for release points A, B, C, D, E, F, G, H, and I; refer to them according to letters in the figure.

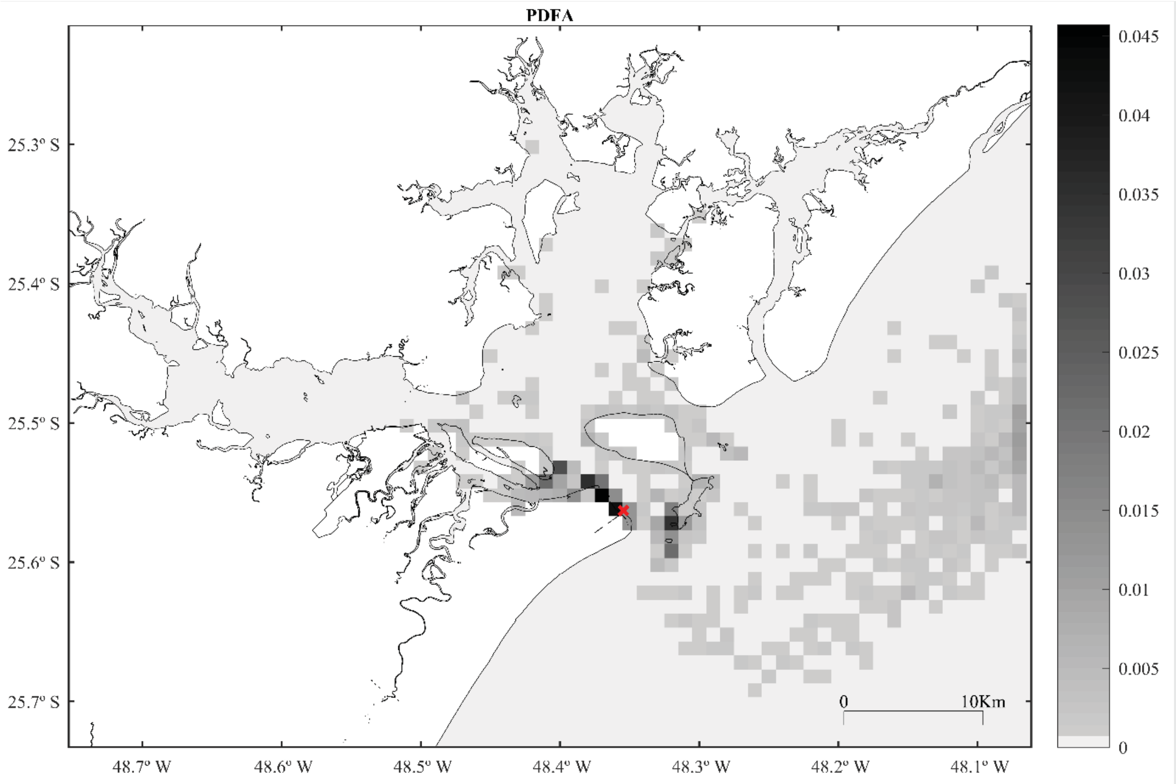

Fig. S2. Probability density distribution (Pv; grayscale bar) for release point A; red 'X' represents the release point location.

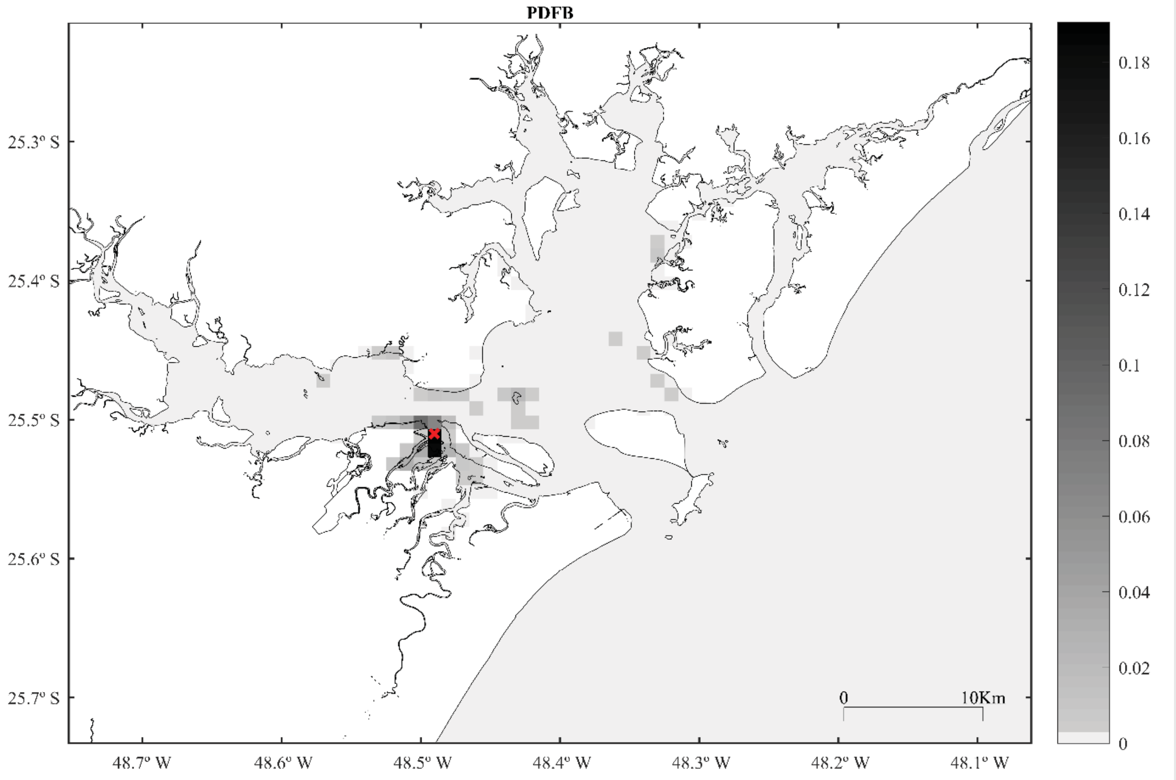

Fig. S3. Probability density distribution (PV; grayscale bar) for release point B; red 'X' represents the release point location.

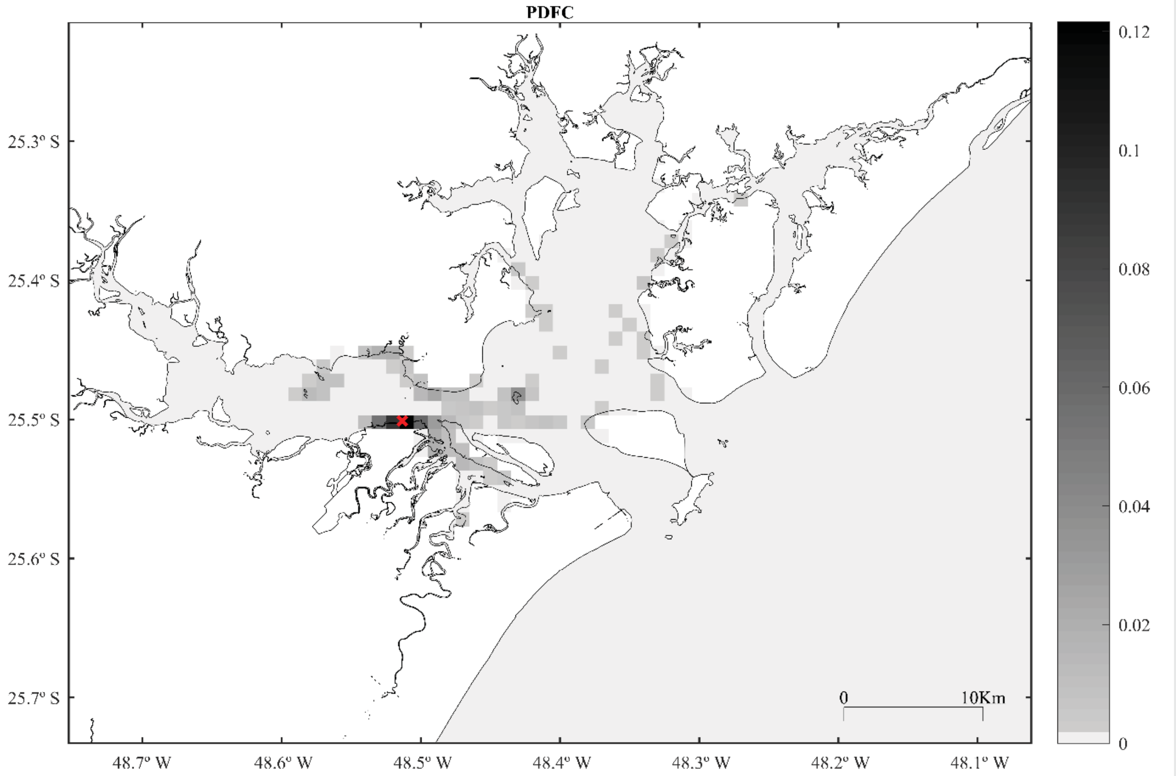

Fig. S4. Probability density distribution (Pv; grayscale bar) for release point C; red 'X' represents the release point location.

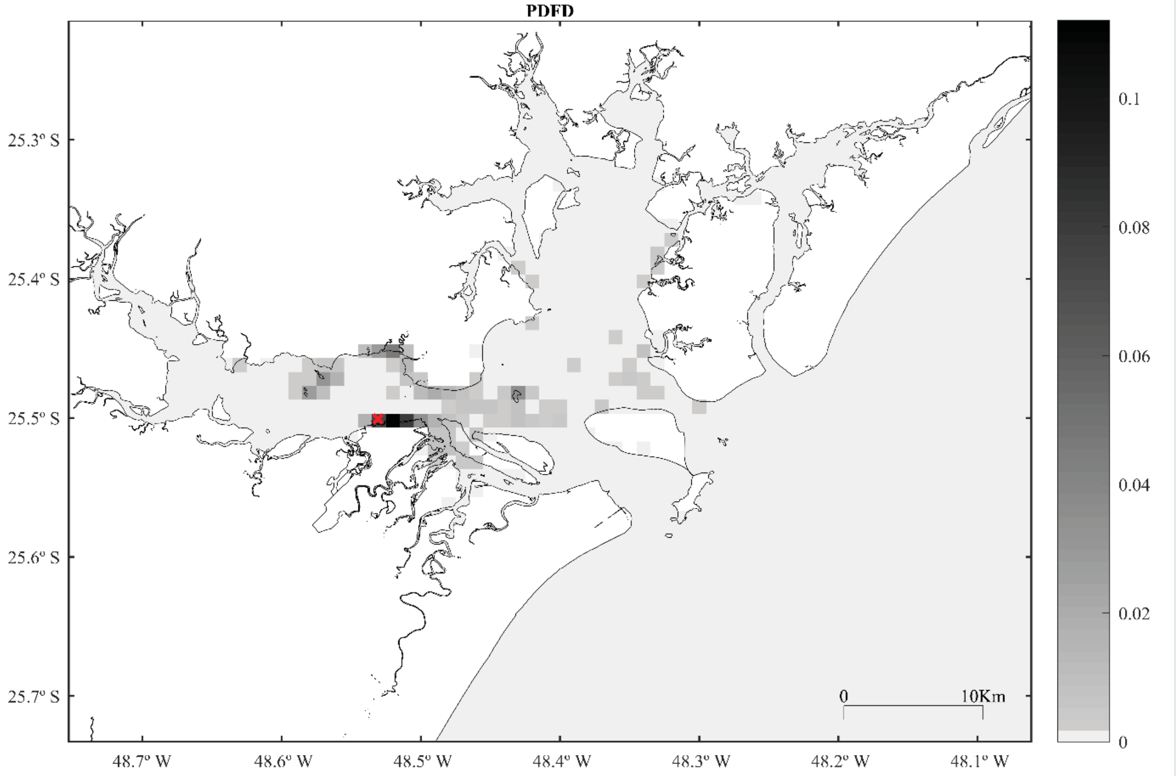

Fig. S5. Probability density distribution (Pv; grayscale bar) for release point D; red 'X' represents the release point location.

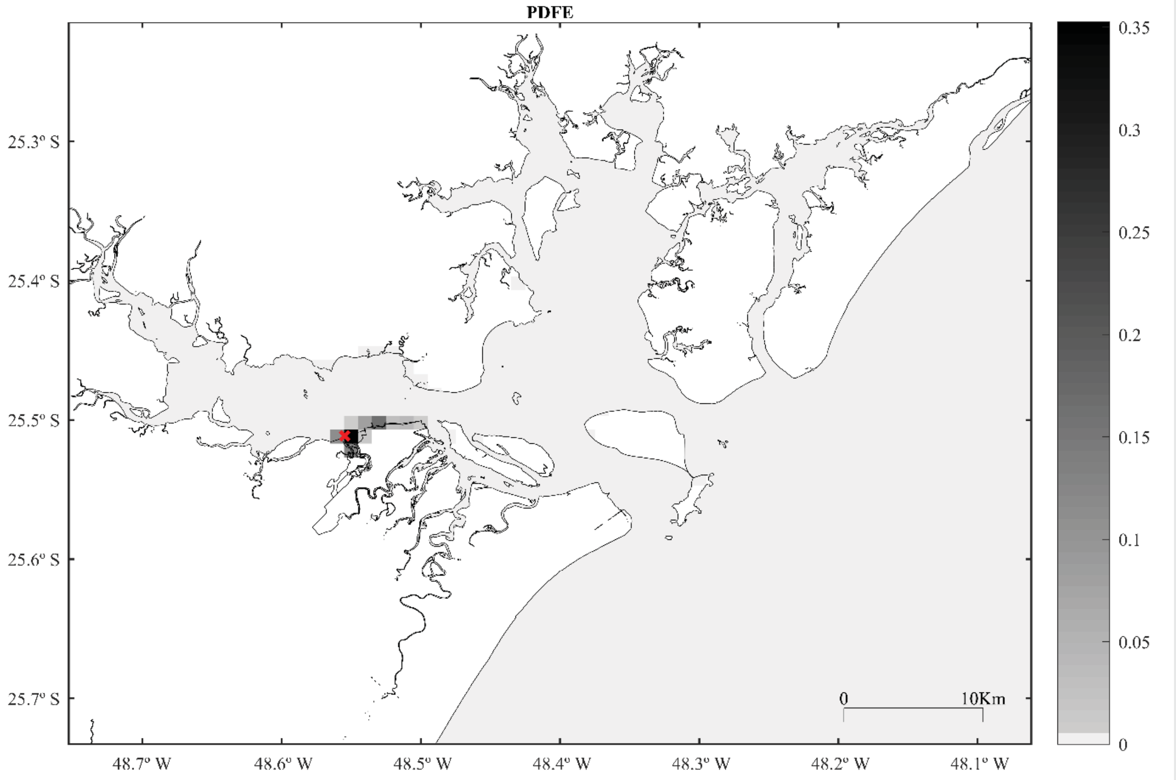

Fig. S6. Probability density distribution (Pv; grayscale bar) for release point E; red 'X' represents the release point location.

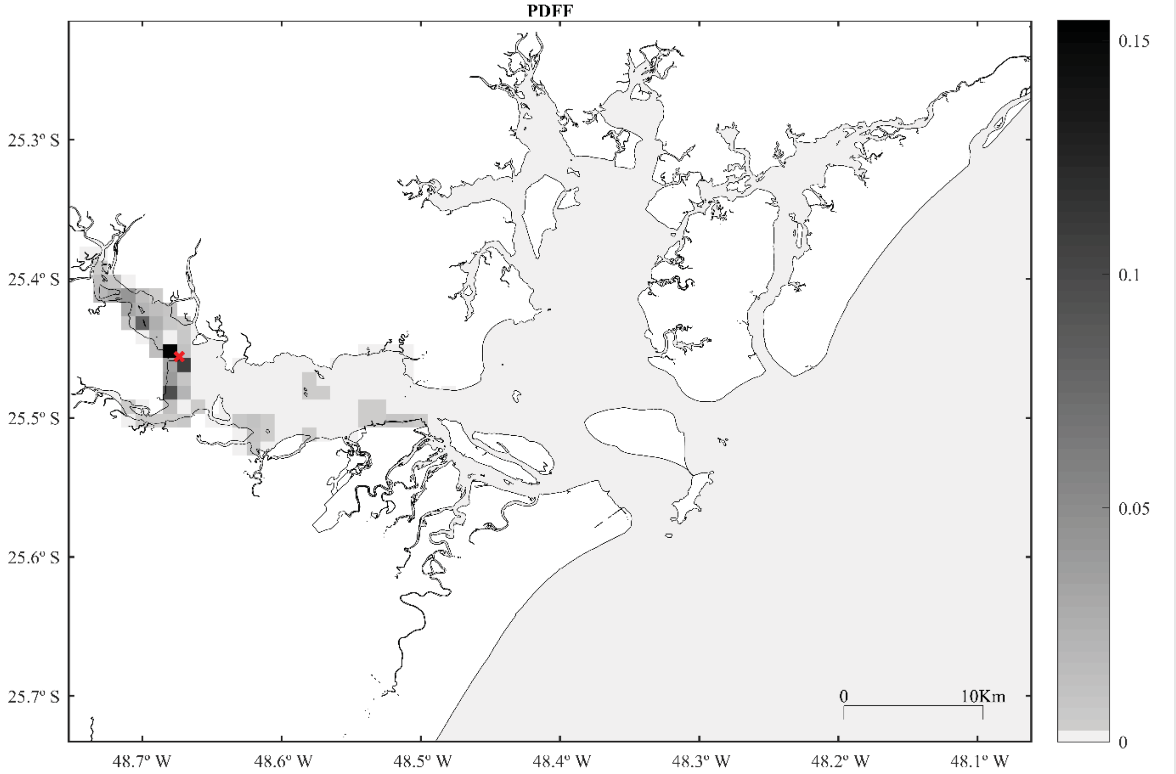

Fig. S7. Probability density distribution (Pv; grayscale bar) for release point F; red 'X' represents the release point location.

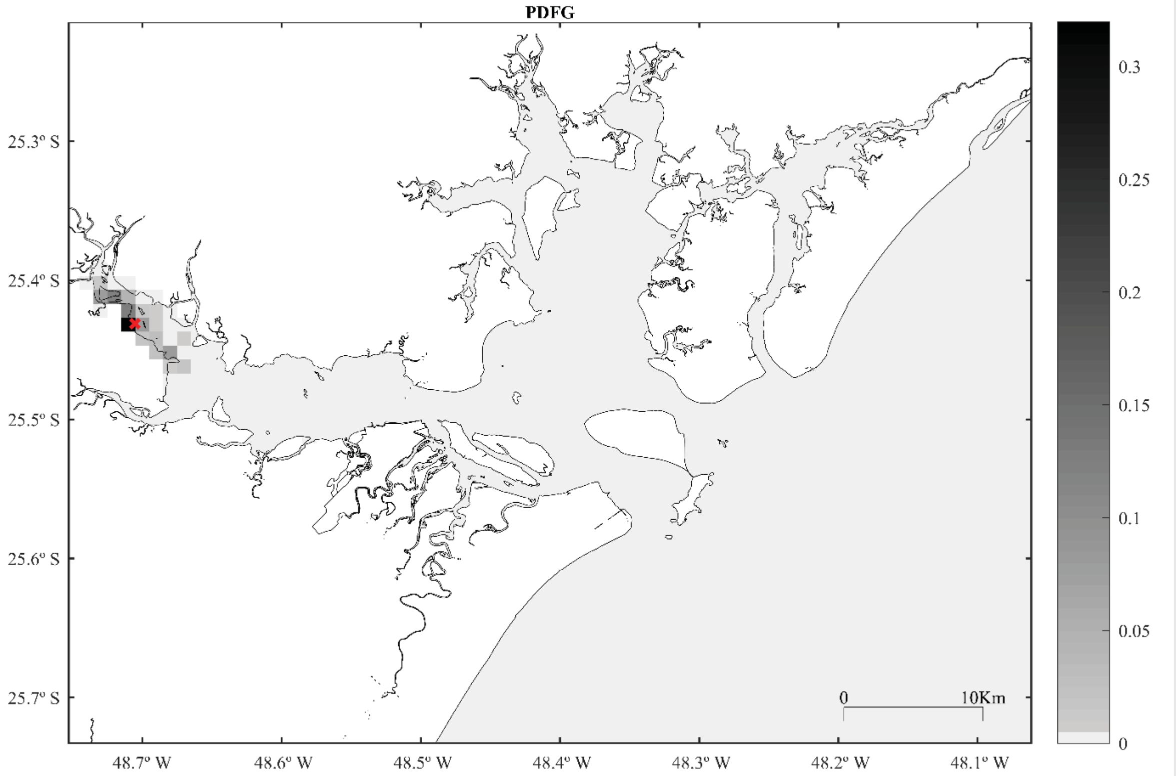

Fig. S8 – Probability density distribution (Pv; grayscale bar) for release point G; red 'X' represents the release point location

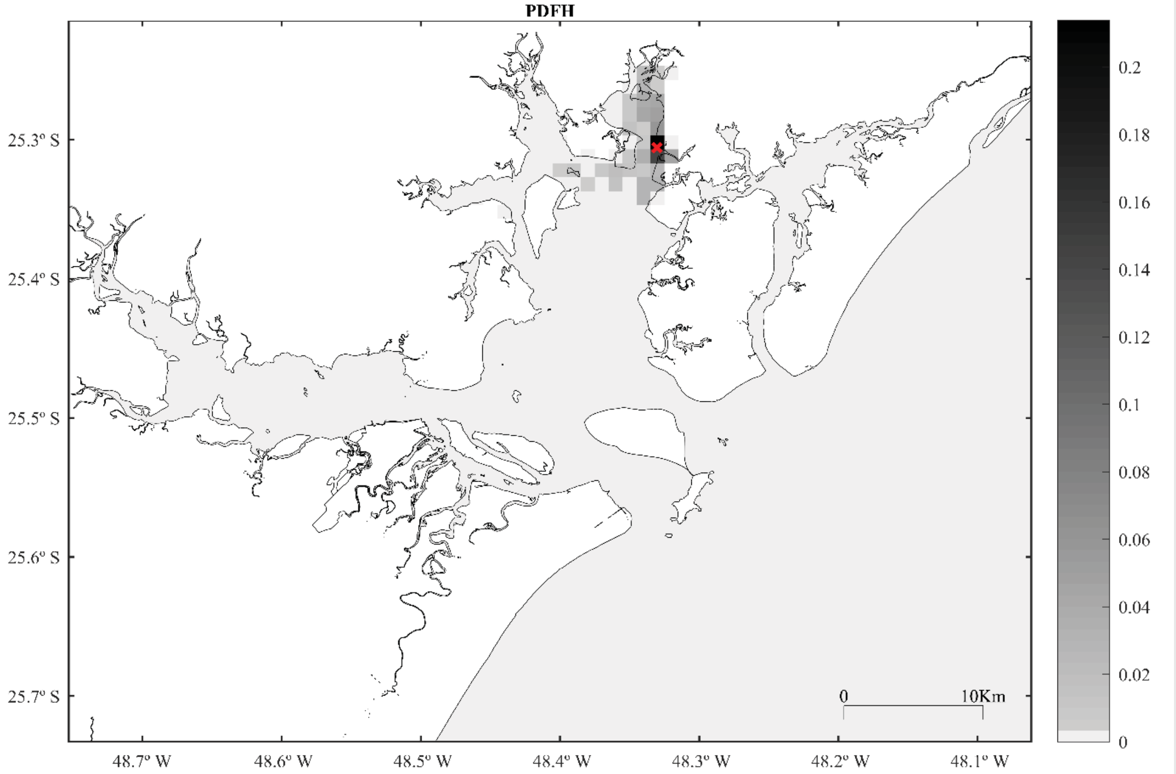

Fig. S9 – Probability density distribution (Pv; grayscale bar) for release point H; red 'X' represents the release point location.

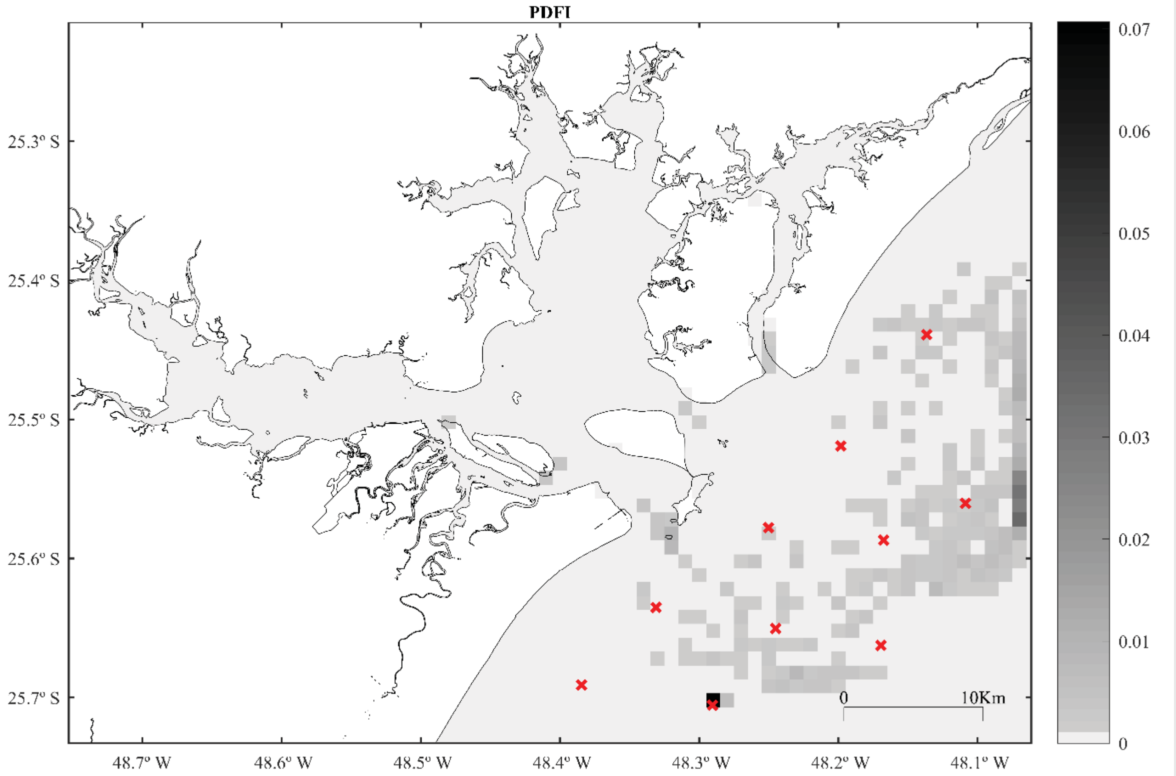

Fig. S10 – Probability density distribution (Pv; grayscale bar) for release point I; red ‘X’ represents the release points locations.

4. CONSIDERAÇÕES FINAIS

A partir da identificação, classificação e abundância de MPs em praias arenosas do CEP, em conjunto com resultados da modelagem de rastreamento de partículas, foi possível gerar uma primeira avaliação da distribuição destes poluentes no CEP e avaliar uma potencial ferramenta de otimização no monitoramento de MPs na região. Nossos resultados indicam que no CEP, os MPs estão presentes na maioria das praias amostradas (em 16 de 19), inclusive naquelas localizadas na APA de Guaraqueçaba, distribuídos espacialmente de forma heterogênea. A presença de MPs primários – *pellets* – nas praias da Baía de Laranjeiras, sugere que as atividades portuárias, transporte e/ou perdas durante carregamentos/descarregamentos, desenvolvidas na Baia de Paranaguá possam contribuir com a poluição por MPs no CEP, podendo impactar as áreas de preservação ambiental da área de estudo. A partir da comparação entre os resultados apresentados no Capítulo I e os resultados obtidos com a aplicação do modelo de rastreamento de partículas – 2D TrackMPD acoplado com o modelo hidrodinâmico MOHID permitiram a avaliação do potencial de aplicação deste modelo no monitoramento da poluição por MPs no CEP. Os resultados desta avaliação, apresentados no Capítulo II, mostram que o modelo ainda precisa ser validado para representar uma ferramenta viável de monitoramento. No entanto, o modelo apresentou dados interessantes a respeito do potencial de exportação de MPs provenientes de fontes localizadas na Baía de Paranaguá para a Baía de Laranjeiras e, consequentemente, para a APA de Guaraqueçaba e para a plataforma oceânica adjacente. Ainda, os resultados do modelo destacam que as principais fontes dos MPs para as praias arenosas do CEP são internas, com uma contribuição mínima de MPs oriundos da plataforma oceânica adjacente. Considerando que a modelagem numérica representa uma ferramenta com potencial de otimizar o monitoramento e subsidiar estratégias de manejo da poluição por MPs em áreas de proteção ambiental e que a validação de modelos numéricos depende de boa correlação entre dados *in situ* e simulados, a ampliação de trabalhos de investigação *in situ* de MPs no CEP representa um próximo passo crucial para a validação da modelagem numérica. Ainda, sugerimos que trabalho futuros investiguem a distribuição espacial e temporal de MPs em diferentes matrizes, abióticas e bióticas, e em ambientes deposicionais diversificados, particularmente em áreas de manguezal. Um arranjo mais robusto de dados observacionais possibilitará um entendimento mais completo do comportamento desses poluentes, possibilitando uma melhor avaliação de modelos numéricos como o apresentado neste trabalho.

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