UNIVERSIDADE FEDERAL DO PARÁ
NÚCLEO DE ECOLOGIA AQUÁTICA E PESCA DA AMAZÔNIA
PROGRAMA DE PÓS-GRADUAÇÃO EM ECOLOGIA AQUÁTICA E PESCA

TESE DE DOUTORADO

DISTRIBUIÇÃO E INGESTÃO DE RESÍDUOS SÓLIDOS, COM FOCO EM MESO E MICROPLÁSTICOS, AO LONGO DA ZONA COSTEIRA DO BRASIL

TAMYRIS PEGADO DE SOUZA E SILVA

Orientador: Dr. Tommaso Giarrizzo

BELÉM-PA
SETEMBRO-2022

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Tese apresentada ao Programa de Pós-Graduação em Ecologia Aquática e Pesca do Núcleo de Ecologia Aquática e Pesca da Amazônia da Universidade Federal do Pará, como requisito para a obtenção do título de Doutora em Ecologia Aquática e Pesca.

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DEDICATÓRIA

AGRADECIMENTOS

Agradeço aquelas pessoas que fazem funcionar a Universidade Federal do Pará (UFPA), o Programa de Pós-Graduação em Ecologia Aquática e Pesca (PPGEAP) e o Núcleo de Ecologia Aquática e Pesca da Amazônia (NEAP), pela formação e por ter agregado boas experiências científicas e pessoais ao longo desse período.

A Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) por fomentar minha formação acadêmica.

Ao meu orientador Dr. Tommaso Giarrizzo pela confiança depositada em mim por mais essa etapa e pelos ensinamentos repassados, tenho grande admiração pelo grande pesquisador com quem tenho o prazer de trabalhar.

Aos colegas da Università degli Studi di Firenze (UniFI) pela disponibilidade e suporte com as análises de polímeros.

Aos meus colegas do GEA, em especial Fabí, Arthur, Érika, Paulo, Thaize, Lúcio, Raque e Juliana. Feliz por compartilhar experiências com pessoas tão prestativas e inteligentes que fizeram a caminhada ser menos difícil. Ao Ryan, pesquisador da UFES que foi muito parceiro contribuindo e guiando para alcançarmos os resultados (artigos) que compõem minha tese.

Aos meus amigos de vida meus mais sinceros agradecimentos. Cada um contribuiu para que eu chegasse até aqui, seja me ouvindo, apoiando, trocando ideias sobre assuntos complexos e/ou desopilando com banhos de praias, rios e igarapés com boas “bibidinhas” e falação de besteira.

A toda minha família, em especial a minha mãe Rita que nunca polpou esforços e colo, a minha avó Lays que sempre tem uma palavra de força e muito afeto, a minha tia/avó Bela e prima/tia Clau por todo apoio, abrigo e cuidados despendidos desde o momento do meu nascimento até hoje. Sou muito grata por ter grandes mulheres ao meu lado, apoiando e que pisaram o caminho para que eu pudesse passar por uma estrada menos tortuosa. Ao meu pai, Alcindo, por todo subsídio, carinho e conversas. Ao meu irmão André por todo apoio e por trazer mais alegria e sabedoria jovem para a minha vida.

Ao Alysson por todo companheirismo e incentivo, tanto para o meu desenvolvimento profissional como pessoal. São anos de caminhada compartilhando os percalços e as boas experiências dessa vida que é muita onda. Sem dúvidas formamos uma boa dupla, os astros já dizem terra (virgem) + água (peixes) são complementares, ele colocando minha cabeça sonhadora demais um pouco no chão e eu colocando um tico de imaginação na cabeça realista demais dele. Agradeço também a família dele que me acolhe tão bem.

A natureza e as deusas, muito obrigada!

EPÍGRAFE

RESUMO

Os resíduos sólidos são definidos de acordo com a ABNT 10004/2004 como sólidos ou semissólidos, que resultam de atividades de origem industrial, doméstica, hospitalar, comercial, agrícola, de serviços e de varrição, cujas particularidades tornem inviável o seu lançamento na rede pública de esgotos ou corpos d’água, ou exijam para isso soluções técnicas e economicamente inviáveis em face à melhor tecnologia disponível. A maior parte da população global vive em centros urbanos, onde os consumidores vêm adotando hábitos e padrões de consumo que favorece a geração de resíduos sólidos, portanto, esta produção está intrinsicamente ligada com o aumento populacional, industrialização e o desenvolvimento econômico.

ABSTRACT

ESTRUTURA DA TESE

A tese está no formato de um capítulo geral integrador e 3 artigos científicos, os quais correspondem a capítulos deste manuscrito. Esta organização obedece ao Regimento Geral do Programa de Pós-Graduação em Ecologia Aquática e Pesca, Resolução nº 4.094/2011 (Art. 66). O capítulo geral apresenta tópicos sobre o estado do conhecimento da problemática da poluição por resíduos sólidos, principalmente os plásticos, formatado de acordo com as regras atuais vigentes da Biblioteca Central da UFPA. Os artigos subsequentes seguem as normas dos periódicos onde 2 estão publicados e 1 a ser submetido.

SUMÁRIO

1. INTRODUÇÃO GERAL

As regiões costeiras estão associadas a grandes crescimentos populacionais e atividades socioeconômicas, e são onde estão inclusas muitas das grandes metrópoles globais (Small e Nicholls, 2003). Os ecossistemas costeiros apoiam o sustento da população como um todo com seus mais diversos serviços ecossistêmicos, tais como reciclagem de nutrientes, habitats para invertebrados e vertebrados ameaçados, além de fornecerem benefícios econômicos para os seres humanos através do lazer e recreação, com alto valor cultural e estético (Rumbold et al., 2001, Agardy et al., 2005, Kotwicki et al., 2005, McLachlan e Brown, 2006, Lucrezi et al., 2009, Krelling et al., 2017).

O Brasil não destoa dessa tendência de concentrar sua população nas áreas costeiras, tendo 50,7 milhões de habitantes distribuídos em 17 estados e 463 municípios costeiros, ou seja, 26,6% da sua população, segundo o último censo demográfico realizado (IBGE, 2011), sendo muito provável um acréscimo nesse percentual nos dias atuais. A grande demanda e presença de atividades antrópicas em ambientes costeiros resultam em conflitos entre o uso dessas regiões e a degradação do meio, trazendo prejuízos a oferta dos seus serviços ecossistêmicos (Yanes et al., 2019).

Ao falarmos de crescimento populacional, estamos falando também da inserção de consumidores nas cidades, portanto, no aumento do consumo de recursos naturais para produzir bens e mais geração de resíduos devido à mudança de hábitos alimentares, padrão de consumo e padrão de vida (Vergara e Tchobanoglous, 2012, Khan et al., 2016). A população urbana apresenta uma taxa rápida de crescimento em relação a população global (Ouda et al., 2016) e atualmente mais da metade da população reside em áreas urbanas, portanto, a geração de resíduos sólidos está intrinsicamente ligada com o crescimento populacional, urbanização e desenvolvimento econômico (Kumar e Samadder 2017).

Os resíduos sólidos são definidos como qualquer material, substância, objeto ou bem descartado que é resultante das mais diversas atividades antropogênicas, onde a destinação final deve estar nos estados sólido ou semissólido e cujas particularidades tornem inviável seu lançamento na rede pública de esgotos ou em corpos d’água, ou ainda exijam para isso soluções técnicas ou economicamente inviáveis (Brasil, 2010). Frente ao tipo de atividade que geram tais resíduos, podemos classifica-los como residencial, comercial, institucional, construção e demolição, serviços municipais, centrais de tratamento, industrial e agrícola (Tchobanoglous e Kreith, 2002).

Em 2020, no Brasil foram geradas 82.477.300 toneladas de resíduos sólidos urbanos, correspondendo a média de 1,07 kg de resíduos/habitante/dia. Desse valor, aproximadamente 60% foi destinado adequadamente, enquanto um pouco mais de 30 milhões de toneladas foram descartadas em aterros ou lixões a céu aberto (ABRELPE, 2021). Matéria orgânica, papel, papelão, tecidos, plástico, borracha, vidro, madeira, metais ferrosos e não ferrosos são os componentes predominantes da composição física dos resíduos sólidos gerados no Brasil (Alfaia et al., 2017).

Descartado no ambiente de maneira inadequada, estes resíduos podem causar impactos negativos na saúde humana, uma vez que podem transmitir doenças, odor, riscos de incêndio, além de causar incômodo estético e perdas econômicas (Yeny e Yulinah, 2012). Além disso, são causadores de enchentes locais, além de contribuírem para a poluição e contaminação atmosférica e aquática, sendo assim considerado um dos poluentes locais mais deletérios (Oteng-Ababio et al. 2013).

O plástico, um dos componentes dos resíduos sólidos, é definido como polímero sintético proveniente principalmente do petróleo, matéria prima esta que tem 8% de sua produção destinada a esse material (Thompson et al., 2009a, Güven et al., 2017). A primeira sintetização de polímero sintético aconteceu no início do século XX, no entanto foi a partir da metade do século que a produção em massa de itens plásticos começou de fato (Thompson et al., 2009b). Desde então o plástico foi introduzido no nosso dia a dia, sendo utilizado nas mais diversas formas e aplicações, como nos transportes, telecomunicações, vestuário, calçado, materiais de embalagem que facilitam o transporte e acondicionamento de uma vasta gama de alimentos, bebidas e outros bens, trazendo avanços médicos e tecnológicos, economia de energia e inúmeros outros benefícios sociais (Andrady e Neal, 2009).

Tanta aplicabilidade se deve ao fato de os plásticos apresentarem características, como baixo custo de produção e transporte, durabilidade, leveza, ser resistente à corrosão, com altas propriedades de isolamento térmico e elétrico (Thompson et al., 2009a). Apesar dos benefícios, atualmente o plástico é encarado como um problema de escala global e precisa ser abordado tanto da perspectiva científica como da tecnológica e socioeconômica (Garcés-Ordónez et al., 2020). Pois, além da grande demanda e produção de plástico que já alcançou 359 milhões de toneladas (Plastic Europe, 2019), estas mesmas características fazem com que esses itens persistam e acumulem no meio ambiente quando descartados de maneira indevida (Barnes et al., 2009). E um dos grandes vilões são aqueles de uso relativamente curto e de uso único, como os descartáveis e sacolas plásticas, pois fazem parte de 40% de toda produção de plástico (Napper e Thompson, 2019).

A maior parte (80%) dos resíduos plásticos presentes nos oceanos e regiões costeiras são provenientes do que produzimos e descartamos no continente, enquanto o restante (20%) é proveniente dos resíduos gerados em atividades em alto mar (Réllan et al., 2023). As principais fontes de plástico para os oceanos são as atividades turísticas, esgoto industrial e doméstico, pesca comercial, aquicultura e acidentes durante o transporte (GESAMP, 2016). E uma vez presente nos ambientes, os plásticos são facilmente transportados pelas ações das ondas e ventos por longas distâncias (Van Sebille et al., 2020), sendo distribuído amplamente ao longo de coluna d'água (Tekman et al., 2020) e presente em todos os oceanos (e.g. Reed et al., 2018, Pabortsava e Lampitt, 2020, Egger et al., 2021, Pattiaratchi et al., 2021, Bergmann et al., 2022).

São principalmente nas áreas costeiras, nos grandes giros e fundos oceânicos que os resíduos plásticos tendem a se acumular (e.g. Rangel-Buitrago, et al., 2021, Egger et al., 2021, Yılmaz et al., 2022). Dependendo do nível de exposição, os plásticos degradam no ambiente de maneira mais lenta ou mais rápida através das ações de fatores químicos, físicos e biológicos, como por exemplo, abração física, fotodegradação através de luz UV, oxidação, hidrólise e biodegradação por espécies de bactérias, fungos e algas (Klein et al., 2018). As praias são os ambientes propícios para

Estes podem ser classificados de acordo com seu tamanho, sendo denominados como macroplásticos (> 25mm), mesoplásticos (entre 5 e 25mm), microplásticos (> 5mm) e nanoplásticos (> 1 μm) (Romeo et al., 2015, Lambert e Wagner, 2016).

Definição de resíduos sólidos

Resíduos sólidos e densidade demográfica

Problemática dos resíduos descartados inadequadamente

Principal componente dos resíduos sólidos – O Plástico

Definição de plástico

Produção global x produção nacional

Distribuição na coluna d'água e nos diferentes ecossistemas

Impactos causados na biota (emaranhamento, ingestão, bioinvasão...)

2. OBJETIVOS

3. MATERIAL E MÉTODOS

4. REFERÊNCIAS

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5. CAPÍTULO I

O capítulo subsequente está formatado de acordo com as normas e publicado no periódico *Marine Pollution Bulletin*

Disponível em: https://www.elsevier.com/journals/marine-pollution-bulletin/0025-326X/guide-for-authors

Anthropogenic litter on Brazilian beaches: Baseline, trends and recommendations for future approaches

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ABSTRACT

Beaches are fundamental habitats that regulate the functioning of several coastal processes and key areas contributing to national and local budgets. In this study we provide the first large-scale systematic survey of anthropogenic litter on Brazilian beaches, covering a total of 35 degrees of latitude, recording the litter type, its use and size. Plastic comprised the most abundant litter type, followed by cigarette butts and paper. Small pieces (< 5 cm) were dominant among litter size-classes and food-related use was associated to most litter recorded types. Generalized additive models showed that proximity to estuarine run-offs was the main driver to beach litter accumulation, reinforcing river drainages as the primary route of litter coastal pollution. Also, the Clean-Coast Index evidenced there was not a pattern of beach litter pollution among regions, which denotes that actions regarding marine pollution must be taken by all state governances of the country.

Keywords: Beach debris, Plastic pollution, Marine pollution, Atlantic Ocean

Baseline

The idea that plastics would become one of the principal environmental problems of the 21st century is not new (Coleman and Wehle, 1984; Bergmann et al., 2015). It is already known that plastics are ubiquitous in the marine environment as they have been found in the most diverse habitats, from deepest oceans to intertidal areas (Mathalon and Hill, 2014; Chiba et al., 2018); and are pervading marine food chains, from tiny plankton communities to large shark predators (Sun et al., 2017; Barreto et al., 2019). Within this scenario, some habitats act as sinks for marine litter pollution. In oceanic waters, denser items tend to accumulate on the seabed (Woodall et al., 2014). Beaches on islands may also act as sinks for drifting litter in regions close to oceanic gyres (Lavers and Bond, 2017; Andrades et al., 2018b; Thiel et al., 2018), while nearshore habitats, such as mangroves and beaches, may accumulate floating litter (Munari et al., 2017; Martin et al., 2019).

Beaches represent an important component of human society contributing to local and national economies through tourism and recreational activities (Silva et al., 2013), as well as providing ecological services such as erosion control and nutrient recycling, and habitats for commercial and threatened species (Schlacher et al., 2007; Defeo and McLachlan, 2018). The presence of litter on beaches can impact its natural features, as well as affect the local fauna and alter ecological processes, which can induce shifts in nutrient cycling across food chains (Provencher et al., 2018). In addition to ecological impacts, marine litter reduce the cultural and economic value of beaches (Domínguez- Tejo et al., 2018; Rangel-Buitrago et al., 2018b), which may have knock-on effects on the economy at local and regional scales, often of the order of millions of dollars (Krelling et al., 2017).

Beaches are key habitats to evaluate human-induced impacts on marine environments due to their accessibility, typically intense human pressure and also their ecological importance. Worldwide, studies involving beach litter has helped researchers to document trends of debris pollution in the oceans and serve as a good proxy to predict the impact of plastics on marine wildlife (Ribic et al., 2010; Schuyler et al., 2012; Schulz et al., 2015; Santos et al., 2016). A broader perspective, that aims to compare patterns of beach littering on regional or continental scales, is nevertheless hampered by a lack of systematic surveys and high variability in methodological approaches. This has led researchers to seek the assistance of NGOs and non-academic volunteers for major monitoring efforts (Hidalgo-Ruz and Thiel, 2013; Bergmann et al., 2017) once they contribute to the development of strategies to reduce the plastic problem (e.g., collaborative campaigns and actions).

Brazil has one of the world's most extensive coastlines (~8000 km), which encompasses a variety of ecosystems, including mangroves, saltmarshes, reefs and beaches. A recent analysis, based on geology, coastal processes and beach types, divided the Brazilian coast into seven regions, which are influenced primarily by the Amazon River Delta, tidedominated and wave-dominated processes (Short and Klein, 2016). Other studies have provided similar classifications based on coastal habitats, invertebrates and fish (Leão and Dominguez, 2000; Barroso et al., 2016; Andrades et al., 2018a). Lastly, it is imperative to consider as important oceanographic drivers of litter carrying and accumulation the surface currents flowing over the Brazilian continental shelf, including the North Brazil current (Fratantoni and Richardson, 2006), the Brazil current and the Brazilian Coast current, and on the outer shelf, the Malvinas current (for details see Peterson and Stramma, 1991; Campos et al., 1996; Souza and Robinson, 2004).

In the present study, we provide the first large-scale assessment of beach litter pollution in the Brazilian coast, covering 35 degrees of latitude (2° N – 32° S). We used a systematic approach to compile data on the abundance, composition and size of the litter found on a selection of beaches representing the whole length of the Brazilian coastline. We also applied generalized additive models (GAM) to investigate the drivers associated with the accumulation of litter along the Brazilian coast.

The study focused on 44 beaches distributed along the Brazilian coast. Data on beach litter were obtained between August and December 2018 through the collection of litter along eight randomly located transects on each beach. The survey transects had a standard width of 4 m, and were conducted at low tide on a sandy portion of the beach, from the edge of the water to the supralittoral zone (beginning of vegetation or pavement). The total amount of litter (items of at least 0.5 cm in size) collected from each transect was counted and classified according to the type of material (plastic, glass, metal, porcelain, processed wood, paper, cloth, cigarette butt and charcoal) and size (the maximum dimension in 5 cm size classes, from 0.5–5 cm to>30 cm). This study was conducted with the participation of dozens of members and volunteers of the Meros do Brasil Project (Projeto Meros do Brasil, PMB, in Portuguese), an educational and scientific project that supports environmental actions to protect the threatened goliath grouper (*Epinephelus itajara*) and its coastal habitats on the Brazilian coast (http://www.merosdobrasil.org). We contacted specialists and members of the PMB at each study location, and with their help we trained volunteers to survey, collect, and classify beach litter, as outlined above. Data were uploaded from the participants' mobile telephones to a free website designed to store beach litter information.

The abundance of beach litter was used as a sample unit. We measured 23 predictive variables, and we used Pearson's correlation coefficient to identify possible collinearity between pairs of variables. The variables were selected for further analysis based on two criteria (collinearity of < 0.7 and their relevance for the description of occurrence of beach litter). Once the above selection method have been applied, seven predictive variables remained for the analyses: (i) distance from the beaches to the nearest estuary (measured against the prevailing coastal current); (ii) number of kiosks on the beach; (iii) number of rubbish bins on the beach; (iv) distance to the nearest major urban centre with>100 thousand inhabitants; (v) number of inhabitants of the nearest urban centre; (vi) the predominant direction of marine currents on the coast adjacent to the beach; (vii) the frequency with which the beach is cleaned.

Based on this, we elaborated three distinct groups of response variables. The first group was referred to the material type of beach litter, which included nine categories (charcoal, cigarette butts, processed wood, metal, paper, plastic, porcelain, cloth and glass). The second group referred to the original use of items, which included 14 categories (building, clothing, drink can, food, fishing, general use, house cleaning, medical waste, ornament, packaging, personal care, plastic bag, recreation and unidentified fragment). The third group was the size of items divided into seven size classes of maximum dimension (0.5–5 cm, 5–10 cm, 11–15 cm, 16–20 cm, 21–25 cm, 26–30 cm,>30 cm). We also verified the Pareto-type distribution of the amount of beach litter found (abundance) by its size, and extracted the angular coefficient from a linear model between litter abundance and size. Thus, the angular coefficient of this analysis represents the distribution of litter by size on each beach.

The response of predictive variables to type, original use and distribution of the litter was verified using a generalized additive models (GAMs) in R Software (R Core Team, 2019). We ran GAMs for each category of beach litter individually within the established groups (type, use and size classes) and one GAM for the Pareto-type distribution of litter abundance and size. We thus elaborated 31 GAMs, 9 for type, 14 for original use, 7 for size classes and one for the size-abundance relationships. In all the models, the predictive variables portray the role of additive factors, avoiding the insertion of interactive elements. The GAM's results were ranked in pre-established groups, using two metrics, the P value and the R^2 of the predictive variables in the models. The relationships were classified as ranging between low ($r^2 = 0$) to high ($r^2 = 1$) importance, while the sign of the GAM linear coefficient also indicated whether the relationship was positive or negative. Finally, we calculated the Clean-Coast Index (CCI) to determine the current status of each study beach in relation to cleanliness (Alkalay et al., 2007).

The 44 studied beaches varied considerably in the frequency of cleaning, infrastructure and other characteristics (Table 1). Altogether, 17,000 items were collected, with > 10% of the total (1757) being collected on a single beach, Baía de Tamandaré, in Pernambuco state. The next largest number of items (1273) was collected on Marudá beach, in Pará state. In contrast, the smallest number of items (21) was collected at Ponta de Nossa Senhora, in Bahia state, followed by Enseada das Garças, in Espírito Santo, with only 27 items. The mean density of items on the studied beaches was 0.42 (±0.53) items/m^2, with the highest densities being recorded at Baía de Tamandaré (2.74 ± 6.53 items/m^2), Praia da Costa, in Espírito Santo state (1.52 ± 3.56 items/m^2) and Calhau, Maranhão state (1.52 ± 3.72 items/m^2). Plastic was the most abundant material in 97.7% of the sampled beaches, with a total of 11,812 plastic pieces collected overall, followed by cigarette butts (1841 items) and paper (783 items). Despite the enormous diversity of materials found on the beaches, plastic, cigarette butts and paper made up>85% of the items collected on most beaches (Fig. 1). A similar predominance of plastic items has been recorded in studies on beaches in Europe (Asensio-Montesinos et al., 2019), Caribbean (Rangel-Buitrago et al., 2018a), North America (Moore et al., 2001; Wessel et al., 2019), Africa (Ryan et al., 2018), Asia (Zhou et al., 2011; Thushari et al., 2017) and Oceania (Hardesty et al., 2017). Plastic waste also predominates in other marine environments, including the open ocean and the seabed (Pham et al., 2014; Galgani et al., 2015). In summary, there is a predominance of plastics among marine litter in Brazilian coast as well as other parts of the world, reflecting the wide use of plastics in the human society.

Table 1

Studied beaches listed from North to South with beach code, location (state and municipality), number of inhabitants of the municipality, and beach characteristics.

Beach Number	Beach code	Beach	State	City	Number of inhabitants	Type beach	Number of kiosks	Number of beach stalls	Number of rubbish bins	Marine current	Cleaning frequency
1	GOI	Goiabal	Amapá	Calçoene	9000	Estuarine	5	0	0	CNB	Biannual
2	ORL	Orla Macapá	Amapá	Macapá	398,204	Urban	15	10	3	CNB	Quarterly
3	COR	Corvina	Pará	Salinópolis	37,421	Estuarine	0	0	0	CNB	No cleaning
4	MAR	Marudá	Pará	Marapanim	26,605	Estuarine	15	40	4	CNB	Weekly
5	PES	Pesqueiro	Pará	Soure	23,001	Estuarine	0	30	0	CNB	Weekly
6	MAT	Mata Fome	Pará	Soure	23,001	Estuarine	5	0	0	CNB	No cleaning
7	GRA	Praia Grande	Pará	Salvaterra	20,183	Urban	15	30	5	CNB	Weekly
8	AJU	Ajuruteua	Pará	Bragança	113,227	Estuarine	30	50	0	CNB	Weekly
9	JOA	Joanes	Pará	Salvaterra	20,183	Urban	0	20	0	CNB	Weekly
10	MOS	Mosqueiro	Pará	Belém	1393.399	Non-urban	10	20	0	CNB	No cleaning
11	ICO	Icoaraci	Pará	Belém	1393.399	Urban	20	0	0	CNB	No

											cleaning
12	CAR	Carimã	Maranhão	Raposa	26,327	Non-urban	0	0	0	CNB	No cleaning
13	CAL	Calhau	Maranhão	São Luís	1014.837	Urban	30	50	30	CNB	Daily
14	PON	Ponta D’areia	Maranhão	São Luís	1014.837	Urban	5	0	0	CNB	Weekly
15	BOA	Boa Viagem	Maranhão	São José de Ribamar	163,045	Estuarine	0	0	0	CNB	Weekly
16	PRI	Praia Principal	Ceará	Jijoca de Jericoacoara	17,002	Urban	10	20	0	CNB	Daily
17	PDC	Praia dos Carneiros	Pernambuco	Tamandaré	20,715	Estuarine	0	0	0	CB	Daily
18	BTA	Baía de Tamandaré	Pernambuco	Tamandaré	20,715	Urban	30	50	3	CB	Biannual
19	BDB	Boca da Barra	Pernambuco	Tamandaré	20,715	Non-urban	0	0	0	CB	No cleaning
20	RID	Riacho Doce	Alagoas	Maceió	932,748	Estuarine	5	20	10	CB	Weekly
21	GUA	Guaxuma	Alagoas	Maceió	932,748	Non-urban	5	30	10	CB	Weekly
22	PVE	Ponta Verde	Alagoas	Maceió	932,748	Urban	30	50	30	CB	Daily

23	PEB	Pontal do Peba	Alagoas	Piaçabuçu	17,203	Estuarine	0	0	0	CB	Biannual
24	SIR	Barra de Siribinha	Bahia	Conde	23,620	Estuarine	10	10	2	CB	Monthly
25	POC	Poças	Bahia	Conde	23,620	Non-urban	5	0	0	CB	No cleaning
26	CON	Praia do Conde	Bahia	Conde	23,620	Urban	15	10	13	CB	Annual
27	PRA	Prainha	Bahia	Candeias	83,158	Estuarine	10	10	1	CB	Monthly
28	PNS	Ponta de Nossa Senhora	Bahia	Salvador	2.675.656	Non-urban	5	40	20	CB	Daily
29	BAR	Porto da Barra	Bahia	Salvador	2.675.656	Urban	5	50	20	CB	Daily
30	REG	Regência	Espírito Santo	Linhares	141,306	Estuarine	0	0	0	CB	Annual
31	EGA	Enseada das Garças	Espírito Santo	Fundão	17,025	Non-urban	0	0	0	CB	No cleaning
32	PCO	Praia da Costa	Espírito Santo	Vila Velha	414,586	Urban	30	50	30	CB	Daily
33	PUR	Puruba	São Paulo	Ubatuba	78,801	Non-urban	0	0	0	CB	No cleaning

34	PIC	Picinguaba	São Paulo	Ubatuba	78,801	Estuarine	0	10	0	CCB	No cleaning
35	PER	Perequê-Açu	São Paulo	Ubatuba	78,801	Urban	15	10	30	CCB	Weekly
36	PVP	Praia Vila dos Pescadores	Paraná	Guaraqueçaba	7871	Estuarine	5	0	0	CCB	No cleaning
37	PDG	Praia de Guarapari	Paraná	Pontal do Paraná	20,920	Non-urban	0	0	0	CCB	No cleaning
38	LES	Praia de Leste	Paraná	Pontal do Paraná	20,920	Urban	0	0	2	CCB	Weekly
39	MDC	Morro dos Conventos	Santa Catarina	Araranguá	61,310	Estuarine	5	0	7	CCB	Biannual
40	MET	Praia da Meta	Santa Catarina	Balneário Arroio do Silva	9586	Urban	0	0	0	CCB	Daily
41	CAÇ	Caçamba	Santa Catarina	Balneário Arroio do Silva	9586	Non-urban	0	0	0	CCB	Daily
42	CA1	Cassino	Rio Grande do Sul	Rio Grande	197,228	Urban	5	0	1	CCB	Biannual
43	CA2	Cassino	Rio Grande	Rio Grande	197,228	Estuarine	5	0	1	CCB	Biannual

			do Sul								
44	CA3	Cassino	Rio Grande do Sul	Rio Grande	197,228	Non-urban	0	0	0	CCB	No cleaning

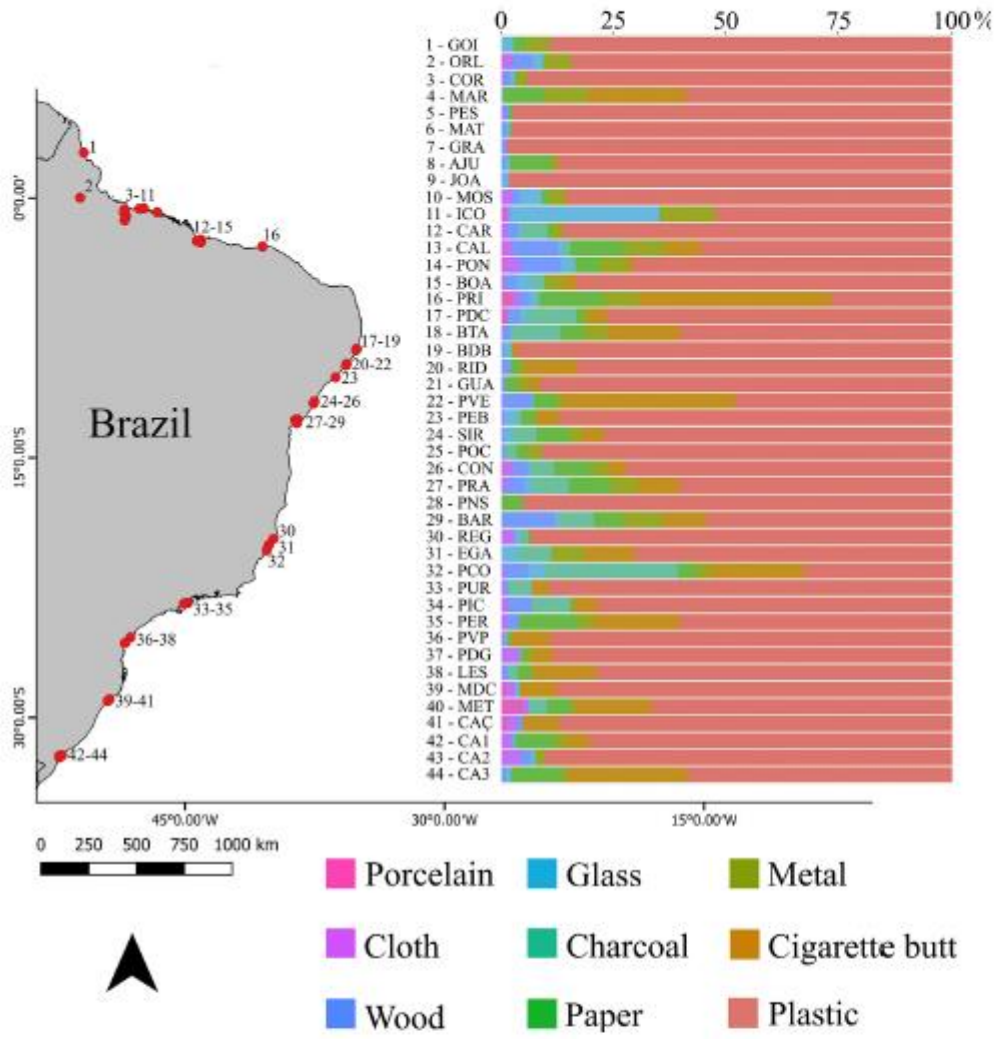

Fig. 1. Relative abundance (%) of different types of beach litter material along the Brazilian coast. Full beach names, numbers and acronyms are depicted in Table 1.

Food packaging was the most common use of the items found on the studied beaches, except for cigarette butts that were assigned to a single use category (Fig. 2). Pollution from food packaging is ubiquitous on Brazilian beaches, irrespective of the degree of urbanization (Andrades et al., 2016), and this kind of waste is also the most common type of plastic ingested by sea turtles in the Brazilian coast (Santos et al., 2015). In general, the majority of litter material found on Brazilian beaches can be linked to the ineffective waste disposal and management programs of most of the country's municipalities (Costa and Barletta, 2016; Barletta et al., 2019). The mean density of items tended to decrease with the increasing size, with the highest density (0.22 items/m2) being recorded for the smallest class of fragments (0.5–5 cm), followed by the 6–10 cm class (0.09 items/m^2), 11–15 cm (0.04 items/m^2), 16–20 (0.03

items/m²), 21–25 cm and>30 cm (0.02 items/m²), and lastly, the 26–30 cm and>30 cm classes (0.01 items/m²). The highest density of the smallest size class may be due to the constant fragmentation of larger pieces as well as for the difficult related to its removal by conventional methods of beach cleaning. However, disposing adequately small beach litter (< 5 cm) is more demanding than the larger due to the ineffectiveness of conventional methods (e.g., cleaning).

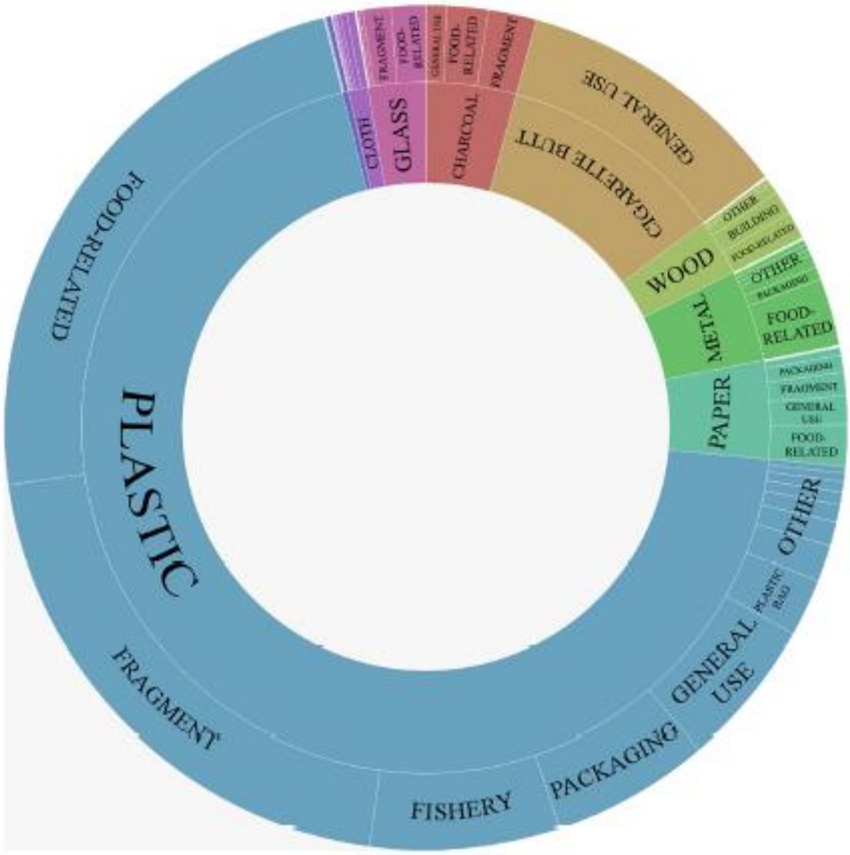

Fig. 2. Predominant types of representative beach litter materials (inner circle) and their most prevalent original use recorded in the Brazilian beaches (outer circle).

Our model indicated that the number of rubbish bins on the beaches may contribute to a reduction in beach litter, mainly for larger (> 6 cm) items. In the same way, beach-cleaning frequency contributed to a reduction in the abundance of certain types of litter. Overall, however, the distance of the nearest estuary appeared to be the most important driver of beach litter pollution, being negatively related to total litter abundance, and to the presence of the predominant types of material, i.e., plastic, cigarette butts and paper (Table 2). The input of estuaries was also correlated significantly with the abundance of smaller items, i.e., 0.5–10 cm (Table 2). Approximately two thirds of the Brazilian population lives near the coast, and most major cities and industries are located either on the coast or adjacent to major rivers (Lacerda et al., 2002; Marques et al., 2009). Given this, the contribution of river drainages that discharge into the sea to the amount of litter found on the studied beaches is understandable. Worldwide, more than two million tons of plastic litter is discharged into the sea each year by rivers (Lebreton et al., 2017). In Brazil, the Amazon estuary is estimated to drain>38 tons of plastic litter per year, which comprises the seventh most polluting river in the world (Lebreton et al., 2017). Our findings indicate that estuaries may have a greater influence on the accumulation of beach litter than urban predictors (i.e., the proximity and size of urban centres), which reinforces the role of estuarine zones as pollution sources of coastal seascapes in general, and not only in relation to plastics (Barletta et al., 2019).

Table 2

Results of the generalized additive models (GAM) expressed by r2. The negative values mean a negative correlation between the variable and each type, use, size litter and abundance-size relationship (AC = Angular Coefficient). The positive ones mean a positive correlation.

	Distance of the nearest estuary	Number of kiosks	Number of rubbish bins	Predominant direction of marine currents	Distance to the nearest major urban centre (> 100 thousand inhabitants)	Number of inhabitants of the nearest urban centre	Cleaning frequency
Type							
Charcoal	-0.19	0.20	-	-	-	-	-
Cigarette butt	-	0.15	-	-	-	-	-
Processed wood	-	0.15	-	-	-	-	-
Metal	-	0.16	-	-	-	-	0.09
Paper	-0.20	0.28	-	-	-	-	-
Plastic	-0.15	-	-0.17	-	-	-	0.20
Porcelain	-	-	-	0.13	-	-	-
Cloth	-	-	-	-	-0.26	-	-
Glass	-	0.21	-	-	-	-	-
Total abundance	-0.18	0.17	-	-	-	-	-

Use							
Food	-	-	-0.14	-	-	-	-
Fishing	-	-	-	0.36	-	-	-
Building	-	-	-	0.34	-	-	-
Ornament	-	-	-	-	-	-	-
Packaging	-	-	-	-	-	-	-
Unidentified fragments	-	-	-	-	-	-	-
Personal care	-	-	-0.11	0.47	-	-	-
Medical waste	-	-	-	-	-	-	-
House cleaning	-	-	-	0.37	-	0.16	-
Recreation	-0.11	-	-	0.32	-	-	-
Drink can	-	-	-0.19	-	-	-	-
Plastic bag	-	-	-0.27	-	-	-	-
Clothing	-0.07	-0.06	-	0.60	-	-	-
General use	-0.19	0.28	-	-	-	-	-
Size							
0–5 cm	-0.19	0.21	-	-	-	-	-
06–10 cm	-0.13	0.11	-0.24	-	-	-	-
11–15 cm	-	0.13	-0.21	-	-	-	-
16–20 cm	-	-	-0.26	-	-	-	-
21–25 cm	-	-	-	-	-	-	-

26–30 cm	-	-	-0.14	-	-	-	-
>30 cm	-	-	-0.12	0.27	-	-	-
Abundance-size AC	-	-0.22	-	-	-	-	-

Recent studies have demonstrated that the estuarine inputs were associated with a higher plastic intake by fishes (Dantas et al., 2012; Ferreira et al., 2019) and sea turtles (Santos et al., 2015). The litter found in the beaches is therefore an important proxy to evaluate the quantity and availability of plastic in coastal waters (Schuyler et al., 2012; Santos et al., 2016; Duncan et al., 2019). Since beach surveys are logistically easier and less expensive than board surveys, and data on this parameter can help stakeholders to implement suitable management efforts to reduce plastic pollution in the oceans. We expect this baseline will collaborate serving as starting point to future comparisons using the standard methodology presented here, as well as with the development of effective measures on a local scale for the mitigation of beach pollution. In fact, CCI did not identify any systematic geographic trend (region or state) in Brazil (Fig. 3), with five beaches in five different states being classified as Extremely Dirty (ED), six as Dirty (D), 11 as Moderate (M), 12 as Clean (C) and 10 as Very Clean (VC). Urbanized (BAR, CAL, PCO and BTA) and estuarine (MAR) beaches were among the most polluted. Among the cleanest beaches, an efficient public and local-mediated trade cleaning system, making it difficult to accumulate solid waste can justify the smallest density of litter. Notably, the PNS beach keep a rigorous daily clean process due to the Blue Flag certification (blueflag.global.com) so that a series of stringent environmental, educational, safety, and accessibility criteria must met and maintained.

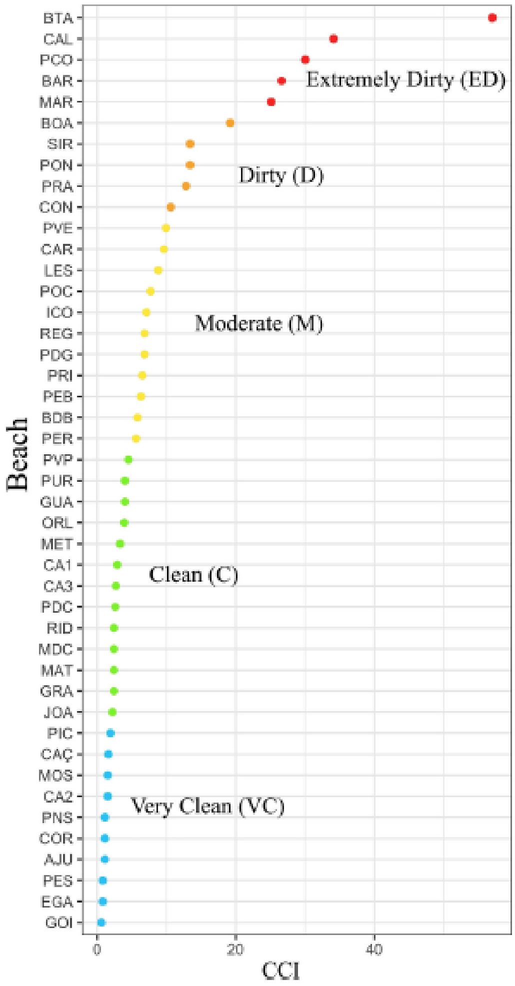

Fig. 3. Mean values of Clean-Coast Index (CCI) recorded for the study beaches on the Brazilian coast classified by cleanliness category (colour-coded).

Local authorities must focus on appropriate measures to solve the specific sources of pollution on the most polluted beaches (ED and D). One important complementary strategy is to enhance local public awareness on the importance of a clean beach environment, and the potential deleterious effects that pollution may pose to marine wildlife. The continuous monitoring of beaches using the CCI will also determine whether cleanliness is increasing or decreasing, indicating whether the measures implemented are appropriate or ineffective (Alkalay et al., 2007). We are aware that the terminology 'Clean' and 'Very Clean' for beaches with small amounts of litter is not the appropriate nomenclature per se since these beaches also recorded litter, which means that may be less polluted than others but still polluted too.

Large-scale beach litter assessments are rare and laborious, but nevertheless offer the opportunity of a broader perspective on key issues. Here we provide the first large-scale assessment of Brazilian beach litter pollution based on a systematic survey approach. We would recommend the long-term maintenance of this monitoring for a more reliable evaluation of the process and of the evolution of this problem on the Brazilian coast. We would also encourage further, complementary research, such as the assessment of the microplastics in beach sediments, to determine whether the drivers observed in the present study also apply to this group of plastic waste.

Author contributions section

R.A. and T.G. conceived and planned the study and sampling idea; All authors supervised or performed the field sampling; R.A., T.P., B.G. and T.G. analysed the data; R.A., T.P. and T.G. wrote the paper with inputs from all authors.

Declaration of competing interest

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

Acknowledgments

We would like to thank all volunteers that contributed to this work (Projeto Meros do Brasil and ICMBio) and the participation of local community residents. VVK express special thanks to Otávio M. Sousa. JLSN thanks to FAPEMA by Productivity Scholarship. Projeto Meros do Brasil is sponsored by Petrobras through the Programa Petrobras Socioambiental.

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6. CAPÍTULO II

O capítulo subsequente está formatado de acordo com as normas e será submetido para o periódico *Science of the Total Environment*
Disponível em https://www.elsevier.com/journals/science-of-the-total-environment/0048-9697/guide-for-authors

Meso and microplastic on Brazilian beaches: large-scale composition, distribution patterns and predictor variables

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ABSTRACT

Plastic pollution is a worldwide problem as climatic changes and exotic species. Due to its proprieties, plastic litter are present in a variety of ecosystems, mainly in aquatic ones, where it accumulates and persists. Plastic litter of 22 sandy beaches along Brazilian coast were assessed by a standardized protocol, where surface sediments were sampled. Meso and microplastics abundance, size, color, type and polymeric composition were quantified, as well as distribution patterns. General Linear Model (GLM) were run to investigate how the predictor variables drove the total concentration and each type of plastic litter. Overall, 3,114 plastic items were found in beach sediments and microplastics comprised more than a half of the total items (54%). Regarding colors and types, white (60%) and blue (13%); Styrofoam (45%) and fragments (39%) were the most common, respectively. Polyethylene (40%) and Polypropylene (32%) composed the main observed polymers. The distribution of plastic litter along the Brazilian beaches is probably ruled by three predictive variables: estuary distance (-), tourism (+) and number of inhabitants in the nearest city (+). Thus, beaches near estuaries and cities with more than 10 thousand inhabitants, and with touristic activities tend to contribute more to meso and microplastics pollution. Is important know plastic's characteristics, as abundance, category, type and color, to understand the magnitude of plastic pollution in the ecosystems. Furthermore, identify distribution patterns is essential/useful to assess source areas and large-scale impacts, assisting thus effective mitigation actions.

1 Introduction

Globally, plastic production reaches ~360 million tons, being single use plastics 50% of total production (PlasticEurope, 2019, PlasticOceans, 2020). Due to its lightweight, low cost and long-lasting properties, plastics are widely used in our daily life and, when improperly disposed, fragment and accumulate in ecosystems, mainly in marine environments impacting wildlife (Kako et al., 2014, Lebreton et al., 2019). Considering this, plastic pollution is a worldwide problem as concerning as climate changes and exotic species (UNEP, 2014), being ubiquitous in the aquatic environments from rivers to deep ocean basins (Chiba et al., 2018, Singh et al., 2021).

Plastics are defined as synthetic polymers derived mainly from petroleum (UNEP, 2018). A wide range of polymers and polymers mixtures exists in commercial production and 80% of them are composed by high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), polyvinyl chloride (PVC), polyurethane (PUR), polystyrene (PS) and polyethylene terephthalate (PET), then, not surprisingly these are the most found polymers in overall marine litter (GESAMP, 2019).

Additionally, plastic litter can be classified according to their size, measuring their largest dimension, as mega- (> 1000 mm), macro- (25 –1000 mm), meso- (1 – 25 mm) and microplastics (< 5 mm) (GESAMP, 2016). Microplastics, in its turn, can be also classified by origin, being primary microplastics those already synthesized with size < 5 mm to fulfil a function (e.g.: microbeads from cosmetics, resin pellets) and secondary microplastics the ones originated by wear and tear of larger objects (e.g.: fragments and synthetic textiles) (GESAMP, 2015).

By definition, microplastics are plastics between 1 µm and 5 mm of diameter (Frias and Nash, 2019) easily incorporated into food webs through ingestion by a wide range of aquatic organisms (Provencher et al., 2019, Setälä et al., 2018). Once ingested, microplastic can act as vector for toxic metals or organic pollutants often added during the manufacture of these polymers or adsorbed from the surrounding environment (Acosta-Coley et al., 2019, Pannetier et al., 2019). Also, coastal ecosystems, such as mangroves and beaches, acts as sinks for plastic litter (Lebreton et al., 2019, Martin et al., 2020), which are originated from anthropogenic activities developed both inland and in open ocean (Jambeck et al., 2015; Karthik et al., 2018). Once in beaches, the combination of high temperature, intense solar irradiation, and strong winds and waves makes sandy beaches ideal environments for degradation of large plastic items into small-sized pieces, therefore increasing microplastic pollution in the marine environment (Browne et al., 2007; Corcoran, 2021). Also, comparing macro- and microplastics litter, is harder track sources areas for the last one, mainly the secondary microplastics, due to their smaller pieces can not be linked to their specific application (Browne et al., 2015, Andrady, 2017).

So, to infer microplastics origins we need to explore their spatial pattern in relation to potential sources (e.g.: urban centers, shipping routes, fishing, recreational uses) (Ryan et al., 2018).The presence of microplastics in beach sediments can disrupt ecological processes provided by the beach meiofauna and macrofauna that are essential to the maintenance of the ecosystem services and goods to society (Browne et al., 2015; Tosetto et al., 2016; Carrasco et al., 2019). In addition, plastic pollution may reduce the cultural and economic values of beaches impairing local and regional economies as well (Krelling et al., 2017). Although microplastic pollution is widespread over sandy beaches of all Earth's coastal landmasses, as in Europe (Urban-Malinga et al., 2020), Asia (Chen and Chen, 2020), Oceania (Bridson et al., 2020), Africa (Vetrimurugan et al., 2020), North and South Americas (Dodson et al., 2020, De-La-Torre et al., 2020), and in remote areas (Kelly et al., 2020), estimates and comparisons in regional and global scales still precluded large-scale comparisons by the lack of standardized sampling and laboratorial procedures among studies (Löder and Gerdts, 2015). Similarly, plentiful studies concerning beach plastic litter were conducted in nations or regions with a continental-scale coastline (e.g., Castro et al., 2020; Maynard et al., 2021; Ryan et al., 2018), but varying in sampling, extraction and determination methods, which hampers to achieve a national picture of microplastic pollution to better assist society and federal management plans.

In this sense, the present work assessed the abundance, distribution, shape, color and size patterns, and polymeric composition of meso and microplastics along the Brazilian beaches applying a standardized protocol along the coast.

2 Material and methods

2.1 Study area

The Brazilian coast (Figure 1) presents a high variety of climatic, geomorphological, oceanographic and ecological characteristics, which encompasses a variety of intertidal ecosystems, including mangroves, reefs and sandy beaches (Schaeffer-Novelli et al., 2000; Amaral et al., 2016; Andrades et al., 2018). Interfaced from land by the biodiversity-rich forests and large estuaries and from ocean by subtidal reef, soft-bottom and macrophyte (seagrass or algal beds) landscapes, Brazil has one of the world's great coastlines, extending approximately 9000 km between latitudes 4ºN and 34ºS, with 4000 km within open coast and bay beach systems (Short and Klein, 2016).

Brazil's coastline is dominated by semi-diurnal tides with the highest tidal amplitude of > 4m in the north, decreasing to less than 1m southwards (Dominguez, 2009). Also, currents flowing over the Brazilian continental shelf are identified as North Brazil Current, which meets the Amazon plume on the north coast, the Brazilian Current off the northeast flowing southwards until its confluence with the Brazil Coastal Current and the Malvinas Current in the south (Peterson and Stramma, 1991; Campos et al., 1996; Fratantoni and Richardson, 2006).

Figure 1: Samples sites along the Brazilian coast.

2.2 Sampling methods and processing

Between December 2017 and January 2018, a total of 22 sandy beaches along the Brazilian coast were surveyed to collect sediment samples. Randomly, six quadrat samples (20 x 20 cm) of each beach were grabbed at the high tide mark, consisting of the top sediment layer (3-5 cm depth), then approximately 1 kg of sediment per sample for each quadrat was collected, stored and taken to the laboratory prior to processing.

In the laboratory, sediment samples were stored in aluminum trays and dried in a stove at 60 ˚C and weighted in a digital scale (0.01 g) for the dry weight (g). A stereomicroscope (Opton Tim-2b) at 6.5× to 50× magnification was used for the visual separation of larger microplastics (0.1 - 5 mm) and mesoplastics (5-25 mm). Plastic particles were separated from the sediment using tweezers and placed in Petri dishes. After that they were counted, classified according to type (cigarette filter, filament, film, foam, fragment, pellet, rubber, silicone, Styrofoam, synthetic fabric) and color categories (black, blue, brown, grey, green, golden, orange, pink, purple, red, silver, transparent, white and yellow). Then were measured (diameter in longest dimension to 0.001mm precision) and photographed using a ZEISS SteREO Discovery V12 stereo microscope with the Zen software (blue edition, v2.0, Zeiss, Oberkochen, Germany). All analysis steps were conducted with caution to avoid cross contamination, with all involved using cotton coat and samples processing being performed in a restricted room under a fume hood and all materials cleaned before and after usage.

The predictor variables assigned were Number of inhabitants, Number of nearest city inhabitants, Human Development Index (HDI), Gross Domestic Product (GDP) and Tourism, obtained from the Instituto Brasileiro de Geografia e Estatística (IBGE) website (IBGE, 2010) (REF do site ou da plataforma); Nearest city distance, Estuary distance, Beach extension and Distance from petrochemical complex taken from Google Earth (Google, 2009) (REF) and Tidal range and River flow from Agência Nacional de Águas (ANA) website (SNIRH, 2018).

A sample of each meso and microplastic shape categories recorded in the study was randomly selected for polymer identification. The particles were analyzed by Attenuated Total Reflectance - Fourier transform infrared spectroscopy (ATR-FTIR), using a Cary 620-670 FTIR microscope equipped with an ATR Ge crystal, acquiring 128 scans with a spectral resolution of 8 cm^{-1}, in the spectral range 4000-450 cm^{-1}.

2.3 Data analysis

A generalized linear model (GLM) was fitted for each of the plastic particle types categories using the mean density of plastic litter of each location, with a Gaussian distribution. For each model, a stepwise forward procedure was used to determine which factors generate the most parsimonious models using the Akaike information criteria (AIC) as a measure of the goodness of fit (Akaike, 1974). Also, the adjusted r-squared coefficient was used to estimate the model’s performance for explaining the observed variance. The model residuals were checked for normality and homoscedasticity assumptions using Shapiro Wilk’s (Shapiro & Wilk 1965) and Breusch-Pagan's (Breusch & Pagan 1979) tests, respectively. The scatter plot of residual values and the predicted values were used to check for linearity. In addition, multicollinearity was checked using the variation inflation factor (VIF) (Kutner et al. 2004).

3 Results and Discussion

Overall, 3,114 plastic items were found in beach sediments, being the majority composed by microplastics (n=1682, 54%) compared to mesoplastics (n=1432, 46%). Regarding the size, plastics litter presented mean size of 6.5 mm (SD ± 1.5 mm) ranging from 0.19 mm to 24.6 mm. Separately, collected microplastics and mesoplastics recorded mean size 3.4 mm (SD ± 0.5 mm) ranging from 0.1 to 4.9 mm and 9.7 mm (SD ± 1.2 mm) ranging from 5 mm to 24.6 mm, respectively. Plastic particles occurred in all the 22 studied beaches, reinforcing their ubiquity in the environment, mainly in aquatic habitats (Maynard et al. 2021).

Ten types of plastic litter were found, being Styrofoam (n=1402, 45%) the most abundant regarding the total number of items, followed by fragments (n=1223, 39%), pellets (n=199, 6%), film (n=125, 4%), cigarette filter (n=65, 2%), filament (n=39, 1%), foam (n=46, 1%), rubber, silicone and synthetic fabric with less than 1% (Figure 2). The plastics presented a great diversity of colors, being white (60%) the most prevalent color, followed by blue (13%), green (7%), yellow (4%), transparent (3%), red (2%), pink (1%), purple (1%), orange (1%), and silver, brown, gray, black and golden with less than 1% (Figure 3). The dominance of Styrofoam in samples also were observed in other studies conducted both over beach and water litter (e.g., Lee et al. 2015, 2017; Cordova and Nurhati 2019), which highlight Styrofoam as one of the main pollutants among marine litter waste. In this sense, some policy initiatives are currently focusing in ban or reduce the use of Styrofoam pollution in the Caribbean countries, including regulations in importation, manufacturing, and commercialization of material (Clayton et al 2021). Although the Brazilian National Plan for Marine litter (Plano Nacional de Combate ao Lixo no Mar in Portuguese; MMA 2019) recognize that Styrofoam items are among the most abundant litter types in the Brazilian beaches, any specific action, target or regulation are planned to Styrofoam pollution. The mean density of plastic litter was 632.1 items/kg (SD ± 128 item/kg) ranging from 1.2 (± 2) to 71.9 (± 15.6) item/kg, which the latter lower and higher ranges represents the Maçarico/Pará and Iracema/Ceará beaches respectively.

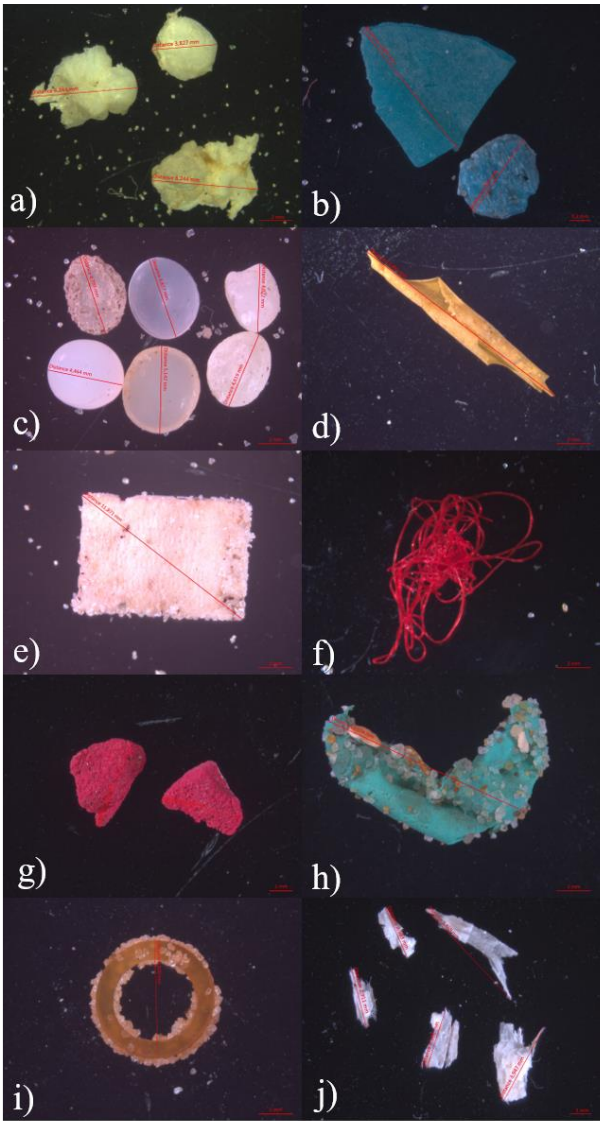

Figure 2: Categories of meso and microplastics found in beach sediments along the Brazilian coast. a) Styrofoam; b) Fragment; c) Pellet; d) Film; e) Cigarret filter; f) Filament; g) Foam; h) Rubber; i) Silicone and j) Synthetic fabric.

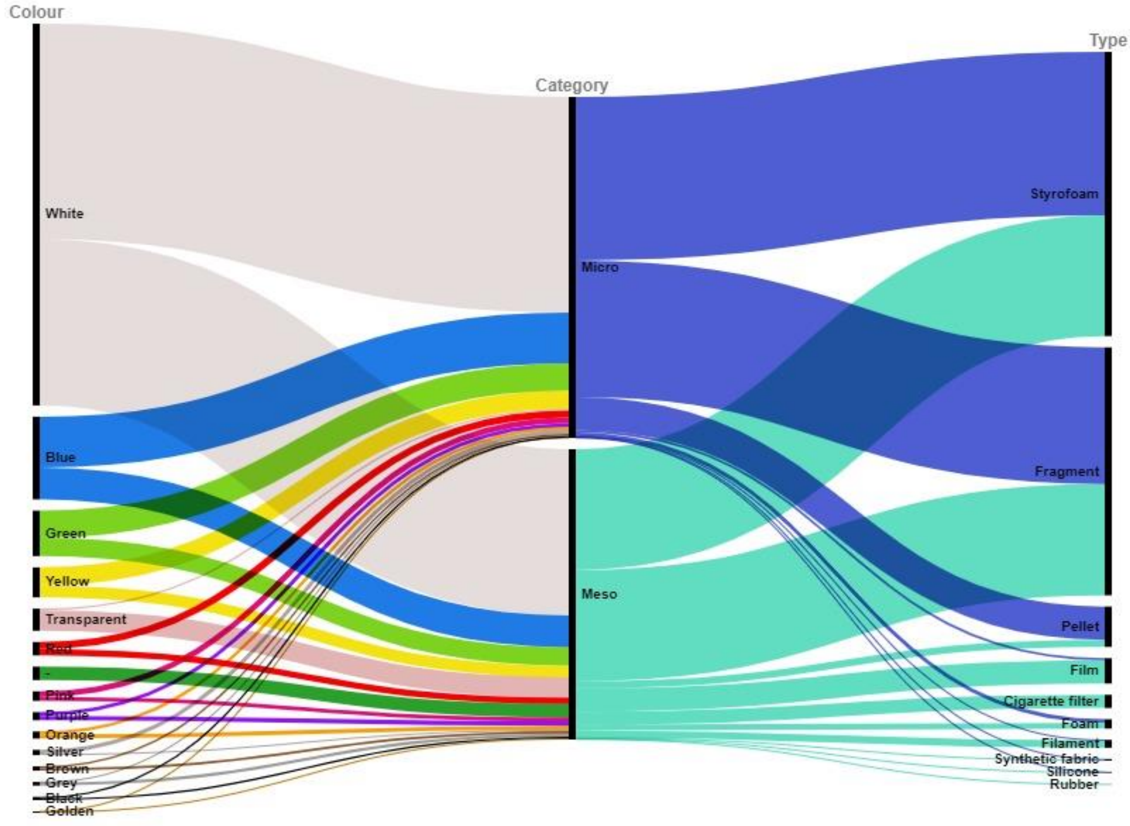

Figure 3: Proportions of plastic litter colors, size, and shape categories.

For polymer identification, 151 particles from at least one type those we previously identify as plastic were selected. Of them, 127 were in fact plastic polymers according to ATR-FTIR analysis, while the remaining were composed by silicates, carbonates and sugar gums. A total of 12 polymers were identified (Figure 4) and Polyethylene (PE) (40%) was the predominant one, followed by Polypropylene (PP) (32%), Acrylonitrile butadiene styrene (ABS) (7%), Ethylene vinyl acetate (EVA) (5%), Ethylene Propylene diene monomer rubber (EPDM) (3%), Epoxy resin (3%), Polyamide (PA) (2%), Polyurethane (PU) (2%), Polyethylene and Polypropylene blend (2%), Polyvinyl acetate (PVAc) (1%) and Polyethylene and Polyamide blend (1%).

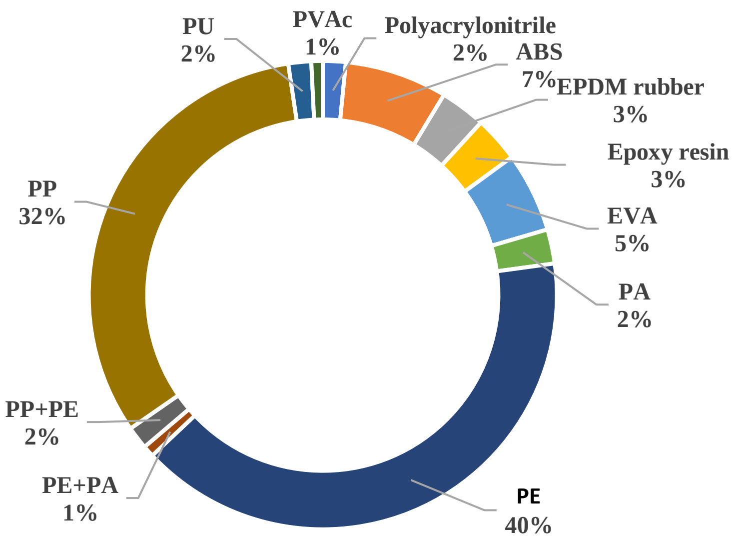

Figure 4: Polymer prevalence identified by ATR-FTIR spectroscopy from Brazilian coastal beach sediments. ABS: Acrylonitrile Butadiene Styrene; EPDM: Ethylene Propylene Diene Monomer rubber; Epoxy resin; EVA: Ethylene Vinyl Acetate; PA: Polyamide; PE: Polyethylene; PP: Polypropylene; PU: Polyurethane; PVAc: Polyvinyl Acetate; PE-PP: Polyethylene and Polypropylene blend; PE-PA: Polyethylene and Polyamide blend.

Regarding the GLM results (Figure 5), four (foam, rubber, silicone, synthetic fabric) of the ten plastic types did not generate a model due to the low abundance occurring just in a few sample sites (Figure 6). The concentration of all types (total) along the Brazilian beaches is probably ruled by three predictive variables (symbols within parentheses denotes positive or negative relations between variables and litter density): estuary distance (-), tourism (+) and number of inhabitants in the nearest city (+). Thus, beaches near estuaries and cities with more than 10 thousand inhabitants, and with touristic activities tend to contribute more to meso and microplastics pollution. Similarly, urbanized and tourism beaches usually are more polluted than rural beaches (Rios-Mendoza et al. 2021). Short distances to estuarine run-offs seem to influence the accumulation of the main types of microplastic (filament, fragment and Styrofoam), as well the overall microplastic pollution pattern in our study (see Figure 5). In tropical nations that holds great freshwater basins such as Brazil, estuarine input into coastal environments can be considered a major driver to coastal macro, meso and microplastic pollution (Andrades et al. 2020; this study), with local tourism and demography/urbanization (population and nearby city) also acting as litter pollution drivers.

Besides estuarine distance, tourism and demography/urbanization seem be related to distribution and accumulation of singular types of microplastic, as well tidal range (+) to filaments, Human Development Index (HDI = IDH in Portuguese) (+) to pellets and river flow (+) to fragments (Fig. 5). Filaments in general is one of the most types of plastics found in coastal environments (Alomar et al., 2016), and often are classified as lines (fishing materials) and textiles fibers, with the main sources ascribed to fishing activities and municipal wastewater drainage, respectively (Li et al., 2016; Cesa et al., 2019; GESAMP, 2019). However, in the present study, we did not separated filaments in these two categories once the majority seems came from fragmentation of ropes, lines and nets (Figure 2f).

Pellet pollution, though not abundant in our study, concentrated mainly in richer regions and was less associated to touristic activities. Plastic resin pellets are the raw material for any plastic objects present in our daily life (Andrady, 2011) and the low abundance in our study may be result of our method since pellet particles often can accumulate below the sediment top-layer (10 cm) (Turra et al. 2014). A study in the Guanabara Bay, Brazil, shows pellets concentrating mainly in beaches, in spite of other marine different matrices (water and bottom sediments) (Castro et al., 2020). Also, Carvalho and Baptista Neto (2016) argued that the high concentrations of pellets in beaches of Guanabara Bay is related to the presence of more than 12 thousand industries, including oil refineries, ports and shipyards. This is in line with the inverse relation between tourism and concentration of pellets and positive relation with cities with a high HDI observed in the present study.

Regarding fragments of meso and microplastics, they were ruled by estuary distance (-), tourism (+), number of inhabitants in nearest city (+) and river flow (+). That is, fragments concentrations were higher in beaches near to estuaries and number of inhabitants in the nearest city. Also, fragments were positively related with tourism activities and with river flow. Fragments is commonly found in beaches (e.g.: Expósito et al., 2021; Rios-Mendoza et al., 2021), and as observed in results earlier, estuarine runoff remains as the main predictive variable contributing to accumulation of plastic in beaches. Tourism and adjacent high populated city probably contribute to concentration of fragments due to incorrect waste disposal and litter breakdown in the environment (De-La-Torre et al., 2020).

Lastly, Styrofoam were ruled by distance of nearest city (+), distance from the main estuary (-), distance of petrochemical pole (-), Human Development Index (-), Gross Domestic Product (GDP = PIB) (+) and tourism (-). As mentioned before, Styrofoam is an abundant component polluting marine habitats worldwide. In our study, the proximity of estuarine run-offs and cities were strongly related to the observed great accumulations of Styrofoam in beaches. In fact, Styrofoam fragments are easily transported and deposited along the shoreline by natural forces (wind, surface currents, and waves). In tropical regions, river discharges, mainly during rainy seasons, are responsible to high transportation and accumulation rates of light litter items such as Styrofoam in beaches and shallow waters in estuarine and coastal environments (Cordova and Nurhati 2019). Nevertheless, Styrofoam pollution in our study seems to be influenced by a myriad of variables in our model, which probably is result by their inherent high dispersal capability due to their relatively low-density feature. In this way, Styrofoam tends to accumulate in any beach regardless of the main driving variable and, with some exceptions, is virtually in all Brazilian shoreline habitats.

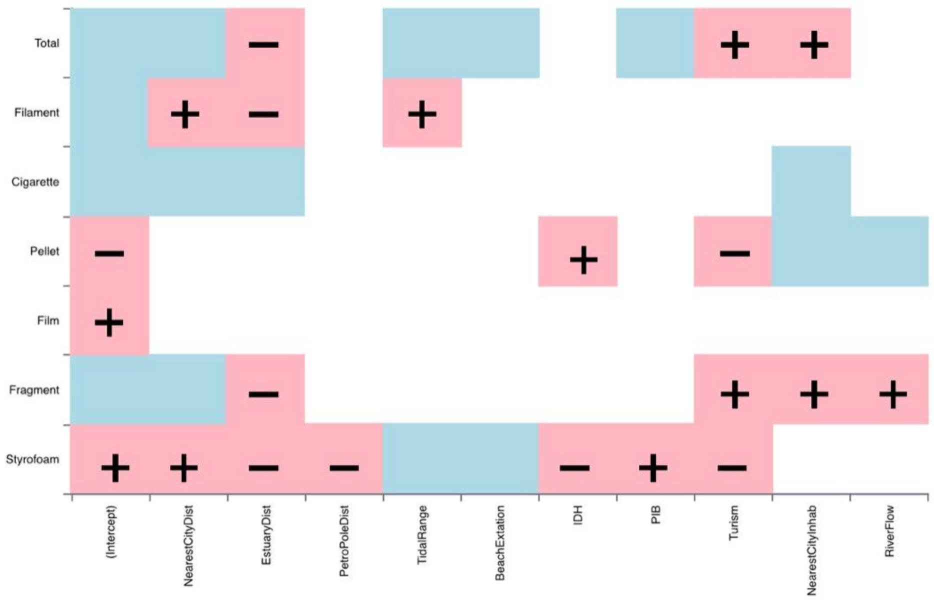

Figure 5: Generalized Linear Models (GLM's) of each predictor variable (NearestCityDist: Nearest city distance; EstuaryDist: Estuary distance; PetroPoleDist: Petrochemical pole distance; Tidal range; Beach extension; IDH: HDI – Human development index; PIB: GDP – Gross domestic product; Tourism; NearestCityInhab: Nearest city inhabitants; River flow) is driving total plastic litter concentration and each plastic type (Filament, Cigarret, Pellet, Film, Fragment and Styrofoam).

4 Conclusions

The present study reports the shape, color, size, concentration, and distribution patterns of meso and microplastics with standardized protocol applied in sandy beaches along the Brazilian coast. Plastic litter was found in all sampling sites, with microplastics comprising more than a half of collected items in comparison to mesoplastics. Styrofoam and fragments were the most common types of plastic with a great variety of colors and the mains polymers was represented by PE and PP. Regarding distribution, beaches near estuaries, near cities with high population density and that are linked with tourism activities presented positive correlation with beach plastic litter accumulations.

It is extremely important discuss on general features, as abundance, category, type and color, of plastic waste in environment to understand the magnitude of plastic pollution, mainly regarding its ubiquity. In addition, identifying plastic litter source areas and large-scale impacts in environment and biota are crucial to generate information on dispersal and potential harmful patterns of plastic pollution to enhance the chances to further achieve guided and effective mitigation actions. Still, standardized protocols are being a demand of scientists that are applying their efforts in knowledge about plastic pollution (Müller et al., 2019; Zantis et al., 2021; Grillo et al., 2022), for the purpose of comparison between studies results and a better understand of plastic dynamic in each different matrix.

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7. CAPÍTULO III

O capítulo subsequente está formatado de acordo com as normas e publicado no periódico *Marine Pollution Bulletin*

Disponível em: https://www.elsevier.com/journals/marine-pollution-bulletin/0025-326X/guide-for-authors

Ingestion of microplastics by *Hypanus guttatus* stingrays in the Western Atlantic Ocean (Brazilian Amazon Coast)

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ABSTRACT

The present study documents, for the first time, the ingestion of microplastics (MPs) by Longnose stingrays in the Western Atlantic Ocean. We examined 23 specimens of *Hypanus guttatus* from the Brazilian Amazon coast and found microplastic particles in the stomach contents of almost a third of the individuals. Fibers were the most frequent item (82%), blue was the most frequent color (47%) and Polyethylene Terephthalate (PET) was the most frequent polymer recorded (35%), as identified by 2D imaging - Fourier Transform Infrared (FTIR). The ingestion of microplastics by Longnose stingray has not been previously recorded. The findings of the present study thus provide an important baseline for future studies of microplastic ingestion by dasyatid rays and other batoid species in the Atlantic Ocean, and contribute to the broader understanding of the spatial and temporal dimensions of the growing problem of plastic pollution in aquatic ecosystems and organisms.

Keywords: Plastic pollution, Elasmobranchii, Longnose stingray, 2D FTIR imaging

Baseline

Microplastics (MPs) are now widely distributed in the environment, reaching even the remotest areas of the oceans, and infiltrating food webs worldwide (Germanov et al., 2019). These particles are potential carriers of persistent organic pollutants (POPs) and metals (Yu et al., 2019). Microplastics are normally defined as plastic particles with a maximum dimension of less than 5 mm (Arthur et al., 2009). These particles can be classified according to their origin as either primary or secondary MPs. Primary MPs are produced intentionally as micro-sized particles for use in cosmetics and a range of other industrial applications (Ogata et al., 2009), while secondary MPs are produced by the physical or chemical degradation of larger plastic waste by the environment (Cole et al., 2011; Godoy et al., 2019). Given their small size and abundance, MPs can be actively ingested by a wide range of organisms (Eriksen et al., 2014; Herrera et al., 2019), when the MPs are mistaken for prey, or passively, through the unintentional ingestion of the particles during normal feeding activities (Campbell et al., 2017; Desforges et al., 2015).

Despite the large number of studies that have focused on the ingestion of MPs by marine teleost fishes (e.g. Markic et al., 2018; Murphy et al., 2017; Pegado et al., 2018), few data are available on elasmobranchs, and most of which refer to sharks or pelagic rays (Alomar and Deudero, 2017; Anastasopoulou et al., 2013; Germanov et al., 2019; Valente et al., 2019). Up to now, only two reports have apparently been published on the ingestion of MPs by benthonic rays in marine environments; Neves et al. (2015) recorded MPs in specimens of Raja asterias, off the coast of Portugal and Pegado et al. (2018) that found MPs in an individual of *Narcine brasiliensis* from Amazon River estuary. However, both studies analyzed less than 10 individuals, which Markic et al. (2020) considered to be a suboptimal sample size for a reliable estimate of plastic ingestion rates.

Elasmobranchs are commercially important fishes, being consumed widely by some Latin American populations, from the Caribbean coast to northeastern Brazil (Feitosa et al., 2018; Rodrigues et al., 2020; Schmid et al., 2019). This suggests that the ingestion of microplastics by stingrays and sharks may eventually also affect human food safety and health (Van Cauwenberghe and Janssen, 2014). The Longnose stingray, *Hypanus guttatus* (Bloch and Schneider, 1801), a species of the family Dasyatidae, is an opportunistic, benthonic predator (Gianeti et al., 2019; Last et al., 2016), distributed from the southern Gulf of Mexico to southeastern Brazil (Bigelow and Schroeder, 1953; Rosa and Furtado, 2016). This species may reach up to 2 m in disc width and is very common as by-catch in the artisanal and industrial fisheries along the northern and northeastern coasts of Brazil (Rodrigues et al., 2020; Tagliafico et al., 2013). The present study investigated the presence of MPs in *H. guttatus* from the southern extreme of the Brazilian Amazonian coast. The study also provides an important baseline for future comparisons of the abundance, shape, and color of the microplastics found in the stomach contents of elasmobranch species.

The Maranhão Gulf is located at the southern extreme of the Brazilian Amazonian coast (Fig. 1) and is formed by the bay of São Marcos and São José, on either side of São Luís Island (Castro et al., 2018; Teixeira and Souza Filho, 2009). São Luís, the capital of Maranhão state, with its population of more than one million inhabitants, is located on this island (IBGE, 2010). This whole area forms an estuarine complex that covers an area of 5414 km2 (Souza Filho, 2005) and has an extreme semidiurnal macrotidal regime, with mean tidal amplitude of 3–7 m (Castro et al., 2018; Teixeira and Souza Filho, 2009). The local climate is tropical humid, with an annual precipitation of approximately 2300 mm (Fisch et al., 1998) and a mean temperature of 26 °C (Castro et al., 2018; Teixeira and Souza Filho, 2009).

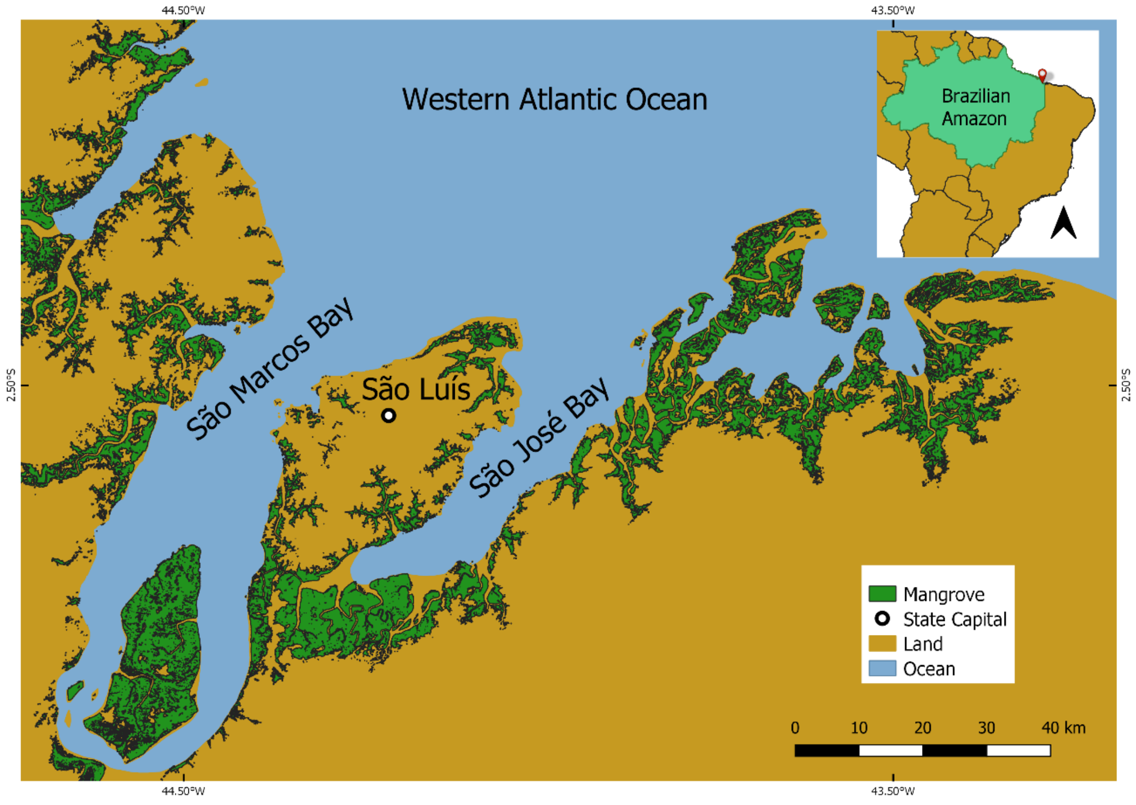

Fig. 1. Map of the Maranhão Gulf estuarine complex, located on southern extreme of the Brazilian Amazon coast in the Western Atlantic Ocean, where the Longnose stingray (*Hypanus guttatus*) individuals analyzed in this study were captured.

The 23 Longnose stingray specimens analyzed in the present study were obtained from local fishers and were captured by longlines and gillnets between August

2018 and March 2019. All individuals were immediately transported to the laboratory on ice in portable coolers. The length and width of the disc of each specimen were measured, and they were then eviscerated through a longitudinal incision in the abdominal area, using surgical forceps and a scalpel. The stomachs were removed carefully, and their contents placed in Petri dishes for analysis under a stereomicroscope (ZEISS Stemi DV4) at a magnification of 8× to 32×. All the MPs identified during this analysis were placed in Petri dishes containing distilled water, dried at 35 °C for 48 h, and then separated according to shape and color. All the material and equipment used during the laboratory processing were cleaned constantly and protected from possible external contamination. Therefore, sample processing (extraction and stomachs contents analysis) was executed under a laboratory fume hood, by personnel using natural fiber clothing and maintaining doors and windows closed. To guarantee the accuracy of the readings, a clean Petri dish was placed beside the stereomicroscope during the analysis of the stomach contents and inspected after the processing of the sample, to identify possible external contamination by MPs existing in the laboratory environment.

The findings of this analysis are presented here through descriptive statistics, including the mean, minimum, and maximum numbers of microplastic items, the percentages of the different categories of shape and color, as well as the polymeric composition of the particles, and the frequency of occurrence (FO%) of the microplastics found in the stomach contents. The FO% was calculated by: FO% = (Ni / N) × 100, where Ni = the number of stomachs that contained microplastic particles, and N = total number of stomachs examined.

Samples of each category of microplastic particle found in the gastrointestinal tracts of the stingrays were separated for 2D imaging-Fourier transform infrared (FTIR) analysis. The FTIR analysis was conducted directly on the dry filters (with no further processing), using a Cary 620–670 FTIR microscope, equipped with a 128 × 128 FPA detector (Agilent Technologies). The spectra were recorded directly on the surface of the samples (or of the Au background) in reflectance mode, with an open aperture and a spectral resolution of 8 cm^{-1}, with 128 scans being acquired for each spectrum. A “single-tile” analysis resulted in a map of 700 × 700 μm2 (128 × 128 pixels), with each imaging map having a spatial resolution of 5.5 μm (i.e., each pixel has an area of 5.5 × 5.5 μm^2).

The discs of the stingray specimens had a mean length of 52.3 (SD ± 8.68) cm, with a minimum of 32.4 cm and maximum of 72.0 cm, and a mean width of 54.6 (SD ±

10.0) cm, ranging from 34 cm to 83 cm (Table 1). Almost a third (FO% = 30.43%) of the samples contained microplastics, a value similar to that recorded in benthonic rays (43%) from the Portuguese coast (Neves et al., 2015). This relatively high incidence of MP ingestion may be related to the foraging strategy of the species (Romeo et al., 2015). The stingray *H. guttatus* is an important predator of benthic and benthopelagic coastal organisms, feeding on a wide range of prey. As a generalist top predator when adult, it seems likely that these individuals were susceptible to bioaccumulated microplastic contamination through the food chain, by passive ingestion (Gianeti et al., 2019).

Table 1

Biometrics of the Longnose stingray (*Hypanus guttatus*) specimens and the characteristics (shape, color, and type of polymer) of the microplastic particles (MPs) found in their stomach contents. The presence of MPs is expressed as the presence (1) or absence (0). The polymers are: ABS = Acrylonitrile Butadiene Styrene; PA = Polyamide; PE = Polyethylene; PET = Polyethylene Terephthalate; PP = Polypropylene; SBR = Styrene-Butadiene Rubber.

Stingray	Disc length (cm)	Disc width (cm)	Presence of MPs	Shape of the MPs	Colors of the MPs	Number of MPs	Polymer
1	55	58	0	-	-	0	-
2	55	59	0	-	-	0	-
3	56.5	60	1	Fiber	Transparent	6	PET, PP, PA
4	56	57.5	1	Fragment	Blue	2	ABS
5	51	54	0	-	-	0	-
6	56.5	54.5	0	-	-	0	-
7	51	56	0	-	-	0	-
8	57	61	0	-	-	0	-
9	57.5	58.5	1	Fiber	Red	1	Blend (PET + SBR)
10	72	73.5	0	-	-	0	-
11	41.5	41	1	Fiber	Blue	3	PET, PE
12	51.5	55	0	-	-	0	-

13	72	83	1	Fiber	Black	2	PA
14	52	55.5	0	-	-	0	-
15	52.5	56	0	-	-	0	-
16	45.5	48	1	Fragment	Blue	1	ABS
17	48.3	52	0	-	-	0	-
18	43.3	45.5	0	-	-	0	-
19	44.8	48.5	0	-	-	0	-
20	49	53	0	-	-	0	-
21	43	45	0	-	-	0	-
22	46	49	1	Fiber	Blue	2	PE
23	32.4	34	0	-	-	0	-

A total of 17 microplastic particles were found in the stomach contents of seven stingrays, with a mean of 2.4 (SD ± 1.7) particles per individual (N = 7 individuals), ranging from one to six particles in a given individual. The majority (82%) of the particles found in our study were classified as fibers and the other 18% as fragments, which were primarily blue (47%) or transparent (35.3%), with some black (11.8%) and red (5.9%) particles (Fig. 2).

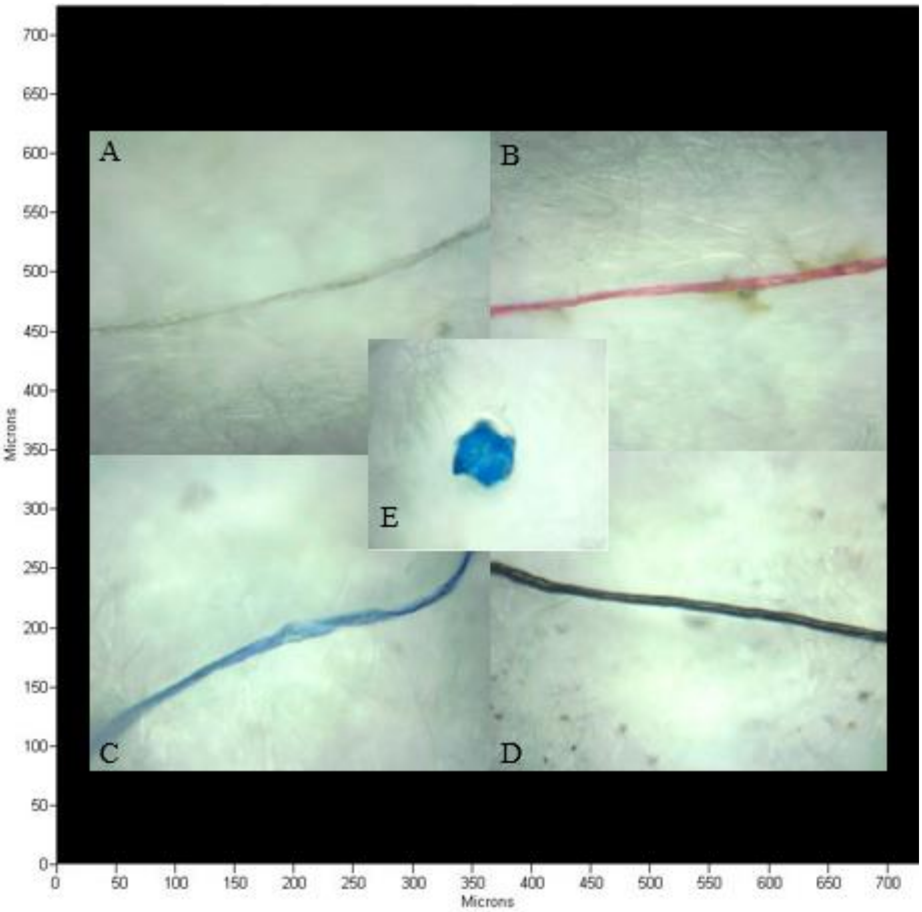

Fig. 2. Examples of the different categories of microplastic found in the stomach content of the Longnose stingray *Hypanus guttatus* specimens collected from the Gulf of Maranhão. A) Transparent Fiber; B) Red Fiber; C) Blue Fiber; D) Black Fiber; E Blue Fragment. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Neves et al. (2015) recorded a mean of only 0.5 (SD ± 0.8) particles per individual in *Raja asterias* and found only fibers in the stomach content of this ray. Many authors have found that fibers are the most abundant microplastic particles in marine environments (Alomar and Deudero, 2017; de Lucia et al., 2018, 2014; Neves et al., 2015; Rochman et al., 2015). Our findings further support that the marine biota, including benthic stingrays like H. guttatus, may be most exposed to microplastic fibers. The distribution of microplastics in the oceans may be influenced directly by anthropogenic processes (Barnes et al., 2009) and large amounts are found in aquatic environments near areas of urban development (Garcia et al., 2020). In Sao Luis, like many other largest cities in the Amazonian region, such as Manaus and Belém, due to the lack of environmental awareness and efficient waste management, more than 19% of the urban solid waste, including plastics, is not collected by municipalities and an unknown fraction of this mismanaged waste is washed into the Gulf of Maranhão (Giarrizzo et al., 2019).

Further, Maranhão is recognized as one of the most important states for artisanal fisheries in Brazil's northern and northeastern regions (Almeida and Isaac-Nahum, 2015). This potentially contributes to the high presence of filaments in the coastal and estuarine ecosystems, originated by the fragmentation of fishing gear (Soares et al., 2017). These particles are introduced into marine environments through ports and fisheries activity, wastewater treatment plants, urban runoff (Peters and Bratton, 2016), and river discharge (Woodall et al., 2014). Strong macro-tidal currents and other oceanographic phenomena (e.g. the permanent east-to-west prevailing winds) found in this region may contribute to the ample dispersal of microplastics through the known accumulating effects of enclosed or semi-enclosed bays within metropolitan urban areas (Auta et al., 2017).

Six types of polymer were identified in the microplastic particles analyzed by 2D FTIR Imaging in the present study (Fig. 3). The most frequent polymer was Polyethylene Terephthalate (PET; 35.3%), followed by Polyamide (PA), Acrylonitrile butadiene styrene (ABS), and Polyethylene (PE), each with a frequency of occurrence of 17.6%, and then Polypropylene (PP) and PET + SBR (Styrene Butadiene Rubber), both with a frequency of 5.9%. The predominance of PET is consistent with the fact that it is one of the polymers most produced by industries, worldwide, and thus more likely than others to be present in the marine environment (Andrady, 2011). This polymer is used in the production of textiles, including clothes, blankets, and fleeces, as well as bottles (Wang et al., 2017). Therefore, PET fibers are common in domestic wastewater, in particular from washing machines, which contaminates river basins and, eventually, oceans (Browne et al., 2011; Napper and Thompson, 2016). As a relatively dense polymer, PET is also more likely to sink to the bottom of aquatic environments, where it can be ingested by benthic organisms (GESAMP, 2015), including the Longnose stingray. The second most common polymers were PE and PA, which could come from the fishing gears, like nets and floats that are often have these polymers in their composition (GESAMP, 2016). Over time, however, lower-density polymers, such as PP and PE, may decompose and sink, and thus become available to a variety of benthic organisms (Long et al., 2015; Morét-Ferguson et al., 2010).

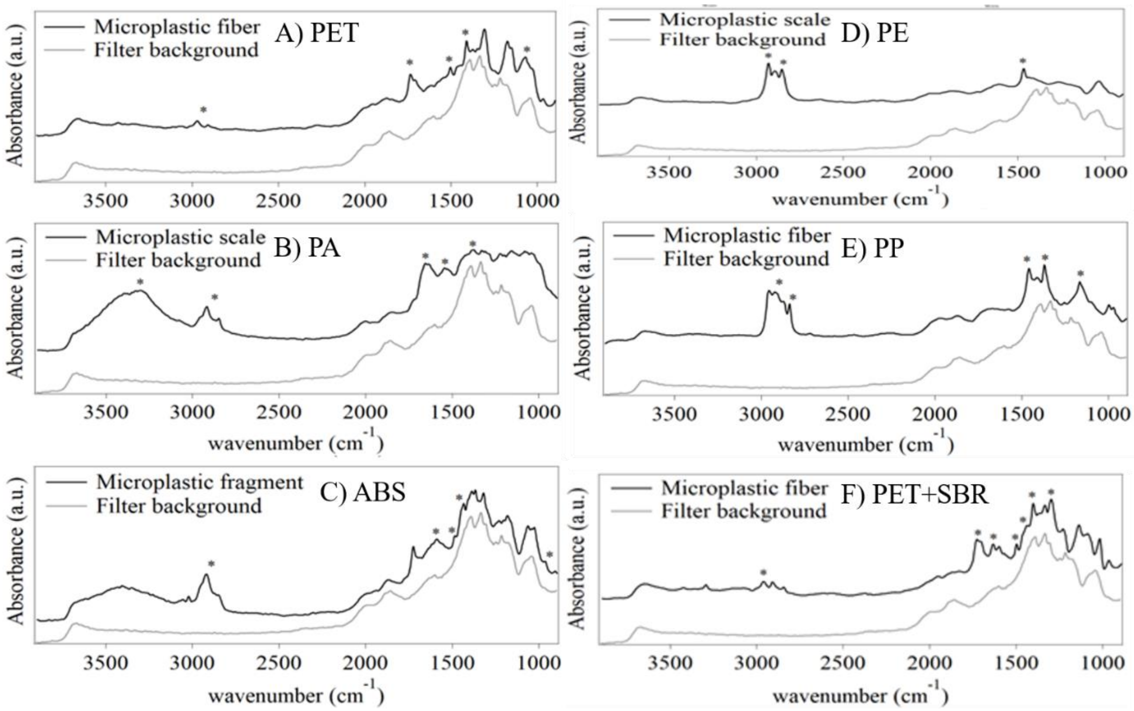

Fig. 3. Representative FTIR reflectance spectra acquired different microplastic polymers, collected from the stomach contents of the Longnose stingray *Hypanus guttatus* from the Maranhão Gulf, Brazil. A) PET: Polyethylene Terephthalate; B) PA: Polyamide; C) ABS: Acrylonitrile Butadiene Styrene; D) PE: Polyethylene; E) PP: Polypropylene; F) Blend of PET (Polyethylene Terephthalate), and SBR (Styrene butadiene rubber).

In the present study, microplastic particles were found in the stomach contents of almost one third of the analyzed *H. guttatus* specimens. This stingray species is an important target of the artisanal fisheries of Maranhão State, at the Latin America and in southern extreme of the Brazilian Amazon coast. Most of the particles were fibers, and the most frequent polymer was PET. With 23 specimens analyzed, the present study provides a more reliable estimate than the previous reports of microplastic ingestion by benthonic rays. Our study provides the first record of ingestion of MPs by *Hypanus guttatus* from the Western Atlantic Ocean, as well as an important database for further comparisons of the exposure of this elasmobranch group to plastic contaminants in the marine environment. Such investigations, specifically for understudied areas and species, are important contributions towards the understanding of spatial and temporal patterns of plastic pollution in aquatic ecosystems and organisms, as well as to support effective prevention and conservation efforts in response to this global problem.

CRediT authorship contribution statement

Tamyris Pegado: Methodology, Formal analysis, Writing – original draft, Visualization, Writing - review & editing. **Lucio Brabo:** Formal analysis, Writing - original draft, Visualization, Writing - review & editing. **Kurt Schmid:** Writing - original draft, Visualization, Writing - review & editing. **Francesco Sarti:** Formal analysis, Resources, Writing - review & editing. **Thaís T. Gava:** Formal analysis, Investigation, Writing - review & editing. **Jorge Nunes:** Conceptualization, Resources, Writing - review & editing. **David Chelazzi:** Methodology, Formal analysis, Resources, Writing - review & editing. **Alessandra Cincinelli:** Resources, Writing - review & editing. **Tommaso Giarrizzo:** Conceptualization, Methodology, Formal analysis, Writing - review & editing, Writing - original draft, Resources, Visualization, Supervision.

Declaration of competing interest

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

Acknowledgments

TP and LB would like to thank the Coordination for Higher Education Personnel Training (CAPES) and the Brazilian National Council for Scientific and Technological Development (CNPq) for financial support. JN received a productivity grant from FAPEMA (02106/18) and is funded by the project FAPEMA 06605/16. TG received a productivity grant from CNPq (#311078/2019-2). DC and AC gratefully acknowledge CSGI (Consorzio Interuniversitario per lo Sviluppo dei Sistemi a Grande Interfase - Center for Colloid and Surface Science) for financial support. Our thanks to the editor

Bruce J. Richardson and reviewers for the constructive comments and recommendations which improved quality of the paper.

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8. CONCLUSÕES GERAIS