



# Littered cigarette butts: links among environmental impacts, demography and market at the highly urbanized Brazilian cities

Ítalo Braga Castro  
Victor Vasques Ribeiro

**ACT**

Promoção da **Saúde**

**Authorship****Federal University of São Paulo**

Ítalo Braga Castro

Victor Vasques Ribeiro

**Collaboration****ACT Health Promotion**

Mariana Pinho

Mônica Andreis

**Institute for Global Tobacco Control (IGTC) at the Johns Hopkins Bloomberg School of Public Health**

Graziele Grilo

Kevin Welding

**National Cancer Institute**

André Szklo

**Funding statement**

This work was supported with an award from the Institute for Global Tobacco Control at the Johns Hopkins Bloomberg School of Public Health with funding from the Bloomberg Initiative to Reduce Tobacco Use

**Layout**

Ronieri Gomes

**Suggested Citation**

Castro, Ítalo Braga Castro; Ribeiro, Victor Vasques Ribeiro.

Discarded cigarette butts: relationships between environmental impacts, demography, and market in highly urbanized Brazilian cities. Rio de Janeiro: ACT Health Promotion, 2023

## Summary

1. Introduction .....	5
2. Methodology .....	9
2.1 Study area .....	9
2.1.1 Urban density levels .....	10
2.1.2 Income levels .....	11
2.1.3 Other urban aspects .....	11
2.2 Sampling procedures .....	12
2.3 Cigarette butt densities .....	13
2.4 Cigarette butt contaminants leakage estimative .....	14
2.5 Cigarette brands and companies .....	15
2.6 Illegal cigarette marketing .....	16
2.7 Cigarette butt experiments .....	16
3. Results and Discussion .....	18
3.1 Cigarette butt densities .....	18
3.1.1 Urban density levels .....	18
3.1.2 Income levels .....	20
3.1.3 Urban density and income levels .....	21
3.1.4 Other urban aspects .....	23
3.1.5 Worldwide cigarette butt densities .....	24

3.2 Cigarette butt contaminants leakage estimative .....	25
3.2.1 Urban density levels .....	26
3.2.1 Income levels .....	27
3.2.3 Urban density and income levels .....	28
3.2.4 Worldwide cigarette butt contaminants leakage .....	29
3.3 Brands of Cigarette butts .....	30
3.4 Illegal cigarette marketing (ICM) .....	34
3.4.1 Cigarette butts .....	34
3.4.2 Cigarette packs .....	37
3.4.3 Cigarette butts and packs viability for future assessments..	39
3.5 Cigarette butt experiments .....	42
4. Conclusions.....	46
References .....	48
Supplementary material .....	57

## Abstract

Annually, over 5.5 and 4.5 trillion cigarettes are globally produced and littered, respectively. Cigarette butts (CBs) are the main anthropogenic litter item, contaminating fresh and coastal waters, leaching contaminants and affecting biota. One CB may contain a complex mixture (>7,000 compounds, of which >150 are toxic), that can contaminate around 1,000 L of water by leaching several hazardous substances, such as toxic metals and polycyclic aromatic hydrocarbons (PHAs). Smokers are more likely to discard CBs than other litter types, but littered CBs occurrence in urban areas are poorly known. In addition, monitoring studies usually ignores the CBs brands, losing valuable information to enable possible reverse logistics and illegal cigarette marketing (ICM) analysis. Brazil has an important role on tobacco mitigation as the leading country on reducing tobacco prevalence in South America. Guarujá city (324,977 inhabitants) presents territorial spaces occupied by environmental protection areas (75%) and highly urbanized zones (25%), with main activities related to port complex and tourism. Guarujá has the 37th best urban cleaning status among 5,568 Brazilian cities. Despite this, our previous study has found high levels of CBs contamination in the 1st (Santos) and 2nd (Niterói) best ranked cities according to Brazilian urban cleaning status. The present study conducted in urban areas of Guarujá, aimed to determine the density of littered cigarette butts (DCBs) in walkways of different urban density zones and income, estimate the contaminants leakage, identify the CBs brands and the illegal cigarette marketing rate. In addition, levels of PHAs and toxic metals were assessed based on CBs leachates. A total of 4,321 CBs were found in 23,694 square meters from Guarujá, with a DCBs of  $0.18 \pm 0.17$  CBs.m<sup>-2</sup>. Considering the obtained DCBs in Santos, Niterói and Guarujá no correspondence with Brazilian ranking of urban cleaning status was seen, indicating that monitoring programs directed to CBs should be adop-

ted. Interestingly, the DCBs was positively related to the number of commercial buildings, stores selling cigarette packs and units, and benches, but not to the number of residential buildings, manholes, bus stops, garbage bins, and local income. The CBs contaminants leakage in Guarujá was overall severe (CBPI=15.4±11.5), which was higher in zones under low urban density. Thus, low urban density zones of Guarujá are the main contributors to CBs contaminants leakage. In Santos, these zones contributed less, while in Niterói, all zones of urban density contribute similarly. Niterói, and especially Santos and Guarujá presented alarming levels of CBs contaminants leakage. The main CBs brands found in Guarujá were Rothmans, Marlboro and Gift, which were similar to those observed in Santos. Based on identified brands, the ICM ranged from 21.7% to 36.7%, indicating that monitoring programs and control actions on illegal cigarette markets must be adopted. Further, DCBs and CBPI were more strongly related to low urban density, while ICM was influenced by the low level of income. Finally, highly toxic levels of polycyclic aromatic hydrocarbons and chemical elements were leached from CBs, indicating concerns about the ecological risks and potential health hazards posed by CBs as hazardous waste. Thus, it is crucial to address and mitigate the environmental contamination caused by CBs to safeguard ecosystems and human well-being.

**Keywords:** Brands; Contaminants; Illegal; Leakage; Walkways.

## 1. Introduction

Cigarette smoking is a fairly common social behavior. It was estimated that 1.14 billion people smoked 7.41 trillion of cigarettes in 2019 (GBD Tobacco, 2021). While smoking is linked to a range of health impacts such as cancer (Hecht and Hatsukami, 2022), stroke (Ikazabo et al., 2022), diabetes (Zeru et al., 2021), birth complications (Gutvirtz and Sheiner, 2022), heart (Sepand et al., 2021) and lung (Li et al., 2021) diseases, the entire production chain of cigarettes (manufacturing, distribution, and disposal) also poses several environmental threats (Araújo et al., 2022; Green et al., 2022). For instance, the manufacturing stage results in natural areas deforestation (Gomersall, 2022), while using harmful pesticides, increasing greenhouse gas emissions and causing labor health issues (Li, 2022; Malahayati and Masui, 2019). Furthermore, cigarettes also become a serious environmental issue after consumption, since littered cigarette butts (CBs) contaminate fresh and coastal waters, leaching contaminants, affecting especially aquatic organisms (Araújo et al., 2022; Torkashvand et al., 2020).

The 5.5 trillion cigarettes produced leads to approximately 4.5 trillion CBs inappropriately discarded annually (Torkashvand and Farzadkia, 2019). Even though cigarette filters might capture some toxic chemicals generated when smoking (Nitschke et al., 2023), CBs may contain over 7,000 compounds, with at least 150 being toxic (Araújo and Costa, 2019). Each CB may contaminate up to 1,000 L of water, releasing hazardous substances such as toxic metals, BTEX, polycyclic aromatic hydrocarbons (PHAs), nitrogenic compounds, aromatic amines, among others (Akhbarizadeh et al., 2021; Dobaradaran et al., 2021, 2019, 2018, 2017; Soleimani et al., 2022). Several studies have found substantial levels of PAHs and toxic elements in CBs littered in natural and urban settings (Ding et al., 2007; Lee et al., 2011; Shimazu, 2016; Vu et al., 2015). Many chemicals present in smoked CBs are persistent in environment,

bioaccumulate in living organisms besides induce mutagenic, carcinogenic, and teratogenic effects (Neira et al., 2017; Qiao et al., 2006; Rochman et al., 2013). Indeed, freshly discarded CBs leak hazardous substances into the environment affecting soils and natural waters (Dobaradaran et al., 2019). Thus, concerns are raised since CBs have emerged as the most common form of anthropogenic litter (Araújo et al., 2022), mainly due to incorrect disposal by smokers compared to other types of litter (de Granda-Orive et al., 2016). Nevertheless, the occurrence and distribution of CBs in urban areas and the associated environmental risks caused by toxic metals and PAHs remain poorly understood (Ribeiro et al., 2022a).

Littered CBs monitoring studies usually analyses the number of items collected by a predetermined area, while ignoring the CBs brands analysis (Araújo and Costa, 2021; Torkashvand et al., 2021) or hiding the brands names (Lima et al., 2021; Santos-Echeandía et al., 2021). However, there are several knowledge gains in assessing the littered CBs brands. For example, reverse logistics strategies may be adopted when cigarette brands and manufacturers are known (Brasil, 2010). Based on such information penalties could be applied to manufacturers saving millions of dollars in costs of collection and management of this waste (Granados et al., 2019; Ranjkesh and Nasouri, 2022). In addition, the identification of littered CBs brands is necessary to adopt reverse logistics actions, which are mandatory to enable integrative evaluations of environmental impacts, demography and market allowing costs estimates (Ribeiro et al., 2022a).

The illegal cigarette marketing (ICM) is one worrying issue related to the cigarette smoking, while tobacco industry amplify its real proportion seeking to exempting then self (Arevalo et al., 2016; Kurti et al., 2020; Nguyen and Nguyen, 2020; Scollo et al., 2014; Szklo et al., 2020; van der Zee et al., 2020). Sold at lower prices, illegal cigarettes are manufactured while avoiding taxes and/or are smu-

gged from countries with smaller taxes (Goodchild et al., 2022). Evaluations of ICM can be performed by using several methods and standardized protocols (Merriman et al., 2000; Ross, 2015; Smith et al., 2019). Overall, surveys, dummy purchases, tax gap analysis, and/or visual analysis of cigarette packs collected in urban walkways or garbage bins are used. Ideally, seeking to produce a reliable panorama of the ICM rate in a specific location, it is needed to apply continuous monitoring samplings using different methods. Littered CBs can represent a complementary strategy to estimate the ICM, despite presenting some challenges such as accurately distinguishing brands as legal or illegal using degraded CBs (Ribeiro et al., 2022a). On the other hand, it can avoid self-reported bias from other methods, providing similar sampling replicates, while could indicate some trends in illicit consumption (Smith et al., 2019).

Brazil is the world's fifth biggest country and the biggest and most populated in Latin America (>212 million inhabitants) (IBGE, 2020). Brazil has an important role on tobacco mitigation in the American continents, as one of the leading countries on reducing tobacco prevalence (Curti et al., 2019; Szklo et al., 2020). While the number of Brazilians who smoke cigarettes has reduced over the past decades, more than 8 million Brazilian smokers still identify themselves as 'loyal', 'satisfied' or 'very satisfied' with the overall "quality" of illegal cigarettes (Dorfman et al., 2017; Gigliotti et al., 2014). Nationwide ICM rates were estimated in Brazil using different methods (Brown et al., 2017; Iglesias et al., 2017; Szklo et al., 2018; Szklo and Iglesias, 2020). In local perspectives, Szklo et al. (2020) analyzed five highly urbanized cities using four different methods (littered cigarette packs in household garbage [1] and walkways [2], and phone [3] and face-to-face surveys [4]). Thus, assessing ICM in local basis play essential role to better estimate the ICM rates, then facilitating cross-referencing methods used nationally. Our previous study in Santos and Niterói was the first to apply CBs identifications to estimate ICM in Brazil (Ribeiro et al., 2022a).

Guarujá is an island that, together with eight other cities including Santos, forms the Baixada Santista metropolitan area, located in the southeast Brazilian São Paulo State (Roveri et al., 2020b). Guarujá has 324,977 inhabitants, 144.8 km<sup>2</sup> and 64 km of extension (IBGE, 2020). Around 75% of Guarujá area is covered by environmental protection areas and the remaining 25% is composed by completely urbanized zones (Roveri et al., 2020b; SMA/CPLA, 2016). The main economic activities held in the region are related to its neighbor city Santos, sharing the biggest Latin American port complex (Puscetdu et al., 2019). Touristic activities are also widely held in Guarujá, being mostly related to the 27 beaches with pleasant climate throughout the year. In this regard, during the high summer seasons (December to February), Guarujá population doubles (Roveri et al., 2020a). According to the Union of Urban Cleaning Companies, Guarujá has the 37th best urban cleaning status among 5,565 Brazilian cities, differently than Santos (São Paulo State) and Niterói (Rio de Janeiro State), which are the first of the ranking (ISLU, 2022). Despite this, our previous study reported high levels of CBs contamination in walkways from Niterói, and especially Santos (Ribeiro et al., 2022a) indicating that even well-ranked cities in public cleanliness indices may have issues related to CBs contamination.

Based on this scenario, the present study aimed to assess the spatial distribution of littered CBs in areas presenting different urban densities and income in Guarujá. This approach, associate to obtention of Cigarette Butts Pollution Indexes, leachate experiments (PHAs and toxic elements) and estimates of ICM, will allow an integrative evaluation of potential socioenvironmental impacts induced by CBs contamination. The results, were compared to previous studies performed in Santos and Niteroi cities seeking guide management decisions on the mitigation of this serious issue in the studied area, also enabling advances in a global perspective.

## 2. Methodology

### 2.1 Study area

The spatial assessment of cigarette butts (CBs) and cigarette packs (CPs) included nine sites in Guarujá city (S1 to S9). The selection of the sites considered three local urban densities classes available in the master plan of the city (*high, moderate and low* urban density) (Guarujá, 2013) (Table 1). Additionally, the sites S1 to S9 were chosen seeking to allow three replicate samplings, using 20 volunteers covering all sites in the same day between 5 and 10 (p.m.). Although urban density was the primary criteria for site selection, analyzes using income were also performed, since the prevalence of people who smoke and ICM rates are often related to urban occupation (Szklo et al., 2020). For this purpose was used available data about Average Nominal Income of Heads of Household (ANIH) in Guarujá city, which in categorized in four classes (*very low, low, moderate, high*) (Guarujá, 2013). Despite equal number of sites for classes of urban density and different number of sites per ANIHs classes (Table 1), the obtained values used were normalized by density (CBs.m<sup>-2</sup> or CPs.m<sup>-2</sup>) or the average percentage when concerning the ICM, thus comparative analysis between different ANIH classes were also possible.

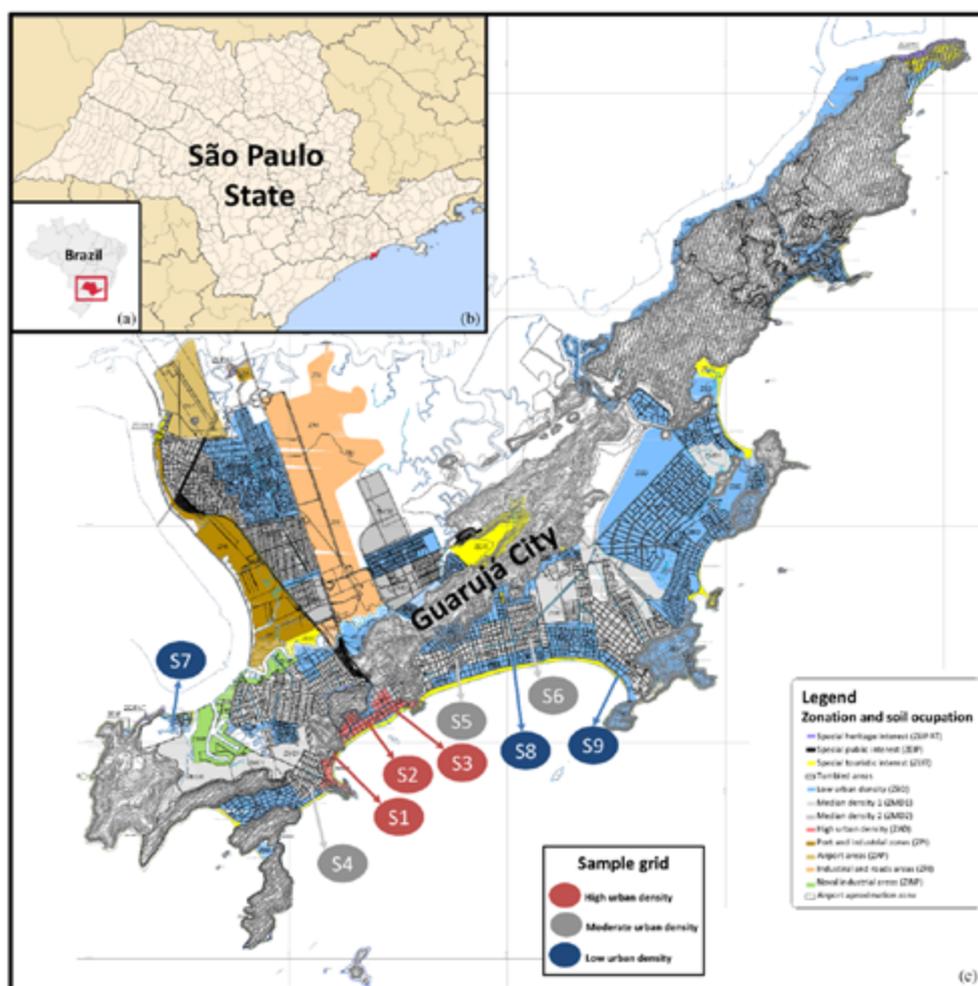
**Table 1.** Sampling sites, area (m<sup>2</sup>), urban density level and Average Nominal Income of Heads of Households (USD) analyzed in the present study.

Site	Area (m <sup>2</sup> )	Urban density level	Average Nominal Income of Heads of Households (USD)	Income level
S1	840	High	606-1,010	Moderate
S2	1,034	High	606-1,010	Moderate
S3	979	High	606-1,010	Moderate
S4	788	Moderate	303-606	Low
S5	1,078	Moderate	303-606	Low
S6	726	Moderate	303-606	Low
S7	734	Low	101-303	Very Low
S8	1,017	Low	101-303	Very Low
S9	705	Low	>1,010	High

## 2.1.1 Urban density levels

The urban density in Guarujá city is divided in *lower*, *moderate* and *high urban density zones*, in addition to *industrial and port activities*, *touristic interest*, *fishing and naval activities*, and others in the same available land occupation map (Fig. 1) (Guarujá, 2013). It was used the three urban densities levels (zones of *high* [S1, S2 and S3 – blue in Fig. 1], *moderate* [S4, S5 and S6 – gray] and *lower* [S7, S8 and S9 – red] urban density) (Fig. 1).

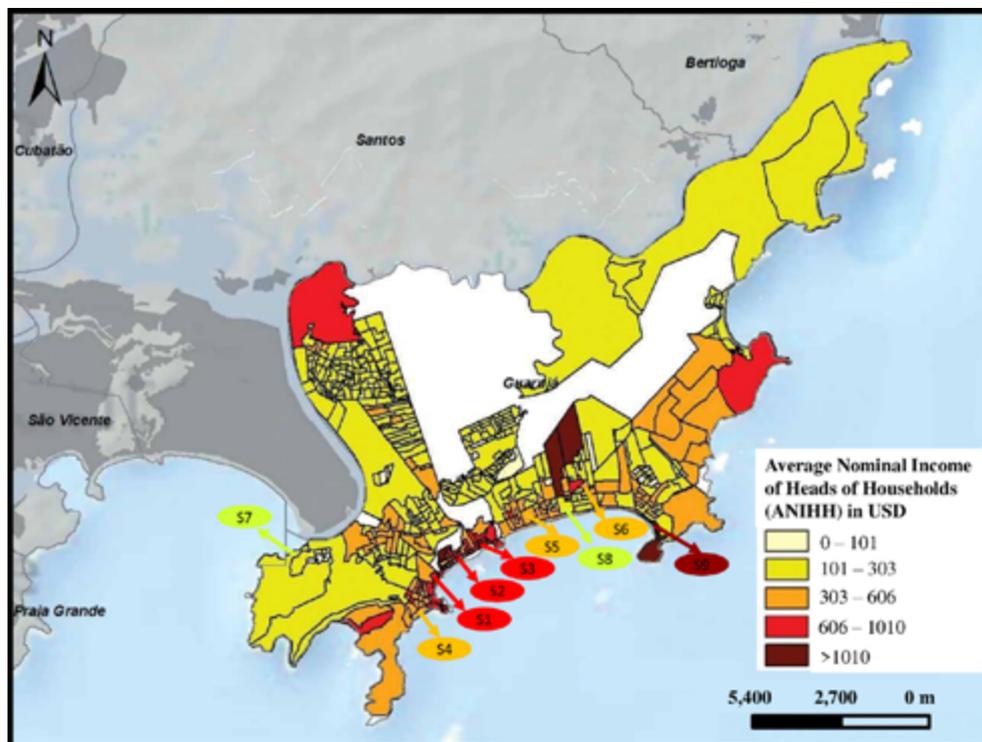
**Figure 1.** Map of Brazil (a), São Paulo state (b) and Guarujá city sampled sites distributed in urban areas of high, moderate and low urban density (c).



## 2.1.2 Income levels

The income of residents was only available data in Guarujá based in the ANIHH per month (in USD). Thus, the spatial evaluation, with the same sites, was carried out based on four classes of ANIHHs (101–303 [S7 and S8], 303–606 [S4, S5 and S6], 606–1010 [S1, S2 and S3] and >1010 USD [S9]) (Fig. 2).

**Figure 2.** Guarujá city sampled sites distributed in urban areas of different Average Nominal Income of Heads of Households (ANIHH).



## 2.1.3 Other urban aspects

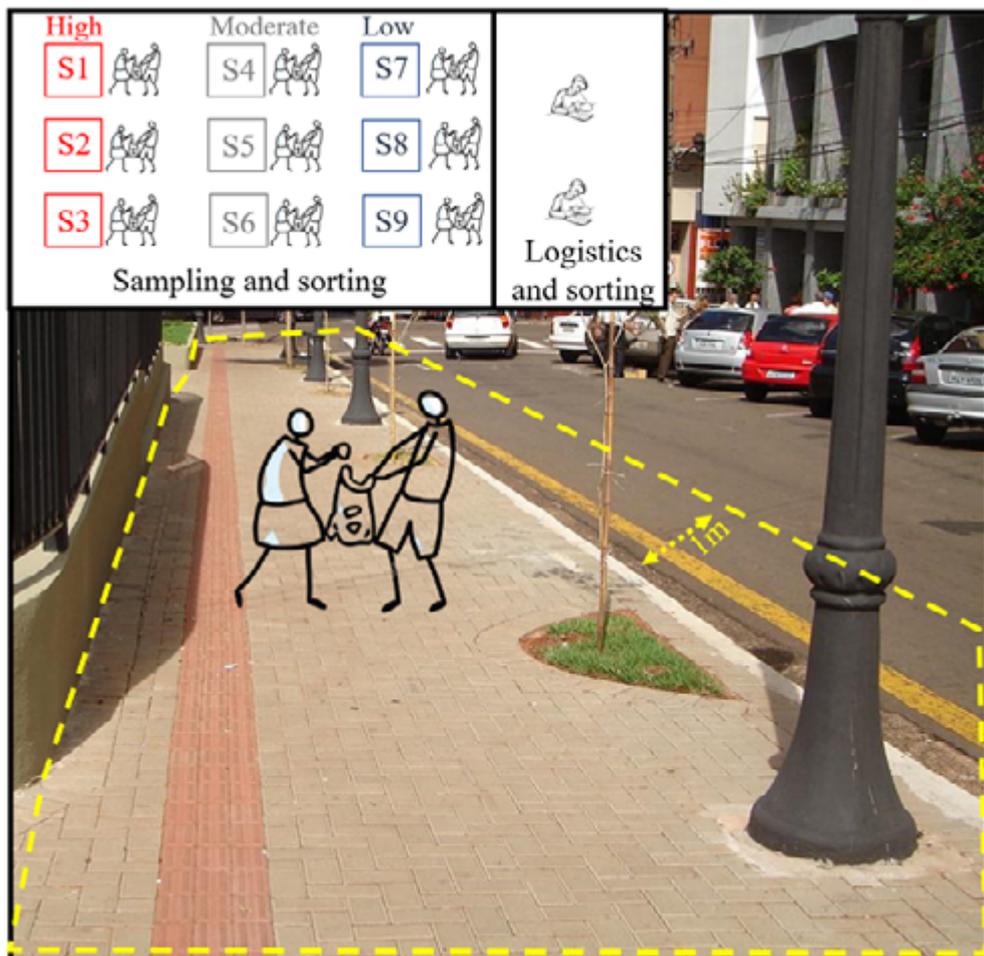
Several other urban aspects are linked with the number of CBs found in urban environments, such as the presence of bus stops (Oliver et al., 2014), tree pits (Green et al., 2014) or entrances to playgrounds and hospitals and educational venues (Valiente et al., 2020).

In the present study, the sample sites were visited before the sampling campaigns, in order to count the number of urban aspects of each sampling site to later correlate with the number of CBs found. More specifically, we considered nine aspects, i.e., the number of: stores that sell cigarette packs (1) and units (2); benches (3); commercial buildings (4); trees and bushes (5); manholes (6); bus stops (7); garbage bins and bags (8); and residential buildings (9) (Fig. S1 to S4 – Supplementary material). Vendors were asked if the store they work sell cigarette packs (1) and units (2) (Fig. S1), and it's worth highlighting that it is illegal but quite common in Brazil to sell cigarette units. Buildings were considered commercial (4) when they sell something or provide any service, and were residential (9) when they were houses or apartment complexes. Often, the same buildings are destined to commercial goals in their lower entrance, and to residential goals in their 2nd floor, being accounted as both in our study (Fig. S2). Additionally, benches (3), trees and bushes (5), manholes (6) and bus stops (7) and garbage bins and bags (8) were accounted by quantity.

## 2.2 Sampling procedures

Quadrants measuring on average 877.5 m<sup>2</sup> (from 705 to 1,078 m<sup>2</sup> - see Table 1) were delimited in each sample site, three sampling campaigns were performed in each quadrant during low season (between March and April 2023) seeking to avoid overestimation of littered CB, i.e., without the massive presence of tourists. Sampling campaigns were performed between 5 and 9 P.M. (Torkashvand et al., 2021), because CBs are most commonly seen in the evening hours (Patel et al., 2013), while avoiding the impact of street cleaning schedules activity (Marah and Novotny, 2011). Briefly, in each sampling site, a quadrant (measuring on average 877.5 m<sup>2</sup>) was drawn in urban walkways, where the littered CBs and CPs were

collected. Based on field visual surveys methods, samplings were conducted on each selected sidewalk and its continuation up to 1 m from the street (Fig. 3) (Cutter et al., 1991; Torkashvand et al., 2021). A total of 20 bachelor students (previously trained for collection of samples in urban and environmental areas) prepared the sample kits (Fig. S5), conducted the sampling campaigns (Fig. S6), and posterior sorting of collected samples (Fig. S7). These steps were supervised by PhD students to assure sample quality control.



**Figure 3.** Sampling strategy considering 20 students (18 for sampling and sorting and 2 for logistics planning) and the sampled quadrant area.

## 2.3 Cigarette butt densities

The density of CBs (DCBs) in each site was obtained by the ratio between the CBs abundance in the previously measured area (m<sup>2</sup>) in each site (Torkashvand et al., 2021). Therefore, the DCBs were expressed in CBs.m<sup>-2</sup>. The DCBs results were compared among classes of urban density and income levels, both separately and simultaneously, in addition to be correlated with the other urban aspects.

## 2.4 Cigarette butt contaminants leakage estimative

The Cigarette Butts Pollution Index (CBPI), proposed by Torkashvand et al. (2021), was calculated using the DCBs while taking into account other local factors in order to estimate the potential pollutants leakage from CBs to the soil. Indeed, the CBs contamination at urban sites may lead to different levels of pollutants leakage into the environment, after being subject to specific anthropogenic and environmental conditions (Green et al., 2014; Nasab et al., 2022; Torkashvand et al., 2020). In this sense, the CBPI was calculated by the equation  $CBPI = DCBs \times E$ , where E is the coefficient that considers simultaneously soil status (concrete, grassy or sandy), kind of pathways (if it had open channels or lines of trees and shrubs), weather conditions (annual rainfall in mm), and distance from groundwater (in meters) (Table 2).

**Table 2.** Cigarette Pollution Index (CBPI) E coefficient calculation.

	Soil status	Kind of urban pathways	Annual rainfall (mm) <sup>a, b</sup>	Distance to groundwater (m) <sup>c</sup>
E = 10x	Concrete, asphalt or high-quality paving = 1	Simple footpath = 1	300 ≥ 1	> 9 = 1
	Concrete, asphalt or low-quality paving = 1.2	Footpath with open channel = 1.5	301 - 500 = 1.5	6 - 9 = 1.2
	<sup>d</sup> Sand and soil roads without surface coating = 1.5	Footpath with a line of trees or shrubs = 1.5	501 - 750 = 2	3 - 6 = 1.5
	Grassy = 2.5	Footpath with open channel and line of trees or shrubs = 2	>750 = 2.5	3 ≥ 2

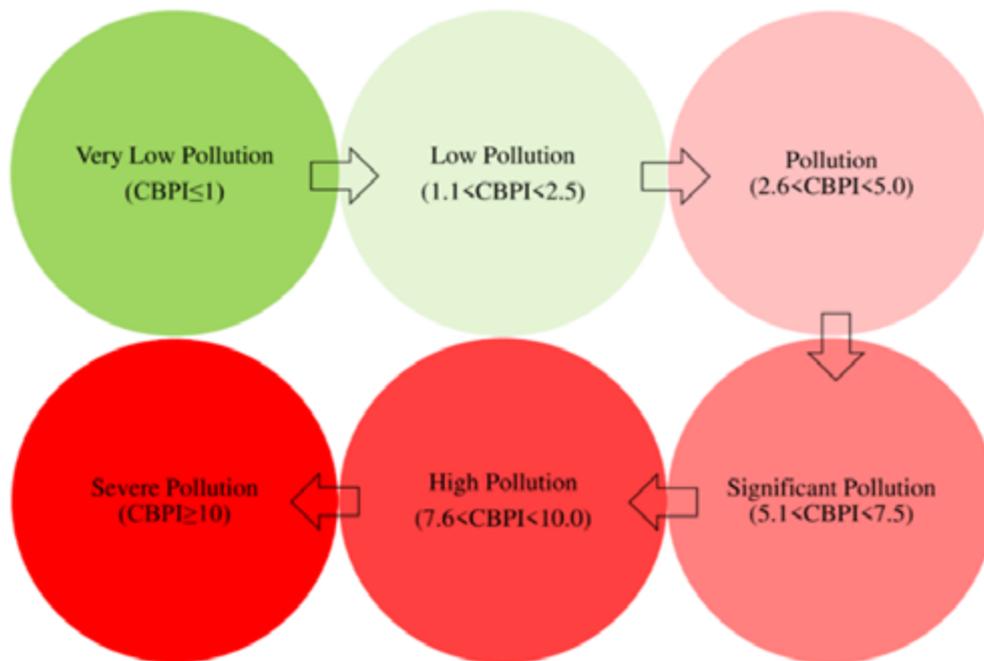
<sup>a</sup>Due to the fact that, up until now, no universal method calculated the CB period in urban places, the annual index was used in this study. However, this parameter is calculated considering seasonal changes or the number of rainy days: months without precipitation = 1, months with less than 8 days of precipitation = 1.5, months with 8–15 days of precipitation = 2, and months with more than 15 days of precipitation = 2.5.

<sup>b</sup>Urban green spaces that are irrigated daily = 2.5.

<sup>c</sup>Adapted from the Oleckno index.

<sup>d</sup>Coastal and sandy areas = 2.

The CBPI indicate more serious environmental impacts in sites with higher CBs density, low quality paving, open channels, wooded areas, annual rainfall regimes higher than 750 mm and areas with shallow groundwater (<3 m). These tend to be ranked worse by this index. On the other hand, smaller CBs densities, paving with good quality, absence of open channels and urban afforestation, deep water tables (>9 m) and low rainfall (<300 mm) suggest less potential for leaching contaminants. In Guarujá, the annual rainfall reach approximately 3,000 mm (then classified as >750 mm) (Roveri et al., 2020b) and the depths of the groundwater were classified as <3 m (São Paulo, 2005). Finally, the obtained CBPI values were classified in levels of CBs pollutants leakage, ranging among *very low pollution* (≤1.0), *low pollution* (1.1–2.5), *pollution* (2.6–5.0), *significant pollution* (5.1–7.5), *high pollution* (7.6–10.0) or *severe pollution* (≥10.0) (Fig. 4) (Torkashvand et al., 2021). Finally, the CBPI results were separately compared between the classes of urban density and income levels, and both aspects simultaneously.



**Figure 4.** Cigarette Butts Pollution Index (CBPI) classes, varying from very low pollution to severe pollution.

## 2.5 Cigarette brands and companies

The brands of all collected CBs and CPs were visually identified by searching for printed designs, logos, or words (Stratton et al., 2016). When the brand of a specific CB was not visually identifiable because loss of its physical characteristics due to a high stage of degradation, it was classified as 'Unidentifiable' (Lima et al., 2021; Ribeiro et al., 2022a). In addition, the identifiable brands were connected with the source company responsible for its marketing (Brasil, 2018). Each CBs and CPs brands were photographed in order to start a guide of identification for further studies.

## 2.6 Illegal cigarette marketing

CBs brands found were assessed for their legality status (given in percentages) by checking them in the list of brand names approved (Anvisa, 2021) on the market by the Brazilian Health Regulatory Agency (ANVISA) (Barkans and Lawrance, 2013; Ribeiro et al., 2022a). In CPs, similar steps were taken, plus the identification of the country of origin, presence (and language) of health warnings and the stamp from Brazilian regulatory agencies (Szklo et al., 2020).

## 2.7 Cigarette butt experiments

The most frequent brand of CBs found during the sampling was used to prepare leachates in ultrapure water (in triplicate). Cigarettes were purchased in local commercial establishments and smoked using a hand-operated vacuum pump (oil-free) connected to a PTFE tubing with 10 simultaneous taps at low-pressure ( $-50$  mmHg), until approximately 1 cm above the filter according to Lima et al., (2021) and Madelli et al., (2023). Then, the CBs were refrigerated ( $-4^{\circ}\text{C}$ ) in glass bottles (250 mL) previously decontaminated (washed using alkaline detergent [10 $\times$ ], immersed in nitric acid [5%] for 24 h, dried and rinsed with acetone and hexane [3 $\times$ ]) until the experiments of settling and leachate preparation for toxicity tests. To prepare CBs leachates were used 4 CBs.liter $^{-1}$  of ultra-pure dilution water. Glass beakers containing CBs solutions were placed on an orbital shaker (100 RPM) for 24 h and stirred under dark conditions. After, the leachates were filtered through a 1.2  $\mu\text{m}$  mesh to remove particulate material and analytically analyzed for identification and quantification of PHAs and toxic elements.

Sixteen PHAs (potentially toxic and considered priority in environmental studies according to United States Environmental Protection Agency), and twelve trace elements (As, Cd, Cr, Cu, Fe, Hg, Mg,

Mn, Pb, Ni, Sn and Zn) were investigated in the leachate solution. After acid digestion, trace elements concentrations were quantified by inductively coupled plasma optical emission spectrometry (ICP-OES) and/or inductively plasma mass spectrometry coupled (ICP-MS) after microwave-assisted acid digestion. The choice of ICP-OES or ICP-MS techniques considered the performance of each specific element and concentration in samples. Quality control of these analyses employed blanks and recovery tests. To investigate PAHs, samples were spiked with 100  $\mu\text{L}$  of deuterated PAH (acenaphthene-d10, phenanthrene-d10 and chrysened12 at 1000  $\mu\text{g L}^{-1}$ ) as surrogate. The samples were submitted to a liquid-liquid extraction using high-purity hexane and cleaned up by alumina-silica gel chromatographic columns. Extracts were injected into a gas chromatography coupled to mass spectrometry system (GC-MS, Agilent Technologies, model 7820A/5975C, Wilmington, USA) in the selected ion monitoring (SIM) mode. Quality control included procedural blanks and recovery of surrogates.

## 2.8 Statistical analysis

The density of CBs and CBPI values obtained for sampled sites were grouped according to categories of urban density (low, moderate and high). Shapiro-Wilk and Levene tests were performed to assess normality homogeneity of variances respectively. After, the differences among categories were assessed by a one-way ANOVA followed by Tukey multiple comparisons. Alternatively, non-parametric statistics (Kruskal-Wallis followed by the Dunn's test) were applied when ANOVA assumptions were not achieved. The results were presented as mean  $\pm$  standard deviation (SD). Pearson or Spearman non-parametric correlation analysis were used to investigate the relationships among density of CBs and the urban aspects in each sampled site. All statistical analyses were performed using Statistica® (version 13.0 (Statsoft)) with a significant level of 0.05.

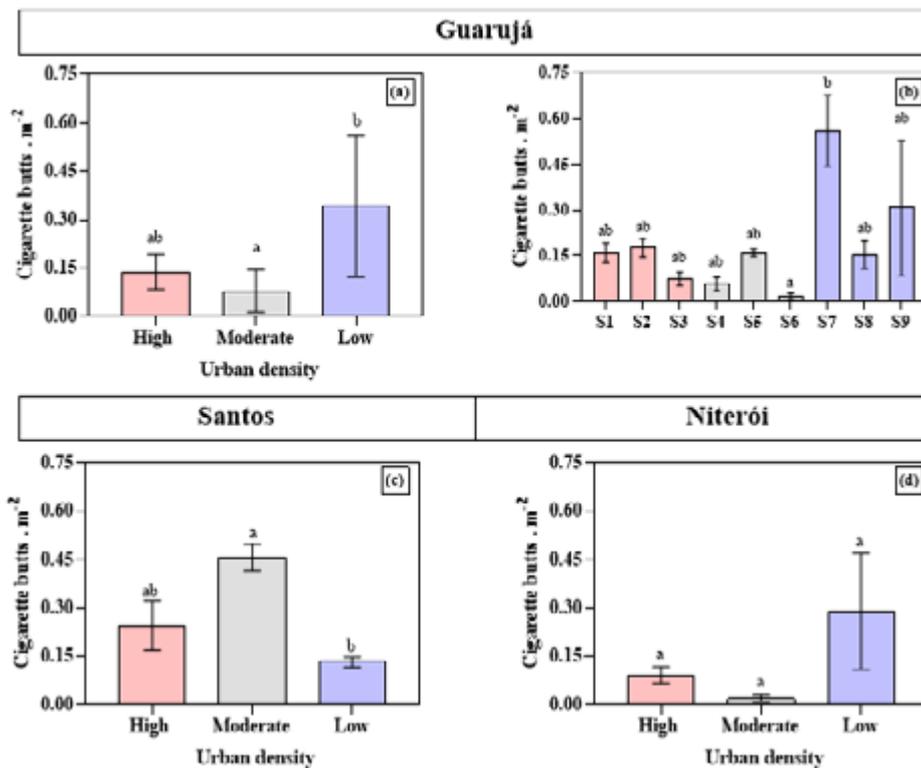
### 3. Results and Discussion

#### 3.1 Cigarette butt densities

A total of 4,321 CBs were found in the sampled sites which covered an area of 23,694 m<sup>2</sup>. Thus, considering all analyzed sites the average density of cigarette butts (DCBs) was  $0.18 \pm 0.17$  CBs.m<sup>-2</sup>. The DCBs was also examined in urban zones of Santos and Niterói cities (Brazil) in 2021, when these cities were ranked as the top 2 cities with the best urban cleaning status among 5,565 Brazilian cities. In this regard, according to 2022 report Guarujá is ranked in 37<sup>th</sup> position (ISLU, 2022; Ribeiro et al., 2022a). Therefore, we expected to find that DCBs in Guarujá would be higher than Santos and Niterói. While this was the case for Niterói ( $0.08 \pm 0.21$  CBs.m<sup>-2</sup>), Santos presented higher DCBs ( $0.25 \pm 0.17$  CBs.m<sup>-2</sup>). Based on analyzed sites and cities, our findings suggest that urban cleaning status may not necessarily reflect the actual prevalence of littered CB in urban environments. Indeed, CBs present a distinct dynamic in comparison to other types of anthropogenic litter items in terms of disposal, distribution and accumulation frequencies (Araújo et al., 2022). Usually, people who smoke tends to get rid of CBs faster, possibly due to the bad smell, and tend to wrongfully discard them due to social acceptance, habitual behavior and lack of awareness. Additionally, the prevalence of people who smoke is huge and CBs tends to remain in the environment after local clean-ups, due to small size and light weight, making them to easily scatter. Therefore, urban cleaning status was not a good indicator for CBs occurrence and local authorities should actively address this environmental issue with specific monitoring programs.

### 3.1.1 Urban density levels

In areas presenting different levels of urban density in Guarujá, sites with *high* and *moderate* levels had no significant differences in DCBs (Kruskal-Wallis followed by Duun's tests,  $p < 0.05$ ), while sites under *low urban densities* accounted for DCBs significantly higher than sites with *moderate* urban densities (Fig. 5a). Such variations were probably caused only by the values obtained in S6 and S7, which were the smallest and higher DCBs sites, respectively (Fig. 5b). Same pattern was also seen in Santos (Fig. 5c) being attributed to wide standard deviations often observed in studies assessing densities of anthropogenic litter (Ribeiro et al., 2022a). Indeed, Santos and Guarujá are neighbor cities in São Paulo state, southeast Brazil, equally sharing the borders of the biggest port complex in Latin America (Ribeiro et al., 2021a). Thus, we expected certain level of similarities between DCBs obtained in Santos and Guarujá. In Niterói, the DCBs along sites under different levels of urban density occupation were similar to Guarujá (*low > high > moderate* urban density), but surprisingly no differences were seen among them in Niterói (Fig. 5d). Such results indicate that CBs contamination in urban areas may not be related to population densities in different cities. Few studies assessing these parameters were so far performed covering just three cities from southeastern Brazil (considering the present study). Therefore, the approach adopted in the present study must be replicated for a larger number of cities in different regions, considering different urbanization levels, alongside with other socioeconomic conditions before a pattern can be identified.

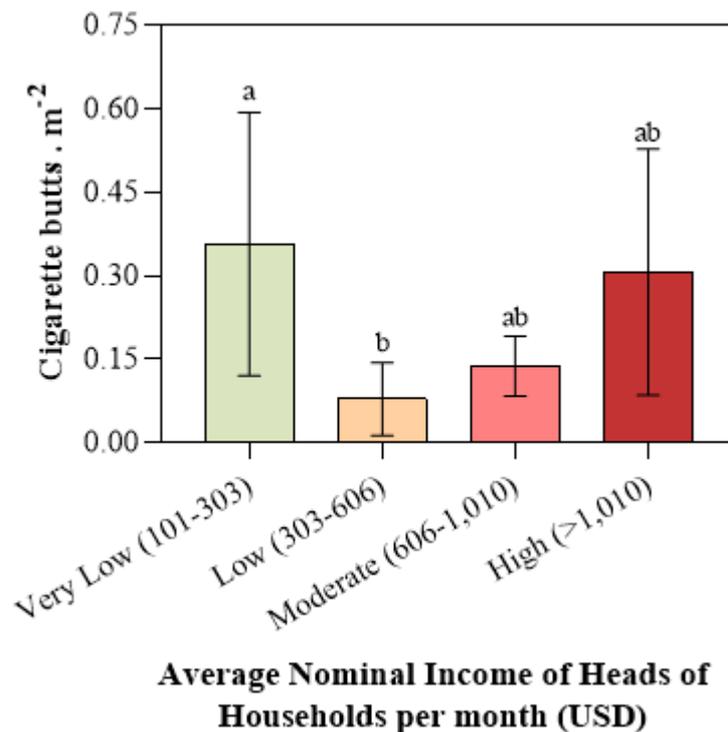


**Figure 5.** Cigarette butts densities (DCBs) in Guarujá city by urban densities (a) and sites (b). Error bars indicate standard deviations. Data from Santos and Niterói were obtained by Ribeiro et al. (2022a). Letters denote significant differences (Kruskal-Wallis followed by Dunn's tests).

### 3.1.2 Income levels

The consumption of cigarettes, and consequent CBs littering, were reduced in high-income countries over the past few years, while increased in middle and low-income countries around the world (Vanapalli et al., 2023). To our knowledge, no study compared the CBs distribution with population income in local, regional or national perspectives. In Guarujá, the DCBs were higher in *very low* (0.36 CBs.m<sup>-2</sup>)

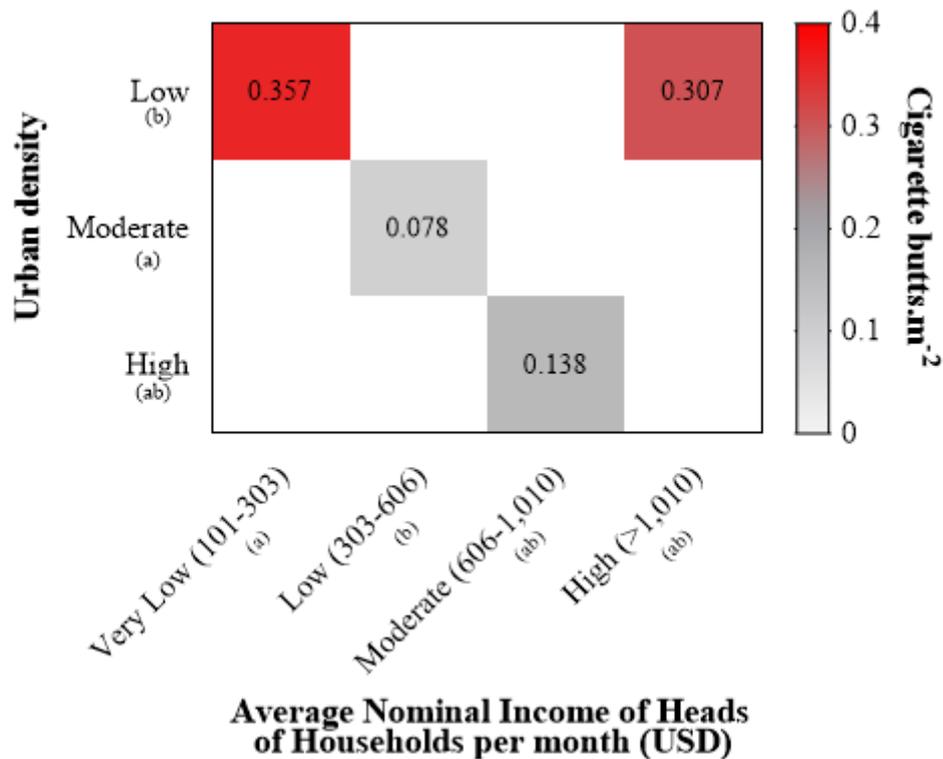
and *high-income* zones (0.30 CBs.m<sup>-2</sup>), i.e., locations in which the Average Nominal Income of Heads of Households (ANIH) is 101–303 and >1010 USD per month, respectively (Fig. 6). Overall, DCBs in Guarujá were similar across locations with different incomes, but significant differences were seen between 101–303 USD and 303–606 USD ANIHs. Thus, CBs contamination in Guarujá city, and possibly its consumption, did not follow the global trend of decrease in high-income countries and increase in low and middle-income countries (Vanapalli et al., 2023). Therefore, more studies are necessary in this topic seeking to identify the trends between cigarette consumption, CBs littering and different income locations.



**Figure 6.** Cigarette butts densities (DCBs) in Guarujá city by Average Nominal Income of Heads of Households (ANIH). Error bars indicate standard deviations. Letters denote significant differences (Kruskal-Wallis followed by Dunn’s tests).

### 3.1.3 Urban density and income levels

The DCBs in zones of low urban density (lowest class) CBs differ from *moderate* zones (second lowest class). Similarly, the DCBs in zones of very low income (lowest class - 101–303 USD per month) differ from *low* zones of income (the second lowest class - 303–606 USD per month). Thus, both aspects showed the same trend in Guarujá. However, low urban density zones had higher DCBs despite the level of income, considering the highest levels found in both richer (>1,010 USD per month) and poorer (101–303 USD per month) income areas of low urban density (Fig. 7). Additionally, other urban density zones (high and moderate, with 303–1,010 USD per month income) had less than half and a third of CBs density, respectively (Fig. 7). Therefore, it is fair to assume that low urban density is more prominently connected with higher CBs densities than income levels. Thus, more CBs are expected to be found in poorer locations, especially under low urban densities. The urban density and income of the heads of households (ANHH) were not simultaneously compared in Guarujá and many other Brazilian and worldwide urban environments, especially when concerning CBs contamination. In this sense, our study can serve as basis for future local urban monitoring initiatives. Thus, more sites should be selected in future studies seeking to produce a more suitable and combine sampling grid covering different income classes (Figure 7). In this sense, our study serves as a preliminary analysis of CBs contamination combining urban density and income levels.

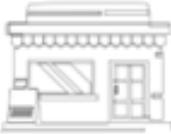
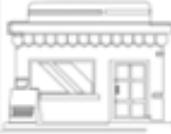


**Figure 7.** Average cigarette butt densities (cigarette butts.m<sup>-2</sup>) in different urban density and income (Average Nominal Income of Heads of Households [ANIH]) levels. Letters denote significant differences (Kruskal-Wallis followed by Dunn’s tests).

### 3.1.4 Other urban aspects

Comparison among spatial distribution of CBs in urban areas is strongly connected with city plans and population maps available (Ribeiro et al., 2022a). For instance, Santos and Niterói city hall’s had available master plans (Niterói, 2022; Santos, 2013), identifying city areas regarding the population densities and soil usage (residential, commercial, public spaces and other zones), amongst several other aspects. In Guarujá, only data on urban population and level of income were previously available (Guarujá, 2013). However, the present study counted how many residential and commercial buildings there were in the sampled sites. The DCBs were not related to the number of residential buildings in each studied site, according to Spearman Correlation ( $p > 0.05$ ) (Fig. 8). On the other hand,

the presence of commercial buildings was positively and strongly related to DCBs ( $p < 0.05$ ,  $r = 0.81$ ), suggesting potential sources of littered CBs. Also, it is known that the CBs occurrence is related to urban aspects distribution, such as bus stops and public transportation and hospitals entrances and other urban aspects (Dobadararan et al., 2019; Green et al., 2014). In Guarujá, the DCBs was higher in locations with more benches and stores selling cigarette in packs and units ( $p < 0.05$ ) (Fig. 8). Moreover, the DCBs was smaller in locations with more trees and shrubs ( $p < 0.05$ ,  $r = -0.73$ ) (Fig. 8). Finally, no relation was observed among the DCBs and the number of manholes, bus stops and garbage bins and bags ( $p > 0.05$ ) (Fig. 8). Thus, our data suggest that more attention from local authorities (i.e., installation of collectors) should be given in these areas.

<p><b>Stores that sell cigarettes packs and units</b> <input checked="" type="checkbox"/> **</p>  <p>Not normal - Spearman <math>r = 0.8751</math> <math>P = 0.0040</math></p>	<p><b>Stores that sell cigarettes packs</b> <input checked="" type="checkbox"/> **</p>  <p>Not normal - Spearman <math>r = 0.8430</math> <math>P = 0.0079</math></p>	<p><b>Commercial buildings</b> <input checked="" type="checkbox"/> **</p>  <p>Normal - Pearson <math>r = 0.8097</math> <math>P = 0.0082</math></p>
<p><b>Benches</b> <input checked="" type="checkbox"/> *</p>  <p>Normal - Pearson <math>r = 0.8262</math> <math>P = 0.0238</math></p>	<p><b>Trees and bushes</b> <input checked="" type="checkbox"/> *</p>  <p>Normal - Pearson <math>r = -0.7344</math> <math>P = 0.0242</math></p>	<p><b>Manholes</b> <input type="checkbox"/> X</p>  <p>Normal - Pearson <math>r = 0.7600</math> <math>P = 0.8458</math></p>
<p><b>Bus stops</b> <input type="checkbox"/> X</p>  <p>Not normal - Spearman <math>r = 0.3677</math> <math>P = 0.3571</math></p>	<p><b>Garbage bins and bags</b> <input type="checkbox"/> X</p>  <p>Normal - Pearson <math>r = 0.0060</math> <math>P = 0.9875</math></p>	<p><b>Residential buildings</b> <input type="checkbox"/> X</p>  <p>Normal - Pearson <math>r = -0.1639</math> <math>P = 0.6691</math></p>

**Figure 8.** Urban characteristics correlation with the density of cigarette butts (DCBs).

### 3.1.5 Worldwide cigarette butt densities

In a worldwide perspective, most studies analyzing the CBs occurrence in urban environments (Iran, Brazil, Japan and Germany) showed higher average DCBs than Guarujá (Table 3). However, Niterói and Qazvin (Iran), had smaller DCBs than Guarujá. However, there are still few studies published assessing occurrence of CBs in urban areas, hindering a proper global snapshot of the DCBs in urban environments. In this sense, analyzing the litter items in freshwater, and mostly coastal environments is more common in the literature (Table 3). Indeed, litter is usually accumulated in those environments, due to a more prominent accumulation caused by local hydrodynamics, wind flow influence and the failings in solid waste management (Ribeiro et al., 2021b, 2022a). Indeed, our previous study found a higher DCBs in Guarujá beaches ( $0.30 \text{ CBs.m}^{-2}$ ) (Ribeiro et al., 2021a) compared to the present study performed in urban environments ( $0.18 \text{ CBs.m}^{-2}$ ). However, there are exceptions as Santos, where DCBs were higher in urban ( $0.25 \text{ CBs.m}^{-2}$ ) than coastal ( $0.20 \text{ CBs.m}^{-2}$ ) environments (Ribeiro et al., 2021b, 2022a), a Mazandaran ( $0.30$  and  $0.11 \text{ CBs.m}^{-2}$ ), Iran, by Nasab et al. (2022) (Table 3).

**Table 3.** Densities of cigarette butts in urban and beach around the world.

Location	Country	Environment	Density of cigarette butts (CBs/m <sup>2</sup> )	Reference
Qazvin	Iran	Urban	0.03–1.20	(Gholami et al., 2020)
Niterói	Brazil	Urban	0.08 (0.00–1.40)	(Ribeiro et al., 2022a)
<b>Guarujá</b>	<b>Brazil</b>	<b>Urban</b>	<b>0.18 (0.01–0.68)</b>	<b>Present study</b>
Qazvin	Iran	Urban	0.21	(Torkashvand et al., 2021)
Behbahan	Iran	Urban	0.23 (0.06–0.53)	(Darabi et al., 2023)
Santos	Brazil	Urban	0.25 (0.06–0.9)	(Ribeiro et al., 2022a)
Mazandaran	Iran	Urban	0.30	(Nasab et al., 2022)
Jiroft	Iran	Urban	0.38	(Sedeh et al., 2022)
Ueda	Japan	Urban	0.70	(Moriwaki et al., 2009)
Berlin	Germany	Urban	2.70 (0.29–5.20)	(Green et al., 2014)
Boiçucanga	Brazil	Riparian forests	0.01	(Ribeiro et al., 2022b)
Boiçucanga	Brazil	Beach	0.04	(Ribeiro et al., 2022b)
Baltic Sea	Lithuania	Beach	0.04 (0.00–0.54)	(Kataržytė et al., 2020)
Northwest	Morocco	Beach	0.06 (0.01–0.09)	(Mghili et al., 2023)
Mazandaran	Iran	Beach	0.11	(Nasab et al., 2022)
36 locations	Baltic Sea	Beach	0.14 (0.00–1.51)	(Haseler et al., 2020)
Búzios	Brazil	Beach	0.14	(Oigman-Pszczol and Creed, 2007)
Santos	Brazil	Beach	0.20	(Ribeiro et al., 2021b)
Guarujá	Brazil	Beach	0.30	(Ribeiro et al., 2021a)
Loung areas	Thailand	Beach	0.36–0.44	(Kungskulniti et al., 2018)
Northeast	Brazil	Beach	0.87	(Aratújo and Costa, 2021)
Baltic Sea	Germany	Beach	1.47 (0.00–29.00)	(Kataržytė et al., 2020)
Pueblo	Uruguay	Beach	0.00–8.00	(Rodríguez et al., 2020)
Recife	Brazil	Beach	1.05–8.85	(Silva et al., 2023)
Bushehr	Iran	Beach	2.00–38.00	(Dobaradaran et al., 2018)

### 3.2 Cigarette butt contaminants leakage estimative

The average CBPI found in Guarujá estimated that the contaminants leakage is overall classified as *severe pollution* (CBPI>10), i.e., 15.4±11.5. Based on the aspects present in the CBPI calculation, all sampled urban walkways had scores of 1.2 for the soil status (Table 4), which means *Concrete, asphalt or low-quality paving*. Indeed, most of Guarujá has uneven and/or drilled walkways of low-quality pavement, providing several spaces to CBs accumulation (Fig.

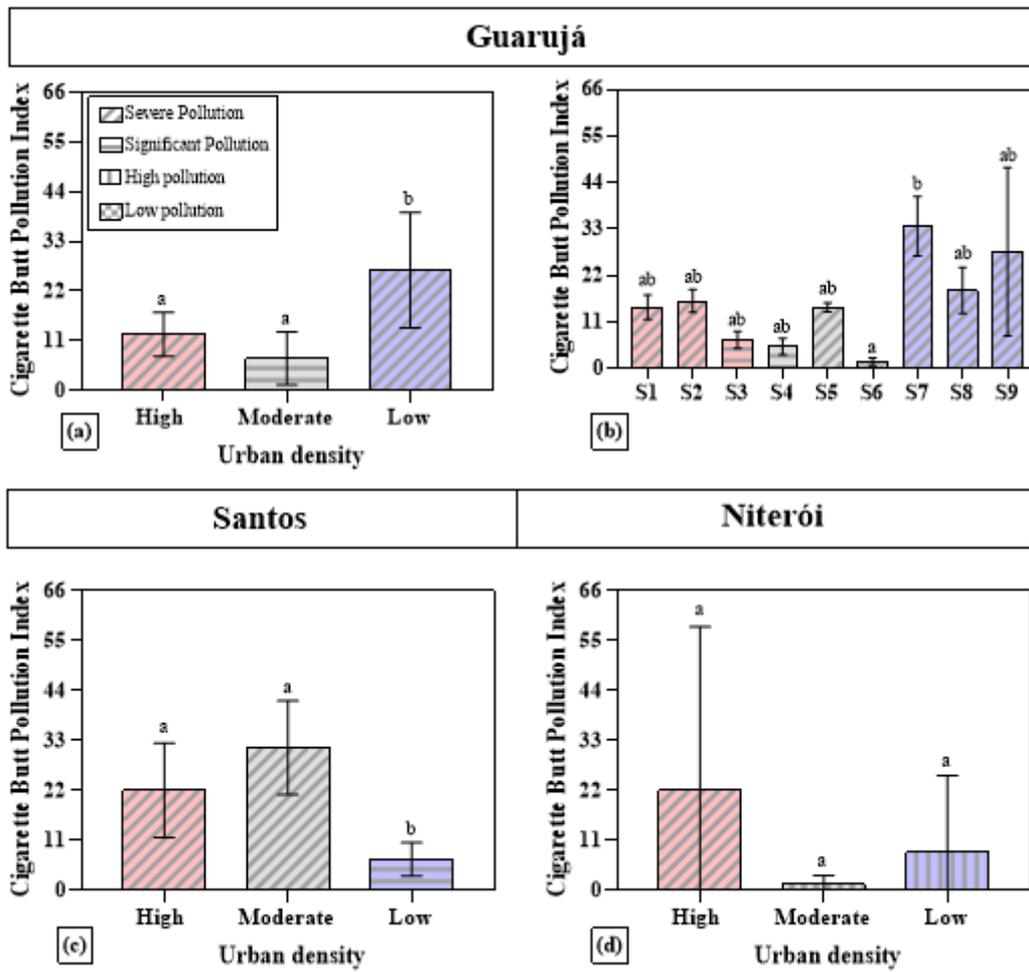
S8). In addition, the annual rainfall and groundwater distance had a score of 2.5 and 2.0, respectively (Table 4), because Guarujá is a coastal city with more than 300 mm of annual rainfall and shallow groundwater with less than 3 m, respectively (São Paulo, 2005). The only CBPI aspect that varied among the sampled walkways was the pathways type. Most sites scored 1.5, meaning *Footpath with a line of trees or shrubs* or *Footpath with open channel*. However, S7 scored 1.0, because it was a *Simple footpath*, while S8 scored 2.0 because it was a *Footpath with open channel and line of trees or shrubs*.

**Table 4.** Cigarette Butts Pollution Index (CBPI) scores, results and classification in Guarujá.

Site	DCBs	E	Soil status	Pathways	Annual rainfall	Groundwater distance	CBPI	Classification
S1	0.198	10	1.2	1.5	2.5	2	14.5±2.9	Severe pollution
	0.146	10	1.2	1.5	2.5	2		
	0.139	10	1.2	1.5	2.5	2		
S2	0.212	10	1.2	1.5	2.5	2	16.0±2.7	Severe pollution
	0.155	10	1.2	1.5	2.5	2		
	0.166	10	1.2	1.5	2.5	2		
S3	0.101	10	1.2	1.5	2.5	2	6.8±2.0	Significant pollution
	0.066	10	1.2	1.5	2.5	2		
	0.059	10	1.2	1.5	2.5	2		
S4	0.082	10	1.2	1.5	2.5	2	5.2±2.0	Significant pollution
	0.052	10	1.2	1.5	2.5	2		
	0.039	10	1.2	1.5	2.5	2		
S5	0.156	10	1.2	1.5	2.5	2	14.5±1.0	Severe pollution
	0.174	10	1.2	1.5	2.5	2		
	0.153	10	1.2	1.5	2.5	2		
S6	0.029	10	1.2	1.5	2.5	2	1.6±0.9	Low pollution
	0.01	10	1.2	1.5	2.5	2		
	0.015	10	1.2	1.5	2.5	2		
S7	0.452	10	1.2	1	2.5	2	33.6±7.0	Severe pollution
	0.685	10	1.2	1	2.5	2		
	0.545	10	1.2	1	2.5	2		
S8	0.206	10	1.2	2	2.5	2	18.5±5.4	Severe pollution
	0.121	10	1.2	2	2.5	2		
	0.136	10	1.2	2	2.5	2		
S9	0.562	10	1.2	1.5	2.5	2	27.6±19.9	Severe pollution
	0.190	10	1.2	1.5	2.5	2		
	0.169	10	1.2	1.5	2.5	2		

### 3.2.1 Urban density levels

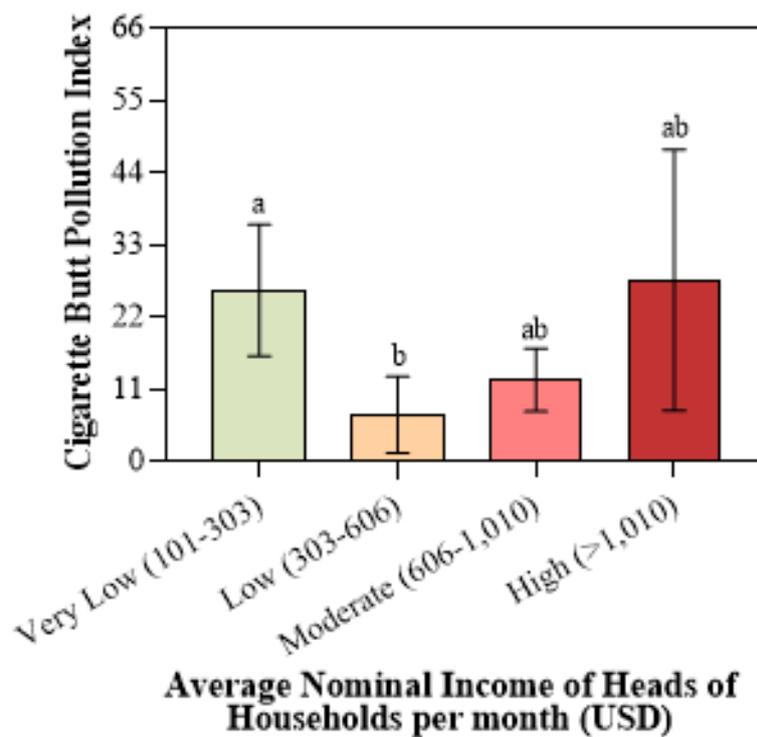
Zones under *low urban densities* presented higher CBPI ( $26.6 \pm 12.0$ ) and significantly different to zones of both *high* ( $12.4 \pm 4.6$ ) and *moderate* urban density ( $7.1 \pm 5.5$ ) (Fig. 8a). As observed for DCBs, the sites S6 and S7 were the main inducers of the spotted differences in CBPI (Fig. 8b). Thus, low urban density zones of Guarujá contribute more with CBs contaminants leakage, differing from other zones. Such difference was also observed in Santos *low urban density* zones, however in an opposite way, contributing less with CBs leakage (Fig. 8c). Indeed, in Santos, the CBs leakage was higher in *low urban density* zones (Fig. 8c), as seen in Niterói (Fig. 8d). Despite this, all zones in Niterói contributed similarly to the CBs leakage (Fig. 8d). Thus, despite few similarities, the CBs leakage in those three cities are different when considering the urban density zones.



**Figure 8.** Cigarette Butts Pollution Index (CBPI) in Guarujá city by urban densities (a) and sites (b). Error bars indicate standard deviation. Data from Santos (c) and Niterói (d) were obtained by Ribeiro et al. (2022a). Letters denote significant differences (Kruskal-Wallis followed by Dunn's tests).

### 3.2.1 Income levels

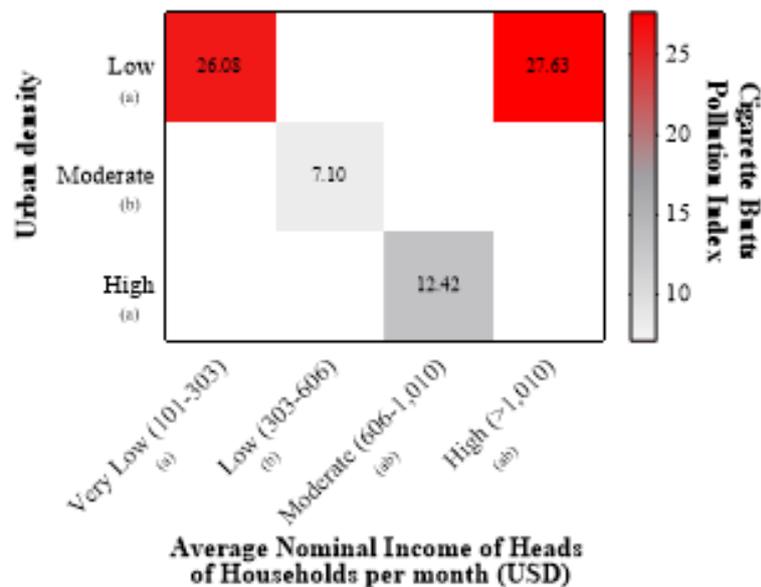
The CBPI was higher in richer and poorer locations, i.e., very low- (101–303 USD per month) and high-income areas (>1,010 USD per month), with  $26.08 \pm 9.14$  and  $27.63 \pm 16.24$ , respectively (Fig. 9). In the *moderate*-income areas, the CBPI was  $12.42 \pm 4.55$ . Finally, the CBPI in *low-income* areas was  $7.10 \pm 5.54$ . Indeed, significant differences between *very low*- and low-income levels were observed (Fig. 9).



**Figure 9.** Cigarette Butt Pollution Index (CBPI) in Guarujá city by urban densities (a) and sites (b). Error bars indicate standard deviation. Letters denote significant differences (Kruskal-Wallis followed by Dunn's tests).

### 3.2.3 Urban density and income levels

The CBPI in zones of *low* urban density (the lowest class) differ from *moderate* zones (the second lowest class). Similarly, the CBPI in zones of very low income (the lowest class - 101–303 USD per month) differ from low-income zones (the second lowest class - 303–606 USD per month) (Fig. 10). Thus, both aspects showed the same trend in Guarujá, as it was also observed for the DCBs (see Fig. 7). Similarly, the CBPI was higher in low urban density zones despite the level of income (Fig. 10). Moreover, other urban density zones (high and moderate, with 303–606 USD per month income) had less than half and a third of CBPI, respectively (Fig. 10). Thus, the DCBs and the contaminants leakage from them, occurred similarly in Guarujá, showing the same patterns, i.e., higher at low urban density zones. However, the number of CBs is higher in poor areas, while its contaminants leakage is slightly higher in poor areas.



**Figure 10.** Cigarette butt densities (DCBs) in different urban density and income (Average Nominal Income of Heads of Households [ANIH]) levels. Letters denote significant differences (Kruskal-Wallis followed by Dunn’s tests).

### 3.2.4 Worldwide cigarette butt contaminants leakage

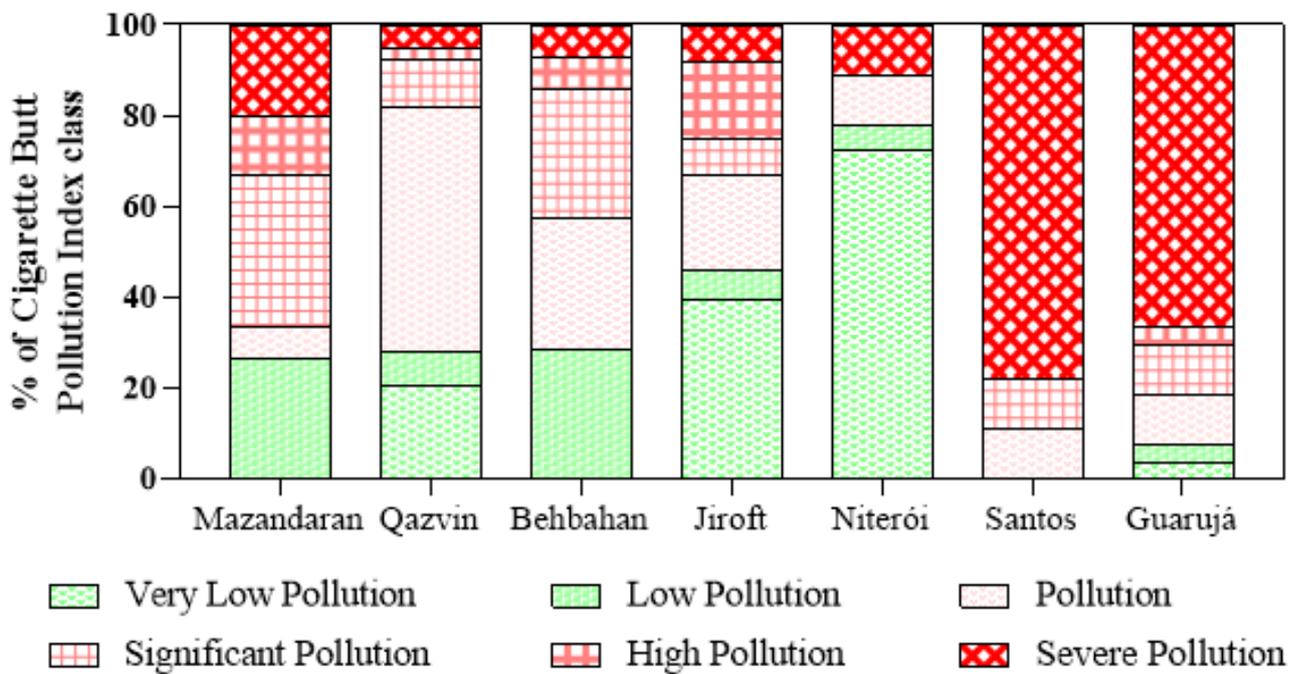
The average CBPI in Guarujá (15.4) was more than twice higher than other six urban areas from Brazil (Niterói) and Iran (Qazvin, Jiroft, Behbahan and Mazandaran) (Table 5). Indeed, the only higher average CBPI was found in the neighbor Santos (17.6) (Table 5). When considering the maximum CBPI, Guarujá (50.6) gets only behind Niterói (101.6) (Table 5). These may represent alarming values for the leakage of contaminants from CBs in Guarujá. However, more studies are necessary in other countries and cities seeking to analyze geographical trends, leading to more assertive patterns for CBs contamination of several urban and socio-economic aspects, such as urban density and the level of income. In addition, the amount of sample sites necessary to validate a specific study is not standardized yet.

**Table 5.** Cigarette Butt Pollution Index (CBPI) around the world's urban environments.

Country	City	Sampled sites	Sampled area (m <sup>2</sup> )	CBPI average	CBPI range	Reference
Iran	Qazvin	39	142,998	3.8	0.1–14.4	(Torkashvand et al., 2021)
Iran	Jiroft	12	Unclear	3.8	0.1–12.3	(Sedeh et al., 2022)
Iran	Behbahan	14	Unclear	4.9	1.3–10.6	(Darabi et al., 2023)
Iran	Mazandaran	15	Unclear	6.2	1.2–13.4	(Nasab et al., 2022)
Brazil	Niterói	18	35,987	6.7	0.1–101.6	(Ribeiro et al., 2022a)
<b>Brazil</b>	<b>Guarujá</b>	<b>9</b>	<b>24,781</b>	<b>15.4</b>	<b>0.9–50.6</b>	<b>Present study</b>
Brazil	Santos	18	13,052	17.6	3.5–42.1	(Ribeiro et al., 2022a)

The CBPI levels differed between *very low pollution* ( $\leq 1.0$ ), *low pollution* (1.1–2.5), *pollution* (2.6–5.0), *significant pollution* (5.1–7.5), *high pollution* (7.6–10.0) or *severe pollution* ( $\geq 10.0$ ), according to Torkashvand et al. (2021). In Guarujá, around 67.0% of the sampled areas had CBs leakage estimated as *severe pollution*. This percentage was higher in Santos (77.8%), while it was significantly smaller

( $\leq 20.0\%$ ) in the other six Brazilian (Niterói and Boiçucanga) and Iranian (Mazandaran, Qazvin, Behbahan and Jiroft) cities under similar conditions of sampling, i.e., in walkways and number normalized by density (Fig. 11). Thus, the neighbor cities of Santos and Guarujá possess alarming levels of CBs pollutants leakage. In this regard, local authorities should be concerned with this significant source of contamination, in order to plan and execute continuous monitoring and mitigating measures (Lima et al., 2021).



**Figure 11.** Cigarette butts Pollution Index (CBPI) classes percentages amongst worldwide studies available (Brazil and Iran only). Green to red colors indicates lowest to highest levels of cigarette butts (CBs) pollutants leakage.

### 3.3 Brands of Cigarette butts

A total of 3,521 CBs found in Guarujá still presented visibly identifiable physical characteristics, representing 81.5% of all collected butts (4,231) (Table 6). In this sense, this percentage was higher than other urban (Santos [77.7%] and Niterói [73.0%]) and coastal (Santos [53.9%] and Recife [33.1%]) environments (Table 6). Thus, the present study reached the best results for brands identification from CBs so far, probably due to elaboration of a visual guide containing all known legal and illegal brands, used to standardize identification procedures (Fig. S9 to S13). Although coastal monitoring result in higher number of collected CBs (10,275–17,845) than urban environments (900–8,611), the percentage of identifiable CBs is higher at urban (73.0–81.5%) rather than beaches (33.4–53.9%) (Table 6). Moreover, more brands may be found in urban (14–26) than coastal (18–22) environments (Table 6). Indeed, coastal environments tend to be harsher towards CBs degradation, and is possible that CBs present in coastal environments may be littered for a longer period of time than the ones found in urban environments (Silva et al., 2023).

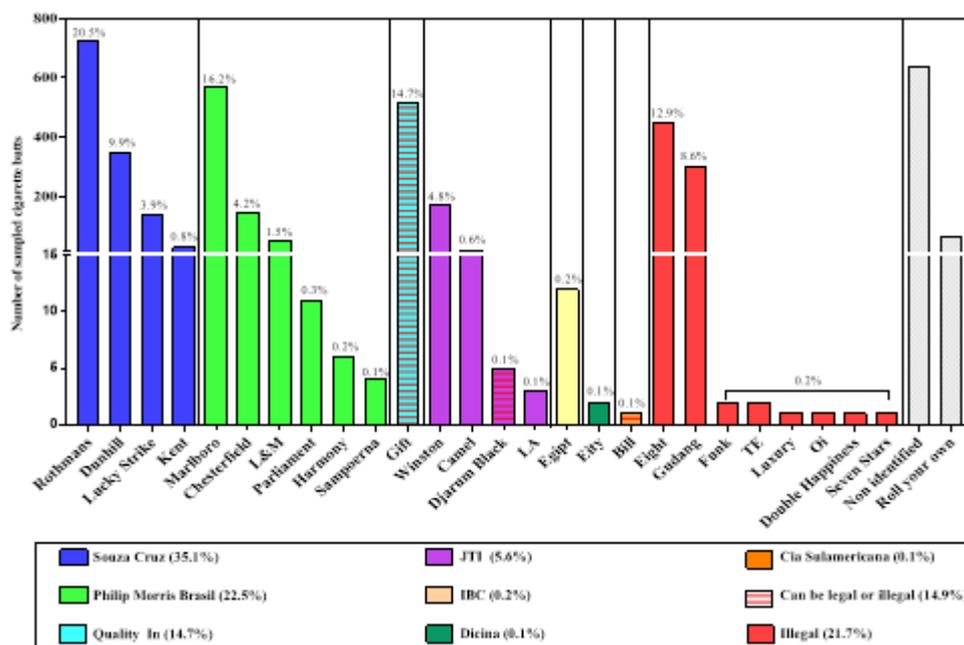
**Table 6.** Percentages and amounts of identifiable and unidentifiable cigarette brands among cigarette butts collected in coastal and urban of Brazil.

City	Guarujá	Niterói	Santos	Santos	Recife
Environment	Urban	Urban	Urban	Beach	Beach
Sampled area (m <sup>2</sup> )	24,781	35,987	13,052	1,600 <sup>a</sup>	1,425
Cigarette butts	4,321	900	8,611	17,845	10,275
Identifiable CBs	3,521	657	6,691	9,619	3,434
Identifiable CBs (%)	81.5	73.0	77.7	53.9	33.4
Unidentifiable CBs <sup>b</sup>	800	243	1,920	8,226	6,841
Unidentifiable CBs (%) <sup>b</sup>	18.5	27.0	22.3	46.1	66.6
Brands	26	14	26	22	18
Reference	Present study	(Ribeiro et al., 2022a)	(Ribeiro et al., 2022a)	(Lima et al., 2021)	(Silva et al., 2023)

a Meaning a 1,600 m of extension, rather than the area (m<sup>2</sup>).

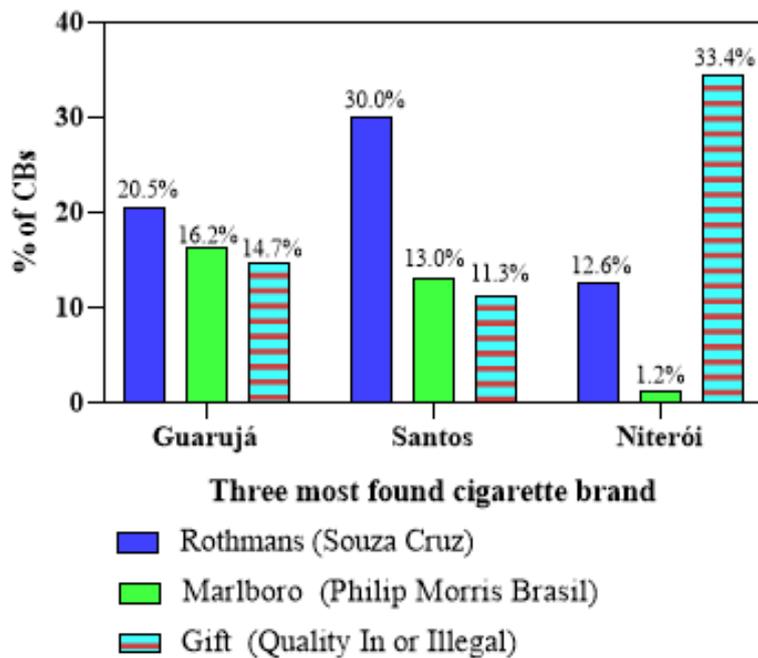
b Unidentifiable CBs due to advanced stage of degradation, plus the roll-your-own cigarettes.

A total of 17.0% CBs (Fig. 12) were in an advanced stage of degradation in Guarujá, and consequently was impossible to identify the brand (see Fig. S14 in supplementary material). This not included the Roll your Own cigarettes (1.5% in Guarujá), in which smokers purchase tobacco, filter and leaf separately (see Fig. S15 in supplementary material) making brand identification impossible (Santos et al., 2023). Indeed, CBs lose their physical identifiable characteristics because of environmental deterioration, caused mainly by the influence of radiation and rainfall. There are no experimental studies assessing degradation rates of CBs improperly discarded in urban areas nor evaluating the loss of identifiable physical characteristics. However, some types of designs printed in the cigarette may become unidentifiable quickly, while other designs may take longer. In Brazil, the cigarette companies are not obligated by governmental authorities to print any design in the cigarettes, but they must print it in cigarette packs. Thus, it should be a demand that companies include information also in cigarettes, as long as it does not become a marketing form (Smith et al., 2017), seeking to facilitate the CBs identification enabling reverse logistics approaches.



**Figure 12.** Percentages and amounts of cigarette brands identified among cigarette butts collected in urban areas of Guarujá.

In urban environments of Guarujá and Santos, the three main CBs brands found followed the same ranking (Rothmans > Marlboro > Gift). The percentages were similar in Guarujá and Santos for Marlboro (16.2% and 13.0%, respectively) and Gift (14.7% and 11.3%, respectively), but varied consistently for the Rothmans brand (20.5% and 30.0%, respectively) (Fig. 13). This shows a similar pattern in these neighbor cities (Guarujá and Santos), but with a higher tendency for Rothmans towards Santos. In the most distant city (Niterói), Rothmans was only the third most found brand (12.6%), while Gift was the most common (34.4%), with Dunhill in second (21.0%). In addition, Marlboro percentage in Niterói was quite smaller (1.2%). Thus, different brands dominate the cigarette marketing in different cities and states of Brazil (Drope et al., 2022).



**Figure 13.** Percentages and amounts of cigarette brands identified among cigarette butts collected in urban areas of Guarujá, Santo and Niteroi. Data from Santos and Niteroi were obtained by Ribeiro et al. (2022a).

There are nine companies allowed to sell 215 different cigarettes brands in Brazil. *Souza Cruz LTDA* is responsible alone for 110 of them, being the most found in Guarujá (29.3%) (see Fig. 8), accounting four brands (Rothmans, Dunhill, Lucky Strike and Kent). In second, *Phillip Morris Brasil LTDA* is responsible for the marketing 37 brands, and it was also the second in Guarujá (18.7%), accounting six brands (Marlboro, Chesterfield, L&M, Parliament, Harmony and Sampoerna). In addition, *Quality In LTDA* is responsible for the selling of five cigarettes brands in Brazil, but only the brand Gift made it the third most found company in Guarujá (12.2%). However, the Gift brand can be also marketed as illegal, as discussed in the next section. Finally, *IBC – Indústria Brasileira de Cigarros LTDA* and *Cia Sulamericana de Tabacos* companies were responsible for 0.2% of all CBs in Guarujá, accounting one brand each (Egipt and Bill, respectively). Indeed, identifying those percentages represents a key step in the reverse logistics application demand.

These companies profit with cigarette selling, but should also be responsible for the costs of controlling the pollution they generate (polluter pays principle), since the implementation of reverse logistics policies are mandatory according to the Brazilian National Solid Waste Plan (Brasil, 2010). Seeking their reuse or other environmentally adequate destination, this instrument involves a set of actions, procedures and measures enabling the collection and return of solid waste to the business sector. Thus, based on obtained data a proportional parcel of the costs with urban cleaning may be directed to cigarette companies as was recently implemented in Spain (Spain, 2023). This is necessary, considering that it was estimated in 2011 that “tobacco product litter” comprises 22 to 36% of all visible litter, and that removal costs range from \$3 million to \$16 million per year for major cities and municipalities (Schneider et al., 2011).

### 3.4 Illegal cigarette marketing (ICM)

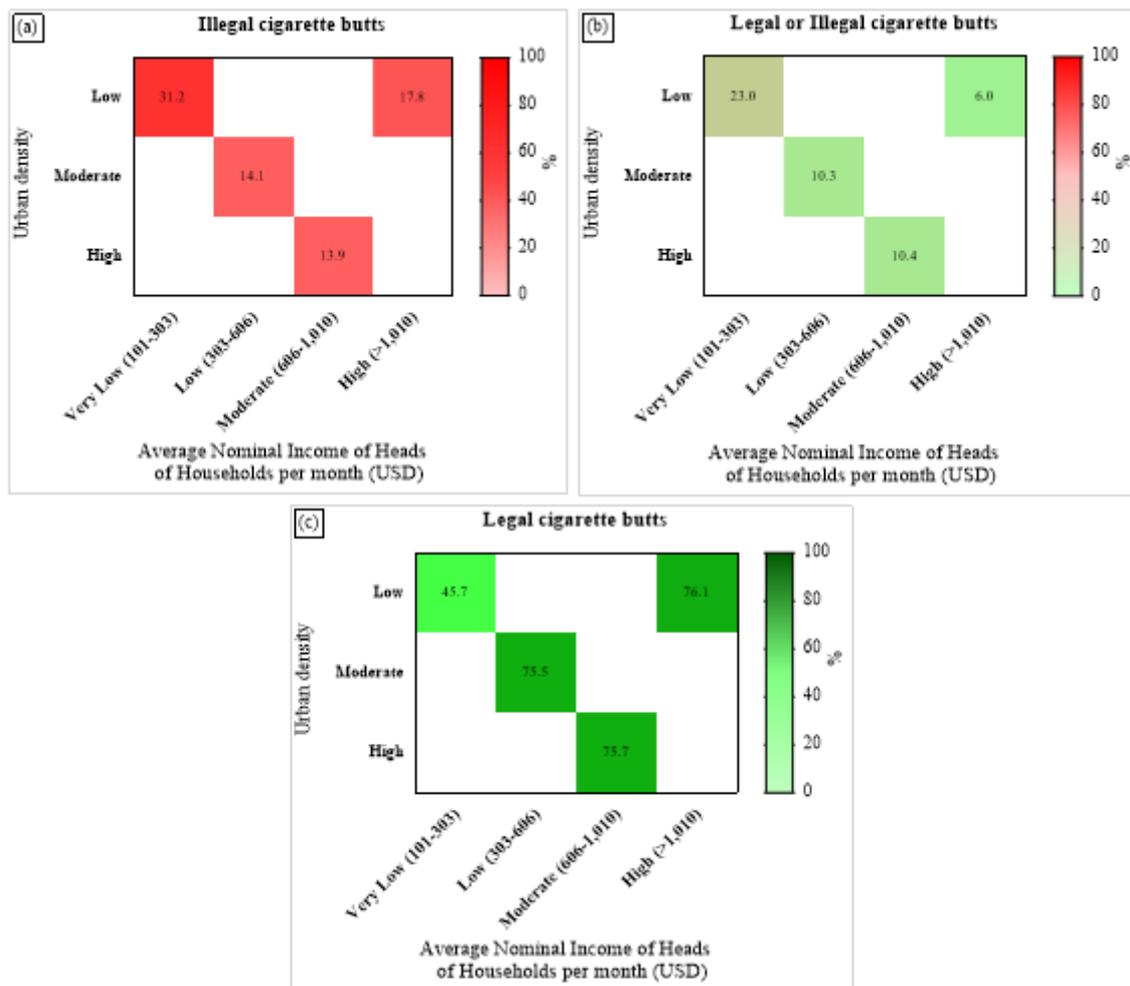
#### 3.4.1 Cigarette butts

The legality status analysis was based on the CBs with identifiable brands (3,521 out of the 4,321 CBs collected) (Table 7), which were checked in the ANVISA lists of known legal brands (Anvisa, 2021). Almost 15% of identifiable brands were listed as both Legal or Illegal (Bill, Djarum Black and especially Gift), since an accurate identification requires checking their packages and may not be concluded based only on littered CBs. For instance, Legal brands represented between 63.4 and 78.3%, while ICM ranged from 21.7 to 36.7% of all CBs collected in urban areas of Guarujá city.

**Table 7.** Percentages and amounts of cigarette brands legality status identified among cigarette butts collected in urban areas of Guarujá.

Cigarette butts legality status								
Sampled cigarette butts (4,231 butts)								
Identifiable brands - Considered (3,521 butts)			Unidentifiable brands - Not considered (645 butts)			Roll your own - Not considered (65 butts)		
Legal (63.4-78.3%)			Legal or Illegal* (14.9%)			Illegal (21.7-36.6%)		
Brand	%	Company	Brand	%	Company	Brand	%	Company
Rothmans	20.5		Gift	14.7	Dicina*	Eight	12.9	
Dunhill	9.9		Djarum Black	0.1	JTI*	Gudang	8.6	
Lucky Strike	3.9	Souza Cruz	Bill	0.1	Cia Sulamericana*	Fuak		
Kent	0.8		<b>TOTAL</b>	<b>14.9</b>		TE		Illegal
<b>TOTAL</b>	<b>35.1</b>					Luxury	0.2	
Marlboro	16.2					Oi		
Chesterfield	4.2					Seven Stars		
L&M	1.5	Phillip Morris Brasil				<b>TOTAL</b>	<b>21.7</b>	
Parliament	0.3					Gift	14.7	Dicina*
Harmony	0.2					Djarum Black	0.1	JTI*
Sampoerna	0.1					Bill	0.1	Cia Sulamericana*
<b>TOTAL</b>	<b>22.5</b>					<b>TOTAL</b>	<b>36.6</b>	
Gift	14.7	Quality In*						
Winston	4.8							
Camel	0.6							
Djarum Black	0.1	JTI*						
LA	0.1							
<b>TOTAL</b>	<b>5.6</b>							
Egypt	0.2	IBC						
Eity	0.1	Dicina						
Bill	0.1	Cia Sulamericana*						

Despite the urban density level, the ICM was higher at poorer areas (31.2%, up to 54.2% when adding the *Legal or Illegal brands*) (Fig. 14a and b), i.e., areas of very low-income (101–303 USD per month). In other *low* (>1,010 USD per month), *moderate* (303–606 USD per month) and *high* (606–1,010 USD per month) income levels, the ICM was similarly significantly smaller (17.8 to 23.8%, 14.1 to 24.3% and 13.9 to 24.3%, respectively) (Fig. 14a) than *very low* income. In this sense, differently from the CBDs and the CBPI (see Fig. 7 and Fig. 10), the ICM seems to be mostly dependent of the level of income, rather than urban density levels since poor areas may have more than the double of ICM than other areas.



**Figure 14.** Illegal (a), Illegal or Legal (b) and legal cigarette marketing rates (c) measured by using cigarette butts of different urban density and income (Average Nominal Income of Heads of Households [ANIH]) levels.

CBs were used as a method to identify the ICM in Sri Lanka (Morais et al., 2018), Brazil (Ribeiro et al., 2022a; Silva et al., 2023) and especially in Canada (Barkans and Lawrance, 2013; CCSA, 2009; OCSA, 2017, 2013; Stratton et al., 2016). Indeed, the ICM is measured with CBs in Canada since 2007, with an average between 9.7% and 58.7% in 2013 and 2014 in Ontario and Quebec cities (Table 8). In Sri Lanka, the ICM average was 15.6%, ranging from 10.4% to 25.7% in six different districts (23 different zones) in 2018. In Brazil, our previous study found that the ICM could reach 25.2% and 36.8% in Santos and Niterói walkways, respectively (Table 7), as in the first study in the country of this sort. Additionally, a study held in Boa Viagem beach located at Recife city by Silva et al. (2023), pointed out 22.0% of the ICM rate. In present study, Guarujá may reach 36.6% of illegal market.

**Table 8.** Illegal cigarette marketing percentage among collected cigarette butts in worldwide assessments.

Country	Location	Sample year	Illegal cigarette market (%)	Reference
Sri Lanka	23 zones within six districts	2018	15.6 (10.4–25.7)	(Morais et al., 2018)
Canada	Quebec and Ontario high schools	2007	36.0	(CCSA, 2009)
Canada	Ontario universities and colleges	2009	14.0	(Barkans and Lawrance, 2013)
Canada	Ontario - 136 public smoking areas	2013	21.0 (10.5–46.0)	(OCSA, 2013)
Canada	Ontario - 135 public smoking areas	2014	22.3 (16.7–27.0)	(OCSA, 2017)
Canada	Ontario – Peel	2013–4	9.7	(Stratton et al., 2016)
Canada	Ontario - Brantford	2013–4	58.1	(Stratton et al., 2016)
Canada	Ontario - 135 public smoking areas	2015	24.6 (15.3–28.4)	(OCSA, 2017)
Canada	Ontario - 135 public smoking areas	2016	32.8 (21.0–54.2)	(OCSA, 2017)
Canada	Ontario - 135 public smoking areas	2017	37.2 (22.9–60.4)	(OCSA, 2017)
Brazil	Recife’s beach of Boa Viagem	2021–22	22.0	(Silva et al., 2023)
Brazil	Santos urban walkways	2022	≤25.2	(Ribeiro et al., 2022a)
Brazil	Niterói urban walkways	2022	≤36.8	(Ribeiro et al., 2022a)
Brazil	Guarujá urban walkways	2023	21.7–36.6	Present study

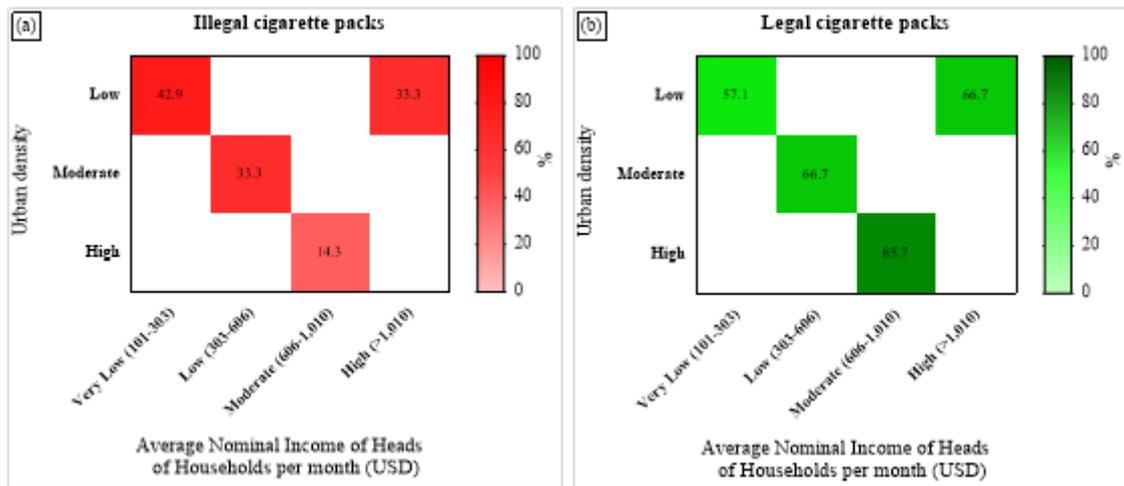
### 3.4.2 Cigarette packs

A total of ten brands were found among 30 CPs collected in Guarujá. The main aspects searched in CPs were country of origin (1), the presence and type of health warnings in Portuguese (2) and the presence of stamps from Brazilian regulating authorities (3) (Table 9). By these parameters, the ICM rate was 33.3% in Guarujá, accounting three brands, i.e., Eight, Gift and Gudang (Table 9). All legal CPs came from Brazil, while illegal Eight and Gift brands were from Paraguay, and Gudang from Indonesia. In a previous study performed in five Brazilian cities, approximately 95% of all Gift CPs were illegal (Szklo et al., 2020). Similarly, 100% of Gift CPs were illegal in the present study. Such evidences suggests that most or CBs of this brand found in the present study may be illegal. The health warnings were present in all CPs, but those legalized had standardized images and captions written in Portuguese, while illegal had written captions in Spanish and Indonesian. However, in one Marlboro CP, only the top portion was found in the site S8. Thus, the printed information was lost and was not possible to check its legality status.

**Table 9.** Illegal cigarette marketing percentage among collected cigarette packs in Guarujá.

Legal cigarette packs (66.7%)					Illegal cigarette packs (33.3%)				
Brand	Country of origin	Health warnings Portuguese	Stamp	Site	Brand	Country of origin	Health warnings Portuguese	Stamp	Site
Dunhill	Brazil	Yes	Yes	S1	Eight	Paraguay	No	No	S7
Dunhill	Brazil	Yes	Yes	S9	Eight	Paraguay	No	No	S8
Dunhill	Brazil	Yes	Un	S4	Eight	Paraguay	No	No	S8
Eity	Brazil	Yes	Yes	S4	Gift	Paraguay	No	No	S1
Lucky Strike	Brazil	Yes	Yes	S1	Gift	Paraguay	No	No	S9
Marlboro	Brazil	Yes	Yes	S1	Gift	Paraguay	No	No	S8
Marlboro	Brazil	Un	Yes	S8	Gift	Paraguay	No	No	S8
Marlboro	Brazil	Yes	Yes	S8	Gudang	Indonesia	No	No	S4
Marlboro	Brazil	Yes	Yes	S8	Gudang	Indonesia	No	No	S9
Marlboro	Brazil	Yes	Yes	S9	Gudang	Indonesia	No	No	S7
Marlboro	Brazil	Yes	Un	S9					
Marlboro	Brazil	Yes	Un	S9					
Rothmans	Brazil	Yes	Yes	S3					
Rothmans	Brazil	Yes	Yes	S7					
Rothmans	Brazil	Yes	Yes	S8					
Rothmans	Brazil	Yes	Yes	S8					
San Marino	Brazil	Yes	Un	S7					
Winston	Brazil	Yes	Un	S8					
Winston	Brazil	Yes	Yes	S1					
Winston	Brazil	Yes	Yes	S1					

The ICM measured based on cigarette packs was higher at *very low*-income level (42.9%) and smaller at *moderate*-income (14.3%) (Fig. 15a). The same patter was seen in the ICM rates measured by cigarette butts, in which *very low*-income level and low urban density areas had higher ICM rates (see Fig. 14a). Indeed, the trend is to be higher at poor areas (Szklo et al., 2020). The number of CPs collected may have been too small in the present study, but it was already able to get this level of geographic comparison.



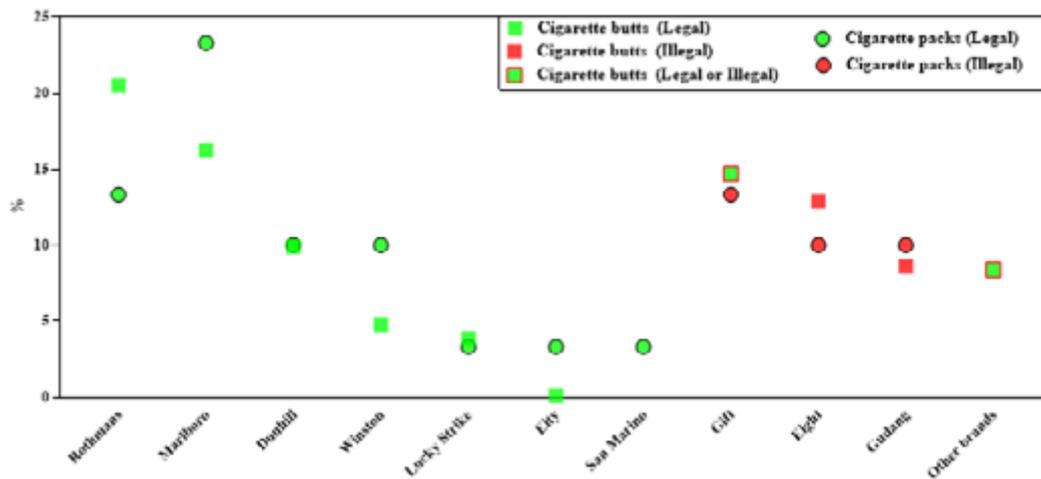
**Figure 15.** Illegal (a) and legal cigarette marketing rates (b) measured by using cigarette packs of different urban density and income (Average Nominal Income of Heads of Households [ANHH]) levels.

### 3.4.3 Cigarette butts and packs viability for future assessments

The methods already used to estimate the ICM levels collected CBs and CPs in urban areas, CPs found in urban garbage bins (Szklo et al., 2020), bought in dummy purchases (Shakya et al., 2023), surveys by phone or in person (Juarez et al., 2021; McDonnell et al., 2021). In addition, media and news reports monitoring (Evans-Reeves et al., 2020), online selling inspections (van der Zee et al., 2023) and tax gap analysis are also used (Goodchild et al., 2020). Most of those methods, present disadvantages and uncertainties. Results generated by a single method need to consider that different countries have distinct number of people who smoke, tobacco taxes and regulations, before comparing obtained outputs. Additionally, different methods may not be directly comparable, despite the possibility of cross-validation during simultaneously applications (Joossens et al., 2014; Juarez et al., 2021; Szklo et al., 2020). In the

present study, we found that CBs and CPs found in urban walkways may be directly comparable, generating one single range of the ICM. The ICM estimated from CBs was 21.7% to 36.6%. Using CPs, the ICM (33.3%) was inside that range. In addition, several other similarities were seen when comparing the CBs and CPs brands.

The ten brands found in CPs and CBs are presented, along with their percentages, in Figure 12. Indeed, the difference between the percentages of each CBs and CPs brand only reached around 7.0% for the two most found brands, i.e., Rothmans (7.2%) and Marlboro (7.1%, respectively). Then, this difference was smaller in the rest of the legal (5.2% for Winston, 3.2% for Eity, 0.6% for Lucky Strike and 0.1% for Dunhill) and illegal brands (2.9% for Eight, 1.4% for Gift and Gudang). Interestingly, the legal San Marino brand was found only amongst CPs (3.3%). Finally, other 15 brands were found only as CBs, adding 8.4%. Therefore, several similarities are observed when comparing brands of cigarettes using CBs and CPs found in urban environments to determine the ICM, despite the low number of CPs found. More legal and illegal brands will be seen in CBs (25 and 8, respectively) (see Fig. 8) than CPs (10 and 3, respectively) (Fig. 12), but the differentiation of brands marketed as legal and illegal is more certain in CPs and can leave a level of uncertainty of 14.9% when using CBs.



**Figure 16.** Cigarette butts and cigarette packs comparison of legal and illegal brands percentages.

The quite high deviation, i.e., level of uncertainty using CBs, is due to presence of brands that can be distinguished between legal and illegal (Djarum Black, Bill and mainly Gift). Indeed, 14.7% of all CBs were Gift, classified as ‘legal or illegal’ when using CBs because of the similar design legal and illegal Gift possess (Fig. 17). However, CPs has different design, and all Gift CPs were illegal, smuggled from Paraguay (see Table 8). Additionally, it may be possible that other brands are also marketed as legal and illegal, but there is a lack of available research on the topic. Thus, the level of uncertainty of the illegal market of CBs dropped from 14.9% to 0.2% if we assume that all Gift CBs are also illegal.



**Figure 17.** Legal and illegal cigarette packs and cigarette units of Gift brand.

To each CP found in the present study, there were 141 CBs, showing a different spatial dynamic of discarding. Thus, future monitoring assessments simultaneously using CBs and CPs should preferentially apply different spatial approaches. In this regard, for each m<sup>2</sup> monitored for CBs occurrence, at least 150 m<sup>2</sup> should be inspected for CPs. Alternatively, separate sampling campaigns can also be planned considering such residues. In the field, sampling higher quadrants for CPs is feasible because lower occurrence. In this perspective, were sampled 6.3 km (23,703 m<sup>2</sup>) in Guarujá for CBs and CPs which is accepted for CBs according to previous studies (Nasab et al., 2022; Ribeiro et al., 2022a; Torkashvand et al., 2021). However, for less frequent items like CPs, the sampled area could be higher. Indeed, studies monitoring the face masks (connected

with COVID-19 pandemic) sampled on total 137,5 km (Cueva, 2023) and 250 km (France, 2022). Therefore, future studies should monitor CBs and CPs, applying simultaneously different methodological procedure, i.e., a higher area for CPs and smaller for CBs, and enabling cross-validation estimates of the illicit market.

The tobacco industry has always been against anti-smoking legislative and educational measures by using the illicit trade narrative as an argument. Often, the industry tends to exaggerate and misrepresent the ICM estimates (Paraje et al., 2022). In this sense, studies of independent sources are important to refute industry's miss-use of the ICM. Our findings may also enable opportunities in Brazil and other countries to assertively implement article 15 of the WHO Framework Convention on Tobacco Control and the Protocol to Eliminate Illicit Trade in Tobacco Products (WHO, 2003). In addition, they reinforce the importance for low-and-middle income countries to implement tobacco control measures to reduce the burden and prevalence of tobacco use. Such countries cannot afford and/or manage the health, social, economic or environmental consequences of tobacco use. In Brazil, it is estimated that approximately 162 thousand people die annually from smoking, representing 125 billion BRL (>25 billion USD) per year in direct and indirect costs (IECS, 2020). Thus, decreasing the overall population of smokers (either those who smoke legal or illegal cigarettes), and consequently, the CBs contamination, is a critical step to achieving sustainable development goals, such as ensure healthy lives and conserve the ocean, seas and marine resources for this and the next generations (Framework Convention Alliance, 2015; UN, 2016).

### 3.5 Cigarette butt experiments

Although limited data exist on the environmental occurrence of CBs in natural and urban environments, their high frequency in walkways (Ribeiro et al., 2022a) and beaches (Araújo and Costa, 2021) raises serious concerns due to potential mobility of associated chemicals across different environmental compartments (Mandelli et al., 2022). Indeed, cigarette filters contain highly toxic substances that pose ecological risks, being classified as hazardous waste (Green et al., 2023). Regarding  $\Sigma$ PAHs (sum of polycyclic aromatic hydrocarbons), a total of 1,069.8 ng L<sup>-1</sup> were leached from four CBs into water during a 24-hour experiment. Among the PAHs found in water leachates, Naphthalene (743.9 ng L<sup>-1</sup>), C2-Naphthalene (146.7 ng L<sup>-1</sup>) and C1-Naphthalene (82.5 ng L<sup>-1</sup>) were predominant, while other compounds were present at lower levels (Table 10). Similar findings were reported by Dobaradaran et al. (2019), suggesting that lighter PAHs with higher volatility and water solubility are easily released from CBs, contaminating water bodies and soils. Naphthalene and their metabolites have been pointed out as cytotoxic compounds inducing chromosomal damages, increased incidence of alveolar/bronchial adenomas, inflammation, tissue damages and potential health hazard over humans and animal species (Schreiner, 2011). Furthermore, the environmental impacts associated related to other PAHs exposure have been extensively documented in scientific literature (Chiovatto et al., 2021). After release into aquatic systems, CBs float for approximately 3 to 20 days before settling into sediments layers (Lima et al., 2021). Similarly, CBs littered on soils are susceptible to leaching by rainwater and runoff. Consequently, several groups of contaminants, including PAHs, are leached from CBs into the water column and soils, potentially causing negative effects, as demonstrated by recent ecotoxicology studies (Lima et al., 2021).

It is worth noting that different environmental conditions (e.g., rainfall regime, soil type, temperature, pH) may influence the intensity of leaching and, consequently, the environmental damage caused by littered CBs. Nevertheless, considering a total of 4,321 CBs found in the present study and the average experimentally determined PAHs levels ( $1,069.8 \text{ ng L}^{-1}$ ), approximately 1,155,652 ng of PAHs would be leached into the soil of the studied areas in just 24 hours. Thus, significant amounts of toxic compounds are expected to reach the natural environment certainly inducing deleterious effects on living organisms. In fact, a recent study pointed out that leached PAHs from CBs may exceed the standards set adopted by Water Framework Directive into force in the European Union, thereby affecting simultaneously aquatic organisms and potentially also humans (Dobaradaran et al., 2019).

Compound	Concentrations (ng.L <sup>-1</sup> )	Element	Concentrations (µg L <sup>-1</sup> )
Naphthalene	743.9 ± 84.1	As	5.4 ± 0.2
C1-Naphthalene	82.5 ± 6.5	Cd	ND
C2-Naphthalene	146.7 ± 15.6	Co	7.8 ± 0.1
Acenaphthylene	14.8 ± 2.9	Cr	ND
Acenaphthene	3.3 ± 0.6	Cu	0.8 ± 0.1
C3-Naphthalene	30.5 ± 4.4	Fe	39.8 ± 1.1
Fluorene	5.6 ± 1.0	Mg	ND
Phenanthrene	24.9 ± 3.5	Mn	0.2 ± 0.0
Anthracene	5.1 ± 0.4	Ni	0.2 ± 0.0
Fluoranthene	<LQ	Pb	0.1 ± 0.0
Pyrene	<LQ	Zn	2.3 ± 0.3
C1-Pyrene	3.7 ± 0.5		
Benz[a]anthracene	2.6 ± 0.6		
Chrysene	<LQ		
Benzo[b]fluoranthene	ND		
Benzo[k]fluoranthene	ND		
Benzo[e]pyrene	ND		
Benzo[a]pyrene	6.1 ± 0.5		
Perylene	ND		
Indeno[123cd]pyrene	ND		
Dibenz[ah]anthracene	<LQ		
Benzo[ghi]perylene	ND		
Sun of PHAs	1069,8		

**Table 10.** Mean and standard deviation of concentrations of individual and total Polycyclic Aromatic Hydrocarbons (ng L<sup>-1</sup>) and chemical elements (µg L<sup>-1</sup>) measured in leachates of cigarette butts prepared in ultrapure water. <LD= below quantification limit, ND= not detected

The levels of arsenic (As), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), lead (Pb), nickel (Ni) and zinc (Zn) were identified in the analyzed leachates, while cadmium (Cd), chromium (Cr) and magnesium (Mg) were not detected (see Table 10). As reported for PAHs, the leaching of metals from BCs into aqueous media can be influenced by several environmental factors. Moreover, chemical

elements often exhibit resistance to biological and photodegradation, resulting in constant leaching rates for longer periods compared to organic microcontaminants (Castro, 2019). In this regard, consistent concentrations, i.e., of similar magnitude order, were reported for Cd, As, Cu, Fe, Mn, Zn and Ni in littered CBs collected along the northern part of the Persian Gulf in Bushehr coastal areas (Dobaradaran et al., 2017). A study assessing potential metals leak from littered CBs, estimated an average of 150 CBs/km/month in road areas generating between 0.02 and 1.7 mg of toxic metals per km by month (Torkashvand et al., 2020). Such findings indicate that CBs can serve as sources of toxic metals to natural environments, presenting a significant and persistent threat to both urban and natural environments.

It is important to highlight that our results were based on a limited number of toxic substances that can be leached from CBs. However, it is crucial to acknowledge that a single CB contains thousands of hazardous substances, which were not analyzed in the present study. These substances include benzene, hydrogen cyanide, pyridine, and nicotine (Green et al., 2014). Their environmental occurrence may pollute up to 1000 liters of water (Green et al., 2014). Thus, CBs represent a serious threat to water quality including drinking and natural waters. In this regard, leachates from CBs contribute to the contamination of aquatic systems and soils by toxic compounds, including polycyclic aromatic hydrocarbons (PAHs) and toxic metals. These compounds have well-documented adverse effects on various organisms, possibly leading to ecological imbalances and potential harm to human health. Such risks extend beyond urban walkways and beaches, highlighting the need for comprehensive management strategies to address CB pollution at their sources. Moreover, further research is warranted to explore the full range of hazardous substances present in CBs and their leachate potential, enabling better assessment and management of the environmental risks they pose. In conclusion, our study provides valuable

insights into the leachate potential of CBs, highlighting their significant contribution to environmental contamination. The findings underscore the urgent need for proactive measures to mitigate CB pollution, protect water resources, and safeguard the well-being of both ecosystems and human populations.

## 4. Conclusions

A total of 4,321 cigarette butts (CBs) were found in 23,694 m<sup>2</sup> of Guarujá urban areas. The CBs density in Guarujá ( $0.18 \pm 0.17$  CBs.m<sup>-2</sup>), Santos ( $0.25 \pm 0.17$  CBs.m<sup>-2</sup>) and Niterói ( $0.08 \pm 0.21$  CBs.m<sup>-2</sup>) were not compatible with the ranking of urban cleaning status of Brazilian cities, indicating that monitoring programs directed to CBs must be carried out by local authorities. In addition, CBs contamination in urban areas seems unrelated to population densities. This pattern, previously observed in Santos and Niterói was also seen in Guarujá. On the other hand, the density of CBs in Guarujá was positively related to the number of commercial buildings, stores selling cigarette packs and units and benches in the studied areas. Interestingly, no correlations were seen for number of residential buildings, manholes, bus stops, garbage bins and local income.

The CBs contaminants leakage in Guarujá was overall severe (CBPI= $15.4 \pm 11.5$ ), and higher in zones under *low urban density* (CBPI= $26.6 \pm 12.0$ ). Areas with *high* (CBPI= $12.4 \pm 4.6$ ) and *moderate urban density* (CBPI= $7.1 \pm 5.5$ ) presented significantly lower CBPI. Thus, low urban density zones of Guarujá potentially contributes more with contaminants leakage from CBs. On contrary, the low urban density zones contribute less to CBPI in Santos, while in Niterói zones presenting different urban densities contributed similarly. In this regard, the average CBPI in Guarujá (15.4) was more than twice higher than six other urban areas from Brazil and Iran, and smaller than Santos (17.6). The Guarujá maximum (50.6) CBPI gets only

behind Niterói (101.6). In addition, the severe pollution represented 67.0% of sampled areas in Guarujá, which was higher in Santos (77.8%), and smaller ( $\leq 20.0\%$ ) in the Brazilian and Iranian cities. Thus, Santos and Guarujá presented alarming potential pollutants leakage from CBs. Local authorities should be concerned regarding this significant contamination source and carry out continuous monitoring and mitigating measures.

A total of 3,521 (83.3%) CBs allowed brand identification based on physical characteristics. This was the highest percentual reached by studies adopting similar methodological approach. The main cigarette brands collected in Guarujá (Rothmans, Marlboro and Gift) were the same found in Santos, considering both, urban and coastal environments. However, they were different from Niterói. The manufacturers of most brands were *Souza Cruz LTDA* (29.3%) and *Phillip Morris Brasil LTDA* (19.7%), which should pay proportional cost of urban cleaning, according to reverse logistic policies issued by National Solid Waste Plan of Brazil. In Guarujá, the illegal cigarette marketing (ICM) ranged from 21.7% to 36.7%, which was higher than Santos and similar to Niterói. In Brazil, a moderate deviation is expected using methods based in CBs collection, because complete information is printed only in cigarette packs and similar designs are seen in legal and illegal CBs. Despite uncertainties, there are evidence that CBs collection and identification is a proper method to estimate ICM, due to easy collection in high amounts providing a representative sample of cigarette consumption. However, a most accurate way to estimate ICM is probably the simultaneous use of CPs and CBs allowing a cross-validation among distinct methods. Nevertheless, in our concern such approach is scarce, and should be the objectives of future studies.

The chemical analysis of leachates from cigarette butts (CBs) provided valuable insights about potential environmental impacts particularly in relation to polycyclic aromatic hydrocarbons (PAHs)

and toxic metals. Our findings revealed that CBs serve as a significant source of PAH contamination, with high levels of naphthalene, C2-Naphthalene, and C1-Naphthalene in the water leachates. These PAHs are known to possess cytotoxic properties, raising concerns about potential adverse health effects on both humans and wildlife. Furthermore, our study highlighted the leaching of metals from CBs into the environment. While certain metals such as arsenic (As), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), lead (Pb), nickel (Ni), and zinc (Zn) were detected in the analyzed leachates, cadmium (Cd), chromium (Cr), and magnesium (Mg) were below detectable levels. The presence of these elements underscores the potential for CBs to contribute to metal contamination in soils and water bodies. Overall, our results emphasize the significant environmental implications associated with CBs, as they serve as sources of both PAHs and toxic elements. These findings raise concerns about the ecological risks and potential health hazards posed by CBs as hazardous waste. It is crucial to address and mitigate the environmental contamination caused by CBs to safeguard ecosystems and human well-being.

## References

Akhbarizadeh, R., Dobaradaran, S., Parhizgar, G., Schmidt, T.C., Mallaki, R., 2021. Potentially toxic elements leachates from cigarette butts into different types of water: A threat for aquatic environments and ecosystems? *Environ. Res.* 202, 111706. <https://doi.org/10.1016/j.envres.2021.111706>

Anvisa, 2021. Consulta a registro - Relação das Marcas de Cigarros [WWW Document]. URL [http://antigo.anvisa.gov.br/en\\_US/tabaco/consulta-a-registro](http://antigo.anvisa.gov.br/en_US/tabaco/consulta-a-registro) (accessed 12.8.22).

Araújo, M.C.B. de, Costa, M.F. da, 2021. Cigarette butts in beach litter: Snapshot of a summer holiday. *Mar. Pollut. Bull.* 172, 112858. <https://doi.org/10.1016/j.marpolbul.2021.112858>

Araújo, M.C.B., Costa, M.F., 2019. From Plant to Waste: The Long and Diverse Impact Chain Caused by Tobacco Smoking. *Int. J. Environ. Res. Public Health* 16, 2690. <https://doi.org/10.3390/ijerph16152690>

Araújo, M.C.B., Costa, M.F., Silva-Cavalcanti, J.S., Duarte, A.C., Reis, V., Rocha-Santos, T.A., da Costa, J.P., Girão, V., 2022. Different faces of cigarette butts, the most abundant beach litter worldwide. *Environ. Sci. Pollut. Res.* <https://doi.org/10.1007/s11356-022-19134-w>

Arevalo, R., Corral, J.E., Monzon, D., Yoon, M., Barnoya, J., 2016. Characteristics of illegal and legal cigarette packs sold in Guatemala. *Glob. Health* 12, 78. <https://doi.org/10.1186/s12992-016-0219-z>

Barkans, M., Lawrance, K., 2013. Contraband tobacco on post-secondary campuses in Ontario, Canada: analysis of discarded cigarette butts. *BMC Public Health* 13, 1–8. <https://doi.org/10.1186/1471-2458-13-335>

Brasil, 2018. Marcas e preços de venda a varejo de cigarros [WWW Document]. *Receita Fed.* URL <https://www.gov.br/receitafederal/pt-br/assuntos/orientacao-tributaria/regimes-e-controles-especiais/cigarros-marcas-e-precos-de-venda-a-varejo-de-cigarros> (accessed 4.26.23).

Brasil, 2010. Política Nacional de Resíduos Sólidos. Lei no 12.305, de 2 de agosto de 2010. [http://www.planalto.gov.br/ccivil\\_03/\\_ato2007-2010/2010/lei/l12305.htm](http://www.planalto.gov.br/ccivil_03/_ato2007-2010/2010/lei/l12305.htm).

Brown, J., Welding, K., Cohen, J.E., Cherukupalli, R., Washington, C., Ferguson, J., Clegg Smith, K., 2017. An analysis of purchase price of legal and illicit cigarettes in urban retail environments in 14 low- and middle-income countries. *Addiction* 112, 1854–1860. <https://doi.org/10.1111/add.13881>

Castro, Í.B., 2019. Improper environmental sampling design bias assessments of coastal contamination. *Trends Environ. Anal. Chem.* 24, e00068. <https://doi.org/10.1016/j.teac.2019.e00068>

CCSA, 2009. Youth Contraband Tobacco Study. Canadian Convenience Stores Association.

Chiovatto, A.C.L., de Godoi, A.V.O., Zanardi-Lamardo, E., Duarte, F.A., DelValls, T.Á., Pereira, C.D.S., Castro, Í.B., 2021. Effects of substances released from a coal tar-based coating used to protect harbor structures on oysters. *Mar. Pollut. Bull.* 166, 112221. <https://doi.org/10.1016/j.marpolbul.2021.112221>

Cueva, A., 2023. Temporal considerations for an effective sampling of personal protective equipment litter derived from the COVID-19 pandemic. *Sci. Total Environ.* 858, 160047. <https://doi.org/10.1016/j.scitotenv.2022.160047>

Curti, D., Shang, C., Chaloupka, F.J., Fong, G.T., 2019. Tobacco taxation, illegal cigarette supply and geography: findings from the ITC Uruguay Surveys. *Tob. Control* 28, s53–s60. <https://doi.org/10.1136/tobacco-control-2017-054218>

Cutter, S.L., Tiefenbacher, J., Birnbaum, S., Wiley, J., Solecki, W.D., 1991. Throwaway societies: a field survey of the quantity, nature and distribution of litter in New Jersey. *Appl. Geogr.* 11, 125–141. [https://doi.org/10.1016/0143-6228\(91\)90039-C](https://doi.org/10.1016/0143-6228(91)90039-C)

Darabi, K., Hassani, G., Alinejad, N., Badeenezhad, A., 2023. Spatial and temporal variation of CBPI and leakage of heavy metals from cigarette butts into the urban environment. *Sci. Rep.* 13, 1424. <https://doi.org/10.1038/s41598-023-28340-6>

de Granda-Orive, J.I., Girón-Matute, W., López-Yepes, L., 2016. Cigarette Butts: The Collateral Effects of Cigarettes on Humans, Animals and the Environment. *Arch. Bronconeumol. Engl. Ed.* 52, 285. <https://doi.org/10.1016/j.arbr.2016.03.007>

Ding, Y.S., Ashley, D.L., Watson, C.H., 2007. Determination of 10 Carcinogenic Polycyclic Aromatic Hydrocarbons in Mainstream Cigarette Smoke. *J. Agric. Food Chem.* 55, 5966–5973. <https://doi.org/10.1021/jf070649o>

Dobaradaran, S., Nabipour, I., Saeedi, R., Ostovar, A., Khorsand, M., Khajeahmadi, N., Hayati, R., Keshtkar, M., 2017. Association of metals (Cd, Fe, As, Ni, Cu, Zn and Mn) with cigarette butts in northern part of the Persian Gulf. *Tob. Control* 26, 461–463. <https://doi.org/10.1136/tobaccocontrol-2016-052931>

Dobaradaran, S., Schmidt, T.C., Kaziur-Cegla, W., Jochmann, M.A., 2021. BTEX compounds leachates from cigarette butts into water environment: A primary study. *Environ. Pollut.* 269, 116185. <https://doi.org/10.1016/j.envpol.2020.116185>

Dobaradaran, S., Schmidt, T.C., Lorenzo-Parodi, N., Jochmann, M.A., Nabipour, I., Raeisi, A., Stojanović, N., Mahmoodi, M., 2019. Cigarette butts: An overlooked source of PAHs in the environment? *Environ. Pollut.* 249, 932–939. <https://doi.org/10.1016/j.envpol.2019.03.097>

Dobaradaran, S., Schmidt, T.C., Nabipour, I., Ostovar, A., Raeisi, A., Saeedi, R., Khorsand, M., Khajeahmadi, N., Keshtkar, M., 2018. Cigarette butts abundance and association of mercury and lead along the Persian Gulf beach: an initial investigation. *Environ. Sci. Pollut. Res.* 25, 5465–5473. <https://doi.org/10.1007/s11356-017-0676-9>

Dorfman, A., França, A.B.C., França, R.F., 2017. Political Commodities and Sovereignty Management: Cigarette Smuggling across Brazil's Southern Borders. *Geopolitics* 22, 863–886.

Drope, J., Figueiredo, V.C., Iglesias, R., Szklo, A.S., Borges, P., Stoklosa, M., 2022. Consumo de cigarros ilegais em cinco cidades brasileiras. *Rio Jan. Cent. Estud. Sobre Tab. E Saúde Esc. Nac. Saúde Pública Fundação Oswaldo Cruz.*

Evans-Reeves, K., Hatchard, J., Rowell, A., Gilmore, A.B., 2020. Illicit tobacco trade is 'booming': UK newspaper coverage of data funded by transnational tobacco companies. *Tob. Control* 29, e78–e86. <https://doi.org/10.1136/tobaccocontrol-2018-054902>

Framework Convention Alliance, 2015. Fact Sheet - Tobacco: a Barrier to Sustainable Development (2015) [WWW Document]. URL <https://fctc.org/resource-hub/fact-sheet-tobacco-a-barrier-to-sustainable->

-development/

France, R.L., 2022. First landscape-scale survey of the background level of COVID-19 face mask litter: Exploring the potential for citizen science data collection during a 'pollution pilgrimage' of walking a 250-km roadside transect. *Sci. Total Environ.* 816, 151569. <https://doi.org/10.1016/j.scitotenv.2021.151569>

GBD Tobacco, 2021. Spatial, temporal, and demographic patterns in prevalence of smoking tobacco use and attributable disease burden in 204 countries and territories, 1990–2019: a systematic analysis from the Global Burden of Disease Study 2019. *The Lancet* 397, 2337–2360. [https://doi.org/10.1016/S0140-6736\(21\)01169-7](https://doi.org/10.1016/S0140-6736(21)01169-7)

Gholami, M., Torkashvand, J., Rezaei Kalantari, R., Godini, K., Jonidi Jafari, A., Farzadkia, M., 2020. Study of littered wastes in different urban land-uses: An 6 environmental status assessment. *J. Environ. Health Sci. Eng.* 18, 915–924. <https://doi.org/10.1007/s40201-020-00515-7>

Gigliotti, A., Figueiredo, V.C., Madruga, C.S., Marques, A.C., Pinsky, I., Caetano, R., e Silva, V.L. da C., Raw, M., Laranjeira, R., 2014. How smokers may react to cigarette taxes and price increases in Brazil: data from a national survey. *BMC Public Health* 14, 327. <https://doi.org/10.1186/1471-2458-14-327>

Gomersall, B., 2022. Big tobacco in Latin America. *Lancet Respir. Med.* 10, 735–736. [https://doi.org/10.1016/S2213-2600\(22\)00252-1](https://doi.org/10.1016/S2213-2600(22)00252-1)

Goodchild, M., Paul, J., Iglesias, R., Bouw, A., Perucic, A.-M., 2022. Potential impact of eliminating illicit trade in cigarettes: a demand-side perspective. *Tob. Control* 31, 57–64. <https://doi.org/10.1136/tobacco-control-2020-055980>

Goodchild, M., Valavan, T., Sinha, P., Tullu, F.T., 2020. Estimating illicit cigarette consumption using a tax-gap approach, India. *Bull. World Health Organ.* 98, 654–660. <https://doi.org/10.2471/BLT.20.251447>

Granados, P.S., Fulton, L., Nunez Patlan, E., Terzyk, M., Novotny, T.E., 2019. Global health perspectives on cigarette butts and the environment. *Int. J. Environ. Res. Public. Health* 16, 1858.

Green, A.L.R., Putschew, A., Nehls, T., 2014. Littered cigarette butts as a source of nicotine in urban waters. *J. Hydrol.* 519, 3466–3474. <https://doi.org/10.1016/j.jhydrol.2014.05.046>

Green, D.S., Almroth, B.C., Altman, R., Bergmann, M., Gündoğdu, S., Warriar, A.K., Boots, B., Walker, T.R., Krieger, A., Syberg, K., 2023. Time to kick the butt of the most common litter item in the world: Ban cigarette filters. *Sci. Total Environ.* 865, 161256. <https://doi.org/10.1016/j.scitotenv.2022.161256>

Green, D.S., Tongue, A.D.W., Boots, B., 2022. The ecological impacts of discarded cigarette butts. *Trends Ecol. Evol.* 37, 183–192. <https://doi.org/10.1016/j.tree.2021.10.001>

Guarujá, 2013. Plano Diretor de Guarujá, São Paulo [WWW Document]. URL <https://leismunicipais.com.br/plano-diretor-guaruja-sp> (accessed 12.6.22).

Gutvirtz, G., Sheiner, E., 2022. Airway pollution and smoking in reproductive health. *Best Pract. Res. Clin. Obstet. Gynaecol.* <https://doi.org/10.1016/j.bpobgyn.2022.09.005>

Haseler, M., Balciunas, A., Hauk, R., Sabaliauskaite, V., Chubarenko, I., Ershova, A., Schernewski, G., 2020. Marine Litter Pollution in Baltic Sea Beaches – Application of the Sand Rake Method. *Front. Environ. Sci.* 8.

Hecht, S.S., Hatsukami, D.K., 2022. Smokeless tobacco and cigarette smoking: chemical mechanisms and cancer prevention. *Nat. Rev. Cancer* 22, 143–155. <https://doi.org/10.1038/s41568-021-00423-4>

IBGE, 2020. Instituto Brasileiro de Geografia e Estatística. Cidades, Panorama. <https://cidades.ibge.gov.br/brasil/sp/guaruja/panorama>.

IECS, 2020. The importance of raising tobacco taxes in Brazil [WWW Document]. URL <https://www.iecs.org.ar/tabaquismo-en-brasil/>

Iglesias, R.M., Szklo, A.S., Souza, M.C. de, Almeida, L.M. de, 2017. Estimating the size of illicit tobacco consumption in Brazil: findings from the global adult tobacco survey. *Tob. Control* 26, 53–59. <https://doi.org/10.1136/tobaccocontrol-2015-052465>

Ikazabo, R.N.N., Bier, J.-C., Jamart, J., Mostosi, C., Mavroudakos, N., 2022. Impact on quitting smoking of cognitive impairment in stroke patients. *J. Neurol. Sci.* 439, 120296. <https://doi.org/10.1016/j.jns.2022.120296>

ISLU, 2022. Índice de Sustentabilidade da Limpeza Urbana | Selur – Sindicato das Empresas de Limpeza Urbana [WWW Document]. URL <https://selur.org.br/wp-content/uploads/2022/10/ISLU-2022a.pdf> (ac-

cessed 12.6.22).

Joossens, L., Lugo, A., Vecchia, C.L., Gilmore, A.B., Clancy, L., Gallus, S., 2014. Illicit cigarettes and hand-rolled tobacco in 18 European countries: a cross-sectional survey. *Tob. Control* 23, e17–e23. <https://doi.org/10.1136/tobaccocontrol-2012-050644>

Juarez, B.S. de M., Reynales-Shigematsu, L.M., Stoklosa, M., Welding, K., Drope, J., 2021. Measuring the illicit cigarette market in Mexico: a cross validation of two methodologies. *Tob. Control* 30, 125–131. <https://doi.org/10.1136/tobaccocontrol-2019-055449>

Kataržytė, M., Balčiūnas, A., Haseler, M., Sabaliauskaitė, V., Lauciūtė, L., Stepanova, K., Nazzari, C., Schernewski, G., 2020. Cigarette butts on Baltic Sea beaches: Monitoring, pollution and mitigation measures. *Mar. Pollut. Bull.* 156, 111248. <https://doi.org/10.1016/j.marpolbul.2020.111248>

Kungskulniti, N., Charoenca, N., Hamann, S.L., Pitayarangsarit, S., Mock, J., 2018. Cigarette Waste in Popular Beaches in Thailand: High Densities that Demand Environmental Action. *Int. J. Environ. Res. Public Health* 15, 630. <https://doi.org/10.3390/ijerph15040630>

Kurti, M.K., Schroth, K.R.J., Ackerman, C., Kennedy, M., Jeong, M., Delnevo, C.D., 2020. Availability of menthol cigarettes in Oakland, California after a partial flavor ban. *Prev. Med. Rep.* 20, 101200. <https://doi.org/10.1016/j.pmedr.2020.101200>

Lee, H.-L., Hsieh, D.P.H., Li, L.-A., 2011. Polycyclic aromatic hydrocarbons in cigarette sidestream smoke particulates from a Taiwanese brand and their carcinogenic relevance. *Chemosphere* 82, 477–482. <https://doi.org/10.1016/j.chemosphere.2010.09.045>

Li, L., Yang, D.C., Chen, C.-H., 2021. Metabolic reprogramming: A driver of cigarette smoke-induced inflammatory lung diseases. *Free Radic. Biol. Med.* 163, 392–401. <https://doi.org/10.1016/j.freeradbiomed.2020.12.438>

Li, Z., 2022. Modeling pesticide residues in tobacco leaves for improving life cycle inventory analysis of pesticides in the cigarette industry. *Sci. Total Environ.* 845, 157267. <https://doi.org/10.1016/j.scitotenv.2022.157267>

Lima, C.F., Amaral dos Santos Pinto, M., Brasil Choueri, R., Buruaem Moreira, L., Braga Castro, Í., 2021. Occurrence, characterization, parti-

tion, and toxicity of cigarette butts in a highly urbanized coastal area. *Waste Manag.* 131, 10–19. <https://doi.org/10.1016/j.wasman.2021.05.029>

Malahayati, M., Masui, T., 2019. The impact of green house gas mitigation policy for land use and the forestry sector in Indonesia: Applying the computable general equilibrium model. *For. Policy Econ.* 109, 102003. <https://doi.org/10.1016/j.forpol.2019.102003>

Mandelli, W.G., Choueri, R.B., Castro, Í.B., Moreira, L.B., 2022. Potential toxicity of contaminants leached from cigarette butts in coastal environments. *Ecotoxicol. Environ. Contam.* 17, 85–98. <https://doi.org/10.5132/eec.2022.02.10>

Marah, M., Novotny, T.E., 2011. Geographic patterns of cigarette butt waste in the urban environment. *Tob. Control* 20, i42–i44. <https://doi.org/10.1136/tc.2010.042424>

McDonnell, B.P., McCausland, R., Keogan, S., Clancy, L., Regan, C., 2021. Prevalence of illicit tobacco use and tobacco tax avoidance in pregnancy. *Ir. J. Med. Sci.* 190, 1445–1449. <https://doi.org/10.1007/s11845-020-02487-x>

Merriman, D., Yurekli, A., Chaloupka, F., 2000. How big is the worldwide cigarette smuggling problem?1. *Tob. Control Dev. Ctries.*

Mghili, B., Lamine, I., Bouzekry, A., Gunasekaran, K., Aksissou, M., 2023. Cigarette butt pollution in popular beaches of Morocco: Abundance, distribution, and mitigation measures.

Morais, D., Colombage, S., Wickramasinghe, D., 2018. A baseline study on the illicit cigarette market - and the resulting tax implications for Sri Lanka. *Imashi Publications.* 978-955-714-030-8.

Moriwaki, H., Kitajima, S., Katahira, K., 2009. Waste on the roadside, 'point-source' waste: its distribution and elution potential of pollutants into environment. *Waste Manag.* 29, 1192–1197.

Nasab, A.Y., Oskoei, V., Rezasab, M., Alinejad, N., Hosseinzadeh, A., Kashi, G., 2022. Cigarette butt littering consequences: a study of pollution rate on beaches and urban environments. *Environ. Sci. Pollut. Res.* 29, 45396–45403. <https://doi.org/10.1007/s11356-022-19155-5>

Neira, C., Cossaboon, J., Mendoza, G., Hoh, E., Levin, L.A., 2017. Occurrence and distribution of polycyclic aromatic hydrocarbons in surface sediments of San Diego Bay marinas. *Mar. Pollut. Bull.* 114, 466–479.

<https://doi.org/10.1016/j.marpolbul.2016.10.009>

Nguyen, A., Nguyen, H.T., 2020. Tobacco excise tax increase and illicit cigarette consumption: evidence from Vietnam. *Tob. Control* 29, s275–s280. <https://doi.org/10.1136/tobaccocontrol-2019-055301>

Niterói, 2022. Mapas de Zonas de Uso. Lei Urbanística. <https://geoniteroi.maps.arcgis.com/apps/webappviewer/index.html?id=b-186888f13ee4955a7d59ac1fc0e6a7e>.

Nitschke, T., Bour, A., Bergquist, M., Blanchard, M., Molinari, F., Almroth, B.C., 2023. Smokers' behaviour and the toxicity of cigarette filters to aquatic life: a multidisciplinary study. *Microplastics Nanoplastics* 3, 1. <https://doi.org/10.1186/s43591-022-00050-2>

OCSA, T., 2017. Contraband Tobacco Levels in Ontario Reaching Alarming Rates According to New Study [WWW Document]. Ont. Conv. Stores Assoc. URL <https://ontariocstores.ca/2017contrabandstudy/> (accessed 12.7.22).

OCSA, T., 2013. Shocking Contraband Butt Study Results [WWW Document]. Ont. Conv. Stores Assoc. URL <http://ontariocstores.ca/shocking-contraband-butt-study-results/> (accessed 12.7.22).

Oigman-Pszczol, S.S., Creed, J.C., 2007. Quantification and Classification of Marine Litter on Beaches along Armação dos Búzios, Rio de Janeiro, Brazil. *J. Coast. Res.* 2007, 421–428. [https://doi.org/10.2112/1551-5036\(2007\)23\[421:QACOML\]2.0.CO;2](https://doi.org/10.2112/1551-5036(2007)23[421:QACOML]2.0.CO;2)

Oliver, J., Thomson, G., Wilson, N., 2014. Measurement of cigarette butt litter accumulation within city bus shelters. *N. Z. Med. J. Online* 127.

Paraje, G., Stoklosa, M., Blecher, E., 2022. Illicit trade in tobacco products: recent trends and coming challenges. *Tob. Control* 31, 257–262. <https://doi.org/10.1136/tobaccocontrol-2021-056557>

Patel, V., Thomson, G.W., Wilson, N., 2013. Cigarette butt littering in city streets: a new methodology for studying and results. *Tob. Control* 22, 59–62. <https://doi.org/10.1136/tobaccocontrol-2012-050529>

Pusceddu, F.H., Sugauara, L.E., de Marchi, M.R., Choueri, R.B., Castro, Í.B., 2019. Estrogen levels in surface sediments from a multi-impacted Brazilian estuarine system. *Mar. Pollut. Bull.* 142, 576–580. <https://doi.org/10.1016/j.marpolbul.2019.03.052>

Qiao, M., Wang, C., Huang, S., Wang, D., Wang, Z., 2006. Composition, sources, and potential toxicological significance of PAHs in the surface sediments of the Meiliang Bay, Taihu Lake, China. *Environ. Int.* 32, 28–33. <https://doi.org/10.1016/j.envint.2005.04.005>

Ranjesh, Z., Nasouri, K., 2022. Facile synthesis of novel porous nickel/carbon fibers obtained from cigarette butts for high-frequency microwave absorption. *J. Environ. Chem. Eng.* 10, 106969.

Ribeiro, V.V., Harayashiki, C.A.Y., Ertaş, A., Castro, Í.B., 2021a. Anthropogenic litter composition and distribution along a chemical contamination gradient at Santos Estuarine System—Brazil. *Reg. Stud. Mar. Sci.* 46, 101902. <https://doi.org/10.1016/j.rsma.2021.101902>

Ribeiro, V.V., Lopes, T.C., Amaral dos Santos Pinto, M., Póvoa, A.A., Corrêa, V.R., De-la-Torre, G.E., Dobaradaran, S., Green, D.S., Szklo, A.S., Castro, Í.B., 2022a. Cigarette butts in two urban areas from Brazil: Links among environmental impacts, demography and market. *Environ. Res.* 213, 113730. <https://doi.org/10.1016/j.envres.2022.113730>

Ribeiro, V.V., Pinto, M.A.S., Mesquita, R.K.B., Moreira, L.B., Costa, M.F., Castro, Í.B., 2021b. Marine litter on a highly urbanized beach at Southeast Brazil: A contribution to the development of litter monitoring programs. *Mar. Pollut. Bull.* 163, 111978. <https://doi.org/10.1016/j.marpolbul.2021.111978>

Ribeiro, V.V., Póvoa, A.A., De-la-Torre, G.E., Castro, Í.B., 2022b. Indexing Anthropogenic Litter as a Contamination Gradient from Rivers to Beaches in Southeast Brazil. *J. Coast. Res.* 38, 1172–1180. <https://doi.org/10.2112/JCOASTRES-D-22A-00005.1>

Rochman, C.M., Manzano, C., Hentschel, B.T., Simonich, S.L.M., Hoh, E., 2013. Polystyrene Plastic: A Source and Sink for Polycyclic Aromatic Hydrocarbons in the Marine Environment. *Environ. Sci. Technol.* 47, 13976–13984. <https://doi.org/10.1021/es403605f>

Rodríguez, C., Fossatti, M., Carrizo, D., Sánchez-García, L., Teixeira de Mello, F., Weinstein, F., Lozoya, J.P., 2020. Mesoplastics and large microplastics along a use gradient on the Uruguay Atlantic coast: Types, sources, fates, and chemical loads. *Sci. Total Environ.* 721, 137734. <https://doi.org/10.1016/j.scitotenv.2020.137734>

Ross, H., 2015. Understanding and Measuring Cigarette Tax Avoidance and Evasion: A Methodological Guide [WWW Document]. URL <https://>

tobacconomics.org (accessed 1.10.22).

Roveri, V., Guimarães, L.L., Correia, A.T., 2020a. Temporal and spatial variation of benthic macroinvertebrates on the shoreline of Guarujá, São Paulo, Brazil, under the influence of urban surface runoff. *Reg. Stud. Mar. Sci.* 36, 101289. <https://doi.org/10.1016/j.rsma.2020.101289>

Roveri, V., Guimarães, L.L., Toma, W., Correia, A.T., 2020b. Occurrence and ecological risk assessment of pharmaceuticals and cocaine in a beach area of Guarujá, São Paulo State, Brazil, under the influence of urban surface runoff. *Environ. Sci. Pollut. Res.* 27, 45063–45075. <https://doi.org/10.1007/s11356-020-10316-y>

Santos, 2013. Revisão do Plano Diretor de Desenvolvimento e Expansão Urbana – LC 731/2011 Diagnóstico Técnico.

Santos, A.M.A.D., Triaca, L.M., Leivas, P.H.S., 2023. How is smoking distributed in relation to socioeconomic status? Evidence from Brazil in the years 2013 and 2019. *Econ. Hum. Biol.* 49, 101240. <https://doi.org/10.1016/j.ehb.2023.101240>

Santos-Echeandía, J., Zéler, A., Gago, J., Lacroix, C., 2021. The role of cigarette butts as vectors of metals in the marine environment: Could it cause bioaccumulation in oysters? *J. Hazard. Mater.* 416, 125816. <https://doi.org/10.1016/j.jhazmat.2021.125816>

São Paulo, 2005. Mapa de águas subterrâneas do Estado de São Paulo: escala 1: 1.000. 000. CPRM.

Schneider, J.E., Peterson, N.A., Kiss, N., Ebeid, O., Doyle, A.S., 2011. Tobacco litter costs and public policy: a framework and methodology for considering the use of fees to offset abatement costs. *Tob. Control* 20, i36–i41. <https://doi.org/10.1136/tc.2010.041707>

Schreiner, C., 2011. Genetic Toxicity of Naphthalene: A Review. *J. Toxicol. Environ. Health Part B* 6, 161–183. <https://doi.org/10.1080/10937400306472>

Scollo, M., Zacher, M., Durkin, S., Wakefield, M., 2014. Early evidence about the predicted unintended consequences of standardised packaging of tobacco products in Australia: a cross-sectional study of the place of purchase, regular brands and use of illicit tobacco. *BMJ Open* 4, e005873. <https://doi.org/10.1136/bmjopen-2014-005873>

Sedeh, M.S., Ehrampoush, M.H., Kashi, G., Hosseinzadeh, A., Gha-

le Askari, S., 2022. Spatial and temporal variations of tobacco waste pollution in our cities. *Arab. J. Geosci.* 15, 1285. <https://doi.org/10.1007/s12517-022-10538-z>

Sepand, M.R., Maghsoudi, A.S., Shadboorestan, A., Mirnia, K., Aghsami, M., Raoufi, M., 2021. Cigarette smoke-induced toxicity consequences of intracellular iron dysregulation and ferroptosis. *Life Sci.* 281, 119799. <https://doi.org/10.1016/j.lfs.2021.119799>

Shakya, S., Lamichhane, A., Karki, P., Gurung, J.K., Pradhan, P.M.S., 2023. Extent of illicit cigarette sales in Nepal: findings from a retail survey. *Tob. Control.* <https://doi.org/10.1136/tc-2022-057619>

Shimazu, H., 2016. Determination of Polycyclic Aromatic Hydrocarbons in Cigarettes and Cigarette Smoke. *Environ. Pollut.* 5, p15. <https://doi.org/10.5539/ep.v5n2p15>

Silva, N.F. da, Christina Barbosa de Araújo, M., Santos Silva-Cavalcanti, J., 2023. Spatio-temporal distribution of cigarette butt contamination in urban beaches with varying levels of use. *Waste Manag.* 168, 179–188. <https://doi.org/10.1016/j.wasman.2023.05.035>

SMA/CPLA, 2016. Zoneamento Ecológico - Econômico - Baixada Santista, São Paulo, Brasil. Secretaria de Meio Ambiente do Estado de São Paulo/Coordenação de Planejamento Ambiental.

Smith, J., Thompson, S., Lee, K., 2019. 'Both Sides of the Argument'? A critical review of existing evidence on the illicit trade in tobacco products in Canada. *Tob. Control* 28, e141–e147. <https://doi.org/10.1136/tobaccocontrol-2018-054687>

Smith, K.C., Washington, C., Welding, K., Kroart, L., Osho, A., Cohen, J.E., 2017. Cigarette stick as valuable communicative real estate: a content analysis of cigarettes from 14 low-income and middle-income countries. *Tob. Control* 26, 604–607. <https://doi.org/10.1136/tobaccocontrol-2016-053148>

Soleimani, F., Dobaradaran, S., De-la-Torre, G.E., Schmidt, T.C., Saeedi, R., 2022. Content of toxic components of cigarette, cigarette smoke vs cigarette butts: A comprehensive systematic review. *Sci. Total Environ.* 813, 152667. <https://doi.org/10.1016/j.scitotenv.2021.152667>

Spain, 2023. Memoria de análisis de impacto normativo del proyecto de real decreto relativo a la gestión de los residuos de los productos

del tabaco con filtros y los filtros comercializados para utilizarse con productos del tabaco. Ministerio para la transición ecológica y el reto demográfico. [https://www.miteco.gob.es/gl/calidad-y-evaluacion-ambiental/participacion-publica/230420maintabacoversionfinal\\_tcm37-561444.pdf](https://www.miteco.gob.es/gl/calidad-y-evaluacion-ambiental/participacion-publica/230420maintabacoversionfinal_tcm37-561444.pdf).

Stratton, J., Shiplo, S., Ward, M., Babayan, A., Stevens, A., Edwards, S., 2016. Assessing contraband tobacco in two jurisdictions: a direct collection of cigarette butts. *BMC Public Health* 16, 622. <https://doi.org/10.1186/s12889-016-3229-0>

Szklo, A., Iglesias, R.M., Carvalho de Souza, M., Szklo, M., Maria de Almeida, L., 2018. Trends in Illicit Cigarette Use in Brazil Estimated From Legal Sales, 2012–2016. *Am. J. Public Health* 108, 265–269. <https://doi.org/10.2105/AJPH.2017.304117>

Szklo, A.S., Iglesias, R.M., 2020. Interference by the tobacco industry in data on cigarette consumption in Brazil. *Cad. Saúde Pública* 36. <https://doi.org/10.1590/0102-311X00175420>

Szklo, A.S., Iglesias, R.M., Stoklosa, M., Figueiredo, V.C., Welding, K., Junior, P.R.B. de S., Machado, A.T., Martins, L.F.L., Nascimento, H., Drope, J., 2020. Cross-validation of four different survey methods used to estimate illicit cigarette consumption in Brazil. *Tob. Control*. <https://doi.org/10.1136/tobaccocontrol-2020-056060>

Torkashvand, J., Farzadkia, M., 2019. A systematic review on cigarette butt management as a hazardous waste and prevalent litter: control and recycling. *Environ. Sci. Pollut. Res.* 26, 11618–11630. <https://doi.org/10.1007/s11356-019-04250-x>

Torkashvand, J., Farzadkia, M., Sobhi, H.R., Esrafil, A., 2020. Littered cigarette butt as a well-known hazardous waste: A comprehensive systematic review. *J. Hazard. Mater.* 383, 121242. <https://doi.org/10.1016/j.jhazmat.2019.121242>

Torkashvand, J., Godini, K., Jafari, A.J., Esrafil, A., Farzadkia, M., 2021. Assessment of littered cigarette butt in urban environment, using of new cigarette butt pollution index (CBPI). *Sci. Total Environ.* 769, 144864. <https://doi.org/10.1016/j.scitotenv.2020.144864>

UN, 2016. THE 17 GOALS | sustainable development [WWW Document]. URL <https://sdgs.un.org/goals>

Valiente, R., Escobar, F., Pearce, J., Bilal, U., Franco, M., Sureda, X., 2020. Estimating and mapping cigarette butt littering in urban environments: A GIS approach. *Environ. Res.* 183, 109142. <https://doi.org/10.1016/j.envres.2020.109142>

van der Zee, K., Filby, S., van Walbeek, C., 2023. When Cigarette Sales Suddenly Become Illegal: Evidence From an Online Survey of South African Smokers During COVID-19 Lockdown. *Nicotine Tob. Res.* 25, 325–330. <https://doi.org/10.1093/ntr/ntac067>

van der Zee, K., van Walbeek, C., Magadla, S., 2020. Illicit/cheap cigarettes in South Africa. *Trends Organ. Crime* 23, 242–262. <https://doi.org/10.1007/s12117-019-09372-9>

Vanapalli, K.R., Sharma, H.B., Anand, S., Ranjan, V.P., Singh, H., Dubey, B.K., Mohanty, B., 2023. Cigarettes butt littering: The story of the world's most littered item from the perspective of pollution, remedial actions, and policy measures. *J. Hazard. Mater.* 453, 131387. <https://doi.org/10.1016/j.jhazmat.2023.131387>

Vu, A.T., Taylor, K.M., Holman, M.R., Ding, Y.S., Hearn, B., Watson, C.H., 2015. Polycyclic Aromatic Hydrocarbons in the Mainstream Smoke of Popular U.S. Cigarettes. *Chem. Res. Toxicol.* 28, 1616–1626. <https://doi.org/10.1021/acs.chemrestox.5b00190>

WHO, 2003. WHO Framework Convention on Tobacco Control [WWW Document]. WHO. URL [http://www.who.int/fctc/text\\_download/en/](http://www.who.int/fctc/text_download/en/) (accessed 3.28.22).

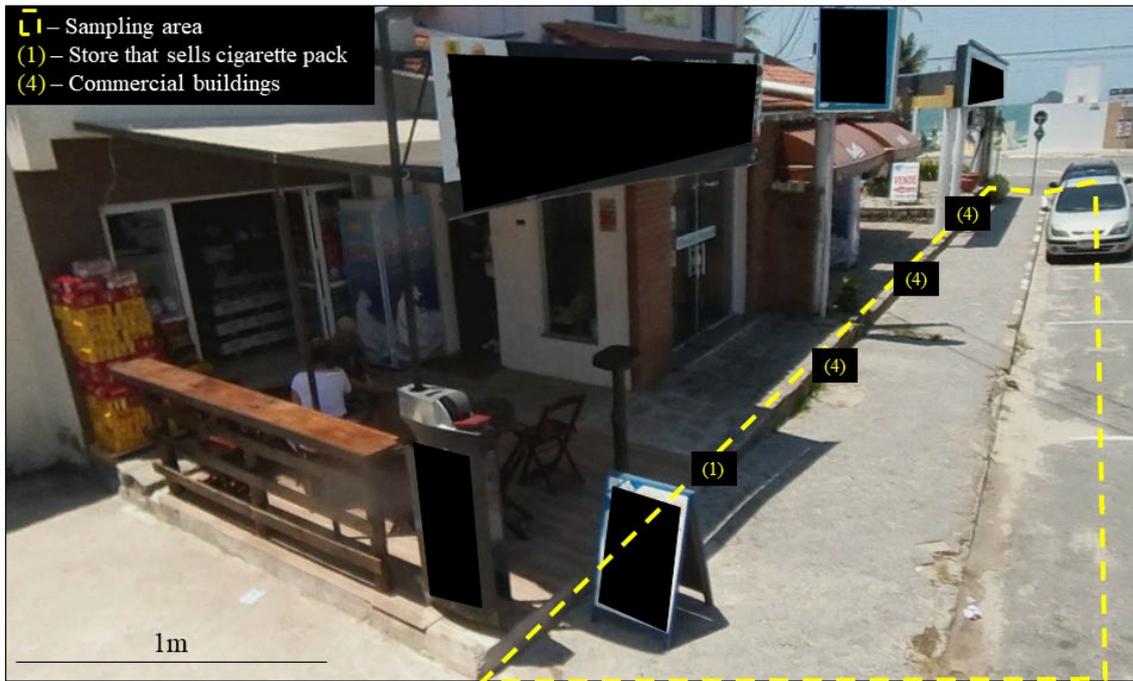
Zeru, M.A., Tesfa, E., Mitiku, A.A., Seyoum, A., Bokoro, T.A., 2021. Prevalence and risk factors of type-2 diabetes mellitus in Ethiopia: systematic review and meta-analysis. *Sci. Rep.* 11, 21733. <https://doi.org/10.1038/s41598-021-01256-9>

## **Supplementary material**

### **Littered cigarette butts: links among environmental impacts, demography and market at the highly urbanized Brazilian cities**

Ítalo Braga Castro

Victor Vasques Ribeiro



**Figure S1.** Accounting of urban aspects inside the sampling site (S9), highlighting in dotted yellow shape the sampling area, the presence of commercial (4) and residential (5) goals in just one building. Garbage bins and bags (8) were also accounted. The name of the commercial establishments and any possible brand that could emerge in Google Earth obtained figure (without copyright) were covered in black.



**Figure S2.** Accounting of urban aspects inside the sampling site (S7), highlighting in dotted yellow shape the sampling area, the presence of commercial (4) and residential (5) goals in just one building. Garbage bins and bags (8) were also accounted. The name of the commercial establishments and any possible brand that could emerge in Google Earth obtained figure (without copyright) were covered in black.



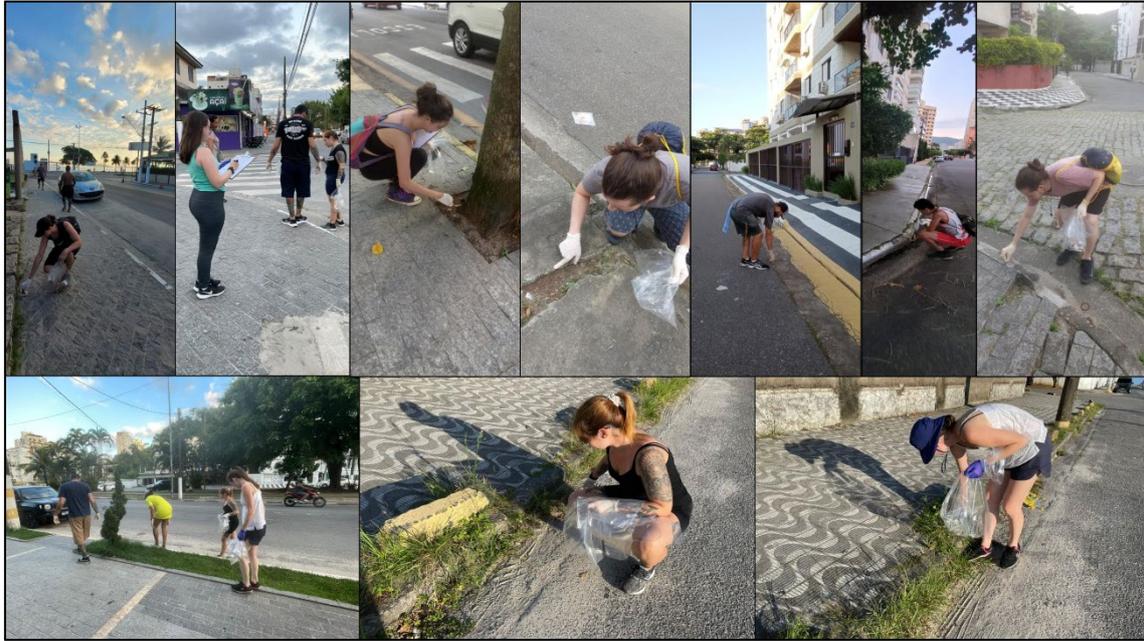
**Figure S3.** Accounting of urban aspects inside the sampling site (S9), highlighting in dotted yellow shape the sampling area, the presence of commercial (4) and residential (5) goals in just one building. Garbage bins and bags (8) were also accounted. The name of the commercial establishments and any possible brand that could emerge in Google Earth obtained figure (without copyright) were covered in black.



**Figure S4.** Accounting of urban aspects inside the sampling site (S9), highlighting in dotted yellow shape the sampling area, the presence of commercial (4) and residential (5) goals in just one building. Garbage bins and bags (8) were also accounted. The name of the commercial establishments and any possible brand that could emerge in Google Earth obtained figure (without copyright) were covered in black.



**Figure S5.** Sample kits preparation in the LECMAR laboratory, containing gloves and labeled plastic bags for volunteers collect the cigarette butts and packs of each site.



**Figure S6.** Cigarette butts (CBs) and cigarette packs (CPs) sampling campaigns in Guarujá city urban walkways, performed between 5 and 10 p.m. Volunteers were always at least paired, and were advised to take pictures of sampling campaigns in the beginning (around 5 p.m.), to avoid putting them at any sort of risk with their cellphones. Volunteers only touched CBs and CPs while wearing gloves. No issues were reported by volunteers during the sampling campaigns. Clipboards were provided to account the urban aspects of each site.



**Figure S7.** Cigarette butts (CBs) and cigarette packs (CPs) sorting in the LECMAR laboratory. Volunteers only touched CBs and CPs while wearing gloves. Sorting was made as soon as possible after sampling to avoid dealing with even smellier samples. Sort was planned to be short (9 to 12 a.m. for one round, and 2 to 5 p.m. to other round of volunteers) with breaks (of at least 5 min to every hour) in order to avoid inhalation due to long exposure. No issues were reported by volunteers during the sorting. As accounting the CBs and CPs, volunteers also separated them between brands.



**Figure S8.** Uneven and/or drilled walkways of low-quality pavement of Guarujá city urban walkways.

Brand	Pack	Butt	Brand	Pack	Butt	Brand	Pack	Butt
Bill			Camel			Chesterfield		
Camel Double Mint & Green			Chesterfield Linea 100 XSL			Chesterfield		
Camel Double Mint & Purple			Chesterfield Terras Brasil. Blue			Chesterfield Remix Beats		
Camel Cretec Option			Chesterfield Terras Brasil. Orange			Chesterfield		
Camel			Chesterfield			Chesterfield		
Camel			Chesterfield			Djarum Black		

**Figure S9.** Initial guide to identification of cigarette butts (CBs) and cigarette packs (CPs) in Brazil (first page – alphabetical order). Diagonal lines denote the brands in which the specific model and/or CP is unknown.

Brand	Pack	Butt	Brand	Pack	Butt	Brand	Pack	Butt
Djarum Black			Dunhill On Red			Gift		
Double Happiness			Dunhill Tob. Lon. Ltd Est 1907 Evoq.			Gift Menthol		
Dunhill Carlton Blend 2.0			Egypt Classic			Gift Red		
Dunhill Tob. London Blue			Egypt Blue Premiere			Gudang Garam Cravo		
Dunhill On Blue			Eight			Harmony by Luxor 100 XSL		
Dunhill On Boost			Funk			Kent		

**Figure S10.** Initial guide to identification of cigarette butts (CBs) and cigarette packs (CPs) in Brazil (second page – alphabetical order). Diagonal lines denote the brands in which the specific model and/or CP is unknown.

Brand	Pack	Butt	Brand	Pack	Butt	Brand	Pack	Butt
Kent			LA Menthol			Lucky Strike		
Kent			Lucky strike			Luxury		
L&M Forward (New Black) KS			Lucky Strike Big Chill			Marlboro Double Fusion		
L&M Forward (New Black) KS			Lucky Strike Double Ice			Marlboro Filter Plus Ks		
L&M Vermelho			Lucky Strike			Marlboro Forest Fusion		
L&M Firm Filter			Lucky Strike			Marlboro Gold Stripes Ks		

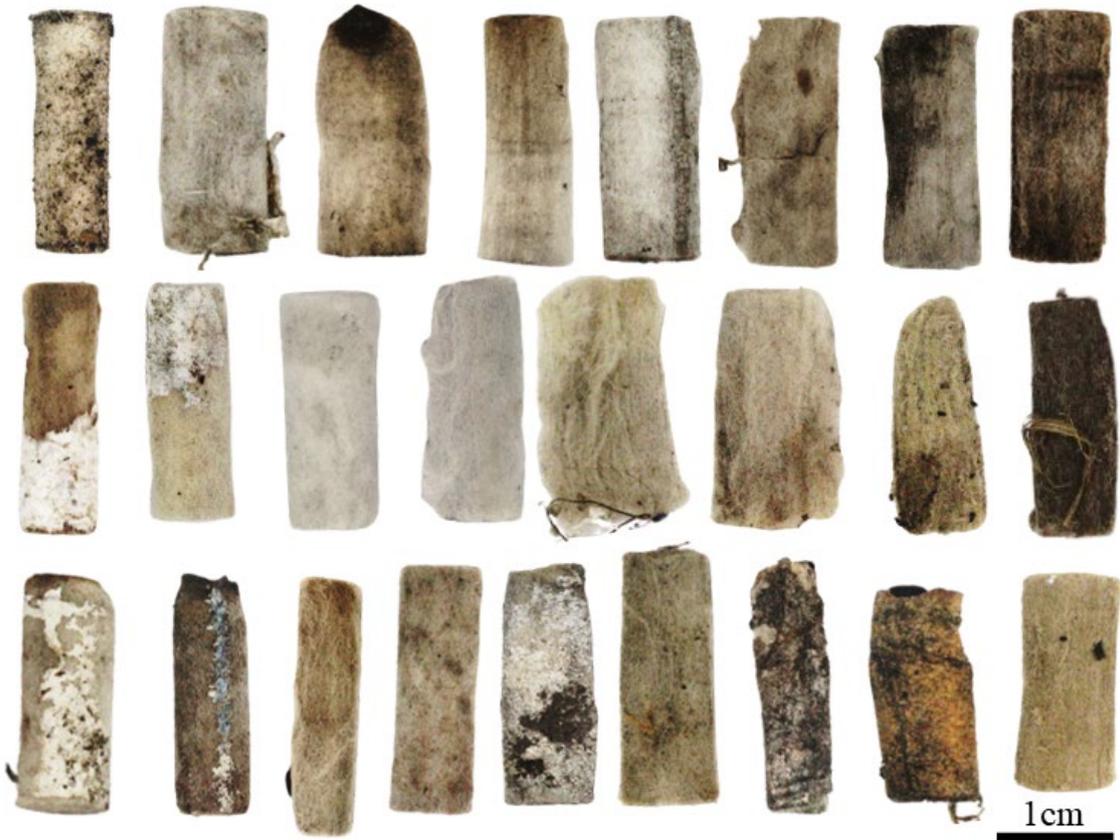
**Figure S11.** Initial guide to identification of cigarette butts (CBs) and cigarette packs (CPs) in Brazil (third page – alphabetical order). Diagonal lines denote the brands in which the specific model and/or CP is unknown.

Brand	Pack	Butt	Brand	Pack	Butt	Brand	Pack	Butt
Marlboro Ice Burst Menthol			Rothmans Lon. Int. Pre. Click Sense			Rothmans		
Marlboro Red Foward Ks			Rothmans Lon Int Wor Dou Click			Rothmans		
Marlboro Silver Blue			Rothmans of London Blue?			Rothmans		
Marlboro Gold Sel. Est 1908			Rothmans			Rothmans		
Marlboro Red Sel. Est° 1908			Rothmans			Rothmans		
Oi Red			Rothmans			Rothmans		

**Figure S12.** Initial guide to identification of cigarette butts (CBs) and cigarette packs (CPs) in Brazil (fourth page – alphabetical order). Diagonal lines denote the brands in which the specific model and/or CP is unknown.

Brand	Pack	Butt	Brand	Pack	Butt	Brand	Pack	Butt
Rothmans			Seven Stars			Winston Sel. Exotic Mint		
Rothmans			TE			Winston		
Rothmans			TE azul			Missing id		
Rothmans			Winston			Missing id		
Sampoerna Kretek Ment Sel. KS			Winston			Missing id		
Sampoerna Kretek Sel. KS			Winston					

**Figure S13.** Initial guide to identification of cigarette butts (CBs) and cigarette packs (CPs) in Brazil (fifth page – alphabetical order and missing identification [id] at the end). Diagonal lines denote the brands in which the specific model and/or CP is unknown.



**Figure S14.** Examples of unidentifiable cigarette butts sampled in Guarujá urban walkways.



**Figure S15.** Examples of roll-your-own cigarette butts sampled in Guarujá urban walkways.