

Danilo Balthazar-Silva

Spatial distribution, input and dispersion of plastic pellets in
coastal zones

Corrected version

Thesis submitted to the Oceanographic
Institute of the University of São Paulo in
partial fulfillment of the requirements for
the degree of Doctor of Science, program of
Oceanography, Biological Oceanography area.

Advisor: Prof. Dr. Alexander Turra

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“There is no “away” because plastic is so permanent and so indestructible. When you cast it into the ocean...it does not go away”.

Sir David Attenborough.

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Abstract

The production and the usage of plastic material increases since the decade of 1950. Nowadays, the elevated production rate, the misusing and the waste turned plastic material in an urgent environmental and economic problem. One of the major environmental problems related to this issue is the contamination of marine environments by microplastics. These constitute plastic particles of size between 1 and 5 mm. Microplastics might occur by breaking of larger plastic pieces or as a manufactured product. The plastic pellets are among this second class, these are small plastic spherules (≥ 5 mm) used in the plastics industry as raw material for the production of manufactured products. It is hypothesized that plastic pellets reach the marine environment due to losses in port terminals or accidental and intentional releases by commercial ships. The present study evaluated the contamination of the coastal zone by microplastics in different spatial and temporal scales. This evaluation approached the dispersion of microplastics in coastal zones, and used the spatial distribution, the stranding and the accumulation of plastic pellets in sandy beaches as a proxy to disclose the behavior of the variation of microplastics in coastal zones. The results of the present paper reveal that microplastics vary both in small and large temporal and spatial scales. Therefore, the present paper brings new insights to the knowledge on microplastics pollution in coastal zones, which might give a new baseline to methodological approaches adopted in management and monitoring programs.

Keywords: marine pollution, plastics, impact assessment, spatial and temporal scales.

Resumo

A produção e o consumo de plásticos vêm aumentando desde a década de 1950. Nos dias de hoje, a taxa elevada de produção, o mal-uso e o desperdício tornaram os plásticos em um problema ambiental e econômico urgente. Um dos principais problemas relacionados à esta questão é a poluição dos ambientes marinhos por microplásticos. Estes constituem partículas de plástico de tamanho que varia entre 1 e 5 mm. Microplásticos podem ocorrer em decorrência da quebra de pedaços de plásticos grandes ou podem ocorrer como um produto fabricado. Os grânulos de plástico estão nesta segunda categoria, estes são pequenas esférulas de plástico (≥ 5 mm) utilizadas como matéria prima para a produção de utensílios variados pela indústria dos plásticos. A hipótese é de que os grânulos de plástico cheguem ao ambiente marinho a partir de perdas em terminais portuários ou após liberações acidentais ou intencionais por embarcações comerciais. O presente estudo avaliou a contaminação da zona costeira por grânulos de plástico em diferentes escalas espaciais e temporais. Esta avaliação abordou a dispersão de microplásticos em regiões costeiras e utilizou a distribuição espacial o aporte e o acúmulo de grânulos de plástico como um modelo para desvendar o comportamento da variação de microplásticos em zonas costeiras. O presente estudo revelou que os microplásticos varia em escalas espaciais e temporais grandes e pequenas. Os resultados apresentados aqui podem conferir embasamento e questões metodológicas para serem adotadas em estratégias de monitoramento e gestão.

palavras-chave: poluição marinha, plástico, avaliação de impacto, escalas locais e regionais.

General Introduction

Management of the solid waste in the Plastics age: a great challenge in the Anthropocene

In the 21st century, plastics' production, usage and waste are widespread and influence the life and the geological processes on Earth in a great variety of ways. According to THOMPSON et al (2009), the influences of plastic upon human life gave birth to the "Plastics age" 70 years ago, whose production increases since the 1950ies. According to UNEP (2014), 1.7 million tons of plastics were produced in 1950 and 288 million tons in 2012, configuring an increase of 8.7% per year. From this amount, the Asian continent is responsible for approximately 45%, followed by Europe, with 20% of the production (UNEP, 2014a). Brazil produced 6.5 million tons of plastics in 2014, representing 2.7% of the global production and the greater in Latin America (ABIPLAST, 2014). The great production of plastic material worldwide is a response to the large usage of plastic items, given their durability, versatility, malleability and low weight (PLASTICSEUROPE, 2015).

In the Declaration of the Global Plastics Associations for Solutions on Marine Litter, associations of plastic industries worldwide pledged to help to spread knowledge regarding waste management and steward the transportation and the distribution of plastic resin pellets in order to prevent accidental losses (GLOBAL PLASTIC ASSOCIATIONS, 2016). This reflects the growing concern of the plastic industry about the management of solid waste. Moreover, this declaration indicates that the plastic industry acknowledges its own specific failures that promote the leaching of plastic waste to natural environments. On the other hand, the excessive plastic production and the lack of conjunct attitudes

between the productive chain and the consumers, regarding sustainable procedures, constitutes one of the main problems for an adequate solid waste management. For example, there is a lack of effective programs regarding recycling of plastic waste originated both by the productive chain and by final consumers (HOORNWEG; BHADA-TATA, 2012).

The constant usage of plastic material occurs in domestic, industrial and recreational activities. In addition, plastics are present in food packaging, clothing, electronic devices and even in cosmetics (DUIS; COORS, 2016; THOMPSON et al., 2009a). The results of UNEP (2014a) indicate that for every 1 million dollars in revenue in the consumer goods industry, 8 tons of plastic are consumed. For example, the population in USA uses between 100 and 380 billion of plastic bags every year (UNEP, 2014a). In the North America and Western Europe, the per capita consumption of plastics reached 100 kg in 2014 (PLASTICSEUROPE, 2015; UNEP, 2014a). In the Asian continent, this value is smaller, reaching 20 kg (GOURMELON, 2015). The Brazilian population consumes about 35 kg per capita of plastics per year (ABIPLAST, 2014). Plastics have a relevant influence on global economy, generating revenues of approximately 600 billion dollars annually (GOURMELON, 2015). However, the total environmental cost of plastic used in the consumer goods industry is more than 75 billion dollars per year, originated from the harmful environmental impacts caused by the leaching of wasted material to natural environments and by landfilling the plastic waste instead of recycling it (UNEP, 2014a).

The wasted plastic material reaches amounts of 25.8 million tons; 29.7% of this amount goes for recycling, energy processes recover 39.5% and 30.8% goes for landfills (PLASTICSEUROPE, 2015). The amount of plastic wasted

might depend on the production rates. For example, the European continent wasted 8 million tons of plastic material in 2014 (PLASTICSEUROPE, 2015). Further, regions of greater production also waste more plastic (JAMBECK et al., 2015). Despite of the relationship between plastics production rates and waste management, wasting also varies according to consumption rates (HOORNWEG; BHADA-TATA, 2012). The inadequate usage has a significant interference on the wasting of plastic material. According to MOORE (2003), the majority of plastic consumed worldwide is wasted just after a single use. Therefore, the wasting also depends on the quality of the consumer goods and on the behavior of the consumers. Further, an adequate management of plastic waste by industries might reduce the amounts of landfilled material (UNEP, 2014a).

The most urgent problem related to the inadequate usage practices and waste management is the leaching of plastic material to natural environments. According to BARNES et al (2009), the strategies of waste retention have been inefficient in preventing environmental contamination, such as in rivers, estuaries and marine environments (HOORNWEG; BHADA-TATA, 2012; ROCHMAN et al., 2013b; UNEP, 2014a, 2014b). One of the responses to the constant leaching, is the entering of the plastic material on the geological processes (CORCORAN; MOORE; JAZVAZ, 2014). According to the review conducted by ZALASIEWICZ et al (2016), the plastic material is a stratigraphic indicator of the Anthropocene. According to the authors, this is a response to the continuous input of plastics in natural environments and to the availability of this material to marine sedimentary processes.

Contamination of marine environments by plastics

According to JAMBECK et al (2015), 80% of marine debris originates from land, representing the main source of plastic material to marine habitats. However, the explanation for the total plastic waste entering in the ocean also relies on the contamination of adjacent environments. Recent estimates reveal the existence of tons of floating plastics in riverine environments. For example, GASPERI et al (2014) estimate that a mean of 27 tons of plastic material might be captured annually by floating debris retention booms established along the river Seine (Paris). MORRITT et al (2014) sampled 8490 submerged plastic items in river Thames (UK), with the most contaminated sampling sites near sewage treatment plants. The abundance of plastic litter in riverine environments might even outnumber the abundance of larval fish (LECHNER et al., 2014). On the other hand, plastic material is also present buried in the sediment column of rivers (CORCORAN et al., 2015). The pollution of rivers by plastics is very important for the dispersion of this contaminant given that they are one of the primary access of plastics to estuarine and, posteriorly, to marine environments (CORCORAN et al., 2015; GALLAGHER et al., 2015; RECH et al., 2014).

The plastic pollution in estuarine environments is a complex issue given the distribution of the particles, the variety of the sources of these material, the responses of the plastic pieces to environmental features and the possible impacts to the biodiversity. LIMA et al (2014) observed that the amount of microplastics might suppress the amount of Ichthyoplankton in tropical estuaries. According to the results of these authors, there might be a great variety of sources of the plastic material in estuarine environments. For example, IVAR DO SUL & COSTA (2013) observed that in a Brazilian estuary located in a protected area,

the main sources of the plastic material are fisheries and local domestic activities. According to their results, the contamination of mangrove systems by plastic varies between rainy and dry seasons. The retention of plastic material in mangrove forests also depends on environmental features, such as the density of the vegetation and local hydrodynamics (IVAR DO SUL et al., 2014). Given this dependence, mangrove forests might retain plastic material for periods that vary from months to years, before the leaching to marine environments (BROWNE; GALLOWAY; THOMPSON, 2010; IVAR DO SUL et al., 2014; SADRI; THOMPSON, 2014).

Once in the ocean, the environmental impacts produced by the plastic material promote an economic cost of approximately 13 billion dollars (UNEP, 2014a). One of the facts that turn the plastic into a hazardous material in the ocean is its massive abundance (ROCHMAN et al., 2013b). An amount between 4.8 and 12.7 million metric tons of plastics outflows from land to marine environments every year (JAMBECK et al., 2015). On the other hand, a recent estimate indicates that there are more than 250.000 tons of plastic pieces floating in the seas (ERIKSEN et al., 2014). However, the dispersion and the distribution of this material is not random in the ocean surface and in the water column. The plastic floating in the open ocean tend to accumulate in the subtropical gyres in response to oceanographic processes (ERIKSEN et al., 2013; LAW et al., 2010, 2014; LEBRETON; BORRERO, 2013; LEBRETON; GREER; BORRERO, 2012). Furthermore, the estimates of abundance indicate the existence of sinking areas of plastic material in the marine environment (CÓZAR et al., 2014). Besides the subtropical gyres, deep-sea and coastal habitats also constitute a sink zone for

plastics (CÓZAR et al., 2014; SHERMAN; VAN SEBILLE, 2016; VAN CAUWENBERGHE et al., 2013; WOODALL et al., 2014).

The plastic material is present in the marine environments in different size classes. Pieces with >25 mm are considered as macroplastics, a size varying from 5 to 25 mm characterize the mesoplastics and the microplastics represent those particles with a size between 1 and 5 mm (LEE et al., 2013). Some studies also consider particles with size <1 mm as microplastics (COLE et al., 2011). However, there are two kinds of microplastics. The fibers and fragments represent the secondary microplastics and their input occurs through breaking of larger pieces (BARNES et al., 2009; BROWNE et al., 2011; O'BRINE; THOMPSON, 2010). The plastic pellets, flakes, microbeads and powder have an industrial origin and represent the primary microplastics. Their input in the sea occurs through losses during shipping and other port activities and sewage discharges (TURNER; HOLMES, 2011; UNEP, 2014a, 2014b).

Microplastics constitute one of the main marine pollutants. Their sources, abundance, distribution and effects on marine and coastal habitats became an urgent environmental concern for researchers and policy makers worldwide in the last decade (BARBOZA; GIMENEZ, 2015; BROWNE et al., 2015; IVAR DO SUL; COSTA, 2014; LAW; THOMPSON, 2014; ROCHMAN et al., 2013b; THOMPSON, 2015). These small plastic particles are present in great abundances in the ocean, VAN SEBILLE et al (2015) estimated a number between 15 and 51 trillion microplastic particles, which corresponds to a weight between 93 and 236 thousand metric tons. The authors also argue that these values correspond to only 1% of the global wasted plastic material. A plausible explanation for this low percentage might rely on the patchy distribution of these

particles. Microplastics are present from the surface to deep layers in the sediment column of coastal habitats and from the surface to deep habitats in the open ocean (KUKULKA et al., 2012; LAW et al., 2010, 2014; TURRA et al., 2014; WOODALL et al., 2014). Moreover, the assimilation of a significant portion of the microplastics present in the seas by the biota constitute an additional sink for those particles (CÓZAR et al., 2014; THOMPSON, 2015).

Once ingested by marine animals, microplastics might promote diverse harmful effects given that these particles assimilate a sort of the chemical pollutants that occur in the natural environment (ROCHMAN et al., 2013a; ROCHMAN; HENTSCHEL; TEH, 2014). The assimilation of microplastics in the basis of marine food webs is a concerning problem related to this issue. COLE et al (2013) observed that, when exposed to assemblages of algae with and without microplastics, the copepod *Centropages typicus* ingests microplastics and shows a decrease in algal feeding. Further, physiological stress and behavioral alterations in zooplanktonic species might occur in response to the ingestion of microplastics (COLE et al., 2015).

There is a growing concern on the transfer of microplastics and/or the assimilated chemicals to superior trophic levels (BROWNE, 2015; ROCHMAN et al., 2016; SANTANA et al., 2016). In addition, recent papers demonstrate the assimilation of microplastics by animals consumed by human populations (ROCHMAN et al., 2015; VANDERMEERSCH et al., 2015). For example, MIRANDA & CARVALHO-SOUZA (2016) reported the existence of microplastics in the stomach of two fishes that humans feed on. Further, VAN CAUWENBERGHE & JANSSEN (2014) found microplastics in soft tissues of mollusks cultured for human consumption. On an extensive review conducted by

THOMPSON et al (2009b) it becomes clear that the constant exposure to plasticizers and chemicals associated to microplastics constitute an urgent risk to human populations. Given all the above issues, studies on the distribution and the behavior of microplastics in marine habitats become crucial for a better understanding of the problem.

There is a growing volume of information on the sinking zones of microplastics. ERIKSEN et al (2015) argue that the weight of microplastics present in the ocean surface might be less than the expected, indicating the existence of mechanisms promoting the removal of these particles to other compartments. Similarly, CÓZAR et al (2014) suggest the outflow of microplastics from the surface for sinking zones. The deep-sea environments might be one of these sinking zones. VAN CAUWENBERGHE et al (2013) observed the occurrence of microplastics in sediments from depths ranging from 1100 to 5000 m. WOODALL et al (2014) evaluated the presence of microplastics in sediment samples collected in depths ranging down to 3500 m from the Mediterranean Sea, SW Indian Ocean and NE Atlantic Ocean. They observed microplastics in all of the collected samples, in abundances ranging from 1.4 to 40 pieces per 50 ml. On the other hand, there is a growing knowledge on the accumulation of microplastics in coastal zones.

CRITCHELL & LAMBRECHTS (2016) modelled the accumulation of plastic particles in coastal zones. The authors observed that the location of the source might be the most important parameter for the velocity of the stranding of plastic litter in sandy beaches. In their simulations, when the source was close to the coast and windward, the majority of particles beached quickly and the particles that did not follow this process remained near the source. However,

physical environmental features such as beach orientation and wind regimes also interfere in accumulation patterns (KABERI et al., 2013). In addition, the accumulation of microplastics varies according to the features of sediments in shorelines (BROWNE; GALLOWAY; THOMPSON, 2010). Further, the ocean dynamics is a determinant factor for the generation of accumulation zones of microplastics in coastlines (SHERMAN; VAN SEBILLE, 2016; VAN SEBILLE; ENGLAND; FROYLAND, 2012b).

The accumulation of microplastics might cause a relevant variety of environmental impacts in the coastal zone. The abundance, widespread presence and the accumulation of small plastic particles in coastal sediments increase the environmental risk promoted by these particles. The abundance of plastic pellets in beaches increases from the lower intertidal to the upper backshore and, in this zone, is higher in deeper sediment layers (TURRA et al., 2014). This pattern of distribution might interfere, for example, on the permeability and on the temperature regulation of the sediment column (CARSON et al., 2011). Given that plastic pellets adsorb chemical and organic pollutants, their spatial distribution interferes on the variability and composition of these contaminants in beaches (ANTUNES et al., 2013; ASHTON; HOLMES; TURNER, 2010; FISNER et al., 2013a, 2013b; TANIGUCHI et al., 2016). The interference of microplastics on the physical environment and their ability to absorb other pollutants promote ecological impacts given their influence on physiological processes of vertebrates and invertebrates (NELMS et al., 2015; NOBRE et al., 2015; RYAN, 2015; VAN FRANEKER; LAW, 2015; WATTS et al., 2014).

Microplastics may impact biodiversity through negative interferences on the dynamics of animals' populations and species' interactions, abundance and distribution (BROWNE et al., 2015; ROCHA-SANTOS; DUARTE, 2014; ROCHMAN et al., 2016; WANG et al., 2016). Therefore, their occurrence in the marine environment is harmful for human societies and promote impacts in activities such as fisheries and even public health. This scenario demands both an increase and improvement of scientific studies and a greater engagement by the productive sector, decision-makers and general community (AMARAL-ZETTLER et al., 2015; IVAR DO SUL; COSTA, 2014). The productive sector still needs a greater engagement on the development of strategies and technologies in order to improve waste management (HOORNWEG; BHADA-TATA, 2012; PEREIRA, 2014; UNEP, 2014a). On the other hand, there is a crescent concerning by independent organizations on the adoption of procedures in order to avoid the stranding of microplastics in the marine environment (PLASTICSEUROPE, 2015; UNEP, 2014a). Further, independent organizations of environmentalists and researchers demonstrate concerning on the development of technologies in order to clean the plastic material already present in the ocean (HIDALGO-RUZ; THIEL, 2015). Moreover, non-governmental organizations engage volunteers in worldwide cleanup events, such as the Ocean Cleanup day (CHESHIRE et al., 2009; UNEP, 2009). Therefore, the development and improvement of the knowledge on the occurrence of microplastics in coastal zones would indicate the hotspots of accumulation and, consequently, promote the improvement of monitoring and management strategies.

Recent papers demonstrated that the monitoring of microplastics depends upon the environmental dynamics of the habitat under study (TURRA et al., 2014), such that the refinement of temporal and spatial scales of the study's conduction is decisive for estimates of distribution and abundance. SMITH & MARKIC (2013) suggest that the environmental problem related to the abundance of plastic debris in coastal habitats might be greater than initially thought given the possibility of biases in estimates conducted in relatively great intervals of sampling. The results observed by RYAN et al (2014) suggest that less frequent samplings do not consider the temporal variation in litter dynamics. Moreover, processes such as the tides dynamics interfere on the input of microplastics in beaches in such a way to promote a great variability of abundance estimates in relatively small spatial scales (MOREIRA et al., 2016a). On the other hand, the interference of physical processes and the scales in which the monitoring should occur in order to avoid biased estimates might depend on the habitat and the type of microplastics under study. However, to fulfill the knowledge gaps and identify solutions to the microplastics problem in coastal habitats it is necessary to find environment-specific variables, such that a standardization of monitoring methods becomes possible (HARDESTY; GOOD; WILCOX, 2015; HIDALGO-RUZ et al., 2012).

Therefore, the widespread pollution of coastal zones by microplastics is a consequence of the "Plastics Age" and the problems related to this period. The urgent fact is that decades of lack of adequate waste management and public policies by human populations promoted a severe contamination of the marine environment by plastics. The environmental impacts already reached the geological processes and the structure of biological communities, even in remote

environments. Moreover, human populations might suffer direct impacts of the plastic pollution by consuming plastic-contaminated seafood. The constant exposure of humans to plasticizers or to chemicals absorbed by the plastic material would result in problems to the public health system. Thereafter, studies on the input, abundance and dynamics of microplastics in coastal zones are crucial to a potential mitigation or even an abbreviation of the problem.

Objectives of the present study.

The present study aimed to disclose processes that are determinant to the accumulation of microplastics in coastal zones through the evaluation of spatial and temporal variation of plastic pellets. This approach improves the knowledge on the dynamics of these contaminants in the coastal zone and indicates how to recognize the priority areas to monitoring and management programs. This survey evaluated the abundance of plastic pellets in different temporal and spatial scales as a proxy in order to get information about how the abundance of microplastics varies in coastal zones. Therefore, the present study has four chapters comprehending the specific objectives:

1. Evaluate the dispersion of microplastics from potential sources to beaches
2. Evaluate the monthly rate of stranding of plastic pellets in sandy beaches;
3. Evaluate the relationship between the stranding and the accumulation zones of plastic pellets in sandy beaches
4. Evaluate the spatial distribution and the accumulation compartments of plastic pellets in sandy beaches.

Chapter 1. Simulation of microplastics dispersion in coastal areas

Abstract

Estimates of the abundance of microplastics in the ocean point to a relevant amount of these particles worldwide. Once present in the water column, microplastics might promote relevant environmental impacts. Recent studies on the dispersion of plastic particles in the ocean indicate that, in open waters, the ocean circulation is a decisive factor for the occurrence of accumulation zones. On the other hand, the abundance of microplastics in the open ocean point to the occurrence of accumulation zones in coastal environments. Numerical models constitute an accurate tool to simulate and predict the dispersion of particulate pollutants in marine environments. The present paper simulated the dispersion of microplastics after their release close to coastal environments in order to predict regional tendencies of dispersion, in regional spatial scales, and posterior accumulation. The simulations contemplated accidental or intentional releases of microplastics by cargo ships during one year in the vicinities of Santos port terminal, southeastern Brazil. The results indicate that the dispersion of particles is monthly variables, with a predominance of dispersion occurring in NE-SW direction. In addition, the particles tend to follow a NE-SW direction during periods of cold fronts. The results might predict where the accumulation zones of microplastics occur in regional scales. This information is important for strategies of risk evaluation and for protocols of monitoring of microplastics contamination.

Keywords: microplastics, numerical modeling, dispersion, accumulation zones.

Introduction

The pollution by microplastics is among the main anthropogenic pressures in the marine environment (THOMPSON et al., 2009a). The abundance of microplastics may reach 100.000 particles/Km² in the water column of the open sea (ERIKSEN et al., 2014), and a relevant amount of these particles may be related to the oceanic gyres (LAW et al., 2014). Samples from deep-sea environments revealed abundances between 1.4 – 40 particles per 50 mL of sediment. Microplastics are present in great amounts and occur in all environments in the coastal zone, as estuaries, sandy beaches, coastal dunes and subtidal habitats (BROWNE et al., 2011; BROWNE; GALLOWAY; THOMPSON, 2010; DEKIFF et al., 2014; LIMA; COSTA; BARLETTA, 2014; TURRA et al., 2014). Given this scenario, recent papers point that coastal zone is a major sinking area for microplastics (CÓZAR et al., 2014; REISSER et al., 2013).

Microplastics contemplate particles of ≤ 5 mm in size (ANDRADY, 2011; COLE et al., 2011; DERRAIK, 2002), however, these particles should be differentiated according to their origin. Particles such as fibers and fragments are generated by the breaking of larger plastic material in marine habitats (ANDRADY, 2011; BARNES et al., 2009; COLE et al., 2011a). Manufactured microbeads, powder polymers and preproduction resin pellets are microplastics generated by the petrochemical industry. Microbeads are small plastic particles present in cosmetics and probably enter the marine environment by discharges of waste water (FENDALL; SEWELL, 2009). The introduction of powder polymers in the marine environment, on the other hand, still needs clarifying. Plastic pellets

reach marine ecosystems probably through transportation losses in port terminals (PEREIRA, 2014). Plastic resin pellets constitute one of the most abundant plastic material present in the coastal zone (ANDRADY, 2011; COLE et al., 2011). The abundance of these particles in sandy beaches is probably related to the proximity to petrochemical facilities and harbors (KARAPANAGIOTI; KLONTZA, 2008; TURNER; HOLMES, 2011). However, there are reports of their presence even in remote localities, which is an indicative of the interaction between their dispersion and coastal circulation (DO SUL; SPENGLER; COSTA, 2009).

Numerical models are a relevant tool for the evaluation of the interference of ocean circulation over the spatial and temporal scales by which the marine anthropogenic debris tend to disperse. Simulations on tracking the source and sinks of marine anthropogenic debris in coastal zones indicate that temporal scales acting on nearshore circulation and tidal dynamics are determinant for the creation of local accumulation zones (CARSON et al., 2013). A recent numerical simulation of microplastics dispersion indicates that local processes, such as tide currents, act as the first dispersal of microplastics in coastal zones (GUTIÉRREZ et al., accepted). Thereafter, the dispersion of microplastics in regional spatial scales occurs according to the interaction between coastal and oceanic processes, such as wind-driven surface currents (LAW et al., 2014; YOON; KAWANO; IGAWA, 2010). It means that hydrometeorological conditions, occurring in different spatial and temporal scales, are determinant for promotion of accumulation zones (DUHEC et al., 2015; LAW et al., 2010; LEBRETON; BORRERO, 2013; LEBRETON; GREER; BORRERO, 2012; MAXIMENKO; HAFNER; NIILER, 2012).

Thus, the interaction between climatic variations and the coastal circulation, such as those promoted by the arriving of cold fronts, might influence the dispersion of plastic pellets in kilometric scales. The revolving of plastic particles would reflect the direction of the currents in response to the wind forcing. VAN SEBILE et al (2012) hypothesize that this process might create regional differences in the abundance of plastic particles in coastal zones. In fact, results presented in recent papers point to the heterogeneity of the presence of plastic material in coastal zones (MOREIRA et al., accepted). There is a probable relationship among microplastics abundance and local inputs, such that sites located closest to the occurrence of input are hypothesized to be more susceptible to receive greater amounts of particles (CLAESSENS et al., 2011). Areas that receive more microplastics are susceptible to a greater environmental risk, given the environmental issues related to these particles (NOBRE et al., 2015; TANIGUCHI et al., 2016). Therefore, the disclosure of the factors involved in the promotion of spatial differentiation still is an urgent issue for evaluations of risk of contamination (HIDALGO-RUZ et al., 2012).

There is still a lack of information on the pathways followed by microplastics after their release in coastal waters. This knowledge would point to the environments more susceptible to the contamination by microplastics in a given coastal region and, therefore, represent a useful information for monitoring and mitigating programs. Once that information on dispersion reveals regions where there is a tendency for plastic pellets accumulation, studies on pathways may constitute an approach for evaluation of risk of contamination. It is evident that the rates of input may play a decisive role in this scenario given that the input of plastic pellets is probable associated to port activities. Predicting the

relationship between seasonal climatic events and the dispersion of pellets indicate the actual interference of plastic particles input in promoting spatial heterogeneity. Thus, studies on the dispersion of plastic pellets in coastal zones would fulfill lacks of information and introduce new perspectives to monitoring programs. Moreover, this approach may explain mechanisms involved in the regional distribution of plastic pellets in coastal zones.

Therefore, the objective of the present work is to simulate the dispersion of microplastics in coastal zones using numerical models. The study test the hypothesis that the interaction occurring between coastal circulation and climatic variations interferes on the pathway followed by microplastics introduced in the marine environment. In this process, the horizontal direction of plastic pellets in the water column would be similar to that assumed by the currents. Further, a second hypothesis states that beaches located closest to the local input would receive greater amounts of microplastics. This regional differentiation would also be reflex of the currents direction; however, the final destination of the particles may be a resultant of the interference of the ocean dynamics over the local inputs.

Materials and Methods

Area of study

The domain of the simulations contemplates the São Paulo state coast, a 400 km² area in southeastern Brazil (23°30' – 25°S 44°30' – 48°W). Both Tropical winds, coming from the Atlantic Tropical Anticyclone system, and polar winds, coming from Migratory Polar Anticyclones, predominate in this area. The

climate in this region is tropical and humid subtropical, with rainy springs and summers and dry autumns and winters (TESSLER et al., 2006).

The main features of the ocean dynamics in the shelf region of São Paulo state accomplishes tides, wind forces and a strong influence from the bathymetry, in both winter and summer seasons. The tidal currents are weak and counterclockwise rotating, with the major axis against the shore and oriented in the Northwest – Southeast direction and turning to the East – Northeast direction, generally more intense, under the influence of cold fronts (HARARI et al., 2006).

The São Paulo state coast is an important site for studies on microplastics pollution given the establishment of the two main sources of production and transportation of plastic pellets (FISNER et al., 2013a, 2013b; TURRA et al., 2014, GUTIÉRREZ et al., accepted). The Cubatão city is characterized by great activity of oil refinery and petrochemical industries. The greater amount of the plastic pellets produced in these facilities is transported by the greater port terminal of Latin America, located at Santos city (PEREIRA, 2014). This import an amount between 350,000 and 400,000 tons and export between 450,000 and 500,000 tons of plastic pellets by year (PEREIRA, 2014).

Numerical Modelling

Dispersion model

The numerical modelling was conducted by the Professor Joseph Harari from the Physical Oceanographic Department of the Oceanographic Institute from the University of São Paulo. The simulation of microplastics' trajectory relied

on the MOCAD, a lagrangean model developed by HARARI & GORDON (2001) to simulate particles' trajectories. In the MOCAD, polynomial interpolations, both two-dimensional (horizontally) and uni-dimensional (vertically) estimate the ocean currents in the exact positions of the particles, which do not necessarily coincide with the grid points of the hydrodynamic model. The advection of particles through their transport by currents is then considered. Subsequently, calculations of diffusion and decay of particles are performed, both being guided by a random number generator with Gaussian distribution.

The random walk method is illustrated with an example in the x direction. If L is the maximum distance that can be traveled by the particle along the x axis in each time step of the model, then at each time step the particle can move between $-L$ and $+L$, in a purely random way. This procedure is repeated in time, and a large number of particles is discharged, each one moving independently in random walks. Therefore, the number of particles per unit distance can be interpreted as the concentration of the material in that distance, and its distribution is equivalent to the effect of diffusion of any other pollutant. In fact, calculations with the random walk approach is equivalent to the diffusion equation when the diffusion coefficient of the equation is $(NL^2 / 2)$, where N is the number of displacements per unit time and L is the maximum displacement allowed in each walk (BOWDEN, 1983).

In relation to decay, at every time step T_p of MOCAD model, a random number is generated for each particle and compared to $(1 - \exp(-T_p / \tau))$ where τ is the "e-folding time" of decay; if the random number is smaller, the particle disappears. Note that, also in MOCAD model, it is assumed, as an initial

assumption, that the particles that undergoes dispersion is inert, completely water-miscible and does not separate from water by gravity.

Hydrodynamic model

The computation of current components relied on hydrodynamic modelling on the Princeton Ocean Model (POM) (BLUMBERG; MELLOR, 1987; MELLOR, 2004). The POM configures a three-dimensional coastal ocean model of primitive equations, with time-varying free surface and sigma vertical coordinates that incorporates a turbulent closure sub-model to provide a realistic parameterization of the vertical mixing processes. The horizontal grid uses curvilinear orthogonal coordinates (Arakawa C-grid) and a variable Coriolis parameter (BLUMBERG; MELLOR, 1987; MELLOR, 2004). Prognostic equations governing the thermodynamic quantities, temperature and salinity account for water mass variations brought about by highly time-dependent coastal upwelling processes as well as horizontal advective processes (ZHANG, 2006). The vertical differencing eliminates time constraints for the vertical coordinate and permits the use of fine vertical resolution in the surface and bottom boundary layers.

The model has a free surface and a split time step. Therefore, the external mode portion of the model is two-dimensional and uses a short time step based on the Courant-Fridrichs-Lewy condition (CFL condition) and the external wave speed. In addition, the internal mode is three-dimensional and uses a long time step based on the CFL condition and the internal wave speed. In this procedure, the computing of free surface elevation occurs in a prognostically manner, so that the simulation of extreme events is possible (ORTON et al., 2012). The model also accommodates realistic coastline geometry and bottom

topography (BLUMBERG; MELLOR, 1983; BLUMBERG; MELLOR, 1987; MELLOR, 2004).

This modeling is frequently used in simulations of pollutant dispersion in marine systems. To evaluate the potential long-term impacts of water discharges derived from offshore oil and gas production operations, ZHAO et al (2008) developed a risk assessment approach based on the POM, random walk and Monte Carlo simulations. RAMSAK et al (2013) simulated an oil-spill occurring during a short-term real-wind event. In this procedure, the authors used the TSPOM model in order to compute the hydrodynamic circulation and the NAPOM to obtain the open boundary condition (MALACIC; PETELIN; VODOPIVEC, 2012). Actually, the TSPOM is a variant of the POM, and its initialization occurs through the operational model outputs of the model NAPOM. HARARI & GORODON (2001) adapted the POM in order to conduct the calculation routines of advection-diffusion for the dispersion of inert seawater mixing substances in which the solution adopts the sigma coordinate coefficients relying on the second order turbulent closure.

In the present study, the model followed the adaptations of HARARI et al (2000) and HARARI & CAMARGO (2003), which concern in making flexible the code for independent processing of the ocean circulation contributors: tides, local and remote winds, density and rivers. Further, the adaptations allow several options of combinations of these contributors and introduces several options in the boundary conditions, such as the exact specification of tides and mean sea level, radiational conditions, no gradients combinations and relaxation schemes. The model grid covers the internal continental shelf of São Paulo State; it has 150 x 450 horizontal points with constant spacing of 1 Km and 11 vertical levels

placed on sigma values. The time steps of the simulations are 180 seconds for the internal modes and 6 seconds for the external modes. Other important parameters adopted in the model processing are: constant in Smagorinsky horizontal diffusivity = 0.01; bottom roughness parameter = 0.002 m; ratio of horizontal heat diffusivity to kinematic viscosity = 1.0 and advective terms of external mode updated at every 5 external time steps. In addition, three points temporal and spatial smoothers are applied to prevent solution splitting and to promote the control of numerical noise.

The present model runs for the year of 2012. It considers mean monthly conditions of temperature and salinity, extracted from LEVITUS & BOYER (1994); typical river discharges (for the summer season) given by FCTH (1997); tidal elevations at the boundaries based on results of the global tidal model of LE PROVOST; GENCO & LYARD (1994) and pelagic tidal measurements in the shelf (DE MESQUITA; HARARI, 2003). The mean sea level oscillations are given by coastal tidal stations of Oceanographic Institute of São Paulo University (IOUSP). The winds at the surface were extracted from the global atmospheric model of NCEP/NCAR, available at <http://www.cdc.noaa.gov/cdc/data.ncep.reanalysis.html>. The bathymetry of the grid was obtained from the global database GEBCO bathymetry (General bathymetric chart of the Oceans - <http://www.ngdc.noaa.gov/mgg/GEBCO/>), which was merged with local bathymetry charts obtained at the Brazilian Geography and Statistics Institute database (<http://mapas.ibge.gov.br/bases-e-referenciais/basescartograficas/>

The interpretation of the particles trajectory according ocean dynamics simulated by the model relied on the synoptic synthesis of each month, available on <http://www.cptec.inpe.br/noticias/noticia/20746>. The present study simulated the releasing of one particle per hour every day of each month contemplated in the period of the model, with a time step of 10 minutes for the dispersion model. The selected releasing point simulates the potential emissions from ships offshore the bay, considering the possibility of emissions derived from cleaning of bilges ships or accidental releases (Figure 1).

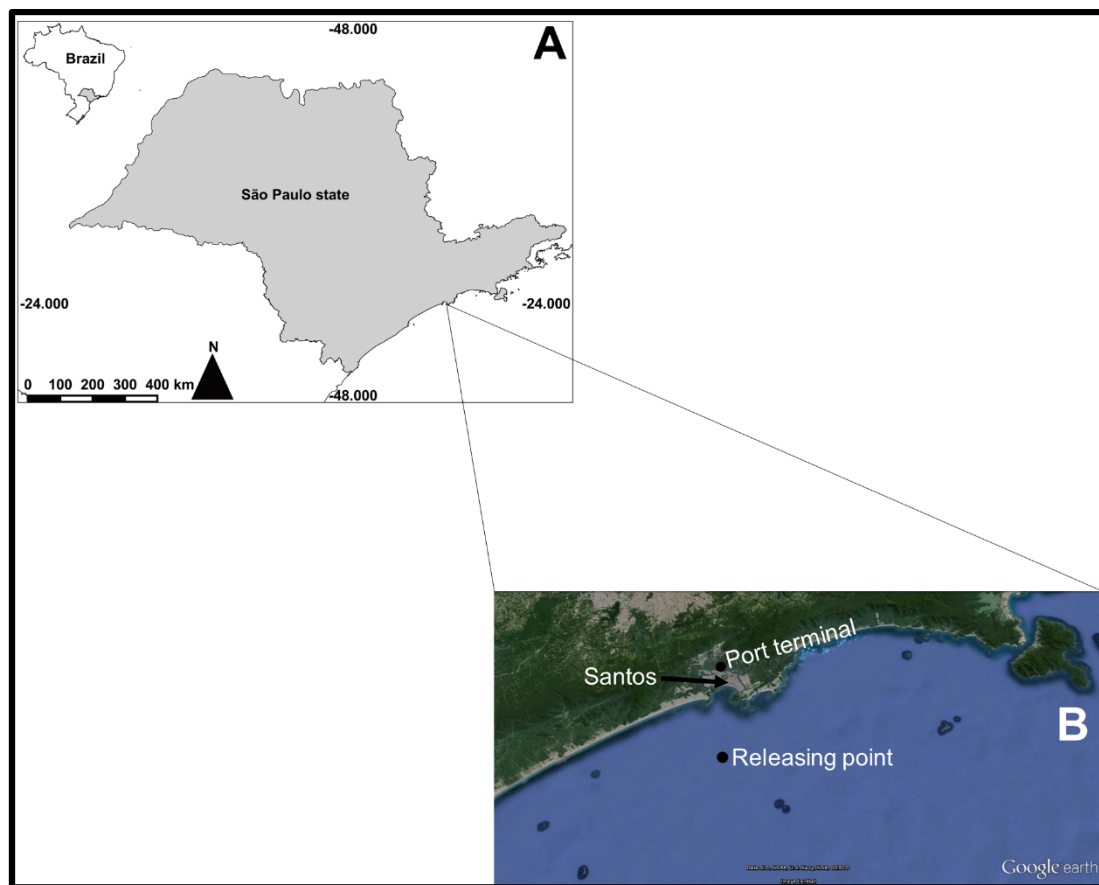


Figure 1. A - São Paulo State coast. B - Location of the releasing point considered in the simulations of microplastics dispersion in São Paulo State coast. B also shows the location of Santos bay and of the port terminal.

Therefore, the simulations contemplate the possibility that ships localized in anchoring region off Santos Bay could act as sources of pellets, especially during the winter season, when winds due to cold fronts cause greater dispersion of the pellets, that may reach the whole coastline.

Results

The simulations pointed to spatial and temporal patterns on the dispersion of microplastics. Once released, these particles tend to disperse to the vicinities of the releasing area. Further, the majority of microplastics disperse in north - south direction. On the other hand, the paths followed by these particles were different among the months considered in the simulations. The movement of particles in the water column corresponds to the predictions conducted for the monthly ocean dynamics in the simulated domain.

The ocean circulation varied along the period considered in the simulations and was more intense closest to the coast. In response to this process, the dispersion of the particles varied according to the month. In January, despite of a revolving of the particles to oceanic regions located southwestward from the releasing point, there was a dispersion to coastal regions located both northeastward and northwestward from the origin. In February, the particles dispersed to oceanic waters located southwestward from the releasing point. On the other hand, the dispersion to coastal regions located southwestward also occurred in March and April (Figure 2).

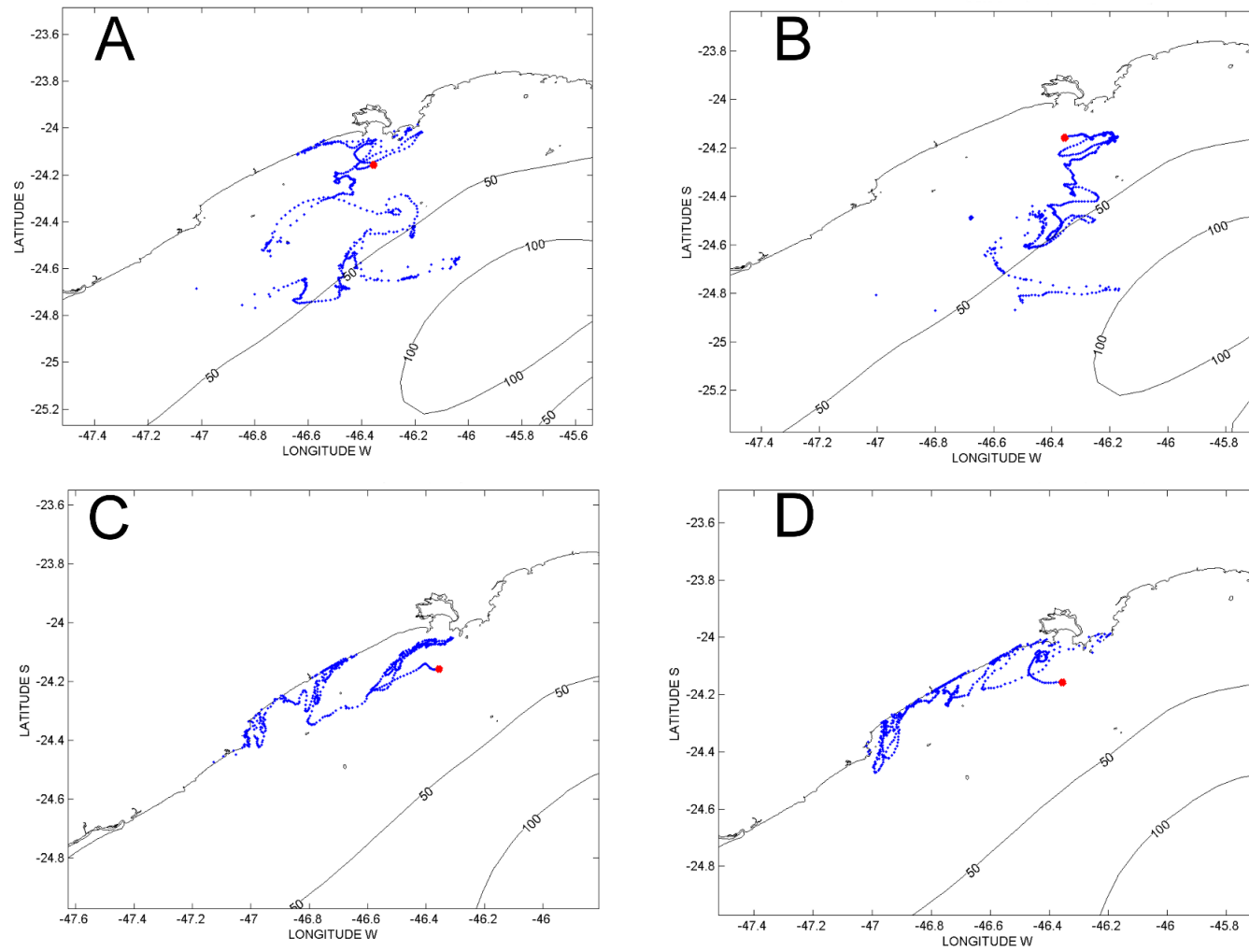


Figure 2. Results of the particles dispersion simulation. A - January, B - February, C - March and D - April. The red dot represents the releasing point

The dispersion of the particles to coastal areas nearby the releasing point, both southwestward and northeastward, persisted in May. In June, despite of a dispersion to adjacent coastal areas, the particles stayed closer to the releasing point. In July, the main dispersion of the particles was to coastal areas northeastward from the releasing point. This process also occurred in August; however, there was a southward dispersion to oceanic waters (Figure 3).

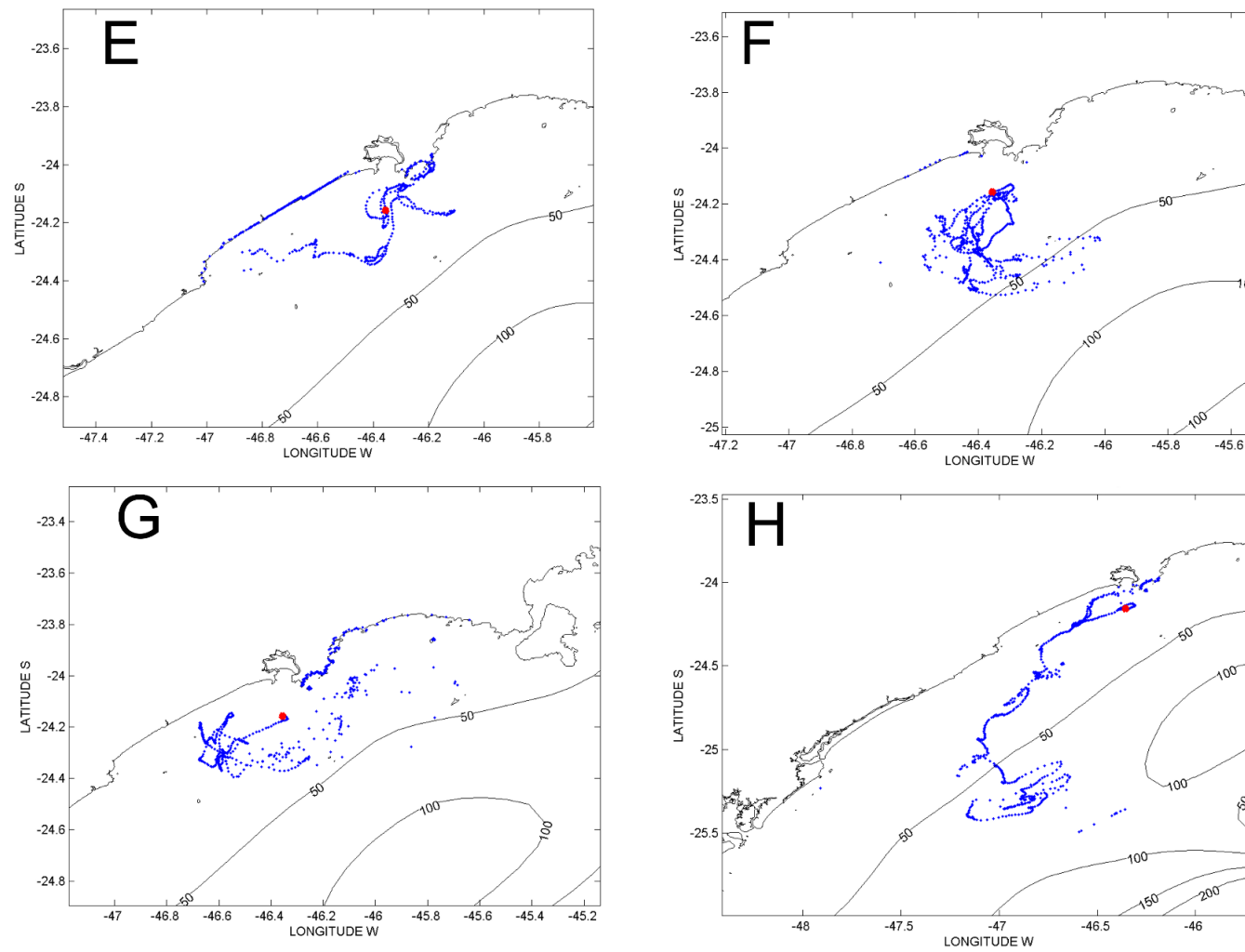


Figure 3. Results of the particles dispersion simulation. E - May, F - June, G - July and H - August. The red dot represents the releasing point.

The process of particles dispersion to coastal areas located southwestward continued in September. However, in this month the particles also dispersed northeastward and to Santos region. In October, the particles continued dispersing southwestward, although not always in direction to the coastline. The southwestward dispersion of the particles continues in November. In this month, there is a predominant revolving of the particles to the coastline. On the other hand, this pattern changes relevantly in December, when there is not a dispersion of the particles to the coastline. In this month, the particles stayed in the vicinities of the releasing point (Figure 4).

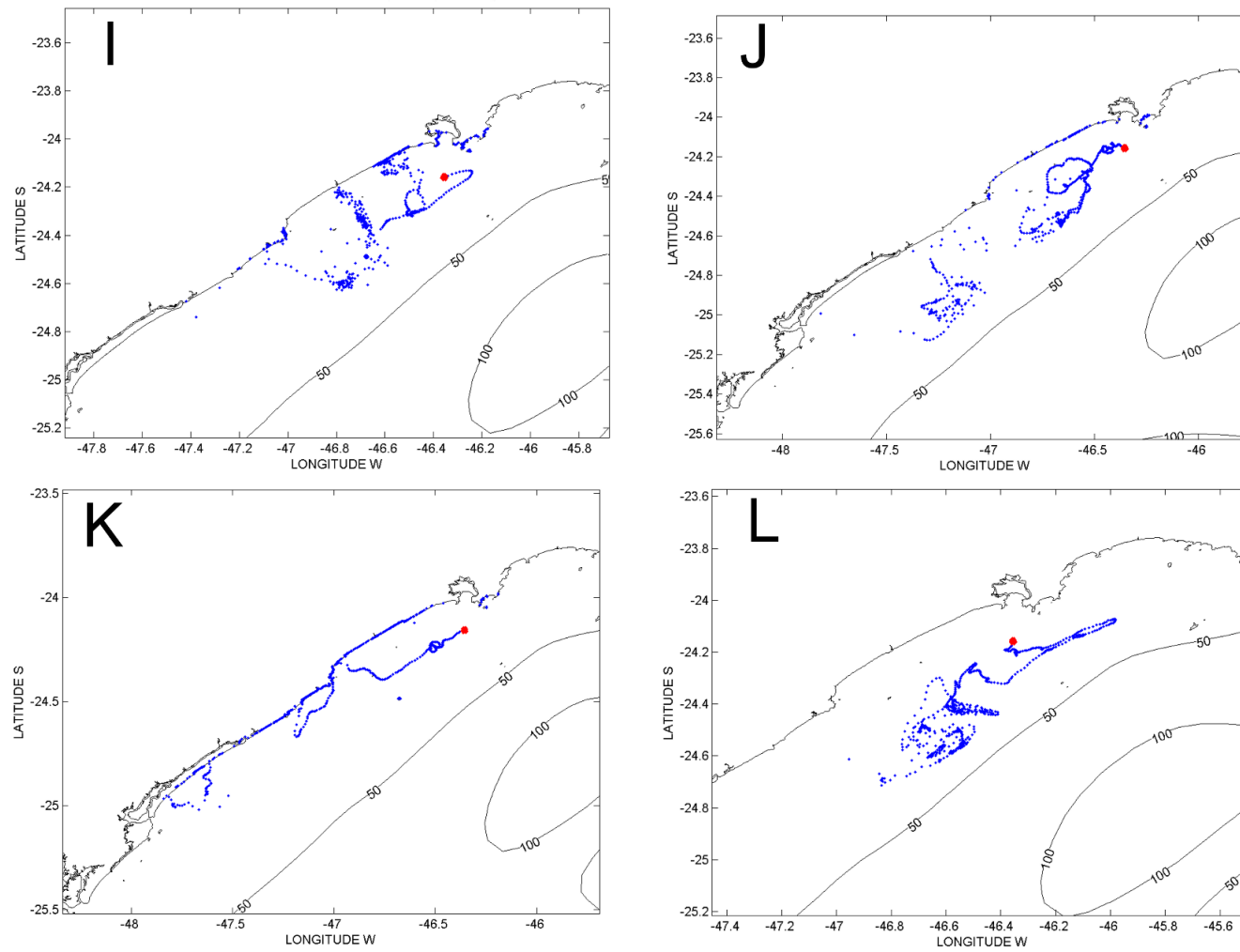


Figure 4. Results of the particles dispersion simulation. I - September, J - October, K - November and L - December. The red dot represents the releasing point.

Discussion

The results of the simulations indicate that, along the year, the microplastics dispersion is variable according to the month, with a greater dispersion to the coastlines in western São Paulo State. The eastward dispersion of microplastics occurs especially in months influenced by cold fronts. The spatial dispersion of plastic particles and their final arriving depends on the water motion that results from the direction and the velocity of the wind (ISOBE et al., 2014). According to the synoptic synthesis conducted by the Brazilian Institute for Space Research, the area of study suffered 27 cold fronts in 2012. The cold fronts occurred every month but in February and December. There were one cold front in January, two in March, May, August, October and November, three in September and June, four in April and six in July. According to CASTRO (1996), the main currents in the internal shelf of São Paulo State occur parallel to the coast. Under the influence of east – northeast winds, the currents follow to the southwestern direction. However, under the influence of cold fronts, when southern winds are stronger, the currents develop to northeastern direction. It is probable that the observed variation of microplastics revolving in the water column result from the response of the ocean circulation to the climatic variation.

The variation in hydrometeorological conditions might be the responsible for the differential arriving of microplastics along the coastline, so that even with a tendency of a greater southwestward dispersion, these particles might also reach northeastward coastal areas. GUTIÉRREZ et al (accepted) point that, under the influence of high-energy hydrometeorological events, even microplastics inputted in estuarine areas might disperse to areas distant from the

original source. The authors simulated the dispersion of microplastics released in the estuarine channels of Santos region; their results indicate that, during cold fronts, the release of microplastics by the estuary is greater and these particles drift to regions that are distant from the releasing point. Further, the simulations conducted by BALLENT et al (2013) indicate that there is a relationship between massive dispersion of microplastics and high-energy meteorological events. It means that hydrometeorological events promote both pulses of microplastics input and dispersion of these particles in large spatial scales. The results of the simulations conducted in the present study indicate that the explanation for relatively great dispersion of microplastics (hundreds of kilometers) from the source of input might rely on the connection between the rate of input and the occurrence of cold fronts. In these situations, the Santos region acts as exporting area of microplastics for the São Paulo State coast. This finding points to the importance of areas of production and transportation of primary microplastics for the spatial differences, in regional scales, in the contamination of marine habitats by these particles.

In addition, the results observed by MOREIRA et al (2016b) indicates that the simulations predicted realistic patterns of plastic pellets distribution. Thus, applying of the POM proved the hypothesis that plastic pellets follow a north – south pathway after their release in the water column close to Santos Harbor. Moreover, this procedure also demonstrated that plastic pellets might follow a south – north pathway during cold fronts and reach beaches located northeastward from Santos city. The results of MOREIRA et al also indicate that the distance from Santos region is important for the promotion of regional spatial differences of plastic pellets abundance. However, the results of the present

study support this hypothesis by explaining the processes that control the plastic pellets dispersion after their release. Therefore, the simulation of the pathways followed by microplastics clarified an important factor involved in the promotion of particles sink in regional scales. This is a relevant finding given the urgency in disclosure of regions that receive relevant amounts of microplastic in the coastal zone (CÓZAR et al., 2014; BROWNE, 2015; THOMPSON, 2015).

Once present in coastal environments, microplastics might harm the physical environment and the biodiversity. For example, the abundance of microplastic might interfere on permeability and heat transfer of beach sediments (CARSON et al., 2011). Microplastics also promote chemical contamination in coastal habitats once that they are carriers of organic pollutants in sandy beaches (FISNER et al., 2013a, 2013b). Given this feature, microplastics might impact the coastal biota in a variety of ways. Factors like size and abundance turn microplastics into an easy catchable item for benthic invertebrates to feed on (WRIGHT; THOMPSON; GALLOWAY, 2013). Feeding on microplastics might input harmful substances to the body tissues of invertebrates (CHUA et al., 2014). Apart from ingestion issues, leaching of harmful substances to sedimentary habitats is also a risk associated to microplastics. NOBRE et al (2015) investigated the toxicity of stranded and virgin plastic resin pellets in the sea urchin *Lytechinus variegatus*. According to the authors, the toxicity of virgin pellets is greater and increases anomalous embryonic development. Thus, the results of the present work brings relevant information to the risk evaluation of the plastic pellets contamination given the indication of beaches that receive relatively greater amounts of those particles.

Studies on risk evaluation of the contamination of the coastal zone by plastic material often rely on field data in order to estimate abundance and chemical contamination (HIDALGO-RUZ et al., 2012). Recent papers on this issue indicate a lack of information on where and in which environments the surveys should occur (BROWNE; JAMBECK; MALLOS, 2015; IVAR DO SUL; COSTA, 2013). Predictions of the pathways followed by microplastics once released in coastal environments would point to areas in which the probability of occurrence is relatively greater. In addition, this knowledge indicates the temporal scales through which microplastics tend to reach a specific site after their input. The present results might have applicability not only for the case of microplastics, but for other classes of floating marine anthropogenic debris as well. Knowledge on debris input in marine environments is increasing (GALGANI et al., 2015), so tracking the pathways followed by this waste in coastal environments would help to explain and predict the final arriving. The present approach is also applicable in the specific case of fragments generated by the breaking of larger plastic material in marine environments. Once fragmented into mesoplastics (≥ 5 mm) and microplastics (< 5 mm), these materials disperse in coastal regions according to the size of the fragments (ISOBE et al., 2014). However, risk evaluations require the prediction of the pathways followed by these fragments in order to indicate the sinking zones specific for each size class (CÓZAR et al., 2014).

Thus, the present work introduces an important approach to the evaluation of the contamination of coastal zones by plastic materials. The presented results explain the promotion of sinking zones of microplastics in kilometeric scales by tracking the pathway followed by these particles after their input in coastal zones. Moreover, the results revealed that the interference of the

meteorological variations on ocean dynamics is responsible for differences in the spatial distributions of microplastics. The model simulated the dispersion of microplastics in a period of one year, however, it pointed out the interference of seasonal variations on the dispersion of these particles. Therefore, the model predicted relevant spatial and temporal trends for risk evaluation; this capability provides theoretical and practical basis to suggest that this approach is applicable in monitoring programs.

Chapter 2. Spatial and temporal variability in the stranding of plastic pellets in sandy beaches

Abstract

The contamination of marine habitats by microplastics is an urgent issue given the impacts of these particles on economy and biodiversity. Once present in marine habitats, microplastics might promote the dispersion of chemicals and entanglement to the biota that feeds on them given the misidentification as a food source. Plastic pellets are among the most abundant microplastics in sandy beaches. Studies on plastic pellets generally focus on their abundance and capacity to absorb organic and inorganic pollutants. However, recent results indicate that the plastic pellets stranding in sandy beaches is influenced by small-scale variability. The present study focused on the variability in plastic pellets stranding according to large spatial and temporal scales in order to introduce a baseline to long-term monitoring strategies. This survey occurred from February 2014 to March 2015. The sampling procedure suffered the interference of the occurrence of rain before and during the fieldwork. The results indicated that plastic pellets input varies in temporal and spatial scales, with interaction between both scales. In addition, Beaches located close to an eventual source show a great variability in the stranding, this processes is diluted with the increase of the distance from the main source. The results suggest that sampling procedures conducted in long-term monitoring should consider a refinement in the temporal and spatial scales of samples acquisition, the distance from the source and the meteorological variation.

Keywords: microplastics, input, large-scale variability, long-term monitoring.

Introduction

The marine environments have been subjected to the introduction of plastic debris worldwide as a consequence of the increasing usage of this material both in domestic and industrial activities (THOMPSON et al., 2009c; UNEP, 2009). A recent estimate indicated that in the year of 2010 the oceans received an amount of eight millions of metric tons of plastic waste from coastal countries (JAMBECK et al., 2015). The significant amount of the plastic material that reach the oceans has ecological and economic consequences. The financial losses promoted by the contamination of the oceans by plastic material may reach approximately US\$ 13 billion per year (UNEP, 2014a). Once present in the ocean, the plastic material promotes deleterious effects both to the physical environment and to the biota (BROWNE et al., 2015; GREEN et al., 2015; NELMS et al., 2015; ROCHMAN et al., 2013b). These deleterious effects suffer an increase given the fragmentation of larger pieces into microplastics (BAKIR; ROWLAND; THOMPSON, 2014; WRIGHT; THOMPSON; GALLOWAY, 2013). The approximate weight of these small particles in the oceans worldwide were estimated between 93 and 236 thousand metric tons (VAN SEBILLE et al., 2015).

Besides the fragmentation of larger pieces, microplastics also have industrial origin. Therefore, these small particles are classified as primary, coming from the petrochemical industry, and secondary, originated through fragmentation (BARNES et al., 2009; FENDALL; SEWELL, 2009; TURNER; HOLMES, 2011). There is not a consensus about the size of a particle considered as microplastic, however, recent reviews estipulate that particles ≤ 5 mm should be contemplated (ANDRADY, 2011; COLE et al., 2011). The chemical

composition of these particles reflects the classes of plastics used in industrial and domestic activities. Therefore, microplastics present in the marine environment are Polyethylene, Polypropylene, Polystyrene, Polyethylene terephthalate and Polyvinyl chloride (ANDRADY, 2011; COLE et al., 2011). The amount of these small plastic particles varies according to the habitat. At the open ocean, the greatest concentrations of microplastics are observed in oceanic gyres (ERIKSEN et al., 2013; LAW et al., 2010, 2014). Besides the accumulation in oceanic gyres, the amount of microplastics increase with the proximity to the shoreline (DESFORGES et al., 2014; SHERMAN; VAN SEBILLE, 2016).

Thereafter, the estimate about the presence of microplastics in the open ocean indicate an amount inferior to that expected (LAW et al., 2010; VAN SEBILLE et al., 2015). A possible explanation of this process is the accumulation of microplastics in coastal zones (BROWNE et al., 2011; LEE; SANDERS, 2014; WANG et al., 2016). Microplastics might accumulate in estuarine systems through inflows coming from the ocean, sewage inputs and by local fragmentation of larger pieces of plastic (BROWNE; GALLOWAY; THOMPSON, 2010; IVAR DO SUL; COSTA, 2013; SADRI; THOMPSON, 2014). Moreover, sandy beaches tend to accumulate expressive abundances of microplastics. FOK & CHEUNG (2015) found a mean abundance of 5595 items/m² in Chinese beaches. The concentration of microplastics might vary according to the beach compartment (TURRA et al., 2014; MOREIRA et al., accepted). In Slovenia, LAGLBAUER et al (2014) found a mean concentration of 155.6 particles/ kg of dry sediment in the intertidal zone and 133.3 particles/ kg¹ of dry sediment in the backshore zone. In fact, those particles are present in the strandline, in the backshore and in coastal

dunes (DEKIFF et al., 2014; HEO et al., 2013; JAYASIRI; PURUSHOTHAMAN; VENNILA, 2013; MARTINS; SOBRAL, 2011).

Plastic pellets represent a relevant amount of the microplastics found in beaches. ANTUNES et al (2013) observed that plastic pellets might represent 53% of the plastic debris present in Portuguese beaches. TURNER & HOLMES (2011), on the other hand, sampled an amount > 1000 granules/m² in central Mediterranean. Once present in sandy beaches, plastic pellets constitute an environmental risk given that they might absorb a relevant variety of contaminants. KARAPANAGIOTI et al (2011) found spatially different concentrations of PAHs, DDTs and PCBs in stranded pellets from Greece. The results of ROCHMAN et al (2013a) indicate that the pellets made from HDPE, LDPE and PP sorb greater concentrations of PAHs and PCBs in comparison to pellets made from PET and PVC. Given this dependence, the authors point that the tendency of increase in the plastics production worldwide, implies in a greater environmental risk for the marine life given that the majority of plastic products is made from pellets with great potential to sorb organic contaminants. Moreover, given their role as vector of pollutants in sandy beaches (FISNER et al., 2013a, 2013b), plastic pellets might also harm the biota through the leached chemicals (NOBRE et al., 2015).

The environmental risk promoted by plastic pellets demands the monitoring of long-term variations in the abundance and in the stranding of these particles in beaches. However, the evaluation of the dynamics of plastic particles in beaches needs to contemplate refined temporal and spatial scales in order to avoid the underestimation of abundance (IVAR DO SUL; COSTA, 2014; KUSUI; NODA, 2003; MOREIRA et al., 2016a). It is probable that the absence of an

adequate refinement promotes biases in estimates because the stranding data of marine litter in beaches varies significantly according to the temporal scale of the sampling procedure (RYAN et al., 2014; SMITH; MARKIC, 2013). Moreover, the stranding of anthropogenic debris in beaches responds to geomorphological and hydrodynamic processes that occur in different scales (KATAOKA; HINATA; KATO, 2013; MOREIRA et al., 2016a). MOREIRA et al (2016) was the first study that evaluated local-scales dynamics of plastic pellets stranding in sandy beaches; the authors observed that beach dynamic processes, such as the overlapping of strandlines might increase the small-scale variability in the stranding of plastic pellets. In order to decrease this variability, the authors suggested that, in evaluations of microplastics stranding, sampling designs should consider the marking of the position of high tide lines between consecutive tidal cycles, cleaning the sampling area prior to sampling and samples acquisition in continuous transects in the surface sediment of the intertidal zone. In regional spatial scales, the dynamics of plastic pellets stranding might respond to different processes, such as the distance from a possible source and oceanographic conditions, such that beaches located in the same region might show different dynamics of plastic pellets stranding.

Therefore, tracking the effect of the source of plastic pellets in the marine environment might also have a relevant impact in the adopted monitoring strategy. The knowledge on the geographic location of the source, as well as the rate of release of plastic materials would determine the temporal and spatial scales considering in the sampling procedure (DUIS; COORS, 2016; NEL; FRONEMAN, 2015; RIBIC et al., 2012). This set of information and procedures would allow the adoption of standardized estimations of plastic pellets stranding

in beaches and, consequently, promote a baseline for long-term monitoring of variations of these particles abundance.

Programs that intend to establish policies of reduction of plastic pollution in the marine environment would rely on long-term monitoring in order to evaluate the effectivity of their protocols. The monitoring of the effects of those programs on the abundance of plastic pellets in beaches should point to the reduction of this material in relatively long temporal scales. It is probable that the evaluation of the dynamics of stranding indicates if the abundance of plastic pellets reaching beaches is decreasing or remains unaltered after the implementation of the Operation Clean Sweep or other similar programs.

Therefore, the present study aims to evaluate the dynamics of plastic pellets stranding in beaches, considering spatial and temporal variations and the interference of a specific source of this material. The stranding might depend on the monthly variations on the oceanographic conditions and on the rates of plastic pellets loss in the source. Therefore, the present paper tests the hypothesis that the stranding is variable along time. In addition, this evaluation test the hypothesis that the stranding varies among the beaches present in the vicinities of the source in the marine environment. It is also probable that there is a link between the spatial variation in the stranding and the distance between the beaches and the source, such that beaches located closest to the source would receive greater abundances of plastic pellets. Therefore, the information presented by this evaluation, as well as the methods implemented in the fieldwork for acquisition of data, might serve as a complement for the establishment of monitoring programs, given that it sets a baseline for long-term monitoring of plastic pellets stranding.

Materials and Methods

Study area

This evaluation occurred from February of 2014 to March of 2015 in the central portion of the São Paulo state coast, between 23°47'37.8"S and 24°01'47.3"S. The presence of petrochemical industries and of one of the greatest port terminals of Latin America turns the region in a relevant site for the study of plastic pellets pollution (FISNER et al., 2013a, 2013b; NOBRE et al., 2015; SANTANA et al., 2016; TANIGUCHI et al., 2016; TURRA et al., 2014). One of the economical features of the region is the great activity of petrochemical industries. There is a relevant relationship between the production and transportation of plastic pellets in this region. The greater amount of the plastic pellets produced in these facilities is transported by the port terminal located at Santos city (PEREIRA, 2014). This terminal imports an amount between 350,000 and 400,000 tons and export between 450,000 and 500,000 tons of plastic pellets by year (PEREIRA, 2014).

According to TESSLER et al (2006), the geological evolution of São Paulo State coast suffers the influence of two processes. The first one is related to the surging of the Serra do Mar, an escarpment located in the southern/southeastern Brazilian coastline (SALGADO et al., 2014), and the subsequence subsidence of Santos Bight in late Cretaceous. The second process is related to the variations of the sea level during the Quaternary. The configuration of São Paulo coastline give baseline to its theoretical division since the late 70s (SOUZA, 2012; TESSLER et al., 2006). The most recent characterization, conducted by SOUZA (2012), described seven morphodynamic

compartments for São Paulo State coast. According to this characterization, the present study contemplated beaches from three compartments.

The beaches selected for the present study are Ocian, Canal cinco, Iporanga and Itaguapé. The compartment that keeps Ocian beach shows the most extensive beaches from São Paulo coastline. These are high-energy dissipative beaches with a NE-SW orientation; therefore, these systems are exposed to high-energy waves from S-SSE. In addition, the morphodynamic state of beaches in this compartment might turn into intermediary in response to meteorological conditions and differences in wave energy (SOUZA, 2012). Canal cinco and Iporanga are in the same compartment, which is characterized by an N-S oriented coastal bay. Beaches located in this bay, such as Canal cinco, show a low-energy dissipative state and an E-W orientation. The occurrence of cold fronts and erosive processes might promote alterations in the morphodynamic state of these beaches. The beaches located in the eastern portion of this compartment, such as Iporanga, are NE-SW oriented and their morphodynamic state is intermediary with dissipative tendencies (SOUZA, 2012). The main feature of the beaches in the compartment that keeps Itaguapé beach is the presence of a narrow coastal plain and an ENE-WSE orientation of the coastline. In the portion of this compartment where Itaguapé is located, the beaches show a dissipative morphodynamic state, with intermediary tendencies. It is also important to highlight that the sampling sites are characterized by semidiurnal tides. The selection of these beaches for the conduction of the present study relied on the fact that they pass through different hydrodynamic and morphodynamic processes and, therefore, constitute a realistic representation of

the variety of beach systems that might occur in regional spatial scales (Figure 5).

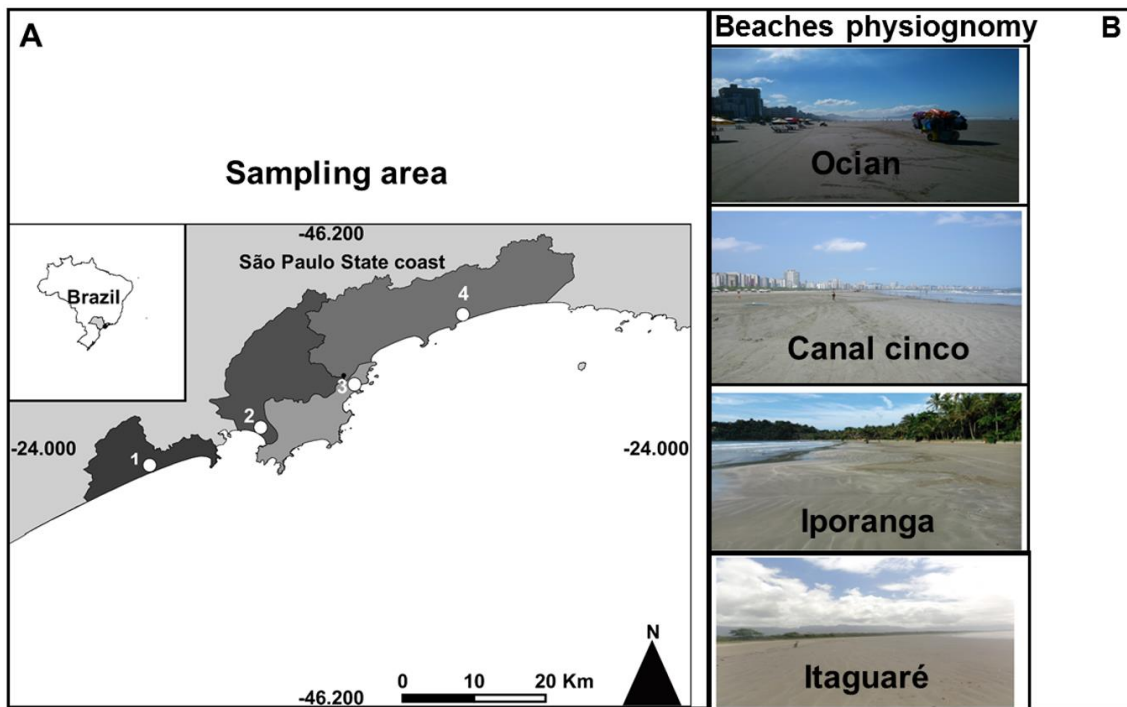


Figure 5. Region of Study. In A, number 1 indicates the location of Ocian beach, number 2 indicates Canal cinco, number 3 represents Iporanga beach and number 4 shows the location of Itaguapé. B shows the beaches physiognomy.

The beach of Canal cinco was not among the sampling sites until June 2014. However, given the great abundance of plastic pellets in this beach and its proximity to Santos port terminal (TURRA et al., 2014), it entered in the sampling procedure in order to improve the representativeness of regional differences and susceptibility to plastic pellets contamination in the evaluation.

Sampling procedure

In order to investigate the spatial and temporal variability in plastic pellets, a one-year period of sampling would detect interferences of seasonal or even monthly changes of the weather and of the plastic pellets movement promoted

by eventual sources (ocean and port terminal) over the average stranding in beaches. In addition, this period of sampling allows the occurrence of a monitoring in a relatively large temporal scale. Consequently, this sampling period brings information on how the applied method reveals the temporal variation of plastic pellets stranding in order to bring a realistic information to programs of long-term monitoring.

In each one of the sampling sites, a 100 meters area along shore was selected at the intertidal zone. At low tide, five across-shore transects with two meter width were randomly established. The across-shore extension of transects varied according to the amplitude of the tides. Each transect was cleaned with a squeegee, this procedure extended from the water line to five meters above the higher strandline. This procedure detached the plastic pellets already present in the sampling area in the beginning of the fieldwork. In the three subsequent low tides, the stranded pellets were detached from transects with a squeegee. This sampling extended from the higher strandline, evidenced by the upper marks leaved in the cleaned transect, to the water line (MOREIRA et al., 2016a). The processing of the detached material occurred on a 0.1 mm sieve and consisted in washing the mixture of sediment and the other materials. Acquisition of pellets from the washed material and posterior packaging occurred at sampling site. (Figure 6).



Figure 6. Sampling procedure. A: Pushing of the plastic pellets present in the intertidal zone with a squeegee. B: Accommodation of the detached sediment in the 0.1 mm sieve for posterior washing. C: Washed material containing plastic pellets.

Further, the replication of the sampling procedure occurred in the four beaches at periods of spring tides. This procedure would minimize the interference of weakly changes in the dynamics of stranding and, therefore, disclose the spatial differences with avoidance of bias in the data. Nevertheless, some samplings coincided with rainy periods. Thus, sampling occurred after rainy days (rainy days 1 or 2 days before sampling), in dry weather and in rainy days. In periods of constant and intense rain, there was accumulation of water on the studied beaches, which turned the surface of the sediment in to a muddy surface. The moist sediment also caused confusion about the extension of the intertidal zone and the location of the previously prepared transects. Further, in those weather conditions the revolving of the sediment promoted by the rain spreads or buries the plastic pellets, avoiding their unbiased sampling. These issues

prevented an adequate sampling of plastic pellets in May and July of 2014 for all the sampling sites and in August of the same year in Ocian and Itaguaré (Figure 7).



Figure 7. Accumulation of water in the surface of the sediment during periods of constant and intense rain, preventing sampling.

Data analysis.

The two-way analysis of variance (Two-way anova) was used to test the hypothesis of spatial and temporal difference in plastic pellets rate of stranding. The data acquired previously to September 2014 did not enter in the analysis given the gaps caused by the weather interference in the sampling procedure. Thus, the data set used in this evaluation consisted in the plastic pellets stranding sampled from September 2014 to March 2015. Two factors entered in the analysis: “Beaches” ($n = 4$), representing the spatial variation and “Months” ($n = 7$), representing the temporal variation. This analysis considered the abundance of plastic pellets, without data transformation. When the analysis of variance

indicated significant differences ($p < 0.05$), the Student-Newman-Keuls test (SNK) was used to conduct an *a posteriori* pairwise analysis in order to check in which levels the differences were present (UNDERWOOD, 1998).

The relationship between the distance from the Port terminal of Santos to the beaches and the monthly rate of plastic pellets stranding, from September 2014 to March 2015, relied on linear regression analysis. The mean abundance of plastic pellets by month in each one of the beaches entered in this analysis as the dependent variable. This data consisted in the summation of the abundance registered in each transect divided by the number of transects (5). The measurement tool from Google Earth enabled the achievement of the distance from the beaches to the port terminal. This data entered in the analysis as the effect variable.

Results

Despite the gaps in the sampling, the results obtained between February and August revealed that the meteorological variation promoted oscillations on the plastic pellets rate of stranding. Given that the fieldwork of the present study contemplated monthly samplings, it occurred in different meteorological conditions. Moreover, it is probable that the sampling frequency detected monthly differences in plastic pellets movement at the Santos port terminal and other facilities located both northward and southward from the study area. Thereafter, it is possible to observe that the mean abundance of plastic pellets was greater when the sampling procedure occurred posteriorly to rainy periods. On the other

hand, the abundance of plastic pellets decreased, in the sampling areas, during dry weather (Figure 8).

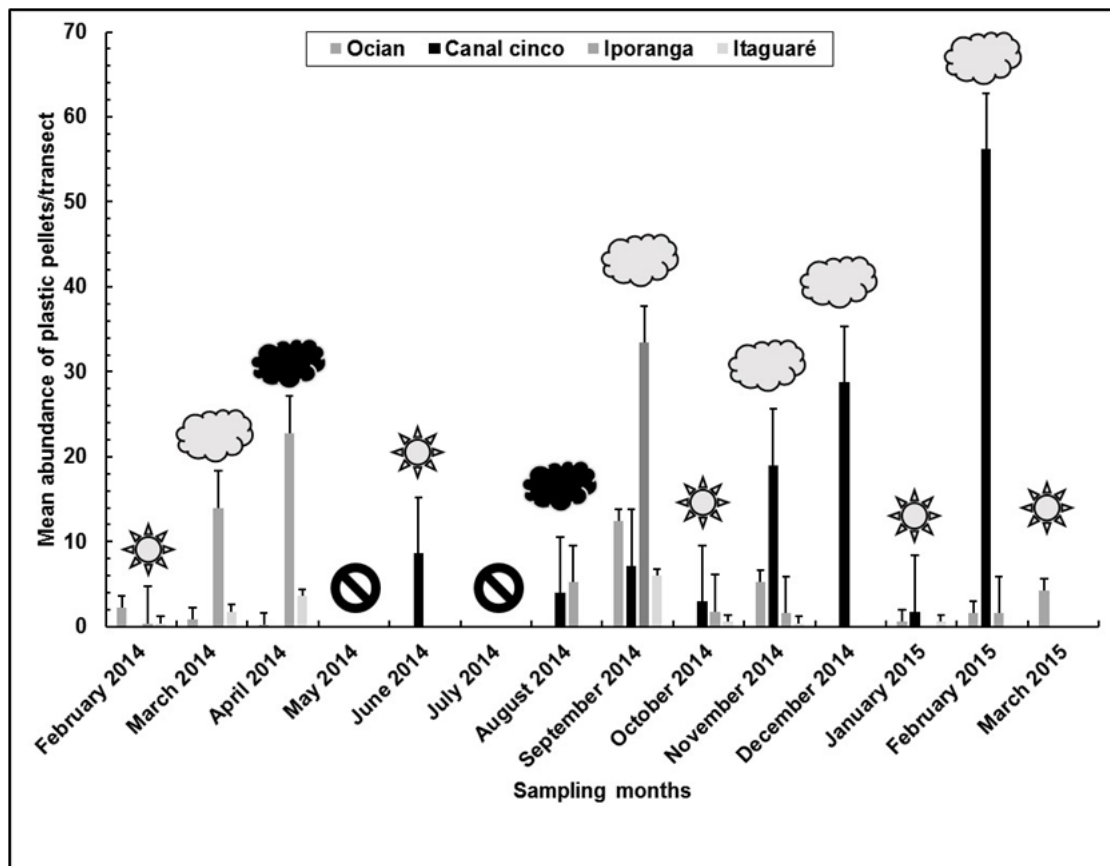


Figure 8. Weather interference in plastic pellets input and in sampling procedure. Weather condition during the monthly sampling where: ☀ = dry weather, ☁ = rain before sampling, 🌸 = rain during the sampling and 🚫 = absence of sampling. Bars represent mean abundance of plastic pellets/ transect \pm SE.

The two-way Anova revealed a significant interaction between Beaches and Months ($p < 0.001$, $F = 4.83$). This result indicates that there is a significant difference on the stranding rate of plastic pellets according to beaches and months; however, this difference does not occur among all the beaches in the months considered in the analysis. The SNK test revealed that the plastic pellets rate of stranding in Canal cinco in February 2015 was significantly greater than the observed in the other beaches during the period of study ($p < 0,05$). Moreover,

the stranding in Canal cinco in November and December 2014 is greater or similar among the possible comparisons ($p < 0,05$). In addition, the rate of stranding was greater or similar in Iporanga in September 2014 (Figure 9).

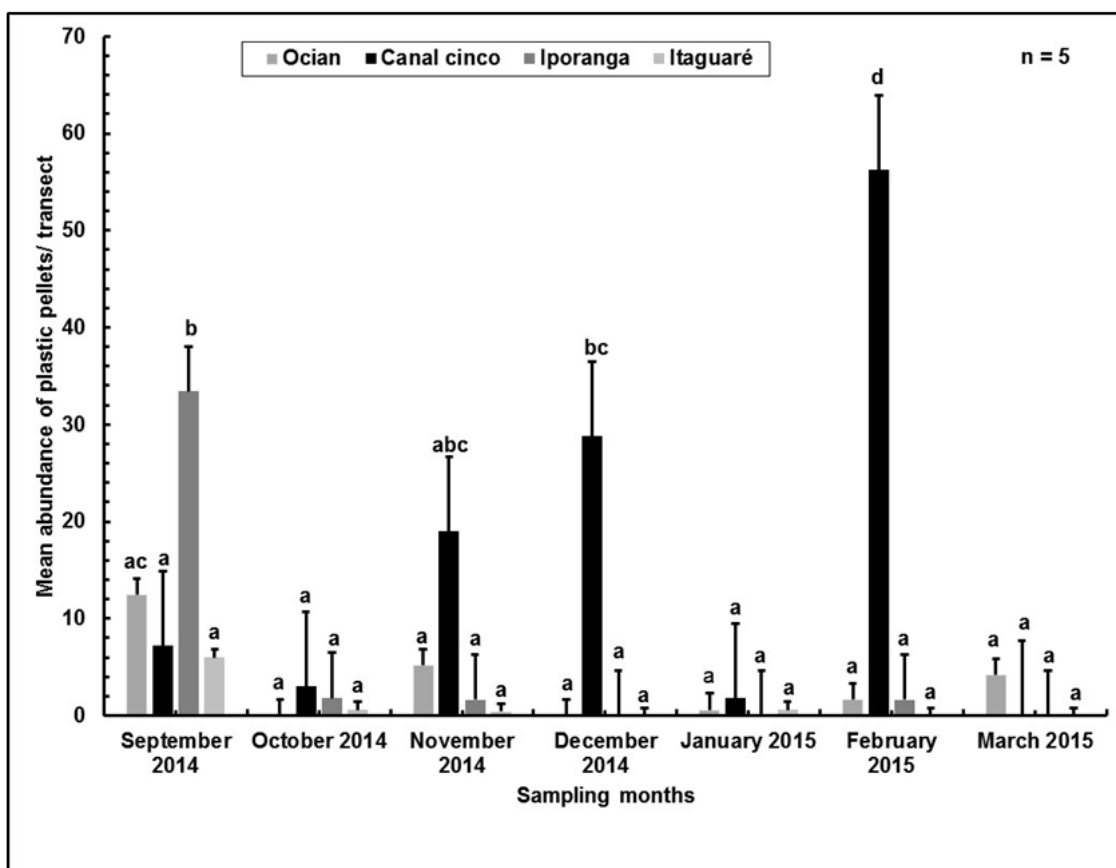


Figure 9. Results of the two-way Anova showing the significant interaction between the factors Months and Beaches. Where letters represent a $p \leq 0,05$ difference.

The results of the linear regression analysis revealed that there is a significant relationship between the distance of the beaches from the Santos port terminal and the monthly rate of plastic pellets stranding ($p = 0.028$). According to this analysis, beaches located closest to this facility receive the greater amount of pellets by month. Further, the results of this analysis revealed that there is a great difference among the stranding registered in each beach by month in

response to the distance from the port terminal ($R^2 = 0.17$). The negative value of the estimated effect of the distance in the variation observed indicates an inverse relation between these variables (-4.60). Therefore, the linear regression revealed that the monthly stranding of plastic pellets increases according to the proximity of an eventual source. It might indicate the occurrence of pulse releases of plastic pellets by the source. Moreover, the variability in the monthly abundance of plastic pellets decreases with the increasing distance from the port terminal (Figure 10).

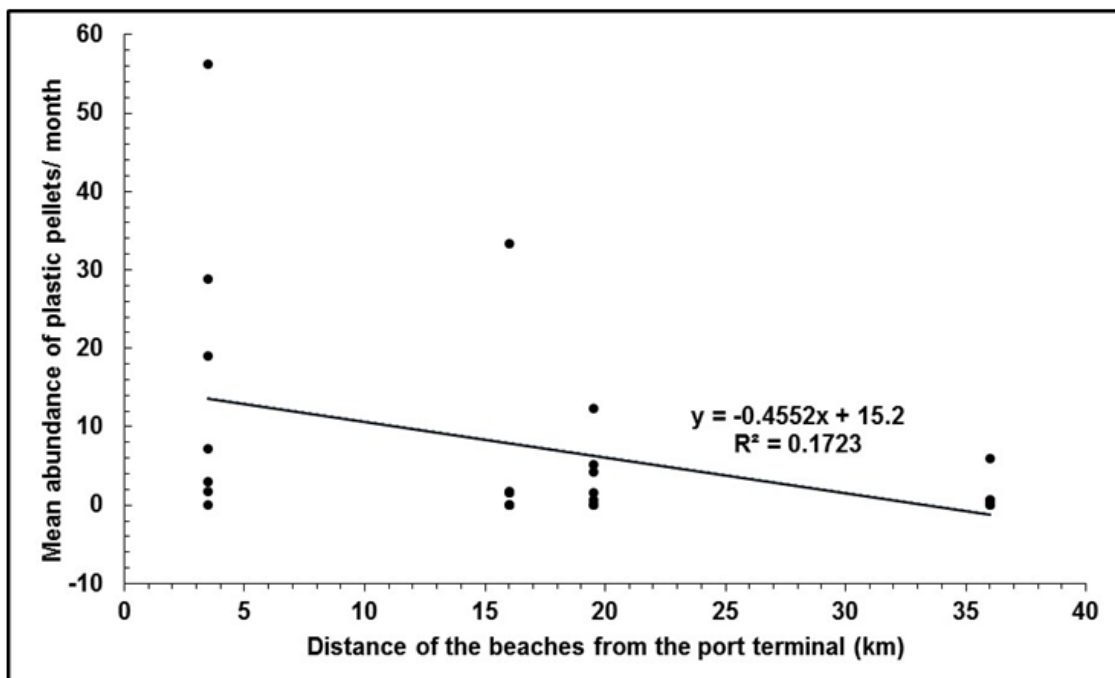


Figure 10. Linear relation between the distance from the beaches to the port terminal and the monthly mean abundance of plastic pellets. The black dots represent the mean abundance of plastic pellets per month and the black line represents the linear fit between the both variables. The R^2 denotes proportion of variance in the mean abundance, predicted by the distance between the beaches and the port terminal. The “y” equation denotes the observed values.

Discussion

The present study suffered the interference of the rain during the fieldwork; however, the data acquired during rainy periods raised important issues to consider in strategies of long-term monitoring of the plastic pellets input in sandy beaches. According to the results, the input of plastic pellets increases after rainy periods. Despite the compromising of the sampling procedure during these periods, a refinement in the temporal scale in the sampling procedure contemplated by long-term monitoring might ensure the samples acquisition in different meteorological conditions without relevant interferences. MOREIRA et al (2016) suggested that evaluations of plastic pellets input during consecutive tidal cycles for relatively long temporal scales might result in estimates that are more reliable. On the other hand, the present study conducted one sampling per month in each one of the sampling sites. This relatively long interval between samplings probably turned the sampling procedure more susceptible to large-scale variability promoted by the weather variation and by the rate of plastic pellets release by eventual sources. Daily temporal scales in the estimation of anthropogenic marine debris in beaches have been advocated in recent papers (RYAN et al., 2014; SMITH; MARKIC, 2013). However, daily sampling might be too much expensive and exhaustive for long-term monitoring. Thus, when there is the intention of monitoring the yearly variation in the stranding of plastic pellets, the fieldwork should consider a refinement in the interval between consecutive samplings that ensures the tradeoff between time spent in the field and samples representativeness. In addition, refined sampling procedures would contemplate environmental variables in small temporal scales.

Further, the increase of plastic pellets abundance in the strandline after rainy periods became clear during the fieldwork. A relationship of the increase in microplastics abundance in the strandline and rainy days has been suggested in recent papers (JAYASIRI; PURUSHOTHAMAN; VENNILA, 2013; MOREIRA et al., 2016a). The intensifying of the sediment revolving during rainy periods might be related to this process (TURRA et al., 2014; ZALASIEWICZ et al., 2016). In addition, the greater intensity of the wind would revolve the microplastics that are present in the water column and bring them to the coast (LEE; SANDERS, 2014; SADRI; THOMPSON, 2014). Further, the incidence of rain might facilitate the releasing of plastic material imprisoned in estuarine systems (IVAR DO SUL; COSTA, 2013; IVAR DO SUL et al., 2014; GUTÉRREZ et al., accepted). Thereafter, there might be a relationship between the plastic pellets rate of stranding and the meteorological variation, such that the abundance of granules entering a given beach increases during rainy periods. In order to estimate the plastic pellets input in long-term temporal scales, monitoring programs should relate the occurrence of rain or historical pluviometric data sets to the period in which the samplings occur.

Therefore, the relationship between the stranding of plastic pellets and the climatic variation highlights the importance of the scales in which the sampling occurs. The analysis of variance proved the hypothesis of temporal and spatial variation in the plastic pellets rate of stranding. On the other hand, the differences in this process do not occur among all the months and beaches studied. The samples acquired in November and December 2014 and in February 2015 in Canal cinco beach were among the main responsible for the observed differences. In the specific case of primary microplastics, recent reviews indicate

that the temporal variability in the abundance of particles in coastal zones might be related to the rate of productivity of the sources (BROWNE, 2015; THOMPSON, 2015). Thus, in months when the accommodation of plastic pellets in the port terminals and their maritime transportation are intensified, the introduction of plastic pellets in the marine environments is greater given the loosing of these particles during these activities (PEREIRA, 2014). In addition, the stranding of microplastics is greater in periods when high-energy climatic events are more frequent, given the intensifying of hydrological and geological processes in the coastal zone (BOWMAN; MANOR-SAMSONOV; GOLIK, 1998; KATAOKA; HINATA; KATO, 2013). It suggests that the temporal scale of the sampling should contemplate information of the transportation activities of port terminals and periods of intensification in the frequency of high-energy climatic events.

Further, spatial differences in the rate of stranding also tend to respond to sources of introduction of plastic pellets in the marine environment. In the present study, the beach of Canal cinco showed the greatest stranding of plastic pellets. The rate of stranding of plastic pellets is greater in this beach given the fact that it is located closer to the Santos port terminal in relation to the other studied beaches. In addition, Canal cinco is located close to the mouth of the estuarine system in which the port terminal is located, such that the hydrological and sedimentary processes that occur in this beach suffer the interference of the tidal currents that come from the estuary (HARARI; CAMARGO, 2003). According to the results of GUITÉRREZ et al (accepted), beaches located in Santos bay receive microplastics both from the estuary and from the ocean, but also export these particles to other regions. Therefore, it is probable that Santos bay receives

plastic pellets prior to the other beaches, which results in the differences observed in the present study. The abundance of plastic debris tend to be related to coastal processes whose interference reaches spatial scales of tens of kilometers, such as winds and coastal circulation (KIM et al., 2015). This interference would result in greater abundances of plastic particles occurring closest to coastal sources (BROWNE et al., 2011; CARSON et al., 2013). On the other hand, the abundances evidenced in Canal cinco are not always greater than that observed in the other beaches of the study, indicating that large-scale processes are also involved in the geographic difference. In fact, plastic pellets have been observed in beaches located far from industrial or urban centers (DO SUL; SPENGLER; COSTA, 2009; MCDERMID; MCMULLEN, 2004). However, the present results suggest that strategies of long-term monitoring should occur in localities where the abundance of plastic pellets is relatively greater than other beaches given its proximity to introduction vectors. On the other hand, the present results also indicate that beaches located far from eventual sources might show a lower variability in the stranding. Therefore, alternatively to the above suggestion, long-term monitoring strategies might occur in localities relatively far from eventual sources, where the stranding is constant.

The negative relation between monthly stranding of plastic pellets and the distance of the beaches from the port terminal highlights the susceptibility of beaches located in Santos bay to stranding that occur in pulses, such that those beaches receive the greater amounts of plastic pellets by month. Despite of the scarcity of information on the relationship between the distance from the source and the stranding of plastic pellets, recent results on the abundance of plastic litter indicate that the temporal dynamics is influenced by the distance of industrial

and urban centers (DAVIS III; MURPHY, 2015; LAGLBAUER et al., 2014; ROSEVELT et al., 2013). In the specific case of plastic pellets, the coastal processes would bring the greater amount to areas located near by the releasing point given the relatively short distance to run. Thereafter, the rate of stranding decreases according to the increasing of the distance from the port terminal. In more rainy months, the input in beaches located closest to the source would be even greater given that the plastic pellets outflow from the port terminal in pulses (ABU-HILAL; AL-NAJJAR, 2009; MOREIRA et al., 2015; TURNER; HOLMES, 2011). This rationale indicates that, besides sampling in the vicinities of eventual sources, long-term monitoring strategies should consider the differences between chronic and episodic stranding.

Studies on beaches contamination by plastic pellets usually focus on the spatial distribution and on the association of these particles to chemical pollutants (ABU-HILAL; AL-NAJJAR, 2009; FERNANDINO et al., 2015; JAYASIRI; PURUSHOTHAMAN; VENNILA, 2015; LLORCA et al., 2014; ROCHMAN et al., 2013a; TURNER; HOLMES, 2011). However, the spatial and temporal variation of plastic pellets stranding and the role of this dynamics in promoting long-term oscillation in the abundance still need clarifying. A possible reason of this lack of information might be the absence of standardization of sampling methods and strategies. Recently, MOREIRA et al (2016) suggested that surveys on the dynamics of stranding should include temporal and spatial small-scale variability in order to identify the existing patterns. Although considering greater spatial and temporal scales, the present study was the first to conduct an estimative of the stranding that considered the methodological suggestions of MOREIRA et al (2016) in order to eliminate biases in sampling. The variability in relatively large

spatial scales observed in the present study (tens of kilometers), as well as the interference of the meteorological variation on the sampling procedure indicate that long-term surveys of the plastic pellets stranding should consider the distance from eventual sources and a refinement in the temporal scale of the sampling procedure. Thereafter, it is plausible to suggest monthly samplings during consecutive days, standardized according to subsequent tidal cycles. In order to improve the feasibility of this procedure, the area to be sampled might be subdivided in transects, which should be cleaned before each sampling (MARTINS; SOBRAL, 2011; MOREIRA et al., 2016a).

Thus, the present study brings important information to the reasoning about methods for sampling the stranding of plastic pellets in sandy beaches (e.g. HEO et al., 2013; HIDALGO-RUZ; THIEL, 2013; HIDALGO-RUZ et al., 2012; MOREIRA et al., 2016). The suggested procedure avoids the interference of the meteorological variation and improves the tradeoff between sampling effort and sample representativeness by targeting highly contaminated areas during a temporal scale adjusted to the dynamics of debris stranding in sandy beaches. For long-term monitoring, this procedure might be applicable to investigate the effectiveness of programs that try to reduce the lost and subsequent stranding of plastic pellets in the marine environment (MOREIRA et al., 2016a).

Chapter 3. Small-scale dynamics of plastic pellets input in sandy beaches

Abstract

The occurrence of microplastics in beaches might promote a relevant variety of environmental impacts. In sandy beaches, microplastics might leach chemical pollutants in the sediment column and promote alterations in physical processes. Among the microplastics present in beaches, plastic pellets might reach relevant abundances in different spatial and temporal scales. Recent studies on the standing-stock of plastic pellets in beaches point that this parameter varies along-shore. The standing-stock of plastic pellets in beaches respond to a variety of environmental factors, such as the occurrence of high-energy oceanographic events. On the other hand, the spatial and temporal dynamics of the rate of input might show a relationship with the standing-stock in local scales. The present paper investigated this relationship and the spatial and temporal dynamics of the input in Santos bay, southeastern Brazil. The investigation of the input dynamics occurred in two periods. Thereafter, these data were used to test the relationship between input and standing-stock. The results indicate that the input varies in temporal and spatial scales, with a significant interaction between both. On the other hand, there was no significant relationship between input and standing-stock. The explanation of the variability in the rate of input might rely on local-scale differences of wave energy and sedimentary processes. On the other hand, input and standing-stock occur in different temporal scales, which prevents the occurrence of a significant relationship.

Keywords: microplastics, input, standing-stock, local scales.

Introduction

The contamination by plastic material is amongst the major environmental problems in coastal zones worldwide (GOLDBERG, 1995; JAMBECK et al., 2015; JUAN et al., 2014). In addition to environmental and biological impacts, the persistence of plastic pollution causes economic problems to coastal zones once that it might interfere on fisheries and tourism (BROWNE et al., 2015; CORCORAN; MOORE; JAZVAZ, 2014; ROCHMAN et al., 2013b; UNEP, 2014b). Amongst the coastal environments, the sandy beaches receive expressive amounts of plastic material, even when protected by environmental policies or located in remote areas (ERIKSSON et al., 2013; HARDER et al., 2015; KAKO et al., 2014; TOPÇU et al., 2013). In response to the sedimentological and hydrological processes that occur in sandy beaches, the plastic material is present from the surface to deep layers of the sediment column and from the strandline to the coastal dunes (DEKIFF et al., 2014; KUSUI; NODA, 2003; MCDERMID; MCMULLEN, 2004; TURRA et al., 2014). In addition to the abundance an occurrence, the plastic material is present in different size classes in sandy beaches. The term microplastics comprehend particles whose size variates from one to five millimeters (ANDRADY, 2011; COLE et al., 2011). Recent papers consider the abundance of microplastics one of the most urgent contaminations in sandy beaches (HEO et al., 2013; HIDALGO-RUZ; THIEL, 2013; LEE et al., 2013).

The plastic pellets are amongst the most abundant microplastics present in sandy beaches (TURNER; HOLMES, 2011). Plastic pellets reach marine ecosystems probably trough transportation losses in port terminals (EPA, 1992;

FOTOPOULOU; KARAPANAGIOTI, 2012; PEREIRA, 2014; TURNER; HOLMES, 2011). In addition, the abundance of these particles in sandy beaches is probably related to the proximity to petrochemical facilities (KARAPANAGIOTI; KLONTZA, 2008; TURNER; HOLMES, 2011). Pellets can be found stranded and/or buried in beach sediment (KUSUI; NODA, 2003; MCDERMID; MCMULLEN, 2004; MOREIRA et al., 2016a; TURRA et al., 2014). Previous works described an irregular distribution of pellets in sandy beaches, such that the abundance increases from the strandline to higher portions of the backshore and from the surface of the sediment to one meter depth (TURNER; HOLMES, 2011; TURRA et al., 2014). Further, plastic pellets distribution might also be spatially different in the along shore direction in sandy beaches, as so as other components of the anthropogenic litter present in these environments (BOWMAN; MANOR-SAMSONOV; GOLIK, 1998). These spatial differences might be related to the dynamics of input (ANDRADY, 2015; THOMPSON, 2015).

Different factors interfere on the dynamics of stranding of marine anthropogenic litter in sandy beaches. BOWMAN et al (1998) showed that the presence of morphological structures such as ridges or tunnels, in the backshore, tend to act as efficient traps for the litter. The same authors also pointed out that wider beaches receive greater amounts of litter, while narrower beaches tend to present a greater trade-off between inflow and outflow of litter and, consequently, lower levels of accumulation. THORNTON & JACKSON (1998) estimated that small plastic particles tend to reach upper portions of the backshore given the susceptibility of these particles to the influence of the wind and wave processes. In the specific case of plastic pellets, the ocean dynamics are usually suggested as possible factors that interfere on the input, given that these granules are

present even in beaches located far from sources (COSTA et al., 2010; DO SUL; SPENGLER; COSTA, 2009).

The coastal processes that occur in estuaries also interfere on the stranding of plastic particles in sandy beaches (BROWNE et al., 2011). The sedimentological and hydrological dynamics of estuaries might interfere significantly on the distribution of microplastics. In estuaries, microplastics are more abundant in downwind areas that receive greater deposition of fine sediment (BROWNE; GALLOWAY; THOMPSON, 2010). The occurrence of seasonal climatic features also interfere on the stranding, given that areas that receive greater amounts of litter might alternate during the occurrence of these events in estuarine environments (RECH et al., 2014). In addition, tidal cycles interfere on the dispersion of microplastics from estuaries to open waters, given that greater amounts of particles leave these environments at spring tides (SADRI; THOMPSON, 2014). However, the input of marine litter in sandy beaches is highly dynamic, such that daily events might be decisive to the amount of particles that reach the coastline (ERIKSSON et al., 2013).

The stranding represents the stranding of microplastics in a given marine habitat after their release by a source and is more likely to occur punctually and in pulses (MOREIRA et al., 2016a). On the other hand, the standing-stock represents a chronical contamination and, therefore, contemplates the microplastics that accumulated in marine habitats (TURRA et al., 2014). Thus, the stranding might constitute one of the processes related to the promotion of the standing stock observed in sandy beaches once that it interferes on the availability of plastic particles for accumulation in the sediment column (BOWMAN; MANOR-SAMSONOV; GOLIK, 1998). However, recent results

indicate that the burial of plastic pellets in sandy beaches respond to the sediment mixing promoted by high energy events (TURRA et al., 2014). WILLIAMS & TUDOR (2001) also suggested that burial of small plastic pieces only occurs in response to a very large depositional phase of wave energy. According to these authors, the dispersive stress of sediment grains controls hydraulic equivalence of deposition and promotes inverse grading of sediments by grain flow. Therefore, both the standing stock and the dynamics of stranding tend to respond to ocean dynamics, beach morphology, weather conditions and to the proximity to sources of plastic particles (RYAN et al., 2009b). Thus, it is reasonable to infer that areas of greater standing stock coincide with areas of greater stranding.

Therefore, the present study aimed to evaluate the along shore stranding of plastic pellets in beaches and to test the connection between this process and standing stock. To that end, the hypothesis that the stranding of plastic pellets is different along shore was tested. In addition, the present study tested the hypothesis that the stranding is different in relatively small temporal scales. Further, the relationship between stranding and standing-stock was evaluated. Given that the stranding is a punctual process and the standing-stock reflects a long-term and chronic contamination, the results presented in this evaluation describe how the spatial dynamics of the stranding promote spatial differences in the standing-stock. Therefore, the study present useful information for evaluations of the environmental risk promoted by the spatial distribution of plastic pellets in beaches to the biota (NELMS et al., 2015) and to the physical environment (ANTUNES et al., 2013).

Materials and Methods

Area of Study

The conduction of the present study occurred in Santos bay ($23^{\circ}56'27''\text{S}$ $45^{\circ}19'48''\text{W}$), located at the central portion of São Paulo State Coast, Brazil. The study area presents 7 km of extension, the coastline is south-southwestward oriented, protected from waves on the east side, partially protected on the southeast side and completely exposed on the south and southwest (ELLIFF et al., 2013). This pattern of exposition suffer the influence of the Urubuqueçaba Island, located in the southwestern portion of the study area (MAGINI et al., 2007). The geographic orientation of this area turns it subjected to the interference of cold fronts, and currents coming from southern regions (HARARI; CAMARGO, 1998; HARARI et al., 2002). The city of Santos receives estuarine waters coming both from the Santos Channel in northeastward and from São Vicente Channel in northwestward directions (GREGORIO, 2009). In addition, the coastline presents seven intern channels, separated by a distance of approximately 1 km, responsible for the pluvial drainage from Santos city (ITALIANI, 2014) (Figure 11).

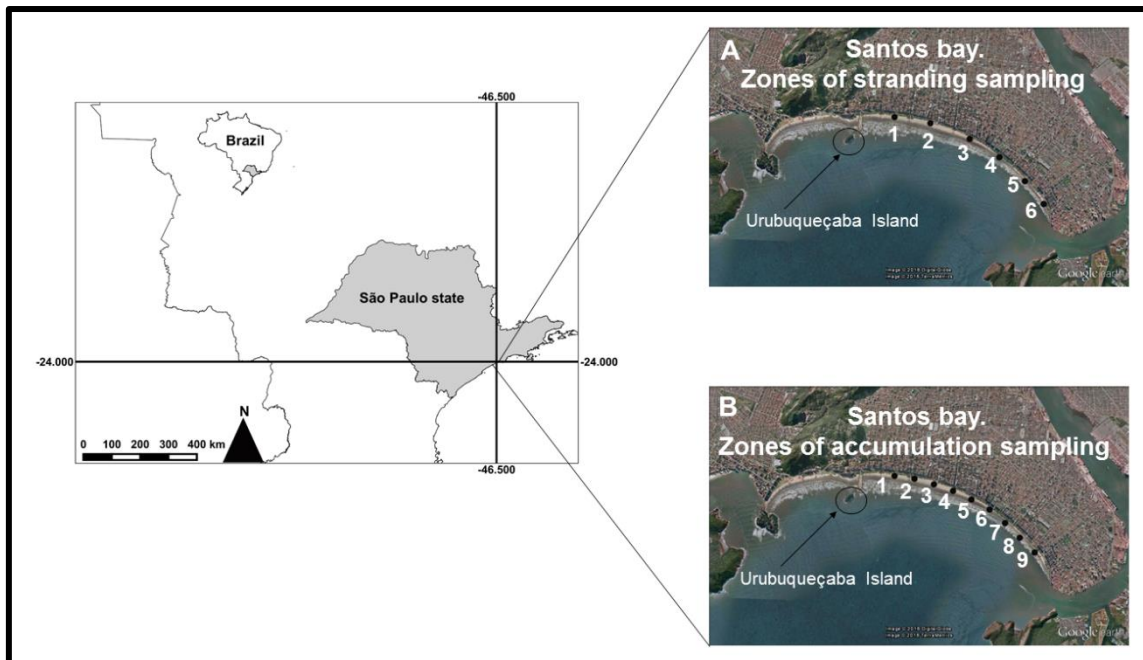


Figure 11. Location of the study area in São Paulo State, southeastern Brazil. A – Zones where the study of the input of plastic pellets occurred in Santos bay. B – Zones where the study of standing-stock of plastic pellets occurred in Santos bay.

The presence of petrochemical industries and of one of the greatest port terminals of Latin America turns the Santos bay in a relevant site for the study of plastic pellets pollution (FISNER et al., 2013a, 2013b; NOBRE et al., 2015; SANTANA et al., 2016; TANIGUCHI et al., 2016; TURRA et al., 2014). One of the economical features of the region is the great activity of oil refinery and petrochemical industries. A relevant amount of the plastic pellets produced in these facilities is transported by the Santos port terminal (PEREIRA, 2014). This facility may import an amount between 350.000 and 400.000 tons and export between 450.000 and 500.000 tons of plastic pellets by year (PEREIRA, 2014).

Sampling procedure

Stranding of plastic pellets

Sampling occurred in November of 2014 and in March of 2015, representing, respectively, the beginning of summer and the beginning of autumn in Brazil. In November, there was no rains or other high-energy climatic event during the period of fieldwork. On the other hand, there were sporadic rains and strong winds during samplings in March, which resulted in an expressive amount of debris in the sampling area (figure 12).



Figure 12. Presence of debris in the study area in rainy and windy weather

In addition, the sampling procedure occurred on cycles of spring tides. In order to evaluate the differences of rate of input along shore the sampling area, the division of Santos bay occurred according to six of the intern channel, such that the study contemplated six sampling plots. The selection of a portion of the bay relied on the improvement of the sampling effort given that sampling occurred simultaneously in the six plots during the tide cycles. In addition, the patterns of sediments deposition and the hydrodynamics of the selected portion of the bay

are well known (ELLIFF et al., 2013; HARARI; CAMARGO, 1998; HARARI; GORDON, 2001; MAGINI et al., 2007). Each one of these plots presented 100 meters on along shore extension at the intertidal zone, established in the center of the distance between two channels.

At low tide, five 2 m wide transects were randomly established on the across-shore direction. The across-shore extension of transects varied according to the amplitude of the tides. The conduction of the fieldwork occurred at the same moment in all sites by different teams. Each transect was cleaned with a squeegee, this procedure extended from the water line to the higher strandline. This procedure detached the plastic pellets already present in the sampling area in the beginning of the fieldwork. The sampling of occurred in the three subsequent low tides in order to adequate the tradeoff between sampling effort and sample representativeness to the number of team members and financial costs of the fieldwork. In these subsequent low tides, the stranding of pellets was recorded from transects with a squeegee. This sampling extended from the higher strandline to the water line. The processing of the detached material occurred on a 0.1 mm sieve and consisted in washing the mix of sediment and the other material present at the intertidal. Acquiring of pellets from the washed material and posterior packaging occurred at sampling site.

Standing-stock of plastic pellets

TURRA et al (2014) studied the standing-stock of plastic pellets in Santos bay. The samplings occurred in 15 zones, equidistantly separated, in the entire bay from July 2008 to December 2009. In each one of these zones, three

transects were placed along a 20-meter strip parallel to the waterline. These transects were sampled at 10 equidistant heights. Sediment was collected at ten 0.2 depth intervals until a depth of 2.0 m was reached. The sediment samples were acquired using a manual auger of 6 inches. However, from the 15 sampling sites present in the study of TURRA et al (2014), nine entered in the present evaluation. This selection ensured that the evaluation of the relationship between input and standing-stock in the same portion of the bay, given the abovementioned adjustments applied on the input sampling (Figure 15).

Data analysis

Stranding data

Given the fact that the meteorological conditions were different between the two sampling periods, data treatment was conducted separately between them in order to avoid a possible interference of the weather features in the input of plastic pellets. Statistical analysis considered the raw abundance of plastic pellets by transect in each one of the sampling zones by tide cycle.

The applying of two-way repeated measures analysis of variance (RM two-way Anova) on the abundance data estimated the differences according to sampling sites and tide cycles. Therefore, for each one of the sampling periods, the analysis considered the factors “Zones”, with six levels, representing the sampling zones and “Cycles”, with three levels, representing the three tide cycles. For both datasets, checking of variance’s heterogeneity relied on Cochran’s test. According to the results of the assumptions checking, it was necessary to

transform the both datasets in $\ln(x+1)$. The significant differences were a posteriori evaluated with the Student-Newman-Keuls test (SNK).

Standing-stock data

In order to evaluate the existence of significant differences in the standing-stock among the nine zones, the abundance data were transformed firstly in pellets by cubic meter by the formula:

$$Y = Ta / V * NS.$$

Where “Ta” represents the total abundance of plastic pellets in a given transect, “V” represents the volume of the auger, calculated considering it’s diameter in meters (0,1524m) in a depth of 2 m, and “NS” represents the total number of sampling points by transect. Further, this data were transformed in abundance of plastic pellets by strip transect (ST). The ST is a strategy of simple spatial interpolation of data that considers the abundance of each zone summed over meters on the area under study (DEFEO; RUEDA, 2002). In addition, the abundance of plastic pellets per strip transect is considered as a more accurate measure of standing-stock in relation to other methods of abundance estimation (MOREIRA et al, submitted). The calculation of the ST comprehends the formula:

$$ST = E * 0.1524.$$

Where “E” represents the extension of the transect and 0.1524 is the diameter of the auger in meters. Furthermore, the density of plastic pellets/ST was calculated for each transect considering the mean density of plastic

pellets/m³, the sampled area for the strip transect and the total sampled area (A*NS) in that transect by the formula:

$$X = Y*ST/ A*NS.$$

The checking of differences of accumulation among the zones relied on one-way analysis of variance (one-way ANOVA). In this analysis, the zones entered as factor (n = 9) and the abundance of plastic pellets/ST entered as dependent variable. Checking of variance's heterogeneity relied on Levene's test and checking of normality relied on Shapiro-Wilk's test. According to the results of the assumptions checking, no transformation was necessary. The significant differences were a posteriori evaluated with the Student-Newman-Keuls test (SNK).

Relation between stranding and accumulation

Firstly, the stranding data were spatially interpolated through the sampled extension on Santos bay (4 km), considering the datasets from the both sampling periods together. This interpolation relied on the Kriging method, which estimate values of a given variable, for an entire area, relying on its known values (FORTIN et al., 2012; GAETAN; GUYON, 2010). In this analysis, the sampling extension was divided in 40 distances, so that the evaluation of the stranding relied on a number of pellets/ 100 m. The same method of interpolation was applied on the data from TURRA et al (2014), so that a value of standing-stock of pellets/ 100 in the same extension was achieved.

Further, the relation between the both processes was evaluated through linear regression. The interpolated values of standing-stock of plastic pellets/ 100 m entered in this analysis as response variable ($n = 40$) and the stranded abundance of plastic pellets/ 100 m entered in the analysis as effect ($n = 40$). Prior to the analysis, the both datasets were standardized, given that they were obtained from data that are in different measurement units (pellets/St and raw abundance of pellets).

Results

Stranding

There was a significant variation of plastic pellets abundance according to the areas and the tide cycles in the samples from November. The RM Anova revealed a significant interaction between the factors. Thus, despite of the existence of a significant variation, a factor's effect depends upon the level of the other factor. This result indicates that the stranding is significantly different; however, it depends upon the portion of the bay and varies according to small temporal scales (hours) (Table 1).

Table 1. Repeated measures analysis of variance (RM Anova) comparing the mean abundance of plastic pellets among tide cycles and among sampling zones in November of 2014. The factor "Zones" had 6 levels and the factor "Cycle" had 3 levels ($n = 5$) Cochran's test evaluated the assumption of homogeneity of variances.

Effect	SS	DF	MS	F	p
Intercept	123.62	1	123.62	592.34	<0.001
Zones	7.76	5	1.55	7.44	<0.001
Error	5.00	24	0.20		
Cycles	5.08	2	2.54	25.99	<0.001
Cycles X Zones	4.39	10	0.43	4.48	<0.001
Error	4.69	48	0.09		

C = 0.47 Cycle 1

C = 0.35 Cycle 2

C = 0.36 Cycle 3

Ln (x+1)

Despite of the interaction between factors, the zone one showed greater or similar mean values in comparison to the other areas. Great abundance values also occurred in the zones 2, 4 and 6. There was a greater value of mean abundance of pellets in the first tidal cycle, the cycles 2 and 3 showed similar values. Given the importance of this finding, the variation of the mean abundance of plastic pellets per transect according to sampling zones and tide cycles was plotted in two different graphs in order to obtain a better visualization of the variation according to the both scales. The results of the SNK test revealed that the mean abundance in zone 1 was significantly greater than the values observed in the zones 3, 4 and 5. In addition, the a posteriori evaluation revealed that the zones 2, 3, 4 and 6 had similar mean abundances, but these values were significantly greater than the one found in the zone 5. The SNK applied to the factor "Cycles" indicated that the abundance values in Cycle 1 was significantly greater than the values of cycles 2 and 3. The cycles 2 and 3, on the other hand, showed similar values (Figures 13 and 14).

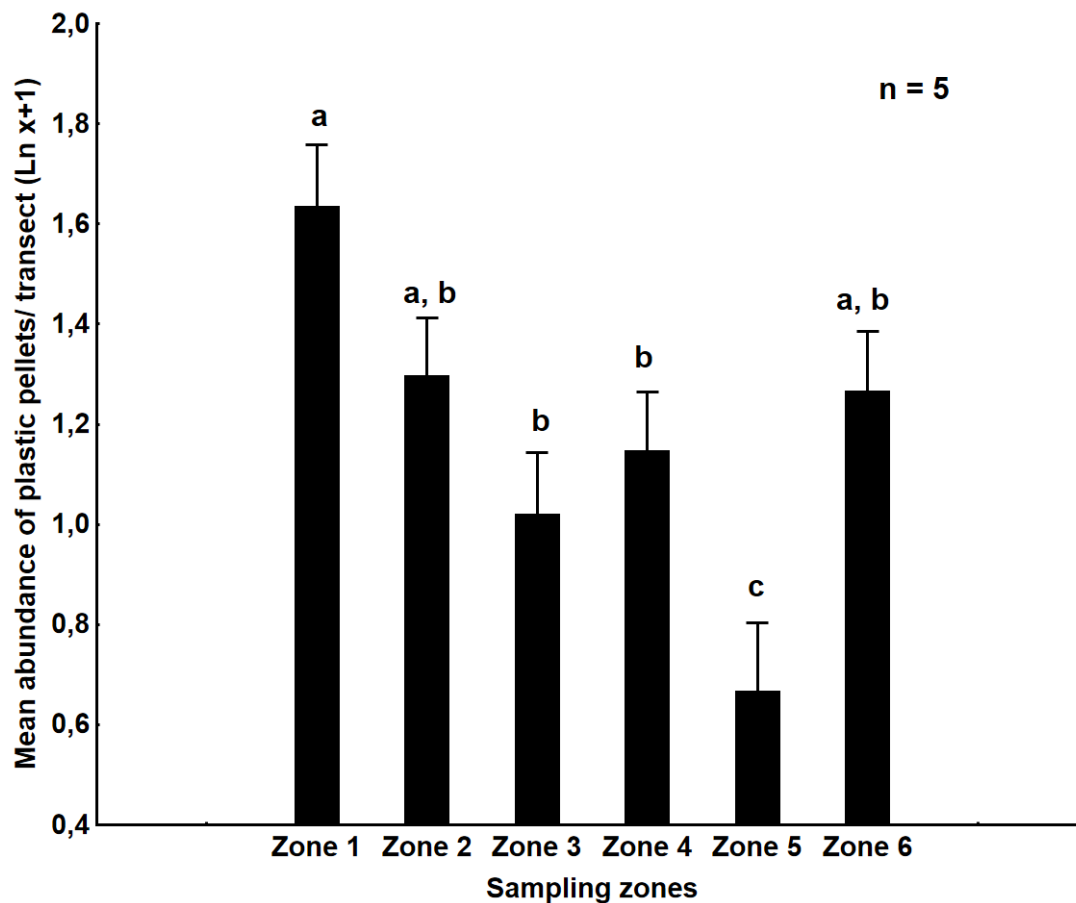


Figure 13. Abundance of plastic pellets according to the factor “Zones” in November of 2014. The bars represent the mean (\pm S.E) abundance of plastic pellets in each one of the sampling zones in Santos bay. The letters represent significant differences ($p < 0,05$) revealed by the *a posteriori* Newman-Keuls test (SNK).

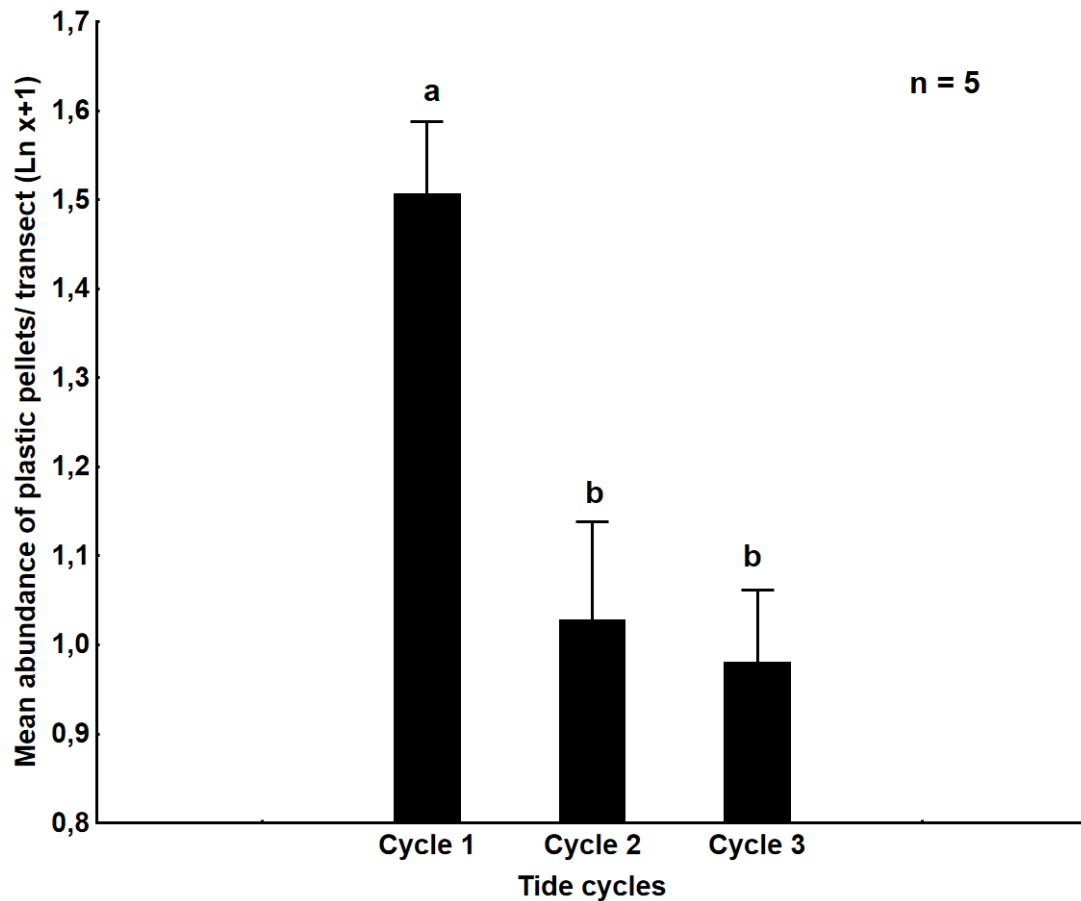


Figure 14. Abundance of plastic pellets according to the factor “Cycles” in November of 2014. The bars represent the mean (\pm S.E) abundance of plastic pellets in each one of the tide cycles in Santos bay. The letters represent significant differences ($p < 0,05$).

A similar process occurred in the samples from March. The zone one showed greater or similar mean values in comparison to the other areas. In addition, the abundance of plastic pellets showed a significant variation according to Zones and Cycles, with a significant interaction between the both factors (Table 2).

Table 2. Repeated measures analysis of variance (RM Anova) comparing the mean abundance of plastic pellets among tide cycles and among sampling zones in March of 2015. The factor “Zones” had 6 levels and the factor “Cycle” had 3 levels (n = 5) Cochran’s test evaluated the assumption of homogeneity of variances.

C = 0.28 Cycle 1 C = 0.40 Cycle 2 C = 0.68 Cycle 3 Ln (x+1)					
Effect	SS	DF	MS	F	p
Intercept	47878.67	1	47878.67	407.74	<0.001
Zones	2003.14	5	400.63	3.41	0.02
Error	2818.17	24	117.42		
Cycles	1838.81	2	919.41	22.85	<0.001
Cycles X Zones	1909.26	10	190.93	4.74	<0.001
Error	1930.94	48	40.23		

The greater values of mean abundance occurred in the zones 1, 2 and 3 and the lower values occurred in the zone 5. The results of the SNK conducted on the factor Zones revealed that there the mean abundance of the zone 1 is significantly greater than the value obtained in the zone 5. The SNK revealed that the mean abundance is similar among the other pairwise comparisons. The a posteriori evaluation of the factor Cycles revealed that the mean abundance in the cycle 1 is significantly greater than the values obtained in the cycles 2 and 3. In addition, this test indicated that the mean abundance of the cycle 2 is significantly greater than the one observed in the cycle 3 (Figures 15 and 16).

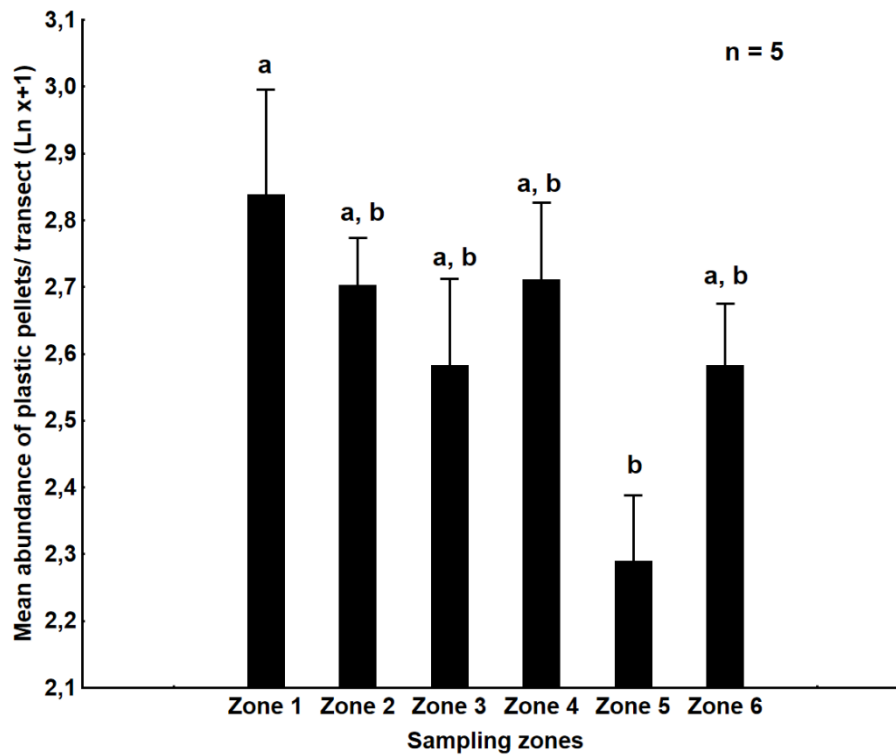


Figure 15. Abundance of plastic pellets according to the factor “Zones” March of 2015. The bars represent the mean (\pm S.E) abundance of plastic pellets in each one of the sampling zones in Santos bay. The letters represent significant differences ($p < 0,05$) revealed by the *a posteriori* Newman-Keuls test (SNK).

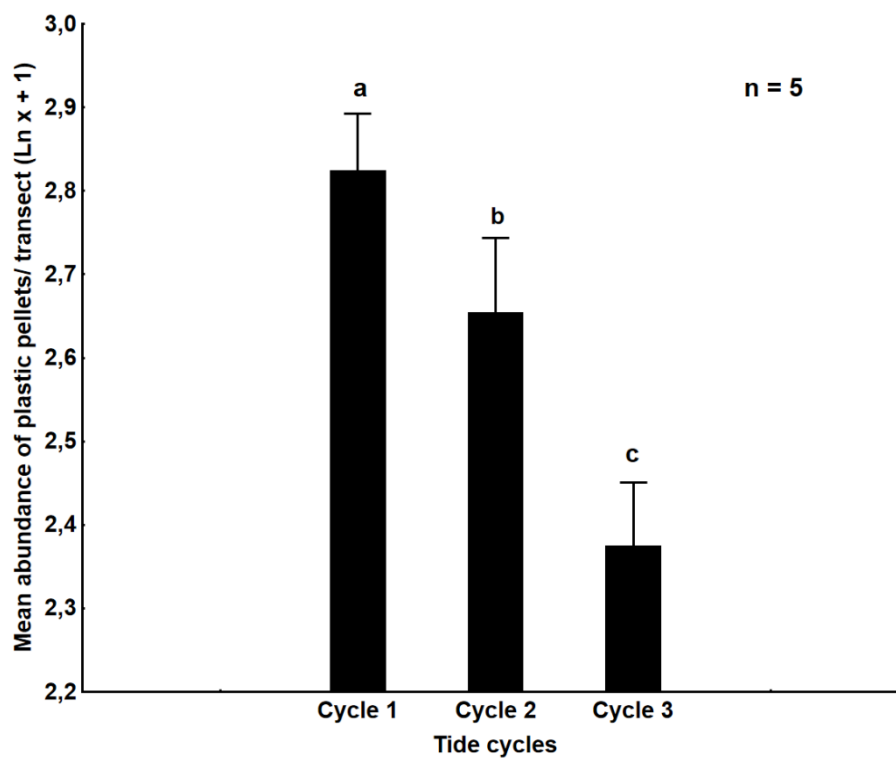


Figure 16. Abundance of plastic pellets according to the factor “Cycles” in March of 2015. The bars represent the mean (\pm S.E) abundance of plastic pellets in each one of the tide cycles in Santos bay. The letters represent significant differences ($p < 0,05$).

Standing-stock

The analysis of variance showed that the standing-stock was significantly different among the sampling zones (Table 3).

Table 3. Analysis of variance comparing the mean abundance of plastic pellets/ ST among tide sampling zones in Santos bay; data were extracted from TURRA et al (2014). In the present evaluation, the factor “Zones” had 9 levels (n = 3). Cochran’s test evaluated the assumption of homogeneity of variances

C = 0.35 Mean abundance of pellets/ ST
Ln (x+1)

Effect	SS	DF	MS	F	p
Intercept	14	1	14	703.01	<0.001
Zones	65	8	81	41.67	<0.001
Error	34	18	19		

In this dataset, the zones 4 and 7 showed the greater mean abundance of plastic pellets/ ST, followed by the zones 5 and 3. The zones 1, 2 and 9 showed the lower values. According to the results of the SNK test, the mean abundance of plastic pellets/ ST is similar between the zones 4 and 7. On the other hand, the values observed in these zones are significantly greater than the observed in the other levels of the factor “Zones”. The SNK also indicated that the mean abundance observed in the zone 5 is significantly greater than the other zones but 4, and 7. In addition, there was similarity among the zones 1, 2 and 9 and in the comparisons among the zones 1, 8, 3 and 6 (Figure 17).

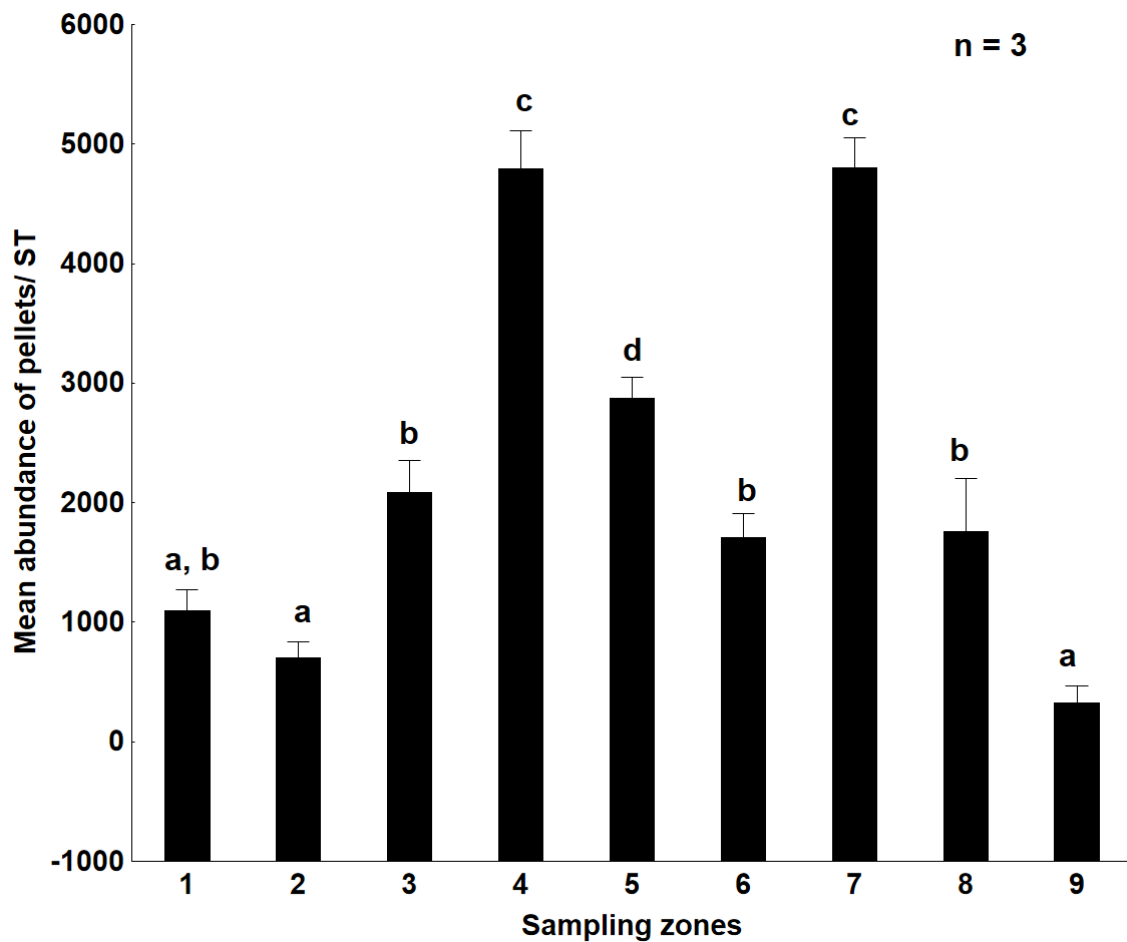


Figure 17. Mean abundance of plastic pellets/ ST according to the sampling zones in Santos bay; data were extracted from TURRA et al (2014). The bars represent the mean (\pm S.E) abundance of plastic pellets in each one of the sampling zones in Santos bay. The letters represent significant differences ($p < 0,05$) revealed by the *a posteriori* Newman-Keuls test (SNK).

Relationship between input and standing-stock

The linear regression analysis revealed that there is no relationship between input and accumulation in the studied extension of Santos bay (Table 4).

Table 4. Results of the linear regression analysis conducted on the relation between input and accumulation of plastic pellets in Santos bay. The input entered in the analysis as effect and the standing-stock as dependent variable. The analysis relied on interpolated values of the both variables for an extension of 4 km in Santos bay. Both datasets were standardized prior to the analysis.

Effect	SS	DF	MS	F	p
Intercept	0.00000	1	0.000000	0.000000	1.000000
Input	0.00000	1	0.001233	0.001202	0.972528
Error	38.99877	38	1.026283		

The regression graph evidenced the absence of relationship between the both parameters given that this figure does not show a clear pattern of distribution of standing-stock values in response to the input effect ($r^2 = 0,00$). In addition, the figure shows an expressive number of outliers in response to the lower values of input. (Figure 18).

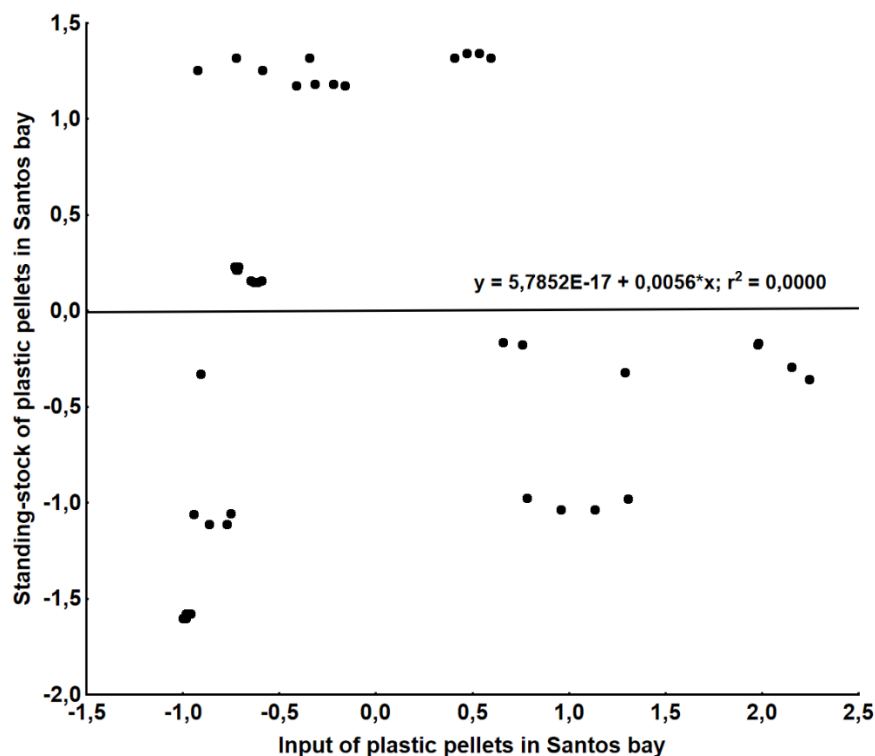


Figure 18. Linear regression graph representing the absence of relationship between input and standing-stock of plastic pellets in Santos bay. The conception of the figure relied on standardized values of the both variables interpolated in order to result estimations for each 100 m in an extension of 4 Km in Santos bay. Therefore, the figure brings the distribution of 40 points and the best fit among them, represented by the black line. The “y” equation predicts the standing-stock values according to the input.

Discussion

Despite the significant interaction between factors in both sampling periods, the hypothesis that the stranding of plastic pellets is different along the shoreline is true. The interaction between factors is more evident in March samples, however, mean values in area one were always greater or similar to the values estimated for the other areas. The area one is located further from the mouth of Santos - Guarujá channel and closest to the sewage emissary. However, the portion of the shoreline contemplated by the areas one, two and three is a site of sediments deposition (MAGINI et al., 2007). In addition, ocean dynamics in this portion of the shoreline is influenced by ebb tide currents coming from the mouth of Santos – Guarujá channel and is completely covered by the sea during flood tides (HARARI et al., 2002; MAGINI et al., 2007). It is probable that the spatial variation of plastic pellets in the shoreline is a response to the influence of these environmental forcing. In this process, the distribution of pellets along the shoreline is a response to the ebb tide currents and the promotion of areas of greater input occurs in response to the flood tides.

In addition, the dynamics of the waves might also constitute a reasonable explanation for the observed spatial differences. The relationship between the spatial distribution of stranded plastic waste in sandy beaches and the wave energy is a widely discussed issue (BROWNE et al., 2015; MCDERMID; MCMULLEN, 2004). The wave's dynamics would influence the stranding of plastic particles with a different intensity along the same sandy beach. Because of this process, the stranding of plastic pellets might be different in local spatial scales (Km). In the area of study, the points of greater hydrodynamic energy

coincide with the areas one, two and three, constituting another environmental process that influences the deposition of sediment (MAGINI et al., 2007). Therefore, the same processes that interfere on spatial differences in the deposition of sediment influence the differential stranding of plastic pellets. Indeed, the accumulation of litter in sandy beaches might be a consequence of waves energy and coastal geomorphology (BOWMAN; MANOR-SAMSONOV; GOLIK, 1998).

The results also revealed the variation in the stranding of plastic pellets according to tidal cycles, indicating that the input is also different in small spatial scales. Greater mean values in the first cycle might indicate that the intensity of the rate of input decreases in the course of time after a pulse release. After their release in the water column, plastic pellets are dispersed from the releasing point to adjacent beaches. The amount of pellets entering the beach environment would decrease according to the decrease in these particles abundance in the water column. In addition, the intensity of the stranding might be greater in regions located close to the releasing point. In Santos region, the stranding of plastic pellets in the beach is greater when the release of these microplastics occurs in the mouth of the estuarine system, given the intensity of ebb tide currents (GUTIÉRREZ et al., submitted). The results of SADRI & THOMPSON (2014) also support the idea that the abundance of microplastics outflowing from estuarine systems is greater during ebb tides. Thus, the plastic pellets would outflow from the estuary during ebb tides and enter the beach environment in flood tides; however, the rate of stranding in beaches decreases with the decreasing in the abundance of these particles in the water column. The present

results indicate that decreases in the rate of stranding might occur in relatively small spatial scales (hours).

The environmental forcing and the intensity of the stranding rate constitute the possible reasons for the observed spatial and temporal differences in the pattern of distribution of the stranded plastic pellets in Santos beaches. However, there was no relationship between stranding and standing-stock. The result of the linear regression indicated that the stranding is not involved in the creation of accumulation zones of plastic pellets in sandy beaches. On the other hand, recent results indicated that the sediment revolving promoted by high-energy events is the decisive factor for plastic pellets burial that results in standing stock (TURRA et al., 2014). Given that this is a chronic process and the stranding is punctual, it is possible that the both processes occur in different temporal scales. Further, the present results also indicate that standing-stock and stranding respond to different environmental forcing.

Thus, the present study has two important results, the spatial and temporal variation in the stranding and the absence of relationship between standing-stock and input. The knowledge on the areas that tend to receive greater amounts of plastic pellets in a given sandy beach point to target places for the monitoring of this contaminant. The indication of these portions in the same sandy beach might result in lowering of costs with field samplings and personnel, and even in a more adequate tradeoff between sampling effort and sample representativeness. In addition, this information indicates the areas more susceptible to the input of anthropogenic litter in coastal environments (BROWNE; JAMBECK; MALLOS, 2015; ROCHMAN et al., 2013b; THOMPSON et al., 2009c). The fact that the areas of greater stranding and greater standing-

stock are not related means that both processes might represent different contamination issues in sandy beaches. For example, PAHs associated to stranded pellets show a great variability in small spatial scales, with high toxicity levels (FISNER et al., 2013b). However, these compounds do not reach toxic levels when present in deep sediment layers and their composition varies from the surface to greater depths (FISNER et al., 2013a). Therefore, risk evaluation on plastic pellets pollution should differ between the rate of stranding and the accumulation of these granules in sandy beaches.

The present study points to new insights on the factors that promote the spatial variation of plastic pellets and, consequently, to the contamination of sandy beaches by microplastics and the harmful environmental issues associated to these pollutants. The spatial difference of stranding rate means that the harmful effects of stranded plastic particles varies in intensity and frequency along a given sandy beach. In addition, given that greater stranding might not represent greater standing-stock, the effects of plastic pellets' contamination are qualitatively different along sandy beaches. In this scenario, the risk evaluation and the monitoring of the ecological effects of plastic pollution should also be different in spatial scales along sandy beaches.

Chapter 4. Accumulation zones of plastic pellets in sandy beaches.

Abstract

Microplastics, such as pellets, are reported worldwide on sandy beaches and have possible direct and indirect impacts on the biota and physical characteristics of the habitats where they accumulate. Evaluations of their standing stock at different spatial scales generate data about the levels of contamination, providing crucial information for the identification of accumulation zones, specific beach habitats and communities that are more prone to the risks associated with their presence. Using a sampling strategy that incorporated across-shore transects from adjacent compartments (berms and backshores) and vertical profiles of the accumulated pellets down to 1 m depth below the sediment surface, the present study evaluated the standing stock of plastic pellets in 13 sandy beaches (São Paulo state, Brazil) and identified accumulation zones at regional (among beaches) and local (between compartments) scales. At regional scale, the pellet density tended to increase at beaches located at the central and southwestern parts of the state, the area located between ports and factories that produce and transport the largest amounts of pellets in the country. At the local scale, berms showed larger accumulations of pellets than the backshores. For both compartments, pellets tended to occur deeper in areas where standing stocks were larger; but most of the pellets were concentrated from the surface to 0.4 m below surface, suggesting that organisms inhabiting this part of the sediment column are more exposed to the risks associated with the presence of pellets. This information can be important for monitoring activities aiming to understand the spatial and temporal patterns of distribution of

microplastics, for studies dedicated to understanding their ecological impacts, and for future evaluations of the effectiveness of worldwide management efforts to reduce the input of pellets into the marine environment.

Key words: microplastics, standing stock, accumulation, spatial scale.

Introduction

Contamination of the coastal zone by plastics is a major environmental issue (ERIKSEN et al., 2015; VAN SEBILLE et al., 2015; ZALASIEWICZ et al., 2016). Plastics in the sea are of different sizes, which are subdivided into macroplastics and microplastics (ANDRADY, 2011; COLE et al., 2011; RYAN et al., 2009a). Microplastics (MPs) have shapes such as spherules, fibers or fragments and are broadly defined to be less than 5mm in diameter (ARTHUR; BAKER, 2009) although clear boundaries have not yet been set. Among these MPs are plastic resin pellets, which constitute the raw material for manufacturing plastic goods. Pellets enter the marine environment by losses during production, transport, transshipment at port terminals, and at plastic processing plants (TURNER; HOLMES, 2011).

In 2014 the world production of plastics materials was 311 million metric tonnes (PLASTICS EUROPE, 2015). In 2014 the world production of plastics materials was of 311 million tonnes (Plastics Europe, 2015). Despite the worldwide management strategies in place to reduce losses along the production chain, such as the 'Declaration of the Global Plastics Associations for Solutions on Marine Litter' (Global Plastics Association for Solutions on Marine Litter, 2011), pellets are still being lost to the environment. Losses occur directly to

marine/estuarine environments or to urban drainage systems and rivers, eventually reaching coastal areas. The lower the density of resin pellets, the greater is their dispersal capability (UNEP, 2014b). Pellets that enter the marine environment are transported by currents and wind, and aggregate in many habitats, such as on sedimentary shores worldwide (COLE et al., 2011). Pellets are hypothesized to accumulate near coastal urban and industrial centers and port areas (ANTUNES et al., 2013; KARAPANAGIOTI et al., 2011; TURNER; HOLMES, 2011), i.e. these materials can originate from local sources. They have been reported on sandy beaches (BROWNE et al., 2011; HIDALGO-RUZ; THIEL, 2013; MALLINSON; TAYLOR; SHEA, 2013; TURNER; HOLMES, 2011; TURRA et al., 2014), where they sometimes constitute more than 80% of the beached plastic items (CAUWENBERGHE et al., 2013). Therefore, sandy beaches can be considered as tractable and suitable model systems to evaluate the accumulation patterns of pellets.

Pellets are found lying on the surface and buried in beach sediments (KUSUI; NODA, 2003; TURRA et al., 2014) and in coastal dunes (DEKIFF et al., 2014; MCDERMID; MCMULLEN, 2004). Pellets are not uniformly distributed across the beach profile (HEO et al., 2013). In the across-shore direction pellets are limited to the surface of the sediment in the intertidal zone, and their concentration increases from the lower to the upper backshore, where they are present to a depth of 2 m below the sediment surface (TURRA et al., 2014). On the backshore, the abundance of pellets decreases from the surface to deeper sediment strata, and the available data suggest that despite gradient differences among beaches, less than half of the standing stock of pellets is accumulated between 0 and 0.2 m depth (TURRA et al., 2014). Thus, the intertidal zone acts

as a zone of transference of pellets from the sea to the backshore, and potentially to berms and dunes, where they may accumulate. Consequently, sediment samples taken from the intertidal zone would be more appropriate to evaluate the recent stranding of pellets to the beach system, while samples taken from different depths in the backshore and berms, would be meaningful to evaluate their accumulation and standing stock (MOREIRA et al., 2016a).

Most of the studies that have evaluated the abundance of pellets have focused on the intertidal zone of sandy beaches (HIDALGO-RUZ et al., 2012; MOREIRA et al., 2016a). Few studies have included the backshore (TURNER; HOLMES, 2011; TURRA et al., 2014) or coastal dunes (LIEBEZEIT; DUBAISH, 2012). The majority of the studies sampled at 0-10 cm below the sediment surface, occasionally down to 0.3 m (CARSON et al., 2011; CLAESSENS et al., 2011), and seldom down to 2 m in the sediment column (TURRA et al., 2014). There is very little information about the standing stock and accumulation of pellets across the beach zones.

Pellets accumulated on beaches can cause impacts from being ingested and from leaching of pollutants to the interstitial water, to the sediment and to organisms (FISNER et al., 2013b; MATO et al., 2001; NOBRE et al., 2015; OEHLMANN et al., 2009). Further plastic pellets might interfere on physical characteristics of the sediment column, such as permeability and heat transfer (CARSON et al., 2011). These factors may cause direct or indirect affects to the sandy beach environment and to the biodiversity. As the biota inhabiting sandy beaches occupies different layers below the sediment surface (CARCEDO; FIORI; BREMEC, 2015; RODIL; COMPTON; LASTRA, 2012), the accumulation of pellets could pose risks to different organisms, depending on the depth where

most of their standing stock is accumulated. Most of the standing stock accumulated down to 0.2 m could be especially harmful to organisms that utilize shallow layers of the sediment, such as the interstitial meiofauna and small-sized macrofauna. On the other hand, accumulation from 0.2 to 0.4 m depth could represent a greater threat to large-sized individuals of the macrofauna. Pellets accumulated deeper in the sediment could influence crustaceans and animals with temperature-dependent sex-determination such as sea turtles (CARSON et al., 2011; NELMS et al., 2015). Thus, evaluation of the distribution of the standing stock of pellets in the sediment and across-shore in sandy beaches could provide information on the specific habitats and communities that are most exposed to their impacts (chronic and/or acute). On the other hand, evaluations at larger spatial scales (i.e. among beaches) could provide information about pellet accumulation hotspots. This sort of information is important to understand where to expect higher chances of contact with the biota, and potentially greater risks, as well as for monitoring and management purposes.

The present study evaluated the presence of areas of accumulation of plastic pellets at different spatial-scales. The hypothesis that the standing stock of pellets would be unevenly distributed at local (between compartments: berm>backshore) and regional (among beaches; southwestern and central >northeastern beaches – see below) scales was tested (Hypothesis 1). There was also the intent to illustrate the importance of considering the accumulation within the sediment column (depth) to identify the accumulation zones and the beach communities that are most exposed to its impacts. For that, the hypothesis that for both compartments there would be no significant correlation between the standing stock of pellets and their depth distribution below the sediment surface

was tested (Hypothesis 2). Further, the present study also described the variation in the percentage of the standing stock of pellets accumulated at different depths below the sediment surface. To determine the most appropriated sampling method to identify areas of accumulation, the hypotheses were tested using two parameters of abundance (pellets \times m⁻² and pellets/Strip Transect).

The sampling strategy was specifically designed to test for differences in spatial scales, but this evaluation also provided additional information on the possible influences of the physical features of the environment and anthropogenic factors on patterns of accumulation.

Materials and Methods

Selection of the study sites

To evaluate the distribution patterns of pellets, a group of 13 beaches was selected along the entire coast of São Paulo state, in southeastern Brazil (Figure 1). Specific beaches were selected with regard to the presence of backshore and berms that were free of artificial structures, and not being mechanically cleaned by municipalities. Due to the pattern of land occupation in this state, many beaches along the São Paulo coast have infrastructure such as hotels, houses, and municipal parks built on the berms and coastal dunes, as well as mechanical cleaning by municipalities. For this reason, the sampling beaches were not equally distributed along the coast. Eight beaches were identified toward the northeastern part of the state and in the Santos region (beaches 1 to 8: Fazenda, Ubatumirim, Prumirim, Félix, Itamambuca, Vermelha do Sul, Lagoinha

and Capricórnio). Two beaches were located in the central part, closer to Santos (beaches 9 and 10: Boracéia and Itaguaré) and three located toward the southwestern coast (beaches 11 to 13: Una, Juréia and Ilha Comprida) (Figure 19).

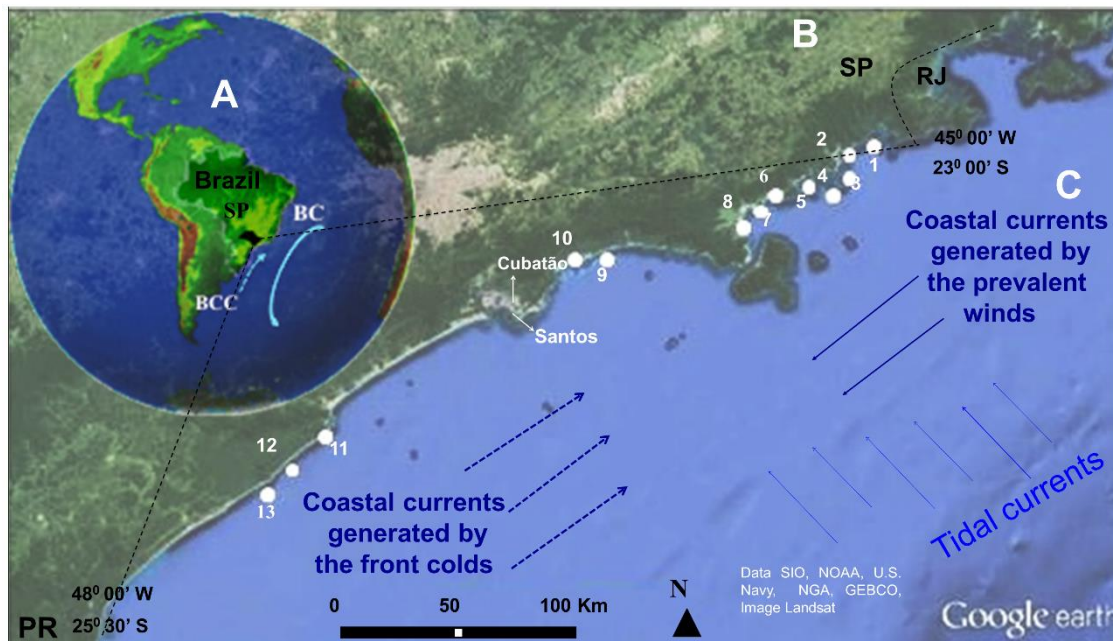


Figure 19. Study area. (A) The Brazilian coast and the location of São Paulo (SP – sampled area), Rio de Janeiro (RJ) and Paraná (PR) states and the Brazilian coastal current and the Brazil Current. Details of the São Paulo coast (B), showing the location of Santos and Cubatão cities, as well as the 13 sampled beaches along the state (1- Fazenda, 2 - Ubatimirim, 3 - Prumirim, 4 - Félix, 5 - Itamambuca, 6 - Vermelha do Sul, 7 – Lagoinha, 8 - Capricórnio, 9 – Boracéia, 10 – Itaguaré, 11 - Una, 12 - Juréia and 13 – Ilha Comprida). Major currents (C), arrows represent the direction of the currents generated by the tides (light blue), by the prevailing winds (dark blue) and by the winds under the influence of systems and wave fronts (dashed dark blue). The thickness of the arrows represents the strength of the currents.

Characterization of the area

São Paulo's coastal physiography differs between the southwestern and northeastern portions of the state. In the southwest the coastal mountain range is located tens of kilometers from the coastline, the coastal plain is wide, the beaches are long and open to the ocean, and islands are few. Toward Santos on the northeastern coast, the mountain range gradually approaches the coastline, the coastal plain and watersheds become smaller, the coastline is more jagged

forming coves and bays, and the beaches are smaller and more sheltered (SOUZA, 2012). The prevailing winds blow from northeast and under the influence of cold fronts that are more frequent during the winter blow from the south/southwest (PIANCA; MAZZINI; SIEGLE, 2010). Thus, the direction of the prevailing winds is either onshore or parallel to the coast.


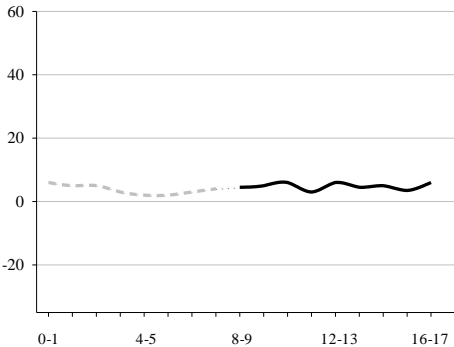

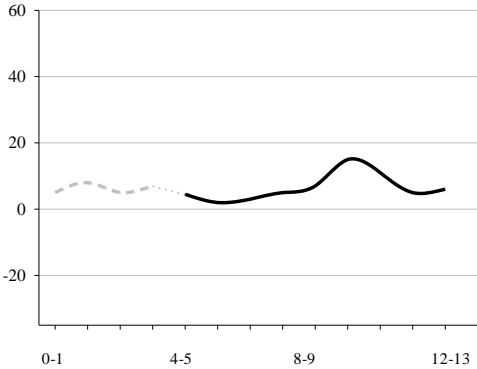
The offshore surface circulation on the Brazilian continental shelf is dominated by the Brazil Current (BC), which flows to the southwest direction (CAMPOS et al., 1995; MESQUITA, 1983; MESQUITA; LEITE, 1986) (Figure 19). In the São Paulo shelf region, both in winter and in summer, tides and wind forces dominate the coastal currents. Tidal currents are weak, with the major axis oriented against the shore. The currents generated by the prevailing winds are steady, somewhat stronger and parallel the coast generally to the southwest, toward Paraná state. Under the influence of cold fronts, currents intensify and turn northeastward, towards Rio de Janeiro state (CASTRO-FILHO, 1996; CASTRO-FILHO; MIRANDA; MIYAO, 1987; HARARI et al., 2006) (Figure 19). From spring to winter, the coastal currents off southern of São Paulo are also influenced by the intrusion of the Brazilian Coastal Current (BCC), which flows from lower latitudes of the South American continent and reaches the southwestern part of São Paulo state (DE SOUZA; ROBINSON, 2004) (Figure 19).

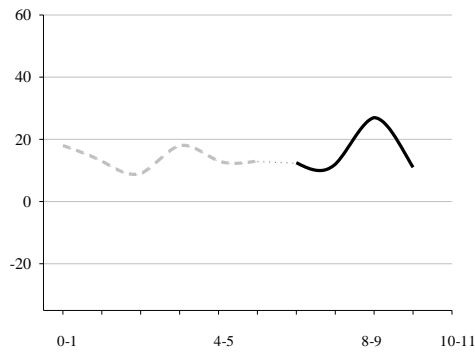
Characteristics of the beaches

The selected beaches were further classified as dissipative, intermediate or reflective based on the available literature on their morphodynamic

characteristics (SOUZA, 2012). This classification and observations on the physical characteristics of the sampling sites (i.e. profile of the sampling area, vegetation characteristics and across-shore pictures of the beaches) were combined in a table to allow further interpretation of the data generated and factors influencing the pellets distribution patterns (Table 5).

Table 5. Physical features of the sampled beaches.

Beach	Profile Y Axis: Slope profile (cm) X Axis: Across shore extension (m) --- Backshore (BS); Buffer; — Coastal dune (CD)	Direction; Morphodynamic (adapted from Souza, 2012); Profile description; Range of transect extensions (RE) & Range of points (RP) sampled per transect in each compartment
 <p>FAZENDA</p>		<p><i>NW / Dissipative</i></p> <p>Narrow herbaceous vegetation, followed by a dense mixture of scrubs and relatively tall woody vegetation. The slope was smooth and the ground almost flat.</p> <p>BS: RE = 6-9m, RP = 7-10 CD: RE = 2-5m, RP = 3-6</p>
 <p>UBATUMIRIM</p>		<p><i>NW / Intermediate</i></p> <p>Herbaceous vegetation covering 2-4m across the shore, followed by subshrubs and shrubs which forms a 'wall'. Presence of woody vegetation. The slope does not vary much across the sampled area, except for the vegetated area which had a sandy bank.</p> <p>BS: RE = 5.8-7m, RP = 6-8 CD: RE = 4.8-8m, RP = 5-9</p>

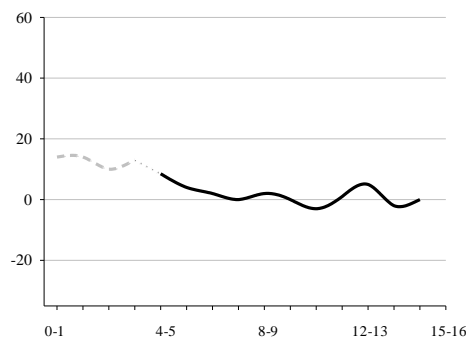


NE / Reflective

Very narrow and sparse herbaceous vegetation, followed by a dense cover of subshrubs and shrubs. The ground was steep and irregular (presence of elevations) and the berm had a high sandy bank.

BS: RE = 4-5m, RP = 5-6

CD: RE = 2-3m, RP = 3-4

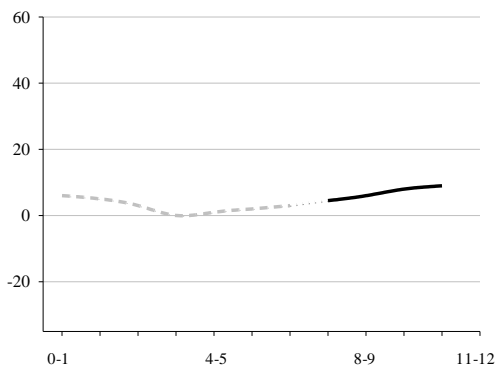


NE / Reflective

The berm had sparse herbaceous, subshrubs and woody vegetation. The beach had a steep positive slope, the backshore sits in a platform and the coastal dune had a negative and irregular ground.

BS: RE = 1-11m, RP = 2-12

CD: RE = 1-10m, RP = 2-11


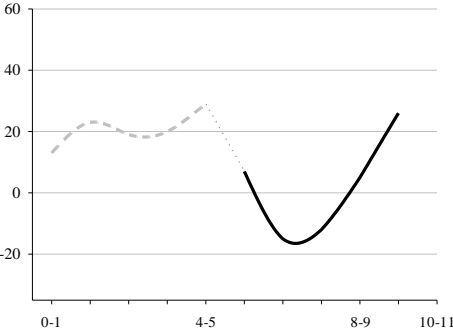

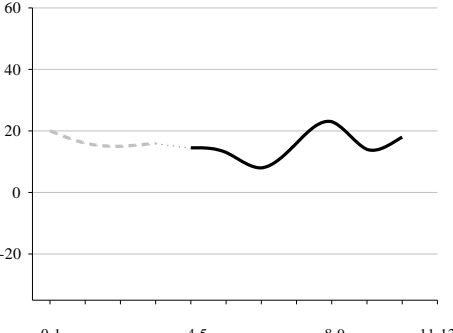

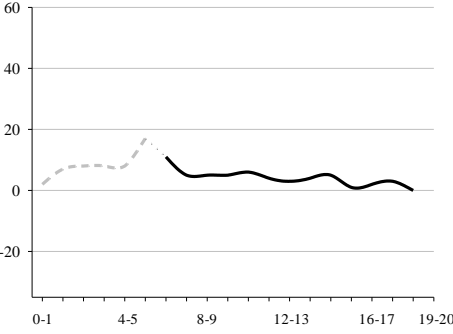



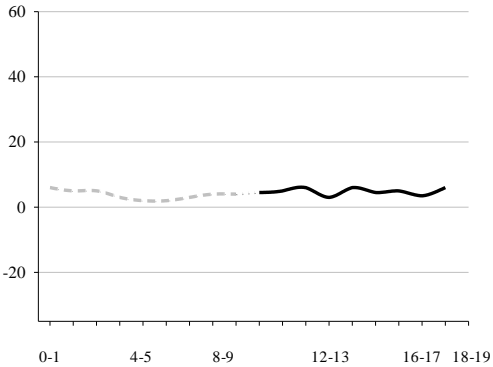

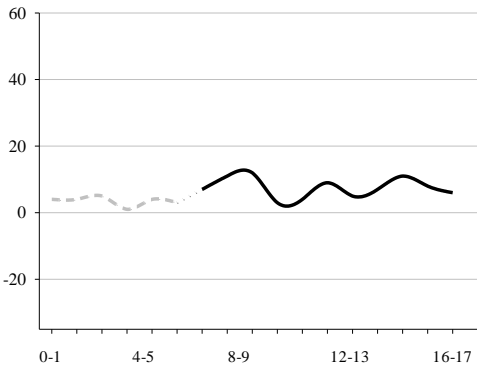

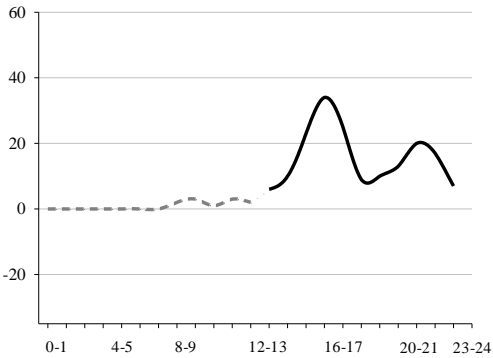
NE / Reflective


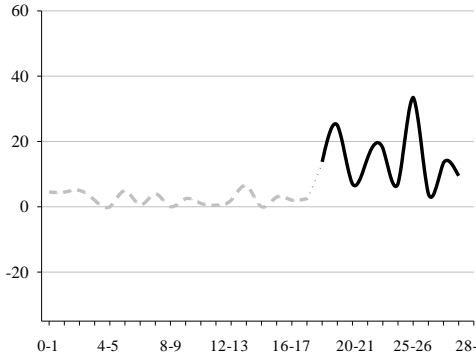

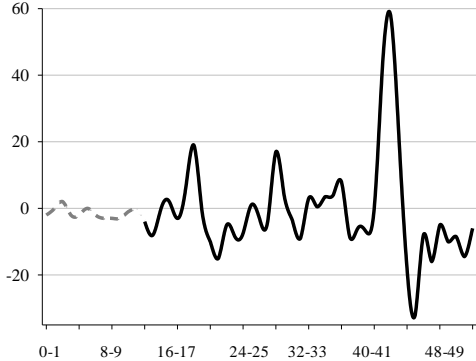
Dense herbaceous vegetation, followed by subshrubs and shrubs. The beach slope was relatively steep, but this increased slow and constantly towards the berm.

BS: RE = 4-10m, RP = 5-11

CD: RE = 3-5m, RP = 4-6

 <p>VERMELHA DO SUL</p>		<p><i>NE / Reflective</i></p> <p>Mostly composed of dense subshrubs and shrubs vegetation. Woody vegetation, although sparse distributed. The beach slope was steep, reaching a maximum elevation at the backshore and was followed by a steep declination, which forms a deep trench along the shore.</p> <p>BS: RE = 3-5m, RP = 4-6 CD: RE = 1-3m, RP = 2-4</p>
 <p>LAGOINHA</p>		<p><i>NE / Intermediate</i></p> <p>Extensive herbaceous vegetation and sparse covers of subshrubs and shrubs. The beach and backshore had an intermediate slope and slightly irregular ground, while the berm present several smooth declinations.</p> <p>BS: RE = 2-4m, RP = 3-5 CD: RE = 2-11m, RP = 3-12</p>
 <p>CAPICÓRNIO</p>		<p><i>NE / Reflective</i></p> <p>Extensive covers of herbaceous vegetation. The beach had a steep slope, reaching a maximum elevation at the backshore and the berm had smooth irregularities and slight negative inclination.</p> <p>BS: RE = 0-5m, RP = 1-6 CD: RE = 10-13.2m, RP = 11-12</p>

		<p><i>NE / Intermediate</i></p> <p>Herbaceous vegetation covering 4-6m across the shore, followed by dense subshrubs and shrubs. The sampled area had a smooth slope, a narrow backshore and extensive coastal dune with slight irregular ground.</p> <p>BS: RE = 6-9m, RP = 7-10</p> <p>CD: RE = 2-5m, RP = 3-6</p>
		<p><i>NE / Intermediate</i></p> <p>Low-density herbaceous vegetation covering 2-4m across the shore, followed by dense subshrubs and shrubs vegetation, which forms a 'wall'. Presence of woody vegetation (Martins et al 2008). The slope was smooth and reached maximum elevation at the beginning of the berm, which had slightly irregular ground.</p> <p>BS: RE = 5-6m, RP = 6-7</p> <p>CD: RE = 8-16.5, RP = 9-12</p>
		<p><i>NE / Intermediate</i></p> <p>Intermediate density of herbaceous vegetation covering extensive area across shore. Followed by subshrubs and shrubs that thickens in the landward direction. The backshore was narrow and slope was smooth. Abrupt slope increase and there were two main sandy banks at the berm.</p> <p>BS: RE = 10-16.5m, RP = 11-12</p> <p>CD: RE = 7-11m, RP = 8-12</p>

		<p><i>NE / Dissipative</i></p> <p>Intermediate density of herbaceous vegetation covering extensive area across shore. Sparse populations of subshrubs. The sampled area had a smooth slope. While the beach and backshore portions were almost flat, the berm had several irregularities sandy banks forming several trenches.</p> <p>BS: RE = 6m, RP = 7</p> <p>CD: RE = 9-15.6m, RP = 10-12</p>
		<p><i>NE/ Dissipative</i></p> <p>Intermediate density of herbaceous vegetation covering extensive area across shore. Sparse populations of subshrubs. The sampled area had smooth slope. While the beach and narrow backshore were almost flat, the berm had several sandy banks forming several steep sloped trenches.</p> <p>BS: RE = 13.2-19.3m, RP = 12</p> <p>CD: RE = 35.8-41.3m, RP = 12</p>

Reflective beaches were only present toward the eastern part of the state, while the dissipative beaches were located at the eastern and western ends of the state (see Fig. 19B and Table 5). Intermediate beaches were scattered along the coast. Reflective beaches had larger grain sizes than dissipative and intermediate beaches (Table 5). Usually, reflective sites had a narrow and

relatively steep profile, reaching greater elevations (i.e. sand stocks on the beach) toward the backshore or the beginning of the coastal dune. Dissipative sites had a wider surf zone, with a gentler profile and larger sand stock accumulated toward the berms, in sandy banks parallel to the water line, which formed berms in the landscape. Beaches characterized as intermediate had characteristics between these stages.

Only two of the beaches were oriented to the northwest (Fazenda and Ubatumirim), while the other beaches were oriented to the northeast. Some dissipative beaches had a narrow strip of herbaceous vegetation, followed by a dense mixture of scrub and woody vegetation (e.g. Fazenda); while others had moderately dense herbaceous vegetation covering an extensive across-shore area and sparse populations of subshrubs (e.g. Juréia and Ilha Comprida). Some reflective beaches had sparse herbaceous, subshrub and woody vegetation (e.g. Felix); others were mostly composed of dense subshrub and shrub vegetation (e.g. Vermelha do Sul) or had only herbaceous vegetation (e.g. Capricórnio). These widely varying vegetation characteristics did not seem to be related to the morphology of the beach or to retention of plastic pellets.

Possible sources of pellets

Plastics production in Brazil in 2012 totalled 6 million metric tonnes, representing The production of plastic in Brazil in 2012 was 6 million tonnes, representing about 2% of world production2% of world production (BRASKEM, 2015). The main losses of pellets in Brazil occur in port terminals, during road transport and at plastic factories (PEREIRA, 2014). No estimates of the amount

of pellets lost at ports and during transport are available. The amount of pellets passing through ports and the number of production factories were used as a proxy of possible sources of pellets to the sampled beaches. São Paulo state concentrates almost 50% of the primary (production) and secondary (transformation) plastic factories of the country (PEREIRA, 2014). The central part of the coast has two large production plants (Cubatão) and Santos, the largest port in Latin America, which ships the largest amount of pellets in the country. The amount of pellets shipped in ports in neighbouring states to the southwest is larger at ports toward the northeastern part of São Paulo (PEREIRA, 2014).

Santos was considered the major source of input, but pellets lost in other states, as well as those originating from oceanic areas, were expected also to influence the regional patterns of distribution. The surface currents would constantly move pellets lost in Santos and those from Rio de Janeiro, Espírito Santo and Bahia toward the southwest, while during cold fronts and winter, pellets lost in Paraná, Santa Catarina and Rio Grande do Sul would be pushed in the direction of São Paulo. Due to these characteristics of the sampling area, we expected that at the regional scale, larger standing stock of pellets would be found on beaches located close to Santos port and at beaches in the southwestern part of the São Paulo coast, than on beaches located toward the northeast.

Sampling methods

Different morphodynamic characteristics (i.e. dissipative, intermediate and reflective) were sometimes present in different sections of the same beach (i.e. Itamambuca, see Souza, 2012). Therefore, specific sections of the sampled beaches were characterized. On each beach, a 50-m stretch was selected based on visual recognition of sediment deposition sections of the beach (more dissipative characteristics). The preference for depositional sections was based on a previous observation that these areas are more susceptible to input and retention of plastic pellets (MCDERMID; MCMULLEN, 2004). Nevertheless, the presence of man-made obstructions precluded the selection of the more dissipative side of some beaches (Ubatumirim, Felix). The selected sections also had clear access to the sea (not blocked by islands) and were located as far as possible from the rocky shore, creeks or rivers, given the potential effect of these features on the input and accumulation patterns.

In the backshore zone, the across-shore width of the study area was defined by the distance between the high tide line and the start of the vegetation on the berms. The immediately adjacent berm compartment was sampled, but a 1-m buffer line between these compartments was established to ensure that the two different compartments were independently sampled. The across-shore width of the sampled area in the berms was limited by the presence of creeks or denser vegetation. To prevent field accidents and the removal of the berm vegetation, sampling was done where vegetation was sparse, so herbaceous species were occasionally removed, but not to the point where scrub or higher plants were dense enough to necessitate their removal for sampling. This

approach was also based on the rationale that the stems and roots of dense vegetation would form a barrier and impeded the movement of sand and pellets.

In each compartment, five transects were randomly allocated in the across-shore direction within the selected 50 m stretch. The number of equidistant sampling points for sediment collection on these transects varied according to the across-shore width. To evaluate differences in the accumulation of pellets between these compartments, the sampling design included transects covering the entire width of the sampled zones. Where a width of 12 m or more was available, the transect length was divided by 11, resulting in 12 sampling points spaced regularly at least 1 m from each other along the transect. For example, where the beach was 12 m or 24 m wide, 12 points were sampled with a distance between points of 1 m or 2 m, respectively. The length of these transects varied from 0 to 19.25 m and 1 to 41.25 m on the backshore and berms, respectively (Table 5). On these transects, sediment was collected at equidistant sampling points with a minimum distance of 1 m between consecutive points. The number of points varied according to the across-shore length of the transect (from 1 to 12 in the backshores and 2 to 12 in the coastal dunes). At these points, the sediment samples were taken using a manual auger (20.32 cm) to take a core of the sediment from 0 to 1 m depth, stratified every 0.20 m. At each 0.20 m, the sediment sample was placed in a bucket filled with seawater. The sediment was vigorously mixed by hand and the plastic pellets were collected by flotation, using a sieve (1-mm mesh size), and then counted manually.

At the middle and lateral ends of the 50-m stretches, sediment samples were also taken (10 cm depth) in the lower, middle and upper backshore zones, resulting in 9 samples per beach. In the laboratory, the mean size (measured in

phi) of the sand grains from each of the 9 samples was measured and an overall mean calculated for the area. The profile of the sampling area was measured in the middle of the sampled area of each beach, from the berms to the low-water mark, using a method based on the principle of communicating vessels. Sequential measurements of the differential elevation between two points 1 m apart were taken on two graduated rods connected by a hose filled with water (ANDRADE; FERREIRA, 2006).

Data analysis and interpretation

Across-shore pellet accumulation at regional and local spatial scales

To evaluate the across-shore accumulation of pellets at regional (among beaches) and local (between compartments within beaches) spatial scales, two different parameters of abundance were used. First, the accumulation was estimated for each transect in terms of numbers of pellets per square meter sampled using the formula:

$$Y = Ta / A * NS$$

Where 'Y' represents the mean density of plastic pellets x m⁻², 'Ta' represents the total density of plastic pellets on a given transect, 'A' the area of the manual auger ($\pi * 0.1016^2$) and 'NS' the number of sampling points within the transect, and thus A*NS represents the total sampled area.

The second parameter described the total number of plastic pellets per strip transects (ST) (pellets/ST) (DEFEO; RUEDA, 2002). The calculation of the

area sampled for each strip transect (ST) used the auger diameter (0.2032m) and the length (E) sampled in each transect, through the formula:

$$ST = E * 0.2032$$

The abundance of plastic pellets per ST (X) was then calculated for each transect, using the mean density of plastic pellets $\times m^{-2}$ (Y), the total area for the strip transect (ST) and the total sampled area (A*NS) in that transect, as:

$$X = Y * ST / A * NS$$

As the auger diameter was the same for all the samples, the analyses and interpretation of data related to the ST mainly considered the length of the strip transect.

Repeated-measures analysis of variance (Anova RM) was used to test the null hypotheses that there would be no significant differences in the distribution of pellets among beaches and between compartments (Hypothesis 1). The factors 'Beach', 12 levels representing the sampling stations where pellets were found (Fixed) and 'Compartment', 2 levels – backshore and berm (Fixed and Orthogonal, $n = 5$ transects in each compartment) were included in the analysis. Cochran's test for heterogeneity of variances was applied prior to the analyses. As variances were heteroscedastic for both compartments in the parameters (pellets $\times m^{-2}$ and pellets/ST), the $\ln(X+1)$ transformation was applied to data. Newman–Keuls tests were done for *a posteriori* comparisons within significant factors.

Patterns of accumulation in depth

To illustrate the importance of considering the accumulation to some depth in the sediment in order to identify accumulation zones, the analysis tested null hypothesis that for both compartments there would be no significant relationship between the distributions of pellets to some depth below the sediment surface and their standing stock (Hypothesis 2). Linear regression analyses were used to evaluate the relationship between the maximum depth where pellets were found and the mean standing stock of pellets, obtained from 0-1 m depth on the 5 transects for each beach compartment. Separate analyses were done for the two estimated parameters of abundance (pellets x m⁻² and pellets/ST). Data on the percentage distribution of number of pellets at every 0.2 m were then used to generate graphs of their depth distribution, in order to evaluate possible patterns in the depth distribution of pellets. The analysis calculated the total standing stock of pellets obtained from 0-1 m on the 5 transects in each compartment. Their concentration in depth was then obtained using their mean percentage density at each 0.2m interval, separately for each compartment. All statistical analyses were done using the Statistica 12.0 software.

Results

Across-shore pellet accumulation at regional and local spatial scales

Among the 13 beaches, pellets were not found in either compartment only at Ubatumirim (beach 2, Figure 20). Seven beaches (beaches 6 to 8 and 1 to 13) had pellets in both compartments, while at five beaches (beaches 1,3-5

and 9) pellets were present only in the berms.

Repeated-measures analyses of variance were done using data from the 12 beaches at which pellets were found. The results showed a significant interaction between beaches and compartments for the two parameters of abundance (pellets x m⁻² and pellets/ST) (Table 6).

Table 6. Repeated measurement analyses of variance comparing the mean abundance of pellets among beaches and between beach compartments. 'Beach' was fixed, with 13 levels. 'Compartment' was fixed and orthogonal to Beach, with 2 levels (backshore and coastal dune) ($n = 5$). Cochran's test was used to test assumption of homogeneity of variances.

Source	df	Pellets x m ⁻²			Pellets/ ST		
		MS	F	p	MS	F	p
Intercept	1	223.61	235.33	<0.001	537.56	252.68	<0.001
Beach Be	12	8.64	9.1	<0.001	19.65	9.24	<0.001
Error	52	0.95			2.13		
Compartment Co	1	111.42	103.64	<0.001	246.37	103.19	<0.001
Co X Be	12	3.53	3.29	0.001	8	3.35	0.001

This indicates that the accumulation of pellets was not consistent between compartments among the beaches. The Newman-Keuls *a posteriori* test indicated that, for both parameters, significant differences between compartments occurred at only in 6 of the 12 beaches (1 - Fazenda, 7 - Lagoinha, 9 - Boracéia, 10 - Itaguaré, 11- Una and 13 - Ilha Comprida, (Figure 20 A-B).

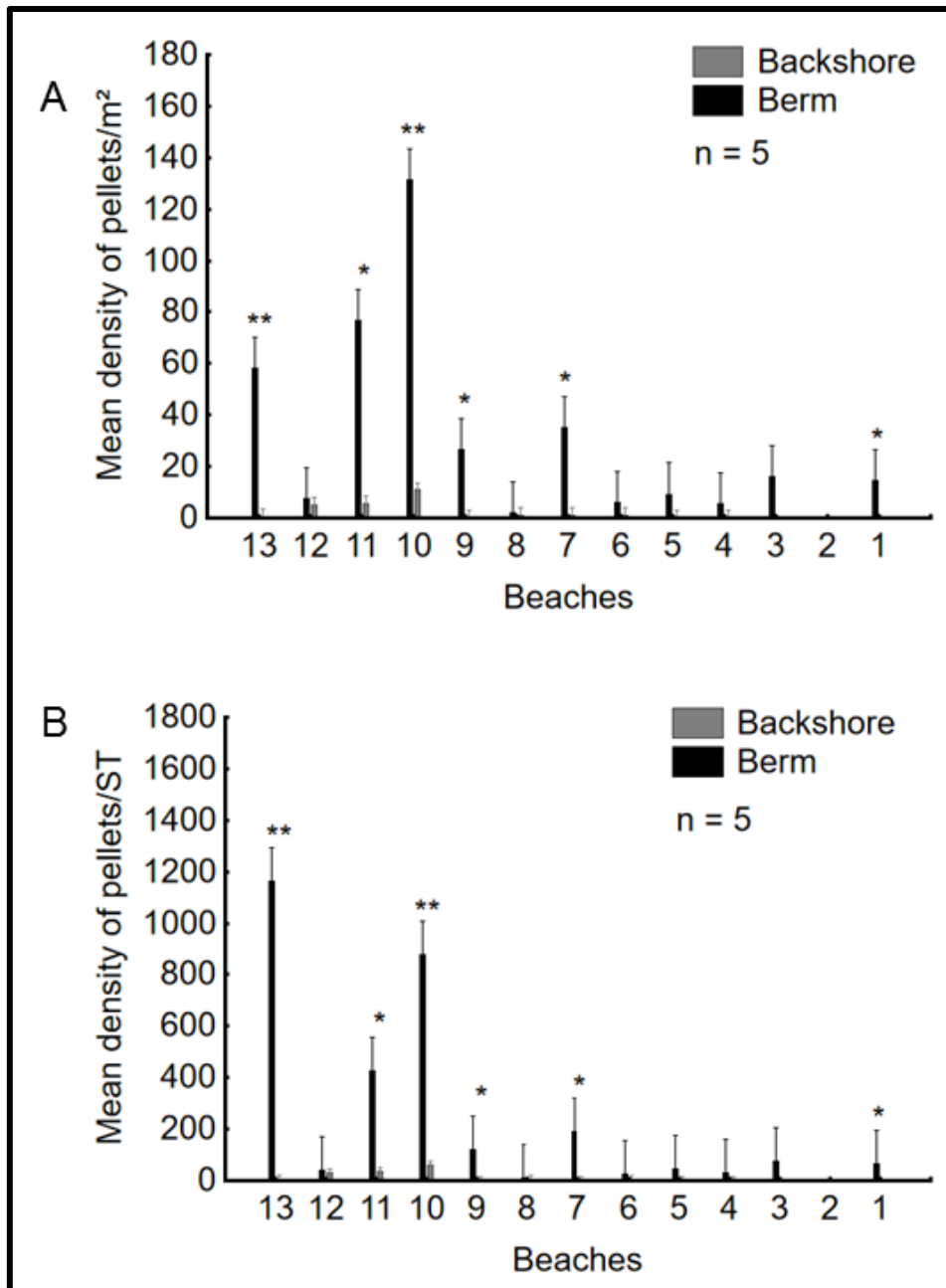


Figure 20. Densities of pellets. (A) Mean abundance of pellets $\times m^{-2}$ and, (B) Mean abundance of pellets/strip transect (ST). The grey bars indicate the mean (\pm S.E.) density of pellets at the coastal dunes and the white bars represent the mean (\pm S.E.) density of pellets at the backshore. Significant differences between compartments (as per Newman-Keuls *a posteriori* tests) are indicated by * ($p < 0.05$) and ** ($p \leq 0.001$).

At local spatial scales, these six beaches showed higher densities of pellets in the berms than in the backshore. No significant differences were detected for the remaining six beaches, which had similar densities between compartments (6 – Vermelha do Sul, 8 – Capricórnio, 12 – Una) or pellets present

only in the berms (3 – Prumirim, 4 – Félix and 5 – Itamambuca). On the other hand, the high variability within replicates prevented significant results. At the regional spatial scale, the highest densities of plastic pellets were found at Itaguapé, Una and Ilha Comprida (beaches 10, 11 and 13 respectively, Figure 20). The estimates of standing stock at the berms varied widely among these three beaches, depending on the parameter analyzed. Calculations for pellets \times m^{-2} showed that the density of pellets was higher at Itaguapé, while calculations for pellets/ST showed that the total abundance of pellets was highest at Ilha Comprida.

Factors influencing deposition and accumulation

The observations on the physical features of the environment (e.g. geometry of the coastline, morphodynamics and vegetation) suggested some factors that possibly influenced the patterns of accumulation. While the proximity to potential sources of input and the major coastal currents influence the distribution of pellets along the coast (WANG et al., 2016; BROWNE et al., 2011), other factors also contributed to their deposition and further accumulation in sandy beaches.

Due to the physiography of the São Paulo coastline, the geometry of the coast near the sampled beaches and the proximity to islands varied. Local patterns of water circulation are influenced by the presence of headlands and islands (BIRD, 2008) and, similar to the situation described by CLAESSENS et al. (2011), the geometry of the coastline may also have contributed to the observed sub-regional distribution of pellets. This could explain the relatively low

pellet density found at Juréia Beach, which is located within the regional area of accumulation (central and southwestern part of the state), but adjacent to a headland (Juréia-Itatins) that could divert the coastal current.

The morphodynamic characteristics of the beaches (SOUZA, 2012) could also play an important role. Small sediment particles and small lightweight plastics are both likely to be transported by currents, settle slowly in the water column, and be deposited in areas where the water movements are slower (BROWNE et al., 2010). Dissipative beaches would be more prone to the deposition of pellets, whereas reflective beaches would be less prone. In addition, the gentler slope of dissipative beaches could facilitate pellet transport from backshores to berms, contributing to a greater accumulation within this compartment. A previous study, using data obtained from point sampling at strandlines, indicated no clear relationship between the abundance of microplastics and the mean size-distribution of natural particulates (BROWNE et al., 2011). This result could, however, be influenced by artefacts of the sampling method that increase minor temporal and spatial variability in the data and can hide patterns (MOREIRA et al., 2016). We observed that at the regional spatial scale, the density of pellets was generally lower in reflective than dissipative or intermediate beaches; and at local scales, differences between compartments were significant for most of the intermediate and dissipative beaches and never for the reflective ones.

A range of physical, biological and anthropogenic factors can influence the burial and depth stratification of pellets in the sediment. TURRA et al. (2014) found that intermediate increases in the abundance of pellets at depth ranges of 0.2 – 0.4 and 0.6 – 0.8 m were related to extreme oceanographic events. The

present results indicate moderate increases in abundance at these depth ranges in some, but not all backshores and berms (Prumirim, Felix, Capricórnio, Itaguaré, Una and Juréia). This indicates that oceanographic events are important factors behind the mechanism of the burial of pellets in both compartments, but also that other factors contributed to the specific stratification, which could be related to the wind action, the local deposition rate of sand, and biological processes, such as burial by crabs (IRIBARNE et al., 2000), which should be further investigated.

Patterns of accumulation in depth

Due to the absence of pellets in some beach compartments (figure 2), Linear regression analyses between depth distribution of pellets and their standing stock were done using $n = 7$ and $n = 12$ respectively for the backshore and for the berm.

Analyses based on estimates per square metre (pellets \times m^{-2}) indicated a significant relationship between the depth distribution of pellets and their standing stock for the backshores ($F_{5,1} = 15.27$, $p = 0.01$, $R^2 = 0.75$) and coastal dunes ($F_{10,1} = 10.1$, $p = 0.01$, $R^2 = 0.50$). Analyses based on extension of the Strip Transect (pellets/ST) indicated significant relationship between those factors for the backshores ($F_{5,1} = 19.2$, $p = 0.01$, $R^2 = 0.79$) and similar patterns, but not significant differences for coastal dunes ($F_{10,1} = 4.7$, $p = 0.05$, $R^2 = 0.32$) (Figure 21).

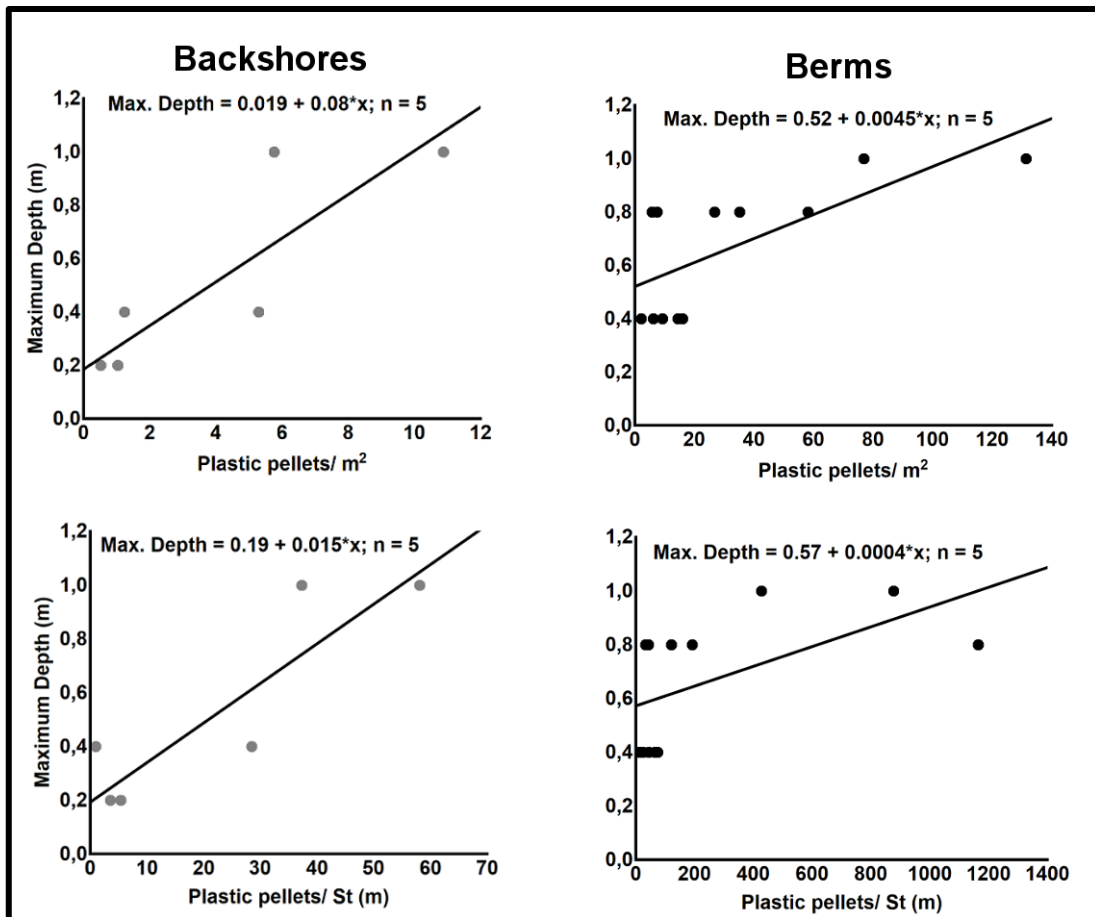


Figure 21. Relationship between distribution in depth with the sediment column and the abundance of pellets. Scatter plots of the Regression Analyses on the relationship between the depth distribution of pellets (m) and their density (pellets $\times \text{m}^{-2}$ and pellets/ST) at backshores (A and C) and coastal dunes (B and D).

The percentage of pellets present down to 0.2 m below the sediment surface varied greatly within compartments (backshore: 0 to 100%; berm: 14 to 67%). For both compartments, most of the standing stock (45 to 100%) was accumulated from the surface down to 0.4 m depth. Within that range, data indicated differences in the concentration of pellets related to the standing stock. In general, beach compartments where 80 to 100% of the pellets accumulated between the surface and 0.4 m depth had relatively low standing stock of pellets (i.e. less than 28 pellets/ST or 5.3 pellets $\times \text{m}^{-2}$ for backshores and less than 191 pellets/ST or 35.15 pellets $\times \text{m}^{-2}$ in the coastal dunes). On the other hand, beach

compartments where 45 to 62% of the standing stock was concentrated within that range had elevated standing stocks (i.e. more than 37 pellets/ST or 5.8 pellets \times m^{-2} in the backshores and more than 877 pellets/ST or 131.3 pellets \times m^{-2}) (Figure 22).

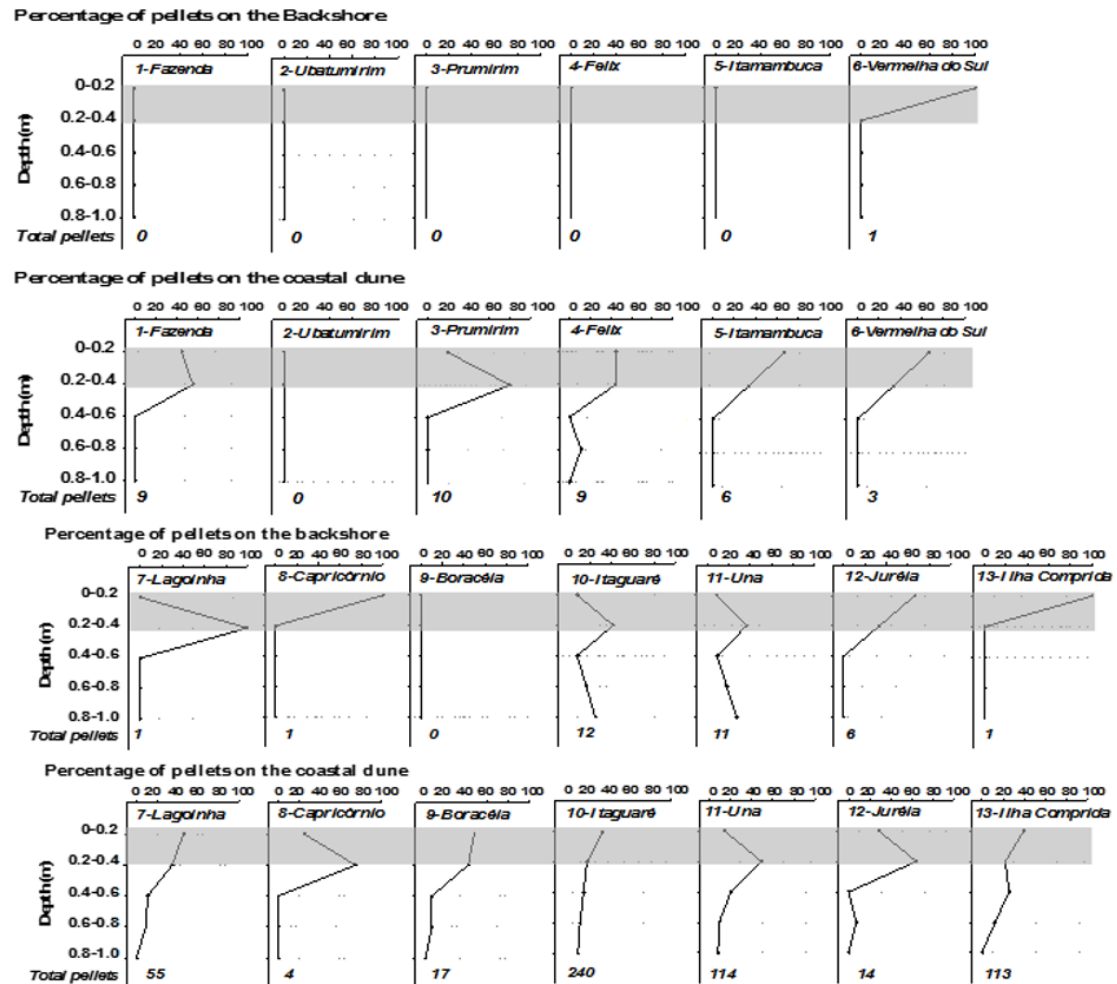


Figure 22. Percentage distribution in depth. Total number of pellets sampled and their percentage distribution down to 1m depth below the sediment surface (stratified at each 0.2m) in the sediment of the backshore (upper panels) and the coastal dunes (lower panels) of the 13 sampled beaches. The grey areas represent the depth range at which forty-five per cent or more of the pellets were concentrated.

Discussion

The main goal of this study was to evaluate the presence of accumulation zones of plastic pellets in sandy beaches at different spatial-scales. The results indicated the occurrence of local and regional accumulation zones of pellets. Further, the results indicated different patterns of accumulation below the sediment surface between areas that were more and less prone to accumulation. The discussion of the results contemplates the identification of specific beach habitats that are more prone to accumulate pellets, and the biological communities for which greater risks associated with their presence could be expected. The present findings have particular implications for studies aiming to understand the ecological impacts of the accumulation of pellets in sandy beaches, as well as for monitoring programs that are crucial to evaluate the efficiency of management strategies in reducing the input of pellets into the oceans.

The sampling strategy included the depth distribution of pellets to calculate their standing stock at the backshore and berms of sandy beaches. The present findings illustrate that, at local spatial scales, pellet accumulation tends to be greater in the berms than in the backshores. This indicates that the identification of accumulation zones necessarily involves the calculation of the standing stock of pellets from adjacent beach compartments, whenever these are connected. As per the initial prediction on the regional scale, there is a trend toward an increase in the density of pellets at beaches located in the central and southwest portions of São Paulo state. The results further indicate that sampling strategies using abundance estimates based on pellets/ST are important to calculate the total standing stocks, because this considers the across-shore width

of each habitat. Nevertheless, the across-shore width of the compartments can vary widely among beaches, especially for coastal dunes, resulting in heteroscedastic variances in the abundance of pellets within this compartment, which increases the probability of Type I error (UNDERWOOD, 1997). Thus, estimates based on pellets \times m⁻² are more meaningful to evaluate accumulation zones, because they use a fixed maximum number of sampling points within each transect and not its entire length.

Patterns at local and regional scales were, however, not consistent among all the beaches within the different portions of São Paulo of the São Paulo State, demonstrating that some areas are more prone to accumulation than others. This suggests that while the proximity to possible sources of input and the major coastal currents influence the distribution of pellets along the coast (BROWNE et al., 2011; WANG et al., 2016), other factors also contributed to their deposition and further accumulation in sandy beaches. The present study includes observations about factors that could have influenced the observed patterns at both spatial scales. However, the sampling strategy was strictly designed to test for differences on spatial scales and not to identify the factors influencing the differences. Thus, further studies designed to evaluate environmental factors influencing the accumulation among beaches and between compartments are still needed. These could benefit from the present observations and use analyses such as General Linear Modelling to determine the relative importance of the factors (BROWNE et al., 2015; RIBIC; SHEAVLY; KLAVITTER, 2012; RIBIC et al., 2010)

As described by TURRA et al. (2014), where pellets were present, they were generally distributed in the sediment column, with gradient differences

among backshores and berms. The results indicate that part of these gradient differences was explained by the standing stock of pellets, as pointed by the overall increase in the depth distribution of pellets in areas with higher standing stocks. Therefore, evaluation of the depth distribution provides further evidence for the identification of accumulation zones of pellets among beaches and between compartments. As the power of those analyses was reduced due to the low number of replicates that it was possible to include in the analytical model, this relationship should be further explored and estimates of density based on pellets \times m^{-2} used for future evaluations. Backshores and berms may undergo erosion or accretion (deposition of sand), due to a combination of geomorphological, oceanographic, meteorological, hydrological and anthropogenic processes (SOUZA, 2012). In areas where sand is accreting, the sediment deposition rates could be used to evaluate the concentration of microplastics within the sediment column over time (CLEASSENS et al., 2011). In this situation, a deeper vertical distribution of pellets in areas with high standing stock would indicate that pellets have accumulated in these areas for a long time (i.e. chronically).

Irrespective of the gradient differences among and within beaches, in areas with the highest standing stocks about half of the pellets were concentrated between 0 and 0.4 m below the sediment surface, while areas with relatively low standing stocks had 80% more of the pellets concentrated within this range. Thus, the proportion of plastics present at different depths also varied among areas that were more or less prone to accumulating pellets, and most of the pellets had accumulated between the surface and 0.4 m, not below 0.2 m depth as found by TURRA et al (2014). These differences in the depth range at where most pellets

had accumulated may be related to the fact that the backshores sampled by TURRA et al (2014) had daily beach cleaning activities (mechanical or raking), which could also influence the burial, re-exposure and depth stratification of pellets.

The present data suggest that within accumulation zones in the São Paulo region, higher risks of ecological impacts could be expected for the berms, when the two beach compartments are connected. However, the standing stock in backshores with seawalls studied by TURRA et al (2014) was up to two thousand or 20 times larger than the present estimates for backshores and berms, respectively. This illustrates that risks could also be expected for backshores of sandy beaches with these artificial structures, which are rapidly replacing coastal dunes worldwide due to increased urbanization of coastal areas (CHAPMAN, 2006). The relationship between the standing stock and the percentages of pellets at different depths demonstrates that, in areas prone to accumulating pellets, benthic communities living from the surface down to 1 m depth are exposed to pellets, but risks of impacts could be greater for those living between the surface and 0.4 m. In areas less prone to accumulating pellets, organisms living between the surface and 0.4 m depth are more exposed than those utilizing deeper layers of the sediment, but the risks of impacts could be lower due to the relatively smaller amounts of pellets accumulated.

The pellets sampled during the current study had a low density, which may be similar to other small plastic debris. Pellets and fragments of polyethylene and polypropylene are similarly distributed in the backshore zone (HEO et al., 2013), and these could be moved to the upper beach levels by wind action (BROWNE et al., 2010). Thus, areas that accumulate more pellets could

potentially also accumulate larger amounts of fragments and other microplastics, increasing the risks to the biota. Despite the evident accumulation of pellets in specific areas, it is difficult to speculate if communities from these accumulation zones are presently experiencing effects from their presence. The reason is that published studies on the pathways of exposure and concentration above which the accumulation of pellets or other MP could exert an impact were conducted in the laboratory (BROWNE et al., 2013; NOBRE et al., 2015). In addition, published studies evaluated the effects of pellets on the physical characteristics of the environment (CARSON et al., 2011), so it is not clear how well they reflect processes occurring in nature.

Finally, there is a lack of monitoring programs to evaluate the spatial or temporal trends of pellets on sandy beaches, which are crucial to evaluate the efficacy of the present management strategies to reduce their input to the oceans. However, an increasing number of individual studies are evaluating the abundance and spatial distribution of pellets and other microplastics on sandy beaches within countries and among continents (e.g. BROWNE et al., 2011; HIDALGO-RUZ; THIEL, 2013; MALLINSON; TAYLOR; SHEA, 2013). These studies used point sampling at the surface or relatively shallow sediment samples from the intertidal zone, where estimates are strongly influenced by beach dynamic processes, which can increase the variability in data sets and impeded the evaluation of patterns in space and time (MOREIRA et al., 2016). Presently available data represent snapshots of the presence of pellets or microplastics in beaches, and methods and sampling strategies currently used cannot provide information about spatial or temporal trends in accumulation on sandy beaches.

The present study demonstrated patterns of distribution of pellets and

accumulation hotspots among beaches, between compartments and in depth within the sediment, by calculating the density of pellets to a depth of 1 m below the sediment surface on backshores and berms of sandy beaches. The results suggest that the used sampling strategy could greatly benefit future monitoring programmes aiming to understand spatial patterns of distribution of pellets and perhaps other microplastics. For monitoring of temporal patterns, accumulation hotspots detected within areas of sand accretion could be used as sites to evaluate long-term trends, and provide information about the effectiveness of the worldwide efforts to reduce the input of pellets into the marine environment. It is important to suggest the research evaluating patterns of accumulation in other geographical areas, to understand the physical, anthropogenic and biological processes that contribute to the establishment of accumulation zones of pellets and microplastics, especially at local scales (across-shore and in depth); and also studies using manipulative field experiments to evaluate the ecological consequences of their accumulation.

Concluding remarks

The present study evaluated variations in plastic pellets abundance according to different spatial and temporal scales as a proxy in order to disclose determinant processes for the promotion of accumulation zones of microplastics. In addition, the set of information given by the present evaluation improves the knowledge on the environments more prone to receive these contaminants in coastal zones. Therefore, this research disclosed new information on local and regional differences of plastic pellets distribution and described possible interferences of the physical environment on the variability of plastic pellets abundance. The information raised in the current evaluation, as well as the interpretation of the results, might be useful for monitoring, mitigation and management strategies. In this approach, the present research brings methodological interpretations and describes key points to consider in future evaluations of contamination of coastal zones by plastic pellets.

It is important to highlight that the present study evidenced the occurrence of accumulation zones both in local and in regional scales. The abundance of plastic pellets varied within sampling sites and among them. This result indicates that monitoring programs should explore the distribution of plastic pellets prior to its conduction. This strategy would indicate the target areas to conduct the sampling design. On the other hand, it is probable that areas that accumulate greater abundances of plastic pellets occur in response to environmental variables. It is important to suggest that the evaluation of the relationship between plastic pellets distribution and environmental variables would indicate the factors responsible for differences in local and regional spatial scales.

The abundance of plastic pellets also varied in different temporal scales. According to the results, the input of plastic pellets in sandy beaches might vary among subsequent semidiurnal tidal cycles and monthly. The variability according to tidal cycles might be a result of the pulse input of plastic pellets in sandy beaches. A similar dynamics might interfere in the monthly differences. On the other hand, the variability both in small and large temporal scales might occur in response to the climatic variation. The behavior of the source is another important factor in the promotion of temporal variability. Recent papers advocated that the release of microplastics by the source occur in pulses during cold fronts (GUTIÉRREZ et al., accepted). This process would increase the temporal variability especially in beaches located close to the source. Monitoring programs should contemplate this information in the selection of sampling sites in order to decrease biases in long-term evaluations.

The present results also highlighted the importance of Santos region as source of plastic pellets in regional scales. As discussed along the present paper, the biggest port terminal of Latin America is located in Santos. In addition, this port commercialize the greatest amount of plastic pellets in Brazil. On the other hand, recent results indicated that Santos bay receives plastic pellets from sources located in the estuary (GUTIÉRREZ et al., accepted). The both information complement each other, such that it is possible to suggest that Santos region accumulates and exports plastic pellets to other areas. Therefore, this region is an important site for plastic pellets contamination in local and regional scales.

Concluding, the high variability of plastic pellets abundance both in small and large spatial and temporal scales was evident during the present research.

Moreover, it is possible to suggest that monitoring and management strategies should contemplate this variability in order to evaluate the contamination by plastic pellets in long-terms. Further, future evaluations should explore the interference of environmental variables in this variability in order to find the accumulation zones.

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