


Can the colors of beach-stranded plastic pellets in beaches provide additional information for the environmental monitoring? A case study around the Port of Santos, Brazil

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Abstract Plastic pellets are granules of different polymers used in the manufacture of various plastic products. Plastic pellets can reach the environment due to losses after their manufacturing process, especially the transportation. Loading in harbour areas and transport by ships are the main sources of plastic pellets to the ocean and coastal areas. After pellets reach the environment, they may adsorb and concentrate chemicals contaminants from different sources. Moreover, the weathering of plastic pellets may result in color changes, from white to brownish. This study aimed to analyse the color pattern of pellets collected on beaches of the central coast of São Paulo, Brazil, as an indicator of weathering and ageing processes. Plastic pellets were collected in four sampling surveys conducted between April 2012 and September 2015, and then separated in five color groups: white, yellowish, orange, brown, and pigmented. All sampled beaches had a consistent pattern of light-toned pellets (white and yellowish). This pattern was also found over time, suggesting a constant supply of plastic pellets to the beaches, coming from the harbour area. We also recommend the use of the color pattern of plastic pellets in citizen science monitoring programs.

Keywords Plastic pellets . Microplastics . Coastal Impact . Colors . Hydrodynamic modeling analysis . Marine Pollution

Introduction

Plastic has become an essential raw material for manufactured products, due to some of its properties, such as resistance, rigidity, and low weight (Hammer et al. 2012; Thompson et al. 2009). The global plastic production is about 400 million metric tons annually, and a consequence of the increasing plastic usage worldwide is the generalized contamination of environmental compartments by plastics of different compositions and sizes. Part of these plastics is improperly disposed of and may reach the coastal environments (Geyer et al. 2017).

Microplastics represent a relevant fraction of plastic litter and are potentially harmful to the ecosystems' health (Geyer et al. 2017). Microplastics can be divided in two groups by their origins: primary - factory

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microplastics in pellets shape, the raw material of plastic products, and secondary - fragments of larger plastics (Andrady 2011; Malankowska et al. 2020). Plastic pellets (or nibs) consist of little spheres of plastic resins, generally smaller than 5 millimeters, representing a dominant and frequent type of microplastic found in the environment. These pellets consist of raw material for practically all plastic products (Mato et al. 2001), and their production and commercialization in small pellets allow using bulk terminals, reducing the overall costs. According to Ogata et al. (2009), plastic granules can be unintentionally lost in the environment during manufacturing and transportation. Thus, pellets can be released directly into the marine environment or carried by runoff, streams, and rivers that eventually reach the ocean (Ogata et al. 2009; Karlsson et al. 2018). Plastic pellets have been found along beaches, and coastal environments worldwide; for example, high densities of plastic pellets were observed in Hawaii beaches (Mcdermid and McMullen 2004) and Antarctic sea surface waters (Lacerda et al. 2019; Jones-Willians et al. 2020), where there are no local sources of these materials.

Once introduced in the marine ecosystems, plastic pellets can interact negatively with the environment and biota. There is a vast literature reporting the ingestion of plastic pellets by marine animals (Moser and Lee 1992; Bjørndal et al. 1994; Tomás et al. 2002; Tourinho et al. 2010; Davison and Asch 2011; Naidoo et al. 2020; Baird and Hooker 2000; Fossi et al. 2012). Ingestion of plastics may cause severe damage to the digestive system, like obstruction of the intestinal canal and reduced appetite, in addition to chemical exposure (Endo et al. 2005; Milić 2001). Some seabirds have color-selective feeding preferences for more transparent colors (Sileo et al. 1990; Vlietstra and Parga 2002), raising a concern about the impacts of plastic pellets in the marine environment.

Another issue about plastic pellets is the adsorption and carrying of contaminants from historically polluted areas to remote sites (Endo et al. 2005; Mato et al. 2002; Taniguchi et al. 2016). Pellets are capable to adsorb hydrophobic substances, such as polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), polycyclic aromatic hydrocarbons (PAHs) (Endo et al. 2005; Fisner et al. 2013; Ogata et al. 2009; Teuten et al. 2009), pharmaceuticals, such as procainamide, doxycycline, triclosan (Islam et al. 2021; Nobre et al. 2020; Prata et al. 2018), flame retardants such as polybrominated diphenyl ethers (PBDEs) and perfluoroalkyl substances (PFAS) (Engler 2012; Hirai et al. 2011; Llorca et al. 2014; Taniguchi et al. 2016), acting as chemical carriers for all these chemicals (Vedolin et al. 2018). Contamination of plastic pellets by anthropogenic substances occurs mainly close to urbanized regions and ports (Wang et al. 2018). From such regions, chemically contaminated pellets can be transported to contamination-free areas by the ocean currents (Heskett et al. 2012; Taniguchi et al. 2016). This flux of chemical contaminants associated with pellets is potentially threatening to marine and coastal ecosystems, as the release of such chemicals may induce toxicity to the marine biota (Izar et al. 2019). Moreover, because plastic pellets are persistent residues, the chemicals adsorbed may remain for extended periods in the environment, even those banned due to their high toxicity (Wang et al. 2018).

Plastic pellets are usually degraded by multiple mechanisms, especially photodegradation and thermooxidative degradation, followed by biodegradation and hydrolysis (Andrady 2011). Besides, plastic pellets stranded on land degrade faster than plastic particles floating on the sea (Pegram and Andrady 1989). Virgin pellets are usually white or translucent (Hammer et al. 2012), while field-collected plastic pellets include yellowish, orange, brownish, and black colors (Endo et al. 2005). In addition to physical degradation, photodegradation caused by the sunlight can directly interfere with pellets' colors, changing the colors of plastic polymers, like polyethylene, into yellowish or brownish (Karapanagioti and Klontza 2007). Thus, pellets of different colors are found in the marine environment, and this factor (color) is commonly used for pellets general characterization (Karapanagioti and Klontza 2007; Pianowski 1997; Day et al. 1990).

Pellets' colors may have a relationship with the presence of contaminants adsorbed from the environment. White, or virgin pellets have a high concentration of industrial chemical additives that can immediately be released into the water as soon as they reach the aquatic environment (Hammer et al. 2012; Koch and Calafat 2009; Nobre et al. 2015). Yellowish and brownish pellets tend to remain for longer periods in the coastal environment (Endo et al. 2005) and may have higher concentrations of persistent organic pollutants (POPs) adsorbed from the surrounding environment (Endo et al. 2005; Gorman et al. 2019; Mato et al. 2001; Taniguchi et al. 2016; Teuten et al. 2009). Fotopoulou and Karapanagioti (2012) showed that the longer polystyrene (PS) pellets stayed in the marine environment, the more polar they became, interacting with more compounds and getting enriched with hydrophobic and polar pollutants. The authors concluded



that the more PS pellets stay in polluted marine environments, the more these materials become polluted. Consequently, they should be regarded as solid waste carrying pollutants.

The colors of plastic pellets may relate to the types of contaminants associated and their potential toxicity (Izar et al. 2019; Nobre et al. 2015; Silva et al. 2016), also indicating for how long the pellets are in the environment. Ogata et al. (2009) and Fotopoulou and Karapanagioti (2012) stated that pellets' colors, from translucent to yellowish and brownish, could indicate the time that they were being weathered after their release in the environment. If this was true, it could be hypothesized that the proportion of pellets of each color would directly reflect their proximity to the sources of plastic pellets. Higher proportion of translucent and white pellets would be expected close to the sources. In contrast, an increasing proportion of yellowish, orange and brownish pellets would be observed at increasing distances from the sources. Consequently, darker pellets are older and present higher levels of organic contaminants. They have a higher potential to release toxic chemicals to the water and sediment, or to transfer these chemicals to the aquatic biota, under certain conditions.

In programs aimed to monitor beached pellets, the color assessment could provide some additional information to analyze their potential impact on the coastal environments. Additionally, if the processes involving the pellets inputs, transport, distribution, and deposition are known, the management of this environmental problem can be improved. In this sense, meteorological events are essential factors that affect the dispersion of pellets. Rainfall, wind speed, wind direction, and sea surface currents can directly influence this particle dispersion (Heo et al. 2013; Kim et al. 2015; Kukulka et al. 2012), and there are forms to predict the behavior of these factors by modeling analysis.

Recently, many researchers have used hydrodynamic modeling analysis to study the behavior and dispersion of microplastics along with coastal environments (Alosairi et al. 2020; Chubarenko et al. 2016; Gorman et al. 2020; Iwasaki et al. 2017; Maximenko et al. 2012; Morét-Ferguson et al. 2010). Hydrodynamic modeling analysis is a powerful tool that can help to better understand local and regional sources, dispersion routes (Gorman et al. 2019) of plastic pellets, and accumulation zones (Iwasaki et al. 2017). It is essential to consider all properties of the particle (i.e., density, shape, and size) that influence the dispersion, buoyancy, and residence time of plastic pellets across the coastal environments (Chubarenko et al. 2016). Cylindrical and rounded microplastics, such as plastic pellets, tend to disperse quickly, rolling over the water surface, under the influence of winds and surface ocean currents. Physical degradation and biofouling also influence the behavior and structure of pellets, increasing their surface area and density, consequently favoring their sinking (Chubarenko et al. 2016; Morét-Ferguson et al. 2010).

This study aims to analyze the variation of the colors of plastic pellets beached along the central coast of São Paulo and verify the existence of color gradients determined by their distance from the sources, considering the hydrodynamic processes related to their input and transport. Pellets were collected on beaches of the São Paulo coast (SW Brazil) and characterized according to the (1) sampling date, (2) area where the pellets were collected, and (3) location of the beach along the coastline since these factors could influence on the distribution pattern of plastic pellets. The hypothesis is that beach-stranded pellets found close to the sources would be predominantly clearer (white or translucent), and those plastic pellets found far from the sources would have a higher contribution of yellowish, orange, and brownish pellets' colors. A dispersion particle model was used to explain the pattern of plastic pellets distribution and verify the influence of weather and oceanographic variables on this pattern.

Materials and methods

Plastic pellets were sampled on 17 beaches (Sonho, Itaquitanduva, Gonzaguinha, Góes, Guaiúba, Tombo, Éden, São Pedro, Conchas, Prainha Branca, Riviera, Itaguaré, Engenho, Paúba, Pitangueiras, Cigarras, and Tenório), covering about 200 km of São Paulo coast, Brazil, at four different times (April 2012, March 2015, June 2015 and September 2015), in two distinct projects (Taniguchi et al. 2016 and Izar et al. 2019) (Fig. 1). Sampling of April 2012, June and September 2015 were focused on beaches located on the central coast of the state (i.e., Baixada Santista Metropolitan Region): Itaquitanduva, Gonzaguinha, Góes, Guaiúba, Tombo, Éden, São Pedro, Prainha Branca, Riviera and Itaguaré beaches. This region comprises two main sources of plastic pellets to the coast of São Paulo (Izar et al. 2019; Turra et al. 2014), the Port of Santos, the largest port of Latin America, and the major industrial complex of Cubatão.



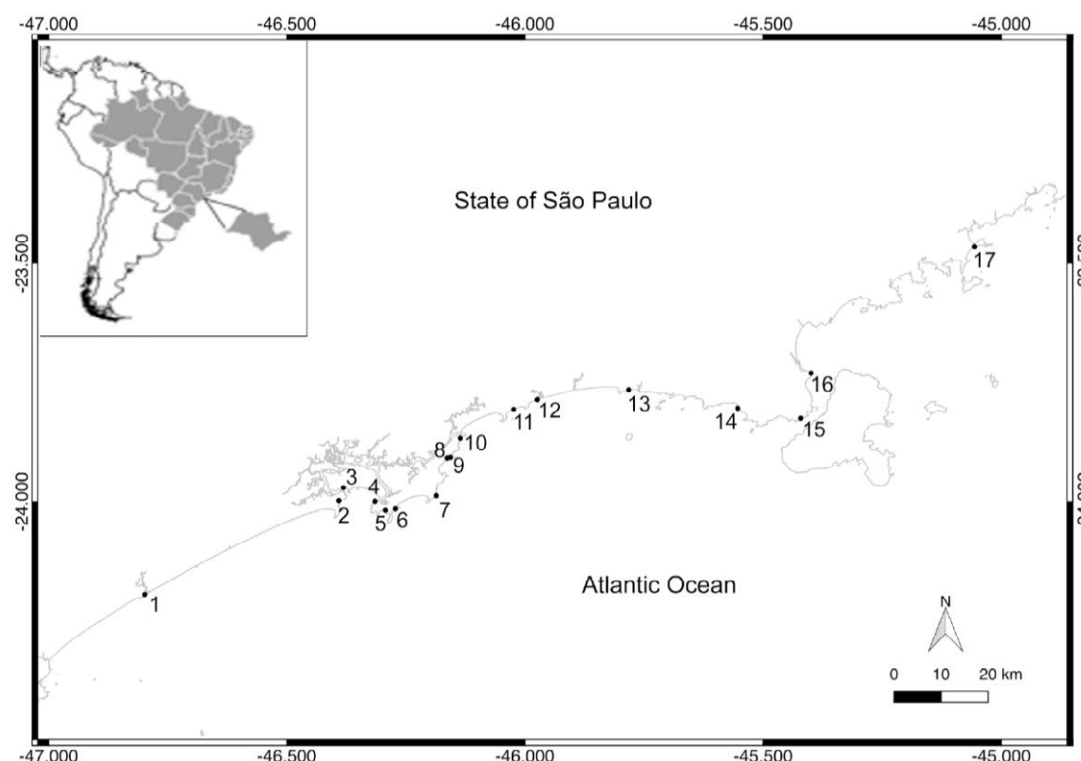


Fig. 1 Map of São Paulo coast showing the 17 beaches sampled between April 2012 and September 2015. Numbers correspond to each beach sampled: 1 - Sonho, 2 - Itaquitanduva, 3 - Gonzaguinha, 4 - Góes, 5 - Guaiúba, 6- Tombo, 7 - Éden, 8 - São Pedro, 9 - Conchas, 10 - Prainha Branca, 11 - Riviera, 12 - Itaguaré, 13 - Engenho, 14 - Paúba, 15 - Pitangueiras, 16 - Cigarras, and 17 - Tenório.

At each beach, we collected pellets using two methods: (1) active search - manual active collection of plastic pellets, finding pellets in the sand by naked eye while walk through the high tide line along the beach, and (2) square flotation - manual collection in a delimited area (square with 0.5 m side) using a bucket with sea water for separating plastic pellets by density difference (Manzano 2009). Both methods were sampled on the high tide line (Table 1), collecting only plastic pellets that have been visually separated from other types of microplastics. Plastic pellets sampled were stored in glass containers and sent to laboratory for color analysis.

Pellets collected in each beach were immediately counted after sampling, and their colors were defined visually between the tones found. Colors were classified by groups: 1) Clear - white, grey and translucent, 2) Yellowish - light and dark yellow, 3) Orange - light and dark orange, 4) Brown - light and dark brown, or 5) Pigmented - that include all colors that are chemically added to pellets (black, pink, blue, green, purple and red). For each sample, the percentage of the different colors was calculated, and for further analysis, plastic pellets' colors were defined as dependent variables.

Data of the March 2015 survey were accounted for the total amount and to describe the pellets' color pattern of each beach sampled by percentage, considering beaches with more than 100 pellets collected. Data from our other surveys followed this beach selection (>100 pellets) to describe the pellets' color pattern of each beach sampled. We excluded data of the March 2015 survey from the multivariate analysis because of the low number of pellets sampled on some beaches.

Data from surveys conducted in June 2015 and September 2015 were analyzed to test the influence of the sampling date and beach location. These data were chosen because both surveys used the same sampling design and method, with the first (June 2015) being conducted in the early winter and the last (September 2015) at the end of the winter. The four beaches sampled were divided into two groups: Inside (Itaquitanduva and Góes beaches) and Outside (Riviera and Itaguaré beaches) of Santos bay to compare the percentages of pellets of each color according to the geographical location. Percentages of the five color groups were compared by using Permutational Analysis of Variance (PERMANOVA: Euclidian) with two factors: Date (Factor 1, random, orthogonal, two levels) and Location (Factor 2, fixed, orthogonal, two levels). The confidence level in this and other statistical analysis was $P \leq 0.05$.



Table 1 Summary of samples made between April 2012 and September 2015, methods used for each sample, number of beaches sampled and total of pellets collected. The last two columns bring information about the studies used to obtain data and the factors tested in the statistical analysis.

Survey	Method	Beaches sampled	Total of pellets collected	Sample paper data	Factors tested
April 2012	Square Flotation	9	4277	Taniguchi et al. 2016	Beach Location and Beach Area
March 2015	Square Flotation	9	2857	Izar et al. 2019 (pilot)	-
June 2015	Active search	4	6104	Izar et al. 2019	Sampling Date and Beach Location
September 2015	Active search	4	5371	Izar et al. 2019	Sampling Date and Beach Location

To statistically test the difference between beaches sampled and beach area where the pellets were collected, we used the April 2012 sampling survey data. In this survey, pellets were sampled in three different beach areas (center, left, and right). In a micro-scale view, this beach area analysis aimed to identify the homogeneity of the pellets' color patterns between beaches and each beach area within the same beach. This analysis aimed to answer if the proportions of pellets of different colors are similar in different beach areas due to oceanographic processes (input, removal, deposition), or if local factors could influence this pattern micro-scale, generating differences of the pellets' colors percentages. Five beaches were chosen, considering the number of pellets >200 collected (Itaquitanduva, Gonzaguinha, Góes, Guaiúba, and São Pedro). In each beach area, pellets were collected, considering three replicates. Thus, factors Beach and Beach Area were compared for the five color group data using a two-way PERMANOVA (Euclidian): Beach (Factor 1, random, orthogonal, five levels) and Area (Factor 2, fixed, orthogonal, three levels).

For both statistical tests (PERMANOVA), Non-metric multidimensional scaling (nMDS) was used to identify the groups formed based on sampling date, beach location, and beach area. A similarity percentage test (SIMPER) was used to determine the percentage contribution of each dependent variable (single color) to each color group in all analysis.

In order to investigate the dispersion of pellets along the coastal zone, we applied the General NOAA oil modeling environment (GNOME) model for oil spill (Beegle-Krause 2001) to simulate the behavior and fate of particles released at two clusters of pellets source in the study area: 1) Santos Bay, and 2) Bertioga Channel, a connection between the inner estuary with the easterly portion of the coastal domain.

The GNOME model is not a numerical hydrodynamic model but instead, it takes a Lagrangian approach to track general particles as plastic pellets (and oil) displacements within continuous flow fields. It needs a proper definition of local currents, from which random walk approach and dispersion definition are combined in a final solution of trajectories.

Plastic pellets were treated as Lagrangian elements where all trajectories collectively represent the fate and transport of particles on the surface. The model is based on the advection-diffusion equation, implementing a forward in time Eulerian scheme for advection and a random walk scheme for diffusion. The Lagrangian elements assume the initial position for each i -particle as $\pi_i(x, y, t)$ within the model evaluated from a provided external flow velocity field. The new i -particle position in time will be $\pi_i(x+\delta x, y+\delta y, t+\delta t)$ where $\delta x, \delta y$ are the new position defined from the flow field and δt is the time step (Zhang et al 2021). The diffusion scheme is based on a stochastic random walk algorithm with a displacement probability such that the mean value is zero, but the variance in position grows linearly with time (Pearson 1905).

The background circulation data was derived from a global dataset run of the hybrid coordinate ocean model (HYCOM) for ocean modeling predictions (Chassignet et al. 2007), from which only the most favorable conditions were retained, to establish a general pattern of plastic pellets distribution in the region. The dataset is freely available from the 1/12 deg HYCOM+NCODA Ocean Reanalysis, which output is publicly available from the U.S. Navy and the Modeling and Simulation Coordination Office at <http://hycom.com>. Although the flow resolution from HYCOM (9.25 X 9.25 km) is relatively coarser than the particle model (0.1 X 0.1 km) it suffices for the pellets fate, which is solved at 30 minutes time steps. The model is based on the Navier-Stokes equations for the ocean, where the momentum equation is represented in isopycnal coordinates,



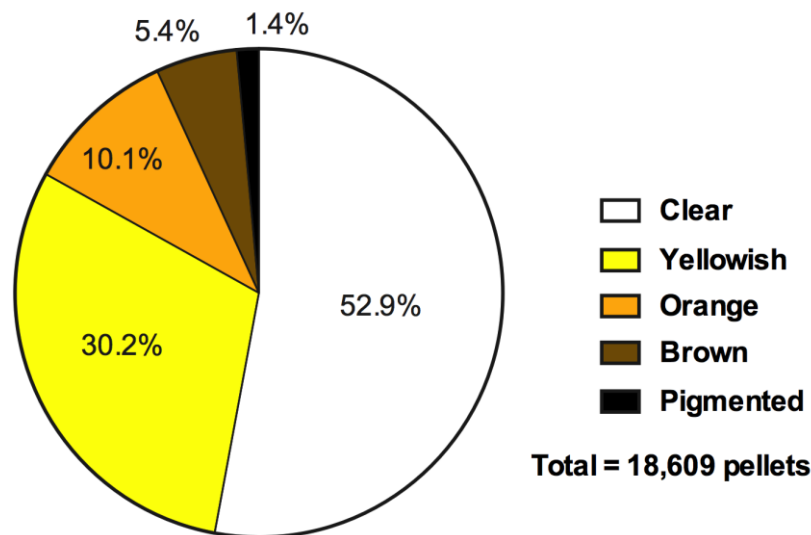


Fig. 2 Classification by colors of the total pellets collected in four samples between April 2012 and September 2015

$$\frac{\partial u}{\partial t} + \nabla \left(\frac{u^2}{2} \right) + (\zeta + f)k \times u = -\nabla M - g \frac{\partial \tau}{\partial p}$$

where ζ the relative velocity, k is vertical unit vector, M is the Montgomery potential, and τ the sum of the external stress and the Reynolds stress.

The use of oceanographic modelling had the purpose of providing some information on the dispersion of plastic pellets from their potential sources, and to indicate the transport direction along the coast based on prevailing conditions. The first cluster of particles was released at Santos Bay, simulating direct pellets' losses from the Port of Santos, considering ebb tides in the Santos Channel, dominated by tidal currents (Fernandino et al. 2016) which are embedded into the HYCOM model. The second cluster was released into the Bertioga Channel, simulating pellets derived inside the estuary due to port terminals' presence. Each cluster consisted of 1000 particles, all of them released simultaneously in both clusters. Then, the model predicted the particles' transport for 15 days, considering the prevailing almost steadily state oceanographic and meteorological conditions.

Results

In this study, a total of 18,609 pellets were collected in the 4 sampling surveys conducted between April 2012 and September 2015, and then they were sorted by colors. Clear tones were the most abundant (52.9%), followed by yellowish pellets (30.2%), orange (10.1%), brown (5.4%), and pigmented colors (1.4%) (Fig. 2).

In April 2012, clearer pellets were predominant in beaches analyzed beaches (Itaquitanduva - 53.4%, Góes - 43.5%, Guaiúba - 51.5%, Tombo - 44.2% and São Pedro - 51.4%). In Gonzaguinha and Prainha Branca beaches, yellowish pellets were the most common (39.1% and 44.4%, respectively) (Fig. 3). In the March 2015 survey, the predominance of clearer pellets remained in Itaquitanduva and Góes beaches (61.1% and 49.4%, respectively). June and September 2015 surveys brought the variation between the beginning and the end of the winter. In June 2015, all the four beaches sampled had more than 50% of clearer pellets (Itaquitanduva - 56.7%, Góes - 50.2%, Riviera - 60.6%, and Itaguaré - 65.3%). In September 2015, this pattern changed in all beaches. The percentages of clearer pellets dropped to 47.6% in Itaquitanduva, 37.9% in Góes, 48.9% in Riviera, and 49.6% in Itaguaré.

In comparison with the sampling surveys conducted in June and September 2015 (data from Izar et al. 2019), a significant difference was found just for the factor Date (Table 2). The proportions of different



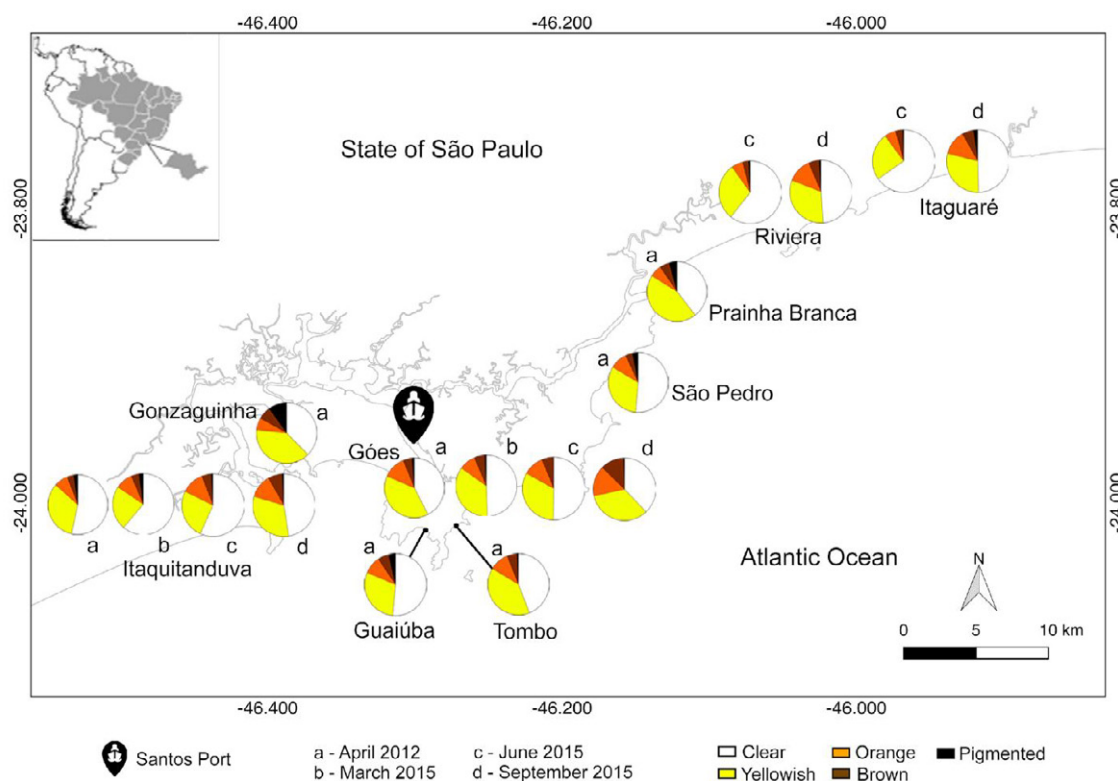


Fig. 3 Map of São Paulo coast showing the percentages of each pellets' color in each beach sampled (>100 pellets). Letters represents the surveys: a - April 2012, b - March 2015, c - June 2015 and d - September 2015; and the pie colors represent the color of each pellets' group.

Table 2 Two-way multivariate PERMANOVA (Euclidian): comparison of five color groups of pellets between dates of samples and beach location in the Santos Bay: Date (Factor 1, random, orthogonal, 2 levels) and Location (Factor 2, fixed, orthogonal, 2 levels). Df = Degrees of Freedom, MS = Mean Squares, F = F-statistic and p = p-value. Bold numbers represent significant p values. Permutation N = 9999.

Source	Df	MS	F	p
Date	1	393.04	10.79	0.005
Location	1	179.98	4.94	0.095
Date x Location	1	15.17	0.41	0.581
Residual	4	36.42		

pellets' color were similar in both sampling surveys, with the predominance of clear pellets, followed by yellowish, orange, brownish, and pigmented ones. However, a reduction of the percentages of clearer pellets was evidenced in all four sites sampled (Itaquitanduva, Góes, Riviera, and Itaguapé beaches). It is possible to observe a clear separation between the two surveys in nMDS graphic (Fig. 4), as indicated on axis 1 of this ordination. Clear pellets were the main contributor to this variation (72.07%), followed by orange (11.11%) and yellowish pellets (9.57%) (Table 3). Even with these differences between samples, each color's relative contribution remained the same as the total in both samples (Fig. 5).

Comparing the five beaches sampled in April 2012, a significant difference was observed for the factor Beach (Table 4). The color distribution in percentage for each sample date was different, but the color pattern was similar. Clearer colors were the primary variable that contributed to this difference (43%), followed by yellowish pellets (23.4%) (Table 5).

Itaquitanduva beach exhibited a different proportion of pellets' colors than Gonzaguinha and Góes beaches (Fig. 7). For the factor Beach Area, no significant differences were found for the pellets' color patterns on all beaches tested, observing more remarkable similarity between Itaquitanduva beach areas (Fig. 6). Comparing the beach area where pellets were sampled for each beach separately, it is possible to identify differences in Itaquitanduva and São Pedro beaches. Both beaches had different pellets' color



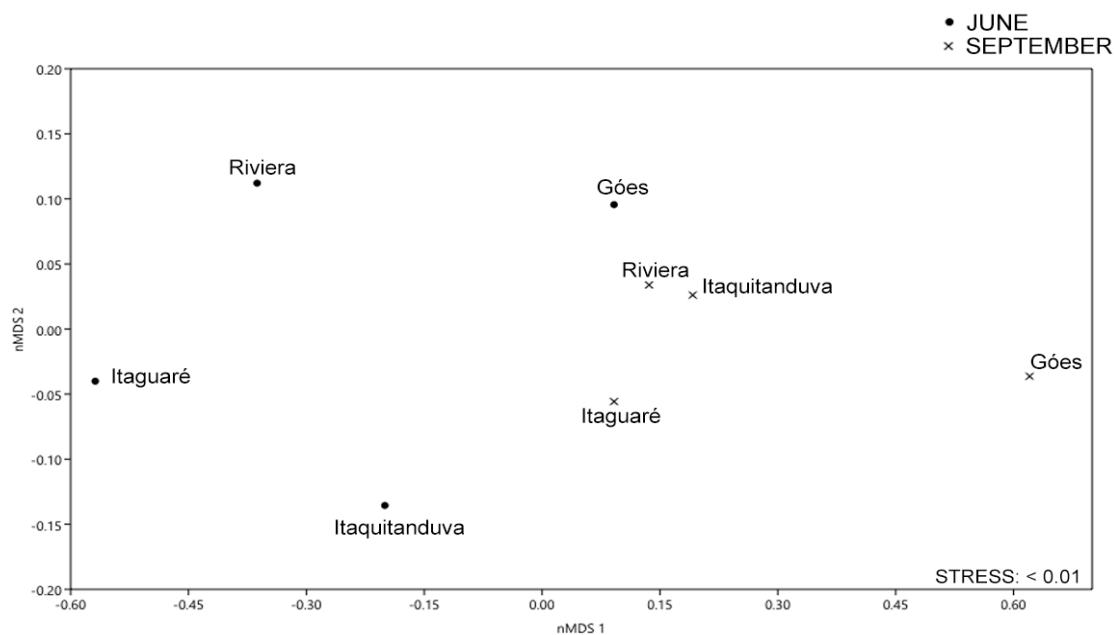


Fig. 4 Ordination of samples based on non-metric multidimensional scaling (nMDS - Euclidian) results for factor Date. Dots represent the sampled beaches on June 2015, and X represents the sampled beaches on September 2015.

Table 3 SIMPER (Euclidian) analysis results showing the most influent colors discriminated by sample dates.

Color	Contribution (%)	Cumulative (%)	Mean June	Mean September
Clear	72.07	72.07	58.20	46
Orange	11.11	83.18	8.59	13.30
Yellowish	9.57	92.75	28.10	31.80
Brown	6.95	99.7	4.47	7.78
Pigmented	0.3	100	0.60	1.16

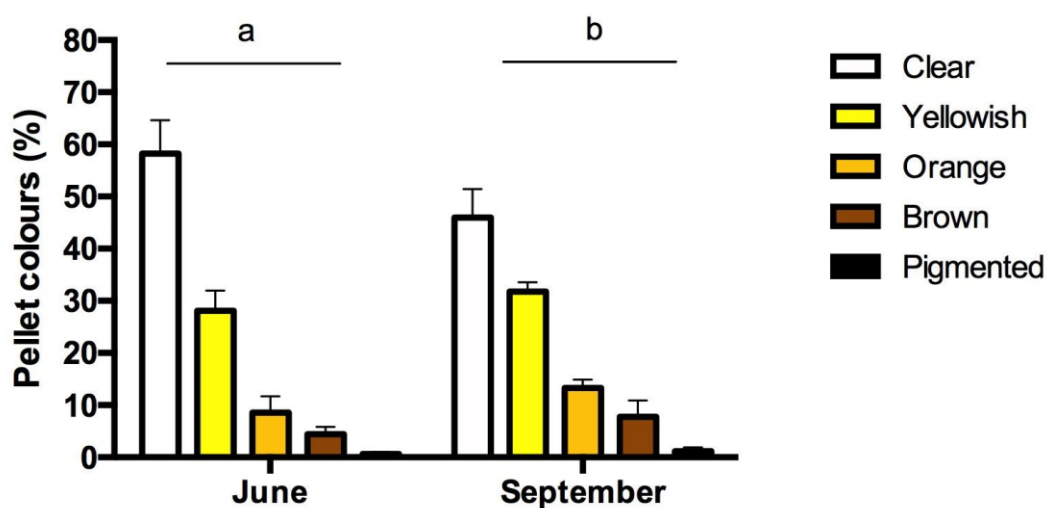


Fig. 5 Percentage of pellets' color distribution at the samples made in 2015, contribution of each pellets' color and comparison between the samples collected in June and September. Samples were obtained by the same method in different months. Different letters indicate significant differences of the pellets' color patterns between samples. Significant differences obtained through a PERMANOVA that compares the color block of each sample and not each separate color.

patterns at the center and left from the beach's right side, with a predominance of clearer pellets in the center and right side of Itaquitanduva and right side of São Pedro. (Fig. 6 and Fig. 8).

Particles from Santos Bay cluster evolve throughout northwest sectors of the bay and with some



Table 4 Two-way multivariate PERMANOVA (Euclidian): comparison of five color groups of pellets between beaches sampled and area sampled on the beaches: Beach (Factor 1, random, orthogonal, 5 levels) and Area (Factor 2, fixed, orthogonal, 3 levels). Df = Degrees of Freedom, MS = Mean Squares, F = F-statistic and P = P-value. Bold numbers represent significant P values. Permutation N = 9999.

Source	Df	MS	F	p
Beach	4	1028.7	2.62	0.0004
Area	2	227.89	0.58	0.5086
Beach x Area	8	- 142.42	- 0.36	0.2597
Residual	30	391.82		

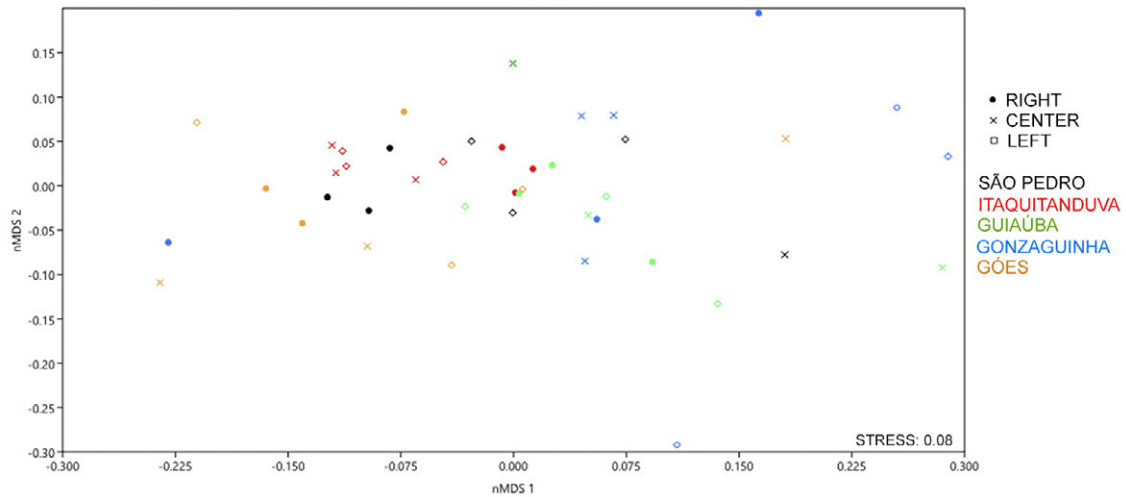


Fig. 6 Ordination of samples based on non-metric multidimensional scaling (nMDS - Euclidian) results for factor Beach (colors) and Area (symbols).

Table 5 SIMPER (Euclidian) analysis results showing the most influent colors discriminated by beach sampled

Color	Contrib (%)	Cumul. (%)	Mean Itaquitanduva	Mean Gonzaguinha	Mean Góes	Mean Guaiúba	Mean São Pedro
Clear	43.02	43.02	52.2	35.7	37.7	52.5	41.3
Yellowish	23.39	66.41	34	36.2	42	28.4	45.5
Orange	12.46	78.88	7.12	7.54	13.1	9.52	7.55
Pigmented	11.1	89.97	2.91	11.7	2.8	3.25	2.14
Brown	10.03	100	3.73	8.82	4.47	6.28	3.49

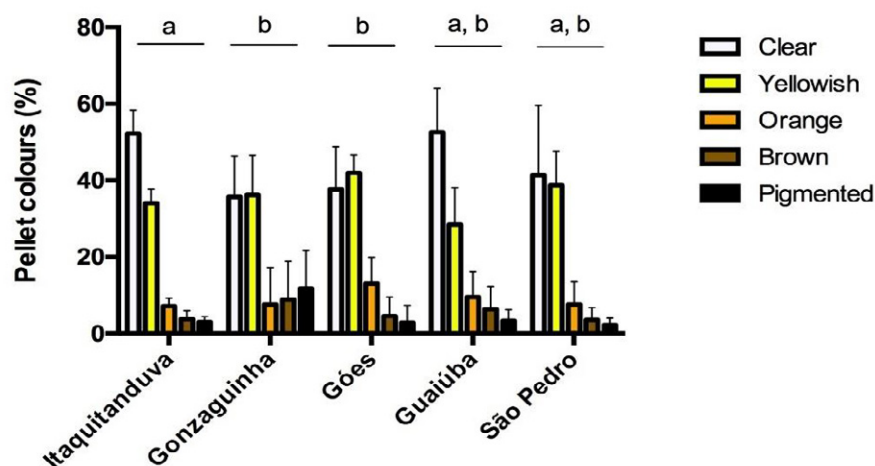


Fig. 7 pellets' color distribution (%) at the beaches in the April 2012 sampling survey. Different letters indicate significant differences of the pellet color patterns between beaches. Significant differences obtained through a PERMANOVA that compares the color block of each sample and not each separate color.



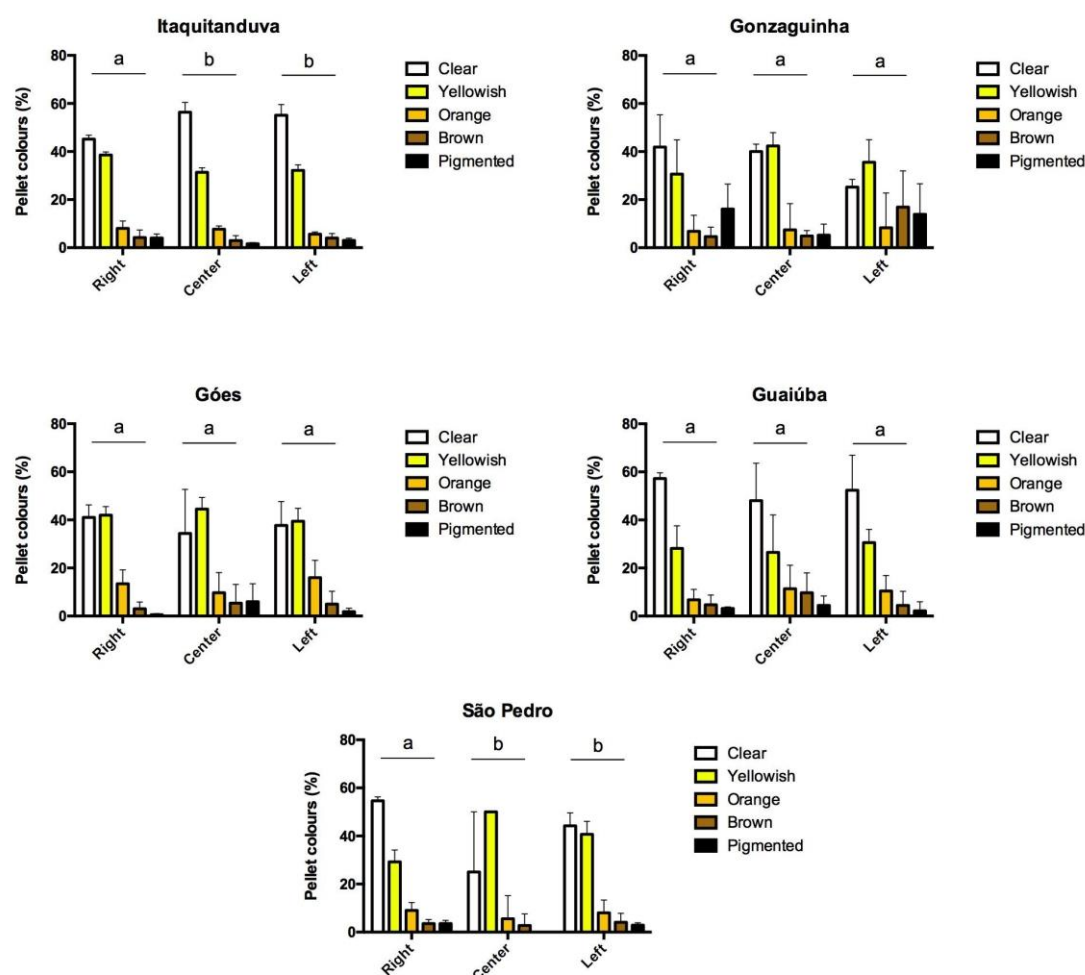


Fig. 8 Pellet's colors (%) distribution at the sample area on 5 beaches in São Paulo coast. Different letters indicate significant differences of the Pellet's colors patterns between sampled areas in each beach. Significant differences obtained through a PERMANOVA that compares the color block of each sample and not each separate color.

particles reaching the open sea. While particles from the Bertioga Channel cluster evolve throughout the north, suggesting that pellets of inside the estuary can easily reach the region of Itaguapé Beach (Fig. 9).

At the end of 15 days simulation, 98% of Santos Bay cluster particles were stranded along the way in the Northwest direction, with a higher concentration on the bay's west side (where Gonzaguinha and Itaquitanduva beaches are located). Some minor depositions can be observed in small islands near Guarujá city, close to Guaiuba, Tombo, Eden, São Pedro, and Prainha Branca beaches. After this 15-days simulation, no particles (0%) reached Riviera and Itaguapé areas, while 1% remained floating. On the other hand, particles from the cluster released at Bertioga Channel reached Riviera and Itaguapé beaches (15.7%), while 1% remained floating and 83.3% stranded along the Bertioga Channel and other beaches around.

Discussion

The color of stranded-beach pellets collected in this study was predominantly clearer, followed by yellowish, orange, and brown. Pigmented colors were less common than others. The contribution of different colors in the total of the four samples was similar to those found in previous studies around the world (Antunes et al. 2013; Carpenter and Smith 1972; Day et al. 1990; Fanini and Bozzeda 2018; Karapanagioti and Klontza 2007; Marti et al. 2020; Pianowski 1997; Ribeiro and dos Santos 2020; Shaw and Day 1994). Clear (transparent, white, and grey tones) and yellowish pellets (light and dark yellow tones) represented more than 80% of the sampled pellets. Besides, these two color groups were the most prevalent in all sampled



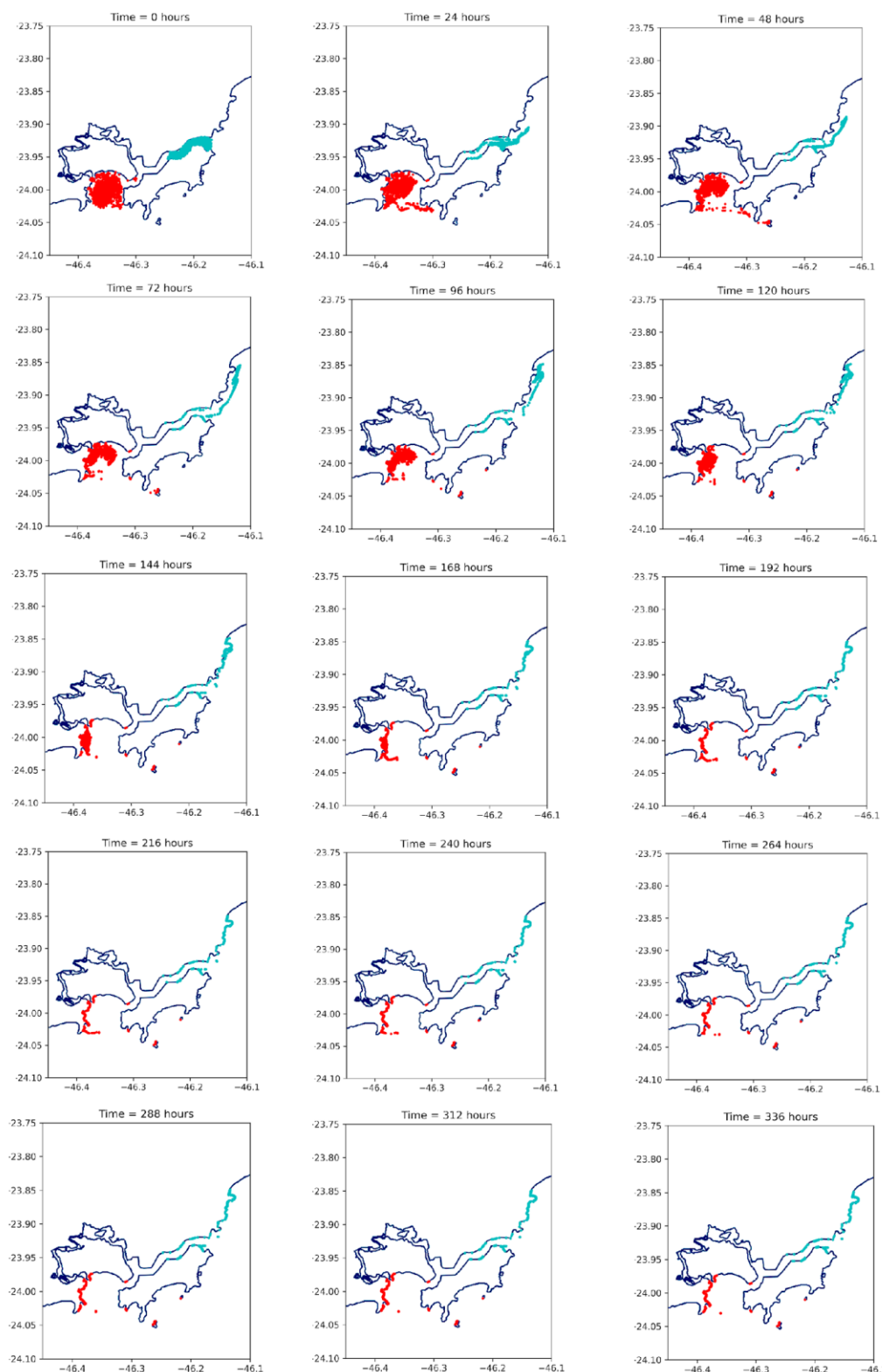


Fig. 9 GNOME oil model for simulation of particles released as two clusters of pellets source in Baixada Santista, São Paulo, Brazil. The model shows the dispersion of both clusters of particles under the most favorable wind and currents conditions. Red dots represent particles released in Santos Bay and Blue dots represent particles released in Bertioga Channel. The GNOME model runs for 15 days.

beaches, individually or combined.

Clearer pellets generally are new or low-aged pellets (estimated time since manufacture) that tend to present a higher concentration of chemical additives, such as vinyl acetate, vinylidene chloride,



bisphenol-A, phthalate esters, aliphatic dibasic acid esters, benzoate esters, trimellitate esters, polyesters, citrates, polybrominated diphenylethers, alkylphenols, and other substances (Teuten et al. 2009). These substances tend to be released and leached from the pellets (Li et al. 2016) and thus can be transferred to the biota and sediments (Teuten et al. 2009). In contrast, the yellowish and dark tones colors in pellets indicate weathering, loss of additives, and adsorption of hydrophobic contaminants such as PAHs, DDTs, PCBs, PBDEs, PFAS, and pharmaceuticals (Endo et al. 2005; Engler 2012; Fisner et al. 2013; Hammer et al. 2012; Hirai et al. 2011; Islam et al. 2021; Karapanagioti and Klontza 2007; Llorca et al. 2014; Nobre et al. 2020; Prata et al. 2018; Taniguchi et al. 2016; Teuten et al. 2009) and even metals (Fotopoulou and Karapanagioti 2012). Yamashita et al. (2018) showed a correlation between the color of plastic pellets and the concentration of PCBs, with increasing concentrations towards darker pellets.

The prevalence of clear pellets, followed by yellowish ones, could be related to the Port of Santos and the grains cargo terminals, which represent sources of continuous inputs of pellets to the adjacent coastal environments (Balthazar-Silva et al. 2020; Gorman et al. 2020; Manzano 2009; Pereira 2014; Turra et al. 2014). The increased percentage of white-translucent pellets during the wintertime (July) suggests the intensification of pellets transport due to waves and currents. Rainfall, wind speed, wind direction, and sea surface currents were similar in both periods preceding the sampling (Marinha do Brasil 2015). However, we can hypothesize that the distribution of plastic pellets along the coast is a mid to long-term process and thus, weather and oceanographic events have an important role. June is the beginning of winter in the southern hemisphere when strong winds, storms, and storm surges are increased, influencing the deposition and removal of pellets on the beaches. Clearer pellets were more abundant in June 2015 sampling (winter), suggesting that the proportions of colors observed are a region's pattern, because it is the pattern most found during the period of greater climatic influence. The abundance of yellowish, orange, and brown pellets increased in September 2015, reducing the percentages of clear pellets, probably due to the intensification of storm surges in the winter, which re-suspend former pellets deposited or buried in the sediments and remobilize them, causing their spread along the coast. Besides, Balthazar-Silva et al. (2020) stated that the entry of plastic pellets on a regional scale could vary considerably in periods over a year. The variation might be more intense after rainfalls, decreasing pellets deposition and increasing pellets' washing out the beach (Fanini and Bozzeda 2018). However, the relatively low variation in the proportions of pellets of different colors in most beaches suggests a balance between their input and removal rates. Thus, at the same time, waves and currents are likely bringing pellets from other places and carrying away pellets. If this is true, the input rates by the sources are probably constant, and in the long term, pellets "jump" from one beach to another during storms in such a way they can be transported for much longer distances. Further studies focusing on these aspects are required to elucidate the way pellets are transported along the shore, particularly the influence of storms.

On the other hand, the higher percentage of clearer pellets has other implications for marine life since the particles' colors may influence the ingestion by several organisms and their predation habits. Animals with prey perception from above eat more clear particles, while animals with perceiving prey from below tend to eat more dark particles (Santos et al. 2016). A high amount of light-colored pellets were previously reported in seabirds' stomach contents (Day et al. 1985; Sileo et al. 1990; Vlietstra and Parga 2002). Ingestion of clearer plastic pellets may also cause transference of some chemicals to the biota, such as stabilizers and plasticizers (Li et al. 2016; Teuten et al. 2009).

Clearer pellets were the most abundant (> 50%) in 3 of 7 beaches sampled in April 2012 (Itaquitanduva, Guaiúba, and São Pedro). That could be related to the proximity to pellets sources (namely, the Port of Santos and industrial complex, as previously mentioned), as both, as located within the Santos Estuarine System. This result confirms our hypothesis even with beaches outside to Santos Bay (i.e. São Pedro) following this pattern. That suggests the pellets introduced into the environment by local sources can be transported to regions beyond the bay's geographical limits (Izar et al. 2019). The predominance of clear pellets in Itaquitanduva beach indicates a constant input of new pellets (Fanini and Bozzeda 2018). This fact raises a concern because Itaquitanduva beach is located inside the Xixová-Japuí State Park. The presence of plastic pellets also concerns Tombo, São Pedro, Prainha Branca, and Riviera beaches, which are part of the Central Coast Marine Environmental Protection Area (APAMLC), and Itaguaré beach, which is also within a state park. More than the physical impacts of the plastic pellets, the presence of yellowish pellets, presenting adsorbed contaminants, may represent a risk to the biota in these areas since yellowish pellets



Table 6 Comparative studies in literature between different pellets' colors and chemical contaminants related.

Studies	Color groups analyzed	Chemical compounds analyzed	Findings
Endo et al. 2005	w - white (non-fouled and non-discolored) f - white (fouled and non-discolored) y - yellowed (discolored) fy - fouled and discolored)	PCB	Discolored and/or fouled pellets had higher PCBs concentrations ($y > fy > f > w$). The highest PCBs concentrations found in those study samples were in a bright orange pellet and the second one in yellowed samples.
Frias et al. 2010	white - translucent white virgin pellets aged - yellow to brown colored - pigmented pellets except black black - black pigmented pellets	PCB DDT PAH	All contaminants analyzed were higher in black and aged pellets.
Antunes et al. 2013	white - translucent white virgin pellets aged - yellow to brown colored - pigmented pellets except black black - black pigmented pellets	PCB DDT PAH	PCBs and DDTs concentrations were higher in aged pellets. PAHs concentrations were higher in aged, colored and black pellets, majority from petrogenic sources.
Fisner et al. 2017	9 color groups: 1–4 - white tones 5–7 - yellow to orange 8–9 - brown and dark tones	PAH	PAHs concentrations in pellets increase across the color spectrum, from light to dark tones.
Yamashita et al. 2018	White Yellowing Orange/Brown	PCB	PCBs concentrations were 29 times higher in orange/brown pellets than in white pellets. In yellowing pellets, PCBs concentrations were lower than orange/brown, meaning that pellets yellowish tones still in uptake phase.

tend to become increasingly contaminated (Ashton et al. 2010; Gorman et al. 2019).

Most of studies of contaminants in plastic pellets focus in yellowish pellets to reduce individual difference in chemicals analysis (Heskett et al. 2012; Jayasiri et al. 2015; Karapanagioti et al. 2011; Mizukawa et al. 2013; Tanigushi et al. 2016), as proposed by the International Pellet Watch (IPW) protocol (Ogata et al. 2009). Nevertheless, a few studies made comparisons of contamination between pellets of different colors (Table 6). Concentrations of chemical compounds adsorbed on pellets were higher in yellowish tones in all studies. Yellowish and dark tones are exposed for longer times in the environment (Endo et al. 2005) and consequently subject to more significant weathering effects, which change the plastic properties and making them more susceptible to adsorption of contaminants. This relationship between color and chemical concentrations is essential for monitoring programs that aim to use plastic pellets as indicators of local contamination, as suggested by Endo et al. (2005). Also, citizen science initiatives can use pellets' colors for the first screening of coastal areas to provide initial estimations of their potential to cause chemical contamination.

White pellets represent virgin and new pellets, the raw material commercialized and shipped in the Port of Santos, where the primary sources of plastic pellets are installed in region. The hydrodynamic modeling indicated that pellets released inside the estuary and in the Santos Bay could be transported toward beaches on the north coast of the São Paulo state. Pellets from Bertioga Channel can reach the beaches of Bertioga city, close to Riviera and Itaguapé beach, faster than those from Santos Bay. On the way, there may be



stranding and constant remobilization until these pellets reach beaches located further north. This long-term transport was not assessed in this study, requiring further studies. Many oceanographic and meteorological conditions may define the paths and fates of those particles after leaving the estuary throughout the coastal domain. Tides, winds, and oceanographic processes in coastal shelves can wipe off particles depending on the intensity and direction of driving forces. In this study, estuarine circulation is far more predictable than the coastal circulation, mostly dependent on the continental shelf dynamics and other oceanographic processes. These are related to wind and ocean pressure fields on the entire Southeastern Brazilian Shelf (Fontes and Castro 2018), where low-frequency oscillations, or subtidal regime, are more energetic than high-frequency movements driven by the tides, meaning that only prevailing NE coastal currents could transport pellets from the estuary to Bertioga Beach.

The harbor region's proximity can explain the higher density of clearer pellets stranded on Itaquitanduva beach in all samples. The ebb tide's influence coming from the estuary's interior, where the Port of Santos is located, might be an essential factor. In this area, pellets are transported into the Santos bay by the estuarine channels (São Vicente channel and Santos channel) and surface currents, especially in the spring tides, the seasonal period that these currents are more intense (Fernandino et al. 2016). Surface currents in the Santos bay turn counterclockwise (Harari and Camargo 1998; Harari and Gordon 2001), pushing pellets to the bay's west side, where Itaquitanduva beach is located. Also, this could explain the clearing color difference between Itaquitanduva and Góes beaches. Both beaches are located inside Santos bay, and Góes is much closer to the Port of Santos. However, in Góes beach, yellowish, orange, and brown pellets had high densities in all samples. The counterclockwise direction of surface currents inside the Santos bay can be related to the predominance of yellowish, orange, and brown pellets in Góes beach. Pellets strand in Góes beach only after going through the entire spin inside the bay. This longer exposure time floating in the environment may increase the chance of yellowing. Also, mangroves' presence in the region could contribute to the yellowing pellets found in those beaches. Pellets can get stuck on sediment and between roots within mangroves areas (Martin et al. 2019; Manullang 2020; Zhou et al. 2020) and be released back after heavy rains and very high tides, increasing pellets exposure time. Beach-stranded pellets on beaches inside the Santos bay can also be removed and transported to furthest beaches, like São Pedro and Prainha Branca, located further north of Port of Santos. Pellets on Prainha Branca beach were more yellowish than clear, suggesting a more significant distance covered by the harbor influence (as a plastic pellets source).

In our samples, clearer pellets were more abundant in the high tide line, representing the new beach-stranded pellets (Moreira et al. 2016). In these areas, plastic pellets usually stay longer and consequently more exposed to photodegradation and adsorb more chemicals due to their uneven surface (Fotopoulou and Karapanagioti 2012). On beaches, pellets are more exposed to photodegradation in the physical process (Corcoran et al. 2009). Both previous information, reinforce our hypothesis of more clear pellets close to the source.

Differences in beach sample areas can reflect those phenomena described above in micro-scale. Beach areas with higher quantities of clearer pellets (e.g., center and left portions of Itaquitanduva beach and right side of São Pedro beach) probably represented the depositional regions at the moment of the sampling. That shows the heterogeneity of the stranding of pellets on beaches, which is very complex and depends on the coastal geomorphology and its interaction with hydrodynamic factors (size, period, and direction of incident waves), coastal currents, the proximity of potential plastic pellets sources, wind direction (offshore/onshore) and rainfall precipitation (Moreira et al. 2016). A heterogeneous deposition of pellets may result in different degrees of ecological impacts in the same beach, with implications to plastic pellets monitoring programs. Since some differences were observed, we suggest more studies regarding the distribution of pellets with different colors on beach areas and their relationship with climatic and oceanographic processes.

Conclusion

This study showed that beach-stranded pellets close to pellet sources had a consistent color pattern: light tones (white and yellowish). This pattern suggests a constant supply of plastic pellets over time. Besides that, the analysis of colors of beached plastic pellets in environmental assessments and monitoring programs might provide additional information on their age, behavior in the environment, and ability to carry contaminants, contributing to the understanding and management of this problem. However, we



believe that the next steps of science in this topic would be an investigation to relate pellets' colors with beach climate factors and hydrodynamic processes, considering the local contexts and particularly the fate of plastic pellets on a long-term scale.

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Availability of data and material All data used in this study are available for consultation and transparency.

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