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GEOSCIENCES

Temporal monitoring of contamination in three sandy beaches from the 2019 oil spill near Cabo de Santo Agostinho, Northeastern Brazil

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Abstract: In 2019, a massive oil spill impacted coastal ecosystems in eleven Brazilian states; these ecosystems were diverse with regard to their different geological settings (sandy beaches, rocky coasts, estuaries, tidal flats, mangroves, and reef systems) being impacted. Here, we present a temporal analysis on the occurrence of remaining contaminants on three beaches (Paiva, Itapuama, and Enseada) of the Cabo de Santo Agostinho City, Pernambuco State. The approach focuses on the systematic mapping of residues, collection of sediment samples, photographic documentation, and qualitative microscopic analysis. Grain size, calcium carbonate, and total organic matter content analyses provided a faciological and energetic characterization of the environment. The objective was to verify the relationship between the remaining contaminants and the geological setting. The results showed intense weathering of the remaining oil residues due to the high energy environment. We found tar balls reworked by seasonal erosion on the three beaches and no tar residues in shallow subsurface sediments. A large quantity of tar residues remains on rock surfaces in Itapuama Beach, which bears the higher permanence of contamination because it is less affected by weathering and beach erosion. This investigation allowed for an impact assessment of the geologicalmorphological characteristics of the beaches.

Key words: beach sediments, environmental disaster, oil spill, Pernambuco State, tropical coastal environments.

INTRODUCTION

From August to November 2019, large quantities of crude oil washed onto the coastal regions of northeastern Brazil. The contamination impacted coastal environments in nine states in the northeast region and two on the southeastern coast, Espírito Santo and Rio de Janeiro. An estimated 1009 locations were affected (IBAMA 2020) (Fig. 1). In the months following, oil traces continued to be reported on the northeastern Brazil coast by the Brazilian Navy, the Environmental Protection Agency (IBAMA), and civilians. The agent responsible for the oil spill has not been discovered despite ongoing investigations carried out by national and international organizations. The IBAMA also suggested that the origin of the spill could have been located 400 to 700 km off the Brazilian margin, and the oil may have floated for approximately 40 days until it reached the coast. Some analyses carried out on the oil suggest that it has characteristics compatible with heavy crude oil produced in Venezuelan oil fields (De Oliveira et al. 2020, De Oliveira Soares et al. 2020). However, no accidents with tankers

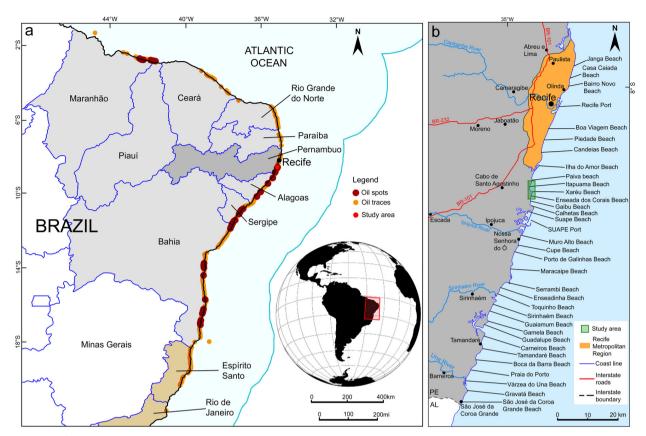


Figure 1. a) Map showing the distribution of contaminated areas along the Brazilian coastal zone until the end of 2019 caused by the offshore oil spill (IBAMA 2020, Gonçalves et al. 2020), which impacted nine states of northeastern Brazil (gray) and two states of the southeast margin (brown). The light blue zone marks the exclusive economic zone. The insert shows the impacted region on the eastern margin of South America. b) The southern coast of Pernambuco state and the study area is shown by the green rectangle.

carrying such types of oil have been reported. De Oliveira et al. (2020) used chemical analyses and multivariate statistics to identify the origin of the oil and suggested that the material has chemical characteristics compatible with some oil types from Venezuela, indicating that it represents altered crude oil due to the time it spent at sea or that it was in a product manufactured with heavy oil produced in this country. Lourenço et al. (2020) showed that oil samples collected from various locations on the Brazilian coast share the same source and that the chemical signatures were not conclusive to determine their geologic origin.

Recent estimates claim that more than 5,000 tons of oil were removed from Brazilian

beaches, mangroves, and coral reefs from August 2019 to January 2020 (Brum et al. 2020). Magris & Giarrizzo (2020) described the extent of the oil spill impacts on coastal marine habitats, as well as a list of threatened coastal species and socioeconomic impacts. The accident damaged the economy due to the suspension of touristic activities and fisheries, which provide the majority of jobs in the region (Magris & Giarrizzo 2020, Câmara et al. 2021).

In the littoral of the Pernambuco state, the oil affected a region with rich marine biodiversity, including benthos, fishes, marine mammals, birds, and a series of complex and fragile ecosystems, such as mangroves and seagrass meadows, which were already battered by anthropogenic pressure (Araujo et al. 2007). Latest reports released by the Pernambuco government claimed that the clean-up operation of the beaches and rivers in the coastal zone, resulted in the removal of approximately 1,650 tons of waste (oil mixed with sand and other materials) since the first oil spots washed up on São José da Coroa Grande beach on October 17th, 2019. The government reported a total of 48 locations in littoral areas and eight locations in rivers/estuaries that were contaminated.

The most affected area in the Pernambuco state was the coastal region of Cabo de Santo Agostinho city (Figs. 2 and 3), where approximately 1,032 tons of waste were removed from the beaches (Figs. 2a to 2d). This region presents a great diversity of coastal depositional systems, such as sandy beaches, rocky shores, tidal flats, estuarine inlets and channels, mangrove strips, and pools in beachrocks and reefs (Manso et al. 2018). The clean-up operation was mainly carried out by mechanical means of removing the oil deposited on the beaches, which was mixed with sediments and seagrass (Fig. 2). Figure 2 shows the intense work performed by volunteers to remove the pollutant from the studied beaches immediately after its arrival.

In the months following the oil spill, tar balls continued to be reported along the Brazilian coast, mainly in July/August 2020, after severe storms exhumed the deposits buried in the previous spring. In Pernambuco, government



Figure 2. Record of the pollution caused by the oil spill event in the study area (August to November 2019). a) Oil spots, Itapuama Beach. b) Cleanup work by government personnel and volunteers in Paiva Beach. c) Plastic bags containing oil removed from the beaches. d) Aerial view of the rocky shores of Itapuama Beach during the climax of the oil arrival on the coast. The topography of the rocky shore is formed by small beachrocks and reef pools with sandy substrates in the foreshore region. e) Emulsified oil patties percolated through naturally formed fractures and joints in the rocks outcropping in the backshore of Itapuama Beach. f) Part of the rocky shore of Itapuama Beach, which is covered by sediments. Yellow arrow - oil floating during low tide, white arrow - oil patties deposited during a receding tide, red arrow - oil patties deposited during high tide in the supratidal zone. White arrow - oil deposited between the rocks in the foreshore, red arrow - oil that overwashed and flooded rock surfaces outcropping in the upper part of the foreshore zone.

officials reported tar residue in Cabo de Santo Agostinho County, south of Recife, and on the beaches of Janga and Paulista, to the north. In Cabo de Santo Agostinho, there were reports of tar residue on Itapuama beach, encrusted in the rocks. In addition to the tar balls found in the subsequent months after the disaster, many areas of the rocky shores of Itapuama Beach remain contaminated because the oil had infiltrated vesicles, fractures and joints in the rock surfaces (Fig. 2e and 2f).

Tar balls and oil-mineral aggregates form during the emulsification and weathering of crude oil and its interaction with minerals (Kiruri et al. 2013). Marine tar residue represents weathered oil conglomerations found in beaches, transitional systems such as estuaries and the open ocean surface, and the seafloor (Warnock

et al. 2015). Marine tar residues can originate from anthropogenic and natural emissions like petroleum seeps. The term tar ball is defined as a round aggregate of weathered oil generally less than 100 mm in diameter. Discrete aggregates larger than 100 mm are referred to as tar patties. Large thick deposits of weathered oil that are partially or completely submerged in water are referred to as tar mats (Warnock et al. 2015). The weathering of crude oil involves a combination of processes such as spreading, evaporation, dissolution, biodegradation, emulsification, sedimentation, dispersion, and oxidation, which leaves a higher concentration of the heavier and more viscous compounds of the oil. Some tar marine residues result from the mixing of oil and sediments, such as quartz-rich sands and biogenic particles. The mixture with sand causes

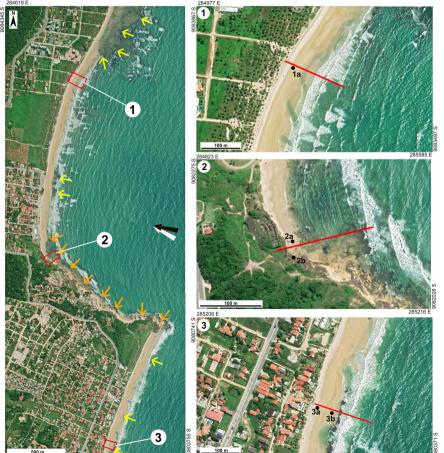


Figure 3. Satellite images of the study area. The larger image shows the study area between the southern portion of Paiva Beach (1), and Enseada dos Corais Beach (3). The black and white arrows indicate the main directions of the winds and currents, respectively (Azevêdo et al. 2021). Yellow arrows indicate beachrocks and orange arrows indicate the rocky coast of Itapuama Beach (2). Insert 1) survey profile 1 (red line) and sampling Station 1a (black dot) in Paiva Beach. Insert 2) survey profile 2 in Itapuama Beach and sampling Stations 2a (sediments) and 2b (rock surfaces). Insert 3) survey profile 3 and sampling Stations 3a and 3b in Enseada dos Corais Beach (image source: Google Earth Pro). Images are from September, 13, 2019.

the oil to lose its buoyancy and sink (Warnock et al. 2015).

In ecosystems severely impacted by oil spills, originating from accidents with wellbores and oil tankers, oil residues are still present several decades after contamination. In the case of the Exxon Valdez oil spill that affected Prince William Sound, Alaska, in 1989, recent studies showed that oil residues were not weathered or biodegraded. Toxic compounds from the Exxon Valdez spill have been found in sediments and water three decades after the accident (Lindeberg et al. 2018). Similarly, Bociu et al. (2019) reported the persistence of tar balls originating from the Deepwater Horizon oil spill buried in sands of the Gulf of Mexico. Their experimental investigation showed that the decomposition of these materials, which includes toxic compounds, will take at least three decades.

This investigation aimed to study the persistence of tar residues in a sector of the Pernambuco coastal zone formed by three beaches (Paiva, Itapuama, and Enseada dos Corais), fourteen months after the detection of the first tar residues in the studied beaches (Figs. 1 and 3). This region is located in the littoral region of Cabo de Santo Agostinho, the most impacted zone in Pernambuco state (Figs. 1 and 3). Furthermore, this area was selected because, in this short coastline sector, which is approximately 7 km long, there are examples of beach profiles formed by different geological substrates and hydrodynamic constraints representing coastal systems commonly found in northeast Brazil. This research tried to determine the relationship between the geodiversity of the small coastline sector affected by the oil spill and the potential they present to tar residue permanence. Geodiversity has been defined as "the natural range (diversity) of geological (rocks, minerals, fossils), geomorphological

(landforms, topography, physical processes), soil and hydrological features. It includes their assemblages, structures, systems and contributions to landscapes" (Gray 2013). The oil spill severely affected the three beaches studied, and the oil was deposited on sandy beaches strongly influenced by marine erosion; in small low tide pools, associated with beachrocks and reef lines; and on the rocky shore formed by outcropping of igneous rocks of Cretaceous age (Figs. 2 and 3).

CHARACTERISTICS OF THE STUDY AREA Geological setting

The study area is located in the coastal zone of the Pernambuco Basin (Buarque et al. 2016), a Cretaceous rift basin of the eastern Brazilian margin (Matos 1999). The coastal zone is bounded to south by the Pernambuco shear zone, and to the north by the Maragogi High (Buarque et al. 2016, Correia Filho et al. 2019). The outcropping rocks in the coastal zone comprise conglomerates and sandstones of the Aptian Cabo Formation; sandstones, shales and claystones of the Lower Albian Suape Formation; sandstones, siltstones and claystones of the Upper Albian Paraíso Formation; limestones, marls, and calcareous sandstones of the Cenomanian-Santonian Estiva Formation; and sandstones, claystones and conglomeratic sandstones of the Miocene-Pleistocene Barreiras Formation (Correia Filho et al. 2019). The basin is also formed by volcanic and subvolcanic rocks, and one alkali-feldspar granite formed by post-rift volcanism comprises the Ipojuca magmatic suite (Nascimento 2003). The volcanic rocks are Upper Albian in age and include basalts, rhyolites, trachytes, trachyandesites, and ignimbrites (Nascimento et al. 2004, 2008). The volcanic rocks occur as sills and dikes, and are secondarily interbedded with sedimentary rocks as spills. The littoral

zone is mainly dominated by Quaternary and Recent sediment cover: marine sands and clay are associated with estuaries, mangroves and tidal flats; Pleistocene and Holocene sandy terraces formed by paleobeaches (Dominguez et al. 1990, Mio & Giacheti 2007, Suguio et al. 2011).

Climatic, hydrological and meteoceanographic constraints

The study area is dominated by a tropical, hot, and humid climate, with an Am type monsoon according to Köppen (1936). The annual rainfall ranges from 1900 to 2200 mm.y ⁻¹, and the annual temperature varies from 24 to 26 °C (Alvares et al. 2013), averaging of 25 °C (Koening et al. 2002). The relative humidity has an annual average higher than 80%, and the average annual evaporation is approximately 170 mm (Silva 2004). Two distinct seasons characterize the climate of the region: a dry season from September to February and a rainy season (winter) from March to August (Passos et al. 2021). During the rainy season, it is not uncommon for precipitation to exceed 400 mm/month, and the hydrological balance is strongly positive. In the dry season, when monthly rainfall is less than 60 mm, the evaporation rate exceeds precipitation (Araújo et al. 1999). This seasonal rainfall pattern affects the local hydrological and sedimentary behavior, as observed by Barcellos et al. (2016) for the Jaboatão estuarine area, located at the northern limit of Paiva's beach. Pardal et al. (2019) observed an increase in sedimentary organic matter and mud during winter in the Capibaribe River estuarine area near Recife city in a temporal study (2010-2017).

On the other hand, the influence of the nearby rivers on the hydrodynamics of the studied beaches is limited due to their low flow rates, which are directly associated with the coastal characteristics of the study area. The transport of sediments occurs from south to north (Fig. 2). The Massangana River estuary, located to the south, is fed by a small coastal river (catchment area of 102.5 km²) with a relatively low discharge (< 2 m³.s⁻¹) (Passos et al. 2021). The Jaboatão River (catchment area of 413 km²) (Barcellos et al. 2016), is located north of Paiva Beach and has an average flow of 2.03 m³.s⁻¹ (Araújo et al. 1999) (Fig. 3).

The prevailing winds are seasonal and regular from the ESE-NNE direction approximately 90% of the time, with a mean speed varying from 3 to 5 m.s⁻¹ (Manso et al. 2018, Azevêdo et al. 2021). Tidal behavior in the region is mesotidal and semidiurnal, with a mean amplitude ranging from 0.7 m to 2.0 m (Schettini et al. 2016). Circulation in the coastline area is controlled by wave-generated currents, which create a northward longshore drift. The waves are influenced by the wind's local and regional regimes, and the current along the coast has a northward trend (Domingues et al. 2017). Between January and April, the maximum significant wave heights remain below 2 m, mainly in the NE-ENE direction. From May to August, they remain above 2 m (Manso et al. 2018), with wave periods from 5.4 to 14.8 s and have a predominant E, ESE and SE direction, influenced by the South Atlantic Polar Anticyclone cold front (Holanda et al. 2020, Azevêdo et al. 2021).

The mesotidal regime dominates the study region (Manso et al. 2018). Coastal morphodynamics are strongly influenced by tidal currents, especially when they are associated with SE winds and spring tides. This association produces intense erosive processes throughout the coastal zone. The current parameters were measured with an ADCP near the Recife and Jaboatão coastal regions and recorded average intensities from 0.09 to 0.23 m.s⁻¹, with peaks reaching 0.64 m.s⁻¹ throughout the year. The higher intensities were associated with the rainy period (winter) and an NNE prevailing direction

of currents, which causes the seasonal increase in river flow (Manso et al. 2018). However, the influence of this increase is limited to the river's mouth (Barcellos et al. 2020), indicated by the deposition of mud lithogenic sediments in the inner shelf adjacent to the local fluvial inlets.

Coastal geodiversity of the study area

The population of Cabo de Santo Agostinho City is approximately 209,000 people (SUAPE 2018). The region has a variable relief with three distinct geomorphological landforms: the hilly domain, colluvium ramps, and the coastal plain (Neumann et al. 1998); the original vegetation on the littoral was the Atlantic tropical rainforest, which was replaced by sugarcane in most of the

area since the 16th century (SUAPE 2018: Oliveira et al. 2020). The coastal zone presents great geodiversity (geological and geomorphological processes and landforms) with sandy beaches, estuaries, hypersaline tidal flats, rocky coasts, mangroves, reefs, pools, and beachrocks. The studied beaches, Paiva (8°17'S, 34°56'W), Itapuama (8°18'S, 34°56'W), and Enseada (8°18'S, 34°57'W), form a continuous shore zone on the Pernambuco central coast, approximately 7 km long (Figs. 3 and 4), and they present narrow depositional profiles. This region is 12 km to the south of Recife city, the capital of Pernambuco state, and just 12 km northward of the SUAPE port complex. Paiva and Enseada represent exposed oceanic beaches (Madruga Filho &

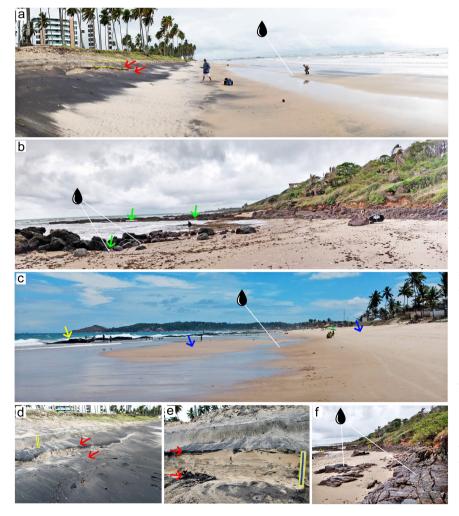


Figure 4. Characteristics of the studied beaches. a) Picture of the southern sector of Paiva Beach, looking toward the northeast. Dashed yellow lines show the scarp limit of the most recent erosive processes. b) Rocky shore of Itapuama Beach, looking toward the southeast. Green arrows indicate small pools. c) Picture of the Enseada Beach, looking toward the south-southeast. Blue arrows indicate the backshore and foreshore zones. The yellow arrow indicates beachrocks. d) and e) Red arrows show scarps formed by recent erosion and two beds containing seasonally deposited heavy minerals (vellow marker - 30 cm ruler). f) Rock surfaces in the upper part of the foreshore of Itapuama and rocks outcropping where the foreshore region is covered by sand. The black drop symbol indicates tar occurrences found during the study. Pictures were taken in March 2021 during low tide.

Araújo 2003; Holanda et al. 2020), and both exhibit an NNE-SSW orientation (Figs. 3 and 4) and faces in the ESE direction. Itapuama beach, a geosite (Nascimento et al. 2012), is classified as a partially sheltered parabolic bay shaped beach (Lino 2015, Holanda et al. 2020), with an ESE-WNW orientation and it faces NNE-SSW (Figs. 3 and 4).

There are no published studies on the marine sedimentology and morphodynamics of Enseada Beach or the sedimentological characteristics of Itapuama Beach. According to Madruga Filho & Araújo (2003), Paiva beach is composed mostly of fine to coarse siliciclastic sands that are moderately sorted, with low calcium carbonate contents. However, on a local low tide terrace in the foreshore, the $CaCO_3$ content was higher than 30% (Fig. 4).

The rocky shore of Itapuama Beach is formed by a large outcrop of hypabyssal trachyte rocks that were emplaced in the region as a sill. These rocks have a porphyritic texture with millimetric phenocrysts of sanidine embedded in a fine-grained matrix (Nascimento et al. 2004; 2012). These rocks present intensely fractured, with planes trending E-W and N-S. The surface of outcrops presents a vesicular texture, with structures varying from 5 to 30 mm in diameter (Figs. 5c and 5d).



Figure 5. a) Example of a square trench excavated to observe the buried sediments in the sampling stations (Fig. 3). The image shows the occurrence of two beds enriched in heavy minerals, Paiva Beach. b) Shallow trench (strip) excavated in Itapuama Beach. c) Survey and collection of tar residues at sampling Station 2b (Fig. 3). Itapuama Beach. d) Example of collecting tar residues from the rock surfaces, Itapuama Beach.

MATERIALS AND METHODS

The study comprised the following procedures: 1- the mapping and visual inspection of the beaches, 2- collecting sediments for qualitative microscopic analysis, 3- a photographic record of the different residues found, and 4sedimentological analyses.

The research involved a qualitative visual survey on the selected areas during three sampling campaigns from January to March 2021. The fieldwork used compasses, scales, pocket magnifiers, a GPS, switchblades, hammers, trowels, and shovels. Information provided by local residents was crucial for selecting the sampling points. The field campaigns occurred during low tide due to better exposure of the beach profiles, including the rocks (trachytes) and beachrock outcrops (Fig. 4). The search for tar residues was carried out in rock outcrops. beachrock surfaces, the bottom surface of the pools, and areas between the rocks in the foreshore exposed during the low tides. The research also performed microscopic analysis supported by a portable UV light of the rock samples to characterize the encrusted tar residues. Analysis of nine samples collected at Paiva, Itapuama, and Enseada beaches was performed in the laboratory using a low-cost device with a 400 nm frequency and 9 watts of ultraviolet light, as described by Rivis et al. (2019).

A series of hand-dug trenches were excavated to verify the occurrence of tar residues entrained/buried in sediments at four sampling stations (1a, 2a, 3a and 3b - Fig. 3). Station 2b is located in the rock outcrops of Itapuama Beach and was used as a reference site to collect tar residues in the rock surfaces (Fig. 3). These trenches allowed an in situ visual analysis of the sediments in the shallow subsurface and sample collection for sedimentological analysis. There

were three types of trenches: 1) 0.40 m square, 0.50 m deep, 2) 1 m long, up to 0.30 m wide, and up to 0.60 m deep, and 3) 0.40 m square, 1.20 m deep (Fig. 5). Sediments were collected 0.05 m, 0.10 m, 0.20 m, and 0.30 m below the surface (Fig. 5) in trenches types 1 and 2 and 0.10 m, 0.50 m, 1.00 m and 1.20 m below the surface in trenches type 3. The locations of the sampling stations were chosen to coincide with the parts of the beaches that were most contaminated during the 2019 oil spill (Fig. 2). The depth of the trenches and sediment collection took into account the average seasonal amount of erosion and redeposition of sediments in the beach profiles (Lino 2015, Holanda et al. 2020). The depth of the trenches was also sufficient to find reworked and entrained tar residues in the sediments deposited since the 2019 disaster. The transects shown in the stations (red lines in Figure 3) represent the position of the schematic profile used to describe the relationship between beach morphology and the temporal effect of contamination. The chosen transect directions considered only the high morphological diversity found in the beaches. Table I shows information about the sampling stations and sample collection.

Eight sediment samples were collected for sedimentological analysis from the three profiles during the March sampling campaign. Grain size analyses were performed by the sieving and pipetting method (Muller 1967). Calcium carbonate (CaCO₃) and total organic matter (TOM) contents were determined by the weight difference before and after acidification with 1 N HCl and 1 N H_2O_2 , respectively (Carver 1971). The grain size parameters were determined according to Larsonneur's (1977) faciological classification, Folk & Ward's (1957) statistical treatment, and Pejrup's (1988) hydrodynamic energy diagram. The organic carbon content was calculated based on TOM contents multiplied by 0.58 (Allen 1974).

RESULTS

Tar balls on sandy beaches

During fieldwork, a few tar balls varying from 5 to 30 mm in diameter were found on the three beaches. The tar balls were found on the foreshore of the sand beaches and represent a small amount of the residue when compared to the volumes of the contaminant that washed up in 2019 (Fig. 6a, b and c). All tar balls were found in the foreshore and swash zones during low tide. Beached tar balls found in Paiva Beach (January 2021) represent scattered particles ranging from 5 to 30 mm in diameter, which washed up with the tidal oscillation and were possibly remobilized during storms. The material is somewhat viscous, but most fragments were hard and crispy. The residue was mixed with sediments of siliciclastic and biogenic origin

(Figs. 6c). Beached tar balls of approximately 10 to 20 mm were also found on Enseada Beach in January 2021 (Figs. 6d and 6e). This material is also somewhat viscous and is more consistent compared to the tar residues found on Paiva Beach. The residue was mixed with sediments and showed almost no fluorescence under UV light. The fragments were rounded and washed up with tidal oscillations.

Tar balls of approximately 10 to 20 mm were found near the rocks on the shoreface of Itapuama Beach (Fig. 6f and 6g). These residues were found scattered and mixed with biogenic particles and also trapped in spaces between the rocks (Figs. 6h). This material has a lower amount of mineral and biogenic particles and has a solid-like consistency. It may represent the remaining residues of the original contaminant, which became trapped in the rocks (Figs. 7 and 8) and was not entrained in the sediments. Thus, the degradation process could be different for these residues than for the tar balls found on

Sampling station	1a (Paiva Beach)	2a (Itapuama Beach)	2b (Itapuama Beach)	3a (Enseada Beach)	3b (Enseada Beach)
UTM Coordinates	25L 0285219E / 9083711S	25L 0284983E / 9082139S	25L 0284987E / 9082114S	25L 0285521E / 9080495S	25L 0285574E / 9080478S
Position	Upper foreshore	Lower foreshore	Upper foreshore	Upper foreshore	Lower foreshore
Substrate	Sand	Sand	Trachyte	Sand	Sand
Trench type 1	Jan and March 2021	Jan and March 2021	-	Jan and March 2021	Jan and March 2021
Trench type 2	-	March 2021	-	March 2021	-
Trench type 3	March 2021	-	_	March 2021	-
Sediment collected for sedimentological analysis	March 2021	March 2021	-	March 2021	March 2021

Table I. Sampling station characteristics, location, and type of trenches excavated in the temporal analysis.

the sandy beaches (Enseada dos Corais and Paiva). These tar balls also did not fluoresce under UV light (Fig. 9). No stranded tar balls, or tar patties, were found in the sediments excavated from the trenches at the sampling stations or in the backshore or foreshore zones of the sandy beaches. Sediment analysis with a portable microscope showed no clear evidence of tar residue particles, but it requires further study.

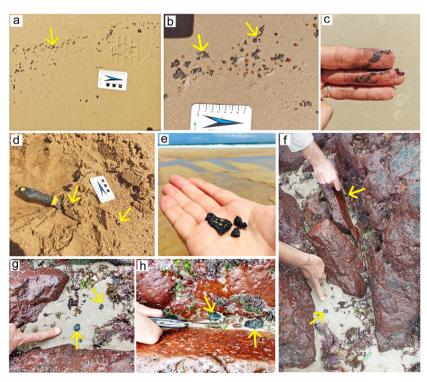
Tar residues encrusted in the rocky shore

A large quantity of tar residue remains encrusted on the surface of trachyte rocks on the rocky shore of Itapuama Beach (Figs. 7a to 7f). Encrustation is also present in the backshore because it is only affected by water erosion during high spring tides. This effect helped reduce the degradation of tar residues that are not exposed to subaqueous conditions. Most of the remaining tar residues are encrusted in natural fractures, joints and in the vesicles of the trachyte rocks (Figs. 7 and 8). As the material breaks down the contaminants percolate deep through the rock joints. After initial degradation, the aggregate material is mixed with sediments in the rock fractures and filled vesicles (Figs. 8a-c). The external surface of the tar residue filling the vesicles and fractures was weathered and oxidized. The tar that filled the vesicles is a millimeter-thick film that can be scraped off with a metal blade (Figs. 7c-f).

Mesoscopic samples collected for analysis showed that the tar residues deeply percolated the joints (Fig. 8). The natural enlargement of the vesicles by meteoric processes created cavities up to 40 mm in diameter and connections between the cavities were filled by the tar residue (Fig. 8). These vesicles within the joints help to protect the tar residues from subaerial weathering. The leaching of this material continues to liberate contaminants for a longer amount of time than the material exposed on the sandy beaches.

Scraping off the tar encrusted in the vesicles revealed that the interior of the film is

Figure 6. Beached tar balls and residue found on the studied beaches. 6a to 6c) Tar residues ranging from 5 to 30 mm in size found in the surf zone of Paiva Beach, January 2021. The isolated tar fragments were washed up with biogenic fragments and presented a viscous consistency. 6d and 6E) Tar balls, 5 to 30 mm in diameter, found in the surf zone of Enseada Beach during low tide, January 2021. The material is solid and is mixed with siliciclastic and biogenic particles. 6f to 6h) Tar balls found in the bottom of a pool during low tide in the surf zone of Itapuama Beach, March 2021. The isolated tar fragments were found with biogenic detrital particles and trapped in natural fractures (6h). The locations where the tar residues were found are indicated in the photos of Figure 4.



shiny with a viscous consistency: this suggests a less altered condition of the material as it is protected in the vesicle cavities (Figs. 9af). Sampled subsurface sediments showed no evidence of hydrocarbons under UV light, and only small spots of fluorescence were observed related to microorganisms and other particles of organic origin (Figs. 9g and 9h). Analysis of the tar residues scraped from the trachytes under UV light showed little signature of light hydrocarbons, which indicates highgrade degradation of the material (Figs. 9i-k). Comparative microscopic analysis of the tar residue scraped from the rocks (Figs. 9c and 9d) and from the tar balls found on the sandy beaches (9e and 9f) showed that the tar balls contain less siliciclastic content.

Sedimentological analysis showed that the beaches contain lithoclastic sand (AL1b) facies (Larsonneur 1977), and the texture varies from fine to medium sands and is moderately sorted (Folk & Ward 1957). Sediments have low organic contents and are subjected to very high hydrodynamics (IVa) according to the Pejrup (1988) diagram. These beach sediments are mostly composed of siliciclastic grains, ranging from 99.46 to 100.00% (mean: 99.87 ± 0.21%). Gravel and mud particles are only observed in Itapuama's beach profile, with 0.44% gravel and 0.10% silt at Station 2B (Fig. 3), and 0.03% gravel and 0.34% silt at Station 2C. Silt was also observed at Stations 1A and 1B in Paiva's beach profile, but was extremely low with 0.10% and 0.03%, respectively (Fig. 3). Clay particles were absent in all the collected samples.

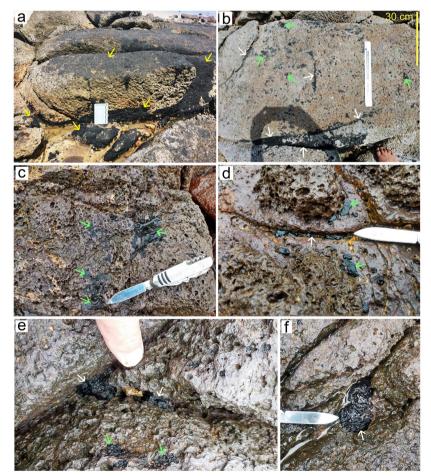


Figure 7. Tar residues in the trachyte rocks of Itapuama Beach. a) Oil residues adhered to the surface of the rocks and trapped within the ioints and vesicles in the backshore in December 2019. b) Rock surfaces in the upper part of the backshore. c) Tar residues trapped in vesicles on a subvertical surface. d) Horizontal surface with tar residue. 7e and 7f) Tar residue trapped in rock joints. Photos c to f were taken in January 2021. White arrows - tar residue encrusted in fractures and joints, green arrows - tar residues encrusted in rock vesicles. The surfaces are wet due to rain. The seawater reaches these areas only during high spring tides.

The geochemical parameters of sediments, such as the calcium carbonate content, are low and vary from 2.9 to 8.4%, averaging of 4.6% \pm 2.2%, and they are classified as lithoclastic (Larsonneur et al. 1982). The (TOM) and the estimated total organic carbon (TOC) contents (Allen 1974) ranged from 0.43 to 3.30% (mean: 1.39 \pm 0.85%) and from 0.25 to 1.00% (mean: 0.80 \pm 0.49%), respectively. These ranges are low and are typical of highly dynamic beach face environments. A relatively enriched value (3.30%) of TOM and TOC (1.00%) was observed at Paiva Beach (1B station); however, Romankevich (2013) also considered a low content to be < 2% TOC.

The sedimentary character of these sandy beaches is directly related to the nature of the coastal fluvial terrigenous sediment inputs and distribution pattern. The prevailing shelf facies are dominated by sandy-gravelly biogenic poorly sorted sediments with low organic matter contents (Barcellos et al. 2020). The quartz-rich sandy beach facies that predominate these shores is the result of the mixing of riverine and shelf palimpsest sediments that are constantly reworked by local longshore currents. These processes keep the sandy siliciclastic deposits along the coastline and the inner shelf (Manso et al. 2018), and mixing with terrigenous mud (Larsonneur 1977) occurs near the river mouths (Barcellos et al. 2020).

DISCUSSION

The oil spill that affected the northeastern coast of Brazil revived the apprehension caused by similar accidents involving oil spills in recent history, such as the Exxon Valdez tanker in Alaska in 1989 (Shigenaka 2014) and the Deepwater Horizon platform in the Gulf of Mexico in 2010 (Parham & Gundlach 2015). Information about the amount and nature of the contaminant washed on the Brazilian coast, as well as its evolution in the environment in the following years is necessary to better understand the long-term impact and the lingering effects of

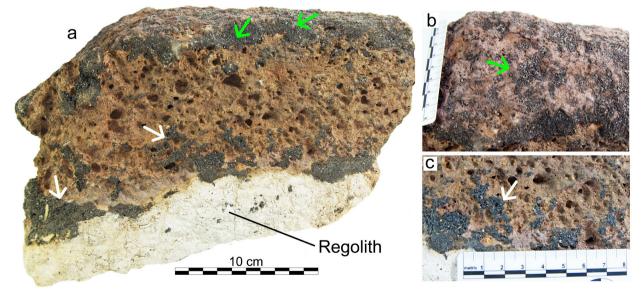
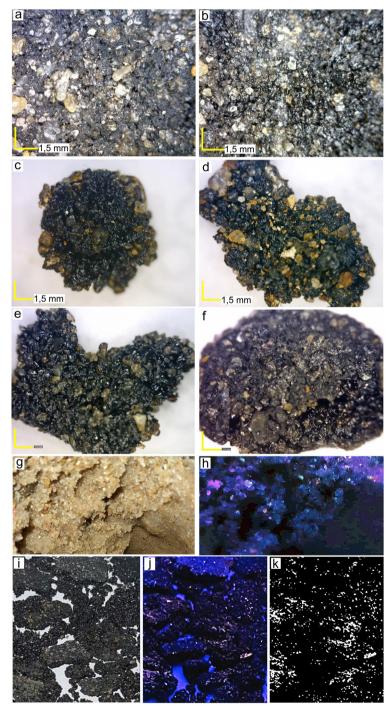


Figure 8. Mesoscopic rock sample collected from the outcrops of the backshore of Itapuama Beach in December 2019. a) Image of a joint face. b) The picture shows that the contaminant encrusted the vesicles in the external surfaces of the outcrops (green arrows - top),) and percolated through the joints, creating accumulations in their contacts (white arrows). The whitish part of the sample marks the buried zone (bottom) which has been leached and altered.

contamination. This will help to compare Brazil's oil spill with similar disasters. The present study provides a qualitative analysis of a small coastal strip with complex geological-morphological diversity that was heavily affected by the oil spill.



The Exxon Valdez accident liberated approximately 35,500 tons of crude oil into the ocean, which mainly reached the Prince William Sound region and spread over 2,100 km of the coastline (Nixon & Michel 2018). Thirty-two years after this accident, studies are continuing to be

> Figure 9. 9a and 9b) Photos of the tar residue adhered within the vesicles of the sample shown in Figure 8. a) Surface of the oxidized and weathered material, and b) the internal view after the surface of the encrusting residue was mechanically removed, which is internally better preserved. The material covering the rocks is highly mixed with siliciclastic minerals and rock fragments. 9c and 9d) Tar residue removed from the vesicles. 9e and 9f) Tar balls collected in Enseada Beach, January 2021 (Fig. 4c). This material is soft and viscous, and the fragments possess minor contents of clastic and biogenic grains. The surface is less oxidized and weathered than observed for the tar residue on the exposed rocks. 9g and 9h) Visible light and UV light photos of sand samples collected at Station 3. Enseada Beach. No fluorescence related to hydrocarbons was detected, and only fluorescence related to biogenic materials was observed. 9i, 9j and 9k) Images of tar residue removed from the rocks. Visible light and UV light images, and contrast processing was used to highlight the fluorescent areas detected. The analysis suggests that the luminescence is related to the high altered tar residue.

produced by government agencies and research institutions and show that environmental damage persists in the Gulf of Alaska. mainly in the Prince William Sound region. The persistence of oil contaminants (toxic compounds) in coastal environments, even decades after the disaster. is discussed by Gundlach et al. (1983), Guterman (2009), Peterson et al. (2003) and Schrope (2010, 2011). The spill caused oil residue to be impregnated on the surface of corals, birds, and mollusks and the massive mortality of some critical groups. The impregnations caused by oil blankets on sandy beach sediments can reappear on the surface decades after the initial spill (Payne et al. 2008). In just one year of cleanup work in the Prince William Sound region, the Exxon Mobil Corporation removed more than 30,000 tons of crude oil (Piper 1993), which cost more than two billion dollars and involved two million man-hours (Rice et al. 2007). However, Wolfe et al. (1994) estimated that less than 20% of the initial volume that was spilled in that region was removed. Lindeberg et al. (2018) confirmed that 26 years after the Exxon Valdez accident, contaminants are still physically present. In the case of Alaska, the morphological and geological characteristics of the coast were revealed to be critical factors affecting the long persistence of the contaminant. The oil reached protected coarse-grained gravel beaches of William Sound, which present armored stable substrates. The oil that percolated through the armored surface and impregnated the sediment was still present at depths greater than 0.25 m in beaches with stable armor eighteen years after the accident (Li & Boufadel 2010). Nixon et al. (2013) pointed to the influence of geomorphic factors, substrate permeability, low exposure to waves, and armoring in gravel beaches as drivers for oil persistence in the subsurface. Lindeberg et al. (2018), suggested that after 14 years of the spill, there was little evidence of change in the

distribution of oiling intensities or the location of the contaminated zones in the William Sound beaches. They demonstrated that oil lingers at 8 of the 9 sites revisited since the accident and that the permanence of oil sequestered in sediments and protected from hydrological washing in low oxygen shallow subsurface conditions inhibits biodegradation (Lindeberg et al. 2018). These conditions will certainly cause the contaminants to persist in the environment for decades.

The Deepwater Horizon explosion resulted in the spill of 22,000 tons of oil in the northeastern Gulf of Mexico and along the shoreline, where it is estimated that there were 965 km of polluted sandy beaches. Bociu et al. (2019) performed a three-year in situ experiment and a survey to quantify the temporal decomposition of oilsediment-agglomerates (tar balls) in Florida buried in the upper 0.50 m of sandy beaches. Analysis of the chemical parameters and the fluorescence response showed that buried tar balls up to 60 mm in diameter would take approximately 30 years to decompose. Bociu et al. (2019) show that tar balls buried deep in beach sands will decompose through rapid microbial oil degradation in tidally ventilated permeable beach sand, what, according to the authors, emphasizes the role of sandy beaches as aerobic biocatalytic reactors. Parham & Gundlach (2015) studied the sedimentary evolution of oil-sand aggregates of the Deepwater Horizon oil spill on sandy beaches of the northern Gulf of Mexico. According to these authors, high wave and tide conditions resulted in the deposition of oil in the supratidal zone and burial at depths of up to 1 m, which produced large volumes of oil-sand aggregates. Effect of higher seasonal (summer) water level variations and wave energy, combined with storms and overwash dynamics. reworked the buried/stranded oil. Subsequent exposure caused by beach hydrodynamics

of buried oil-sand aggregates led to its rapid degradation through in situ weathering and breakdown under surface, subsurface, and subtidal conditions. The authors emphasized that continuous-wave energy/overwash and weathering will be effective in the breakdown of long-term remaining residue.

Regarding the ethnology of the diverse types of sediment-oil aggregates (SOAs) (Gustitus & Clement 2017), we suggest that the material found in the beaches can be classified as tar residues (tar balls or tar aggregates), formed due to the erosion of oiled sands on beaches polluted by heavy oil (Warnock et al. 2015). However, the remaining residues may have formed pelagic tar balls, resulting from the mixture of sunk oil and its entrainment in sediments that were subsequently reworked and washed ashore. We consider the residues adhered to the exposed rocks as a type of tar residue that results from in situ mixing with sediments transported by wind and waves. This encrusting material was not the result of reworking in the beaches or the effect of pelagic transported aggregates. This kind of tar residue is not well described in the literature and deserves further study.

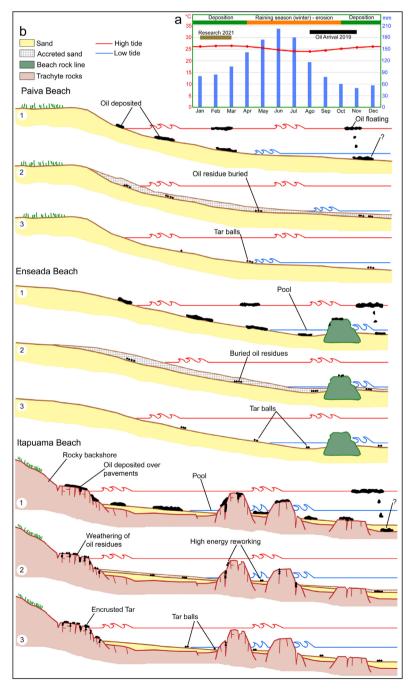
The finding of dozen beached centimetric tar balls and the apparent absence of macroscopic residues (tar patties and sand-oil aggregates) up to 0.60 m deep in the supratidal and intertidal sediments suggest that the processes of weathering and breakdown due to the highenergy conditions were effective in decomposing most of the contaminants left behind after the clean-up operation.

As it is clear from the comparison to other disasters that occurred in different marine environments, some parameters, such as wave energy, morphology, climate and geodiversity, are key factors that influence lingering pollution. Processes of erosionaccretion, which can remobilize large volumes

of sediment make the residues bioavailable and expose them to weathering. Yim et al. (2020) described the rapid recovery of the Hebei Spirit oil spill on the western coast of South Korea. The spill released 10,900 tons of three types of crude oil and contaminated 200 km of coastline, which severely affected various ecosystems. The authors showed that after ten vears of monitoring, the concentration of the oil residue in seawater, sediments, and oysters rapidly dropped to background levels at 16, 75, and 33 months, respectively. Damaged benthic intertidal and subtidal communities recovered after six years. Yim et al. (2020) showed that high tidal mixing (~9 m) and an intensive cleanup operation (~1.2 million volunteers) during the very initial period of contamination contributed to rapid recovery, which was 4-5 times faster than the Exxon Valdez case.

The results of the present study showed a large degree of homogeneity in terms of the composition and pattern of facies distribution across the beach profiles. The study area is mostly characterized by siliciclastic sands with heavy mineral grains as observed in Paiva beach, which agree with the pattern already described by Madruga Filho & Araújo (2003). The characteristics are similar on the Enseada dos Corais Beach, where the profile is sheltered by beachrock outcrops located on the upper shoreface (Fig. 4c). It was also found granular marine components dominated by calcium carbonate grains and silts in the Itapuama profile.

In Paiva Beach (1a in Fig. 3) sediments were collected at the surface and at a depth of 1.2 m (trench) in the upper foreshore region. This area was severely affected by the deposition of oil patties (Fig. 2b) in October 2019. Analysis of these samples showed that the sediments represent fine (1A) to medium sand (1B), moderately sorted, and dominantly lithoclastic (> 99.90% of CaCO₃) (Larsonneur et al. 1982). Similar faciological characteristics were reported by Madruga Filho & Araújo (2003). These authors indicate that the faciology is directly related to the local wave energy fluctuations. The analysis of the chemical composition of the sediments from Paiva Beach (1.2 to 1.3 m depth) also revealed relatively enriched values of TOM (3.30%) and TOC (1.00%)



in the sandy deposits. This unexpectedly high organic matter concentration could be related to the subsurface decomposition of oil residue deposited in 2019 before the deposition of the sampled sediments (Fig. 10). However, this conjecture needs a more sophisticated approach to determine the nature of the compounds present in these sediments.

> Figure 10. Schematic model of oil contamination evolution. a) Climograph with monthly average temperatures (red) and precipitation (blue) for Cabo de Santo Agostinho City (source: climatedata.org/Copernicus Climate change Services. Weather data were collected between 1999-2019). The rainy season (winter) marks the period of seasonal erosion. b) The proposed three stages for the evolution of oil residues in the studied beaches. 1 -Deposition of oil. The tidal variation allowed the contaminants to reach the foreshore and parts of the backshore zone. This stage occurred after the erosive period when the sandy beaches presented a concave profile. 2 - Residues left after the cleaning operation were reworked and buried due to the high-energy process of sand remobilization. During this period, oil residues underwent intense weathering (chemical, mechanical, biochemical) and breakdown. 3 - The following erosive stage allowed the exhumation of sandoil aggregates (beached tar balls). The weathering of the oil residues encrusted in the rocky shore underwent a different evolution, which prevented its degradation.

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The results allowed us to determine that the three studied beaches are subjected to similar wave energy influences dominated by high hydrodynamics, as indicated by 100% of the grain size analysis (Pejrup 1988). Holanda et al. (2020) and Lino (2015) classified Paiva and Itapuama beaches as moderately to highly vulnerable to erosion, according to the Coastal Vulnerability Index (CVI) (Martins et al. 2016). We determined the same features for Enseada Beach. However. the relatively sheltered conditions and the different orientation of Itapuama Beach (Figs. 3 and 4b) are possibly responsible for the higher CaCO₂ content and the beached gravelly tar balls found in the intertidal zone, which suggests comparatively lower hydrodynamic conditions.

Lino (2015) and Holanda et al. (2020) described the seasonal profile variation for Paiva and Itapuama beaches and demonstrated that these beaches present annual effects of accretion and erosion with up to 1 m of vertical variation, especially in the swash zone's low tide terraces (Fig. 4). Beach erosion increases in autumn-winter (March to June) due to the higher wave energy. The studied sand beaches present convex profile shapes during summer (December to March) and concave shapes in winter (Manso et al. 2001). During winter, low tide terraces form, and in summer, the beach profiles exhibit an alternation among rhythmic bar and transverse bar and rip stages (Paiva Beach) (Holanda et al. 2020). Low tide terraces also form in Itapuama Beach during winter (Lino 2015).

During the climax of the oil arrival on the coast, from 17 to 25 October 2019, the beach profiles were eroded and concave in shape, created by seasonal sand remobilization (Fig. 10). During the following summer (December 2019 to March 2020), the oil residues left were buried by remobilized sands, and the beach profiles were convex in shape (Figs. 4a, 4c, and 4d). During the subsequent morphodynamic stage of the annual cycle (July to August 2020), beached tar balls washed ashore in several localities following severe storms that hit the littoral zone. This material, probably buried during the last spring-summer, was eroded, exhumed and remobilized by tidal currents.

This monitoring study allowed us to propose that seventeen months after the beginning of the spill, most of the residues in the sandy beaches were degraded by weathering, influenced by the high-energy hydrodynamic of the area. We need to emphasize that the enormous mobilization of volunteers in the clean-up operations possibly represent a key aspect for the rapid recovery of beaches, as highlighted by Wang et al. (2020) and Yim et al. (2020) on recent oil spills. The energy level and fast remobilization of sediments represent other key parameters for fast recovery. Despite the weathering and breakdown of residues in sandy beaches, Itapuama's rocky beach is a different situation, where a large amount of tar residue encrusts the rocks. At present, the occurrence of stranded-buried oil-sand aggregate patties in these beaches is probably rare, but we cannot discard this possibility. In the future, the main risk in the study area is the seasonal exhumation and redistribution of residues stranded in the sandy beaches and the leaching of residues in the rocky shore of Itapuama Beach.

The information gained from the monitoring work allowed suggesting a model, consisting of three temporal stages, that summarizes the fate of oil residues on the studied beaches (Fig. 10):

Stage 1 - shows the extent of oil contamination in the intertidal zone influenced by tidal variation and in the backshore zone due to overwash. We did not survey the subtidal zone, and the remaining oil contamination in this zone and the inner shelf is unclear. The clean-up operation removed most of the oil that washed up on the shore. The oil spill occurred during the seasonal process of sand erosion on the beaches (Fig. 10).

Stage 2 - The remaining residues were mixed with sediment and formed sand-oil aggregates, which were buried by the subsequent seasonal process of sand mobilization in the beaches (accretion) (Fig. 10). The fast weathering and breakdown were driven by the high permeability and oxygenation of the substrate.

Stage 3 - This stage represents the first summer period after the spill. The oil residues stranded in the sandy beaches were exhumed and reworked. Beached tar balls are evidence of the persistence of the contaminants. Large quantities of tar residues are not expected to be found on these beaches. However, the tar residue encrusted in fractures and vesicles of the rocky beach (Itapuama) are protected from marine erosion and remain a risk (Fig. 10).

CONCLUSIONS

Qualitative monitoring showed that the immediate and massive clean-up operation supported by volunteers removed most of the oil residues on the sandy beaches. This was a crucial aspect of the rapid recovery observed in this study. The remaining oil residues that were mixed with sand resulted in small quantities of sand-oil aggregates that were buried and intensely reworked on the beaches, and eventually exhumed in the seasonal process of sand remobilization. Fast degradation and breakdown of these aggregates were due to the high-energy environment (waves), climatic parameters, and the high permeability/ oxygenation conditions of the substrates. Fourteen months after the spill, beached tar balls are evidence of residue persistence on the beaches, which will be rapidly reduced in the next few years. The tar residues formed in the

rocks of Itapuama beach underwent a different evolution due to the protection created by the substrate, and it will probably persist longer than expected compared to the residues on the sandy beaches. Morphological characteristics, geological diversity, and dynamic parameters were revealed as key factors to critically understand the fate of oil residues in the studied beaches, and as demonstrated in other oil spills.

We suggest that further studies should be carried out in the subtidal zone and in the adjacent inner shelf. Collecting sediments of the surface and subsurface can show the persistence level of residues in these areas. It is also strongly recommended to continue investigating the beaches with new field campaigns of excavation and monitoring, especially of the contaminants in the rock surfaces to verify the persistence of tar residues and aggregates and disseminated toxic compounds associated with hydrocarbon degradation. Other studies need to consider the chemical characterization of the oil residues found on these beaches to discard confusion from contaminants produced by ships navigating near the littoral zone due to the intense traffic related to the SUAPE Complex port.

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Bontempo Filho, EB conceived the research, wrote the manuscript, performed lab analysis and fieldwork; Coutinho, RB conceived the research, discussed the results, revised the manuscript; Barbosa, JA conceived the research, wrote and revised the manuscript, performed lab analysis and fieldwork, Barcellos, RL performed fieldwork and lab analysis, wrote and revised of the manuscript; Giachetti, HL conceived the research, lab analysis, discussed the results and revised the manuscript; Ramos, GMS performed fieldwork and lab analysis, and revised the manuscript.

