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Occurrence, distribution, and associated pollutants of plastic pellets (nurdles) in coastal areas of South Texas

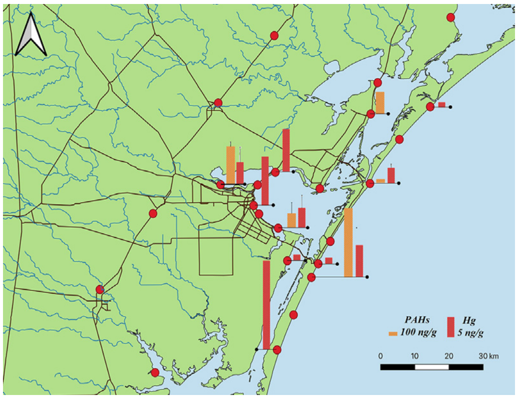
Xiangtao Jiang, Niki Conner, Kaijun Lu, Jace W. Tunnell, Zhanfei Liu *

University of Texas Marine Science Institute, 750 Channel View Drive, Port Aransas, TX 78373, United States of America

HIGHLIGHTS

- Nurdles were collected in both land-based and ocean-based regions in south Texas.
- Land-based nurdles were made of a broader range of polymer types than ocean-based ones.
- PE nurdles from shoreline sites had a broader range of oxidation indices or weathering degrees than those from railway sites.
- PAHs and mercury were detected only on ocean-based nurdles, and no PCBs were found on nurdles.

GRAPHICAL ABSTRACT



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ABSTRACT

Nurdles, also known as plastic resin pellets, are now a major source of plastic pollution on beaches globally, thus it is important to elucidate their weathering patterns and environmental fates as well as the associated pollutants. In this study we collected nurdles from 24 sites in the coastal bend region of south Texas, covering areas from the near shore railway stations to the adjacent bays and barrier islands. The morphologies of nurdles and associated pollutants including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and mercury, were investigated. The results showed that the nurdles varied greatly in color, shape, polymer composition, and oxidation degree. More than 80 % of the nurdles were made with polyethylene, and the rest with polypropylene, polyester, polystyrene, polyethylene-vinyl acetate, and polyvinyl chloride based on Fourier Transform Infrared Spectroscopy (FTIR) analysis. PCBs were not detected on nurdles. PAHs and mercury on nurdles were detected at 12 % and 20 % of the sampling sites. The total concentrations of detectable PAHs ranged from 92.59 to 1787.23 ng/g-nurdle, and the detectable mercury concentrations ranged from 1.23 to 22.25 ng/g-nurdle. Although the concentrations of these pollutants were not at the acute toxic effect level, the presence of PAHs and mercury suggested the potential risk of pollutant exposure to marine organisms in ecosystems, given the fact that nurdles are persistent in the environment.

1. Introduction

Large quantities of trash/plastic debris have been found along the shoreline of the Gulf of Mexico (Tunnell et al., 2020; Wessel et al., 2019). Among them, nurdles with about 4 mm in diameter, as pre-produced plastic pellets for final plastic products (Fotopoulou and Karapanagioti, 2012), are released unintentionally into the environment at all stages of the plastic supply chain (Karlsson et al., 2018), and they are a significant source of plastic pollution along the beaches of south Texas. Since September 2018, a large number of nurdles have been observed on Mustang and North Padre islands. According to the surveys of the Gulf of Mexico citizen

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* Corresponding author.
E-mail address: zhanfei.liu@utexas.edu (Z. Liu).

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science program, Nurdle Patrol, over 150 nurdles are frequently recorded in 10-min surveys on beaches along the Texas coasts since 2019 (Tunnell et al., 2020). Moreover, a growing body of literatures have shown that nurdles can be ingested by hundreds of marine species, such as turtle, fish, and birds (Graham and Thompson, 2009; Ryan, 1988). Evidence of various adverse effects after ingestion such as intestinal blockage, starvation, and pollutants exposure have also been reported (Besseling et al., 2015; Gregory, 2009; Jiang et al., 2019; Ma et al., 2016).

Nurdles are typically made of hydrophobic polymers, such as polyethylene and polypropylene (Endo et al., 2005; Jiang et al., 2021). Once dispersed in the environment via wind, surface currents and tides, hydrophobic nurdles have the potential to adsorb organic pollutants from the ambient environment (Jiang et al., 2021; Ogata et al., 2009). Launched in 2005, International Pellet Watch Program has been monitoring the pollution status of the plastic pellets in the ocean, focusing on persistent organic pollutants (POPs; http://pelletwatch.org/). A variety of POPs including PAHs, PCBs, polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane, and dichloro-diphenyl-trichloroethane (DDT) have been detected on nurdles collected from natural waters and beaches (Antunes et al., 2013; Chen et al., 2018; Fisner et al., 2017; Jiang et al., 2019; M. Rios et al., 2010; Ogata et al., 2009; Zhang et al., 2015). For example, concentrations of total PAHs and PCBs were in the ranges of 53–44,800 ng/g and 2–223 ng/g, respectively, on nurdles collected along the Portuguese coastline (Antunes et al., 2013); concentrations of total PAHs were in the range of 737 to 39,763 ng/g on polyethylene (PE) pellets and 871 to 9252 ng/g on polypropylene (PP) pellets collected from the Santos Bay in Brazil (Fisner et al., 2017).

As the sixth-largest port in the United States, the Port of Corpus Christi accepts around 70 million metric tons of petroleum annually (City of Corpus Christi data). The well-developed petrochemical industries here also bring in a huge amount of networks of pipelines from wells to refineries, which all increase the potential release of petroleum hydrocarbons to the environment and thus the adsorption of petroleum hydrocarbons to the dispersed nurdles. In addition to petroleum hydrocarbons, heavy metals and organochlorine pollutants have been continuously detected in Corpus Christi and Nueces Bays since 1976 (Custer et al., 1998; Stunz et al., 2022; White et al., 1980), and these pollutants could also be absorbed by nurdles. Once the contaminated nurdles are ingested by marine organisms, the pollutants can be assimilated and accumulated in the organisms (Besseling et al., 2013; Jiang et al., 2019; Ma et al., 2016), and further passed along the food chain to humans through seafood consumption (Smith et al., 2018), thus having detrimental effects to estuary ecosystems and public health. Pollutants on nurdles, including PAHs and PCBs, have been recently documented along the shoreline of Texas coasts (Jiang et al., 2021). However, there is very little information on the common pollutant levels on the vastly dispersed nurdles along the shorelines and inlands of the Coastal Bend region, and the detailed characteristics of nurdles, including size, polymer types and physical appearance, also remain unclear. The objectives of this study were to: (1) determine the polymer type, basic morphology, and adsorbed pollutants of nurdles collected within the Gulf beaches, bays, and adjacent terrestrial areas of the Coastal Bend, and (2) identify potential spatial trends of nurdles between near shore regions and shorelines.

2. Materials and methods

2.1. Nurdle sampling

Nurdles were collected from March to April 2021 at 24 sampling sites along railways and shorelines within bays and Gulf beaches around the Coastal Bend region in south Texas (Fig. 1). The geographic coordinates of the study sites are listed in Table 1.

The sampling methods employed in this study were based on previous publications (Jiang et al., 2021; Tunnell et al., 2020), and the experience of the team with the nurdles. At each sampling site, nurdles were collected in a sampling area of ca. 2 m × 5 m from the nearest/first high tide line (wrack line) for the shoreline region, and 1 m × 10 m from the thin layer of sands or rocks near the railway junction for the railway region, using dichloromethane-rinsed forceps, stored in 25 mL pre-combusted amber glass bottles at 4 °C in a cooler with ice in the field, and stored in a 4 °C

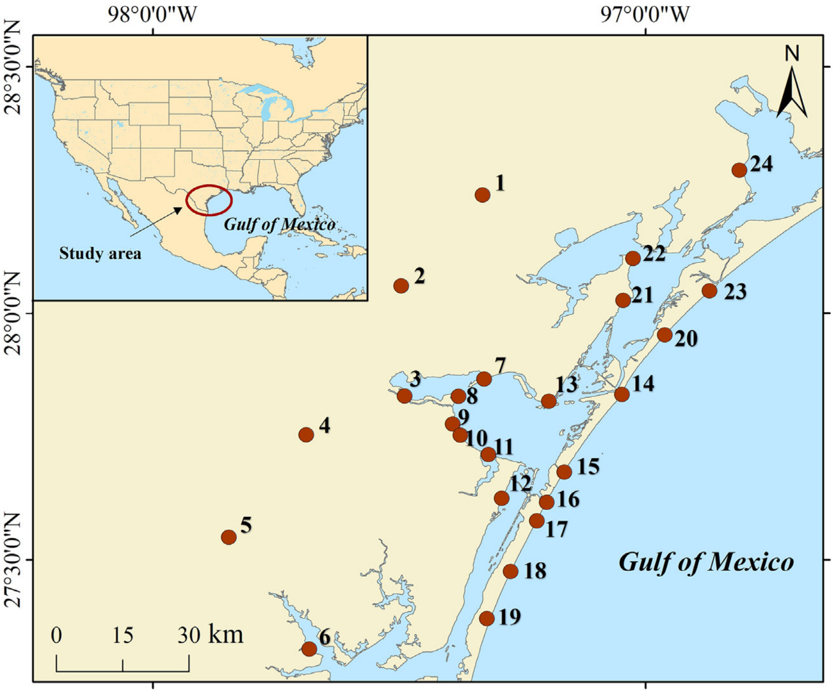

Fig. 1. Locations of sampling sites, as shown by red points on the map. The sampling sites are: 1. Woodsboro, 2. Sinton, 3. Nueces Bay, 4. Robstown, 5. Kingsville, 6. Baffin Bay, 7. Portland, 8. North Beach, 9. Cole Park, 10. Ropes Park, 11. TAMUCC beach, 12. Laguna Road, 13. Ingleside, 14. Port Aransas, 15. Mustang Island State Park, 16. North Packery, 17. Bob Hall Pier, 18. PINS- Malaquite Beach, 19. PINS- South, 20. San Jose Island, 21. Rockport, 22. Blackjack Peninsula, 23. Cedar Bayou, 24. Aransas National Wildlife Refuge (ANWR).

Table 1
Sample information of nurdles collected from the South Texas region.

Sites	Locations	Latitude	Longitude	No. nurdles
1	Woodsboro	28.240689	−97.330941	419
2	Sinton	28.056224	−97.496571	455
3	Nueces Bay	27.833068	−97.489715	276
4	Robstown	27.7543564	−97.689059	90
5	Kingsville	27.546299	−97.845983	309
6	Baffin Bay	27.31919	−97.682876	5
7	Portland	27.867021	−97.32781	298
8	North Beach	27.832366	−97.380179	322
9	Cole Park	27.776108	−97.391957	310
10	Ropes Park	27.753253	−97.376146	304
11	TAMUCC beach	27.714255	−97.319601	311
12	Laguna Road	27.625033	−97.292409	283
13	Ingleside	27.822392	−97.196418	297
14	Port Aransas	27.836152	−97.048402	306
15	Mustang Island State Park	27.67814	−97.165437	359
16	North Packery	27.616833	−97.20124	413
17	Bob Hall Pier	27.579582	−97.221391	433
18	PINS- Malaquite Beach	27.477228	−97.274093	518
19	PINS- South	27.38142	−97.322669	391
20	San Jose Island	27.95699	−96.961544	270
21	Rockport	28.026518	−97.046215	293
22	Blackjack Peninsula	28.111984	−97.025666	39
23	Cedar Bayou	28.046222	−96.870551	300
24	ANWR	28.290624	−96.810041	336

refrigerator in dark upon returning to the lab within the same day. The goal was to collect enough nurdles (~300) at each site for the chemical analysis, but when the number of nurdles was not enough, the sampling areas were expanded. Photos of all samples collected, including a picture of each container used during a sampling event, with all nurdles collected and a visible label, were taken at each site using the app "Theodolite" which stamps photos with geographic coordinates, time, date, and direction (Fig. S1).

2.2. Nurdle characterization

In the laboratory, nurdles at each sampling site were transferred into a pre-combusted beaker with 500 mL ultrapure water and cleaned in an ultrasonic bath for 30 min. This cleaning process was repeated 3 times. The cleaned nurdles were then dried in pre-cleaned glass bottles and stored in a desiccator with silica gel before further analysis.

The shapes, colors, and weathering patterns of nurdles at each sampling site were recorded and measured. To identify the polymer composition, 30 of the total nurdles at each sampling site were randomly selected and analyzed using Fourier Trans-form Infrared Spectrometry (FTIR, AIM-9000 FTIR Microscope, Shimadzu). The polymer type of a given nurdle was identified by comparing the sample spectrum with standard spectrum from the library (LabSolutions IR Software, Shimadzu), as well as through comparisons with NIST standards. When comparing samples to the library, the match acceptance criteria ranged from 650 to 800 score (1000 as the highest match score), depending on the algorithm used to examine the sample. After characterization, nurdles from each site were transferred into 25 mL pre-combusted glass bottles and stored at 4 °C in the dark, until further analysis.

The nurdles identified as polyethylene were further used to evaluate the weathering degree. Specifically, the oxidation index for each nurdle was calculated as the sum of all four indices (Brandon et al., 2016), including hydroxyl index (R-OH index), carbonyl index (C═O index), carbon-oxygen index (C—O index), and double bond index (C═C index) based on its IR spectrum. Specific indices were calculated as the ratio of the maximum peak absorbance (specific bond) to the value of a reference peak (polyethylene) as follows:

$$R-OH\ index = {}^{A_{3300-3400}}\!/_{A_{2908-2920}}$$

$$C-O\ index = {}^{A_{1000-1200}}\!/_{A_{2908-2920}}$$

$$C=O\ index = {}^{A_{885-925}}\!/_{A_{2908-2920}}$$

$$C=C\ index = {}^{A_{1550-1810}}\!/_{A_{2908-2920}}$$

where A stands for the absorbance, and the subscript numbers stand for wave numbers (cm^{-1}).

2.3. Chemical analysis

For the chemicals associated with nurdles, the EPA 16 priority polycyclic aromatic hydrocarbons (PAHs) were analyzed, including naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, benzo[*g*,*h*,*i*]perylene, indeno[1,2,3-c,d]pyrene, and dibenz[*a*,*h*]anthracene. Also analyzed were 7 common commercialized polychlorinated biphenyls (PCBs) aroclors, including aroclor 1016, 1221, 1232, 1242, 1248, 1254 and 1260, and 1 typical heavy metal-mercury. Three empty 25 mL pre-combusted amber glass bottles were carried along the sampling process to serve as control blanks for chemical analysis. For each group of associated chemicals (i.e., PAHs, PCBs, and mercury), 30 nurdles (~0.5 g) were randomly selected to constitute a composite sample representing the total nurdles collected at each site, and triplicate batches of nurdles from each site for each chemical were selected and sent to a National Environmental Laboratory Accreditation Program accredited commercial Lab (A & B Environmental Services) for analysis. Concentrations of the selected chemicals were measured according to EPA methodologies (i.e., EPA 8270D, EPA 8082A, and EPA 7470A) with strict quality controls. Briefly, for PAHs and PCBs, around 0.5 g of nurdles of each composite sample with different surrogate standards (nitrobenzene-d_5, 2-fluorobiphenyl, 2,4,6-tribromophenol, *p*-terphenyl-d14, and phenol-d6 for PAHs and tetrachloro-m-xylene and decachlorobiphenyl for PCB aroclors) were extracted using 1:1 hexane: acetone under microwave extraction at 100–115 °C for 20 min with the pressure of 100 psi, and the extracts were then analyzed after cooling down to the room temperature. For mercury, around 0.5 g of nurdles of each composite sample was added to 100 mL mill-Q water and then mixed with 5 mL of H_2SO_4, 2.5 mL of concentrated HNO_3 and 15 mL of potassium permanganate solution. After mixing for 15 min, 8 mL of potassium persulfate was added, and the bottle was heated for 2 h in a water bath maintained at 95 °C. After the bottle was cooled to room temperature, 6 mL of sodium chloride-hydroxylamine sulfate was added and set for 30 s. Finally, 5 mL of stannous sulfate was added and the bottle was attached to the aeration apparatus, through which the mercury vapor was analyzed. The qualification and quantification of associated organic chemicals (i.e., PAHs and PCBs) were analyzed by gas chromatography–mass spectrometry (GC/MS) and mercury by cold vapor atomic fluorescence spectroscopy (CVAFS). The detection limit of PAHs was 50 ng/g-pellets for all individual PAH, PCBs was 1.67 ng/g-pellets for all individual aroclor, and mercury was 1 ng/g-pellets. The final concentrations of all target compounds were corrected by the measured recoveries (90–110 %).

3. Results

3.1. Nurdle characterization

A total of 24 sites were sampled (Fig. 1 and Fig. S2), with 4 sites next to railway stations (sites 1, 2, 4, 5), 8 along with the barrier islands (sites 14, 15, 16, 17, 18, 19, 20, 23), and the rest 12 in the inner bay area (sites 3, 6, 7, 8, 9, 10, 11, 12, 13, 21, 22, 24). Nurdles were sampled near the railway stations or from the wrack line for the bay and barrier island beaches using solvent-rinsed forceps. Among all the sites, an insufficient amount of nurdles for chemical analysis was collected at 3 of 24 sites due to their low abundance. Specifically, only 5 nurdles were found at the Baffin Bay site (site 6), and 39 and 90 were found at the Blackjack Peninsula site (site 22) and the Robstown site (site 4), respectively. For the other sites, 270–518 nurdles were collected per site (Table 1).

Among all the sites except for the Baffin Bay site, 30 nurdles were randomly chosen to identify the polymer type by FTIR (Fig. S3). Among the 690 nurdles tested, 566 nurdles were made with PE, followed by 70 with PP, 23 with polyester (PES), 16 with polystyrene (PS), 8 with polyethylene-vinyl acetate (EVA), 6 with polystyrene-*co*-acrylonitrile (SA), and 1 with polyvinyl chloride (PVC). The polymer composition of nurdles from different sites is shown in Fig. 2. For all the sites except for the Woodsboro and Sinton sites, the nurdles were dominated by PE, followed by PP, comprising greater than 70 % and 10 % of the samples, respectively. Nurdles from the Woodsboro site (site 1) were dominated by PES (63.3 %), followed by PS (13.3 %) and SA (13.3 %), whereas nurdles from the Sinton site (site 3) were dominated by PE (43.3 %), followed by PS (30 %), PES (13.3 %), and PP (13.3 %).

In addition to the polymer identification, the morphology of nurdles, including shape, color and weathering patterns, was also recorded and measured. The photo collection of nurdles samples from all the sites is shown in Figs. 3 & S2. Nurdles from the railway sites (sites 1, 2, 4, and 5) differed greatly from those of the bay and barrier island beaches (all the other sites). Nurdles from the railway sites had the unique colors of pure white, green, grey, black, and blue, indicating various manufacture batches. However, nurdles from the bay and barrier island beaches had common colors from clear to yellow, orange, or dark brown (Fig. S3), which are signals of discoloration due to natural weathering (Endo et al., 2005; Jiang et al., 2021). Some of the beach nurdles had more or less black or green flecks, which indicated biofouling on these nurdles due to the constant contact with water.

Weathering-related functional groups were observed from the IR spectra of PE nurdles, including R-OH, carbonyl, C—O, and C═C groups. The C—O and C═O peaks were readily identifiable from the spectra, however, the C═C peak and R-OH peak were less distinctive. Oxidation indices of the PE nurdles varied between different sites, from 0.16 to 46.51 for railway nurdles and 0.02–127.98 for beach nurdles (average value, Fig. 4); from 0.46 to 1.36 for railway nurdles and 0.32–2.39 for beach nurdles (median, Fig. 4). There was no significant difference of the oxidation indices between railway nurdles and beach nurdles. However, the beach nurdles had a wider range of oxidation indices and thus weathering degrees than the railway nurdles.

3.2. Chemical concentration levels

The PCBs on the nurdles were not detectable at any sites, whereas PAHs and mercury were detectable at some sites (sites 3, 11, 14, 17 and 21 for PAHs and sites 3, 7, 9, 11, 12, 14, 16, 17, 19, 23 for mercury, Fig. 5). No targeted chemicals were detected on control samples.

The total concentrations of PAHs ($\Sigma - 16$ PAHs) in individual composite samples at the detected sites varied up to 2 orders of magnitude, ranging from 92.59 to 1787.23 ng/g-nurdle, with the highest concentration found in one composite sample from the Bob Hall Pier site (site 17). No PAHs had been detected at the railway sites, and all nurdles associated with PAHs were found at the beach sites.

The concentrations of mercury in individual composite samples at the detected sites ranged from 1.23 to 22.25 ng/g-nurdle, with the highest concentration at the Yarborough Pass site (site 23). Similar to PAHs, mercury was detected only on nurdles from the beach sites.

4. Discussion

4.1. Nurdle occurrence and distribution

The difference in the polymer compositions of all nurdles was dependent on sampling locations, railway vs. shoreline regions, based on the distribution data (Fig. 2). Once spilled, nurdles could enter the environment through different pathways. Sites Woodsboro and Sinton are located near the railway junctions, and the nurdles from these sites were commonly encountered in the grooves of rocks or buried in the thin layer of sands near the railway junction. The dispersive distribution of nurdles indicated the unintentional leakage rather than intentional spillage or waste disposal. The polymer composition of these railway nurdles was more diverse, including all the polymer types identified in this study, as compared to the beach nurdles dominated by PE and PP. This result indicated that the

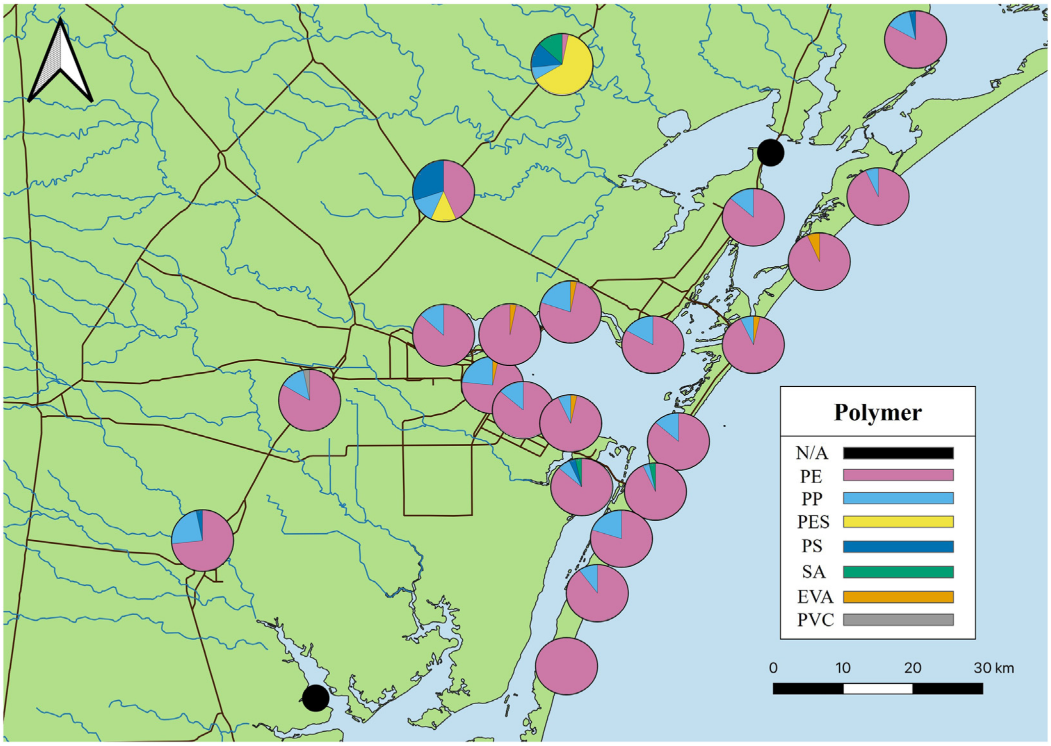

Fig. 2. Polymer composition of nurdles sampled from all the sampling sites, as shown by the pie graph in different colors. The whole black pie graphs represent lack of nurdles for FTIR analysis.

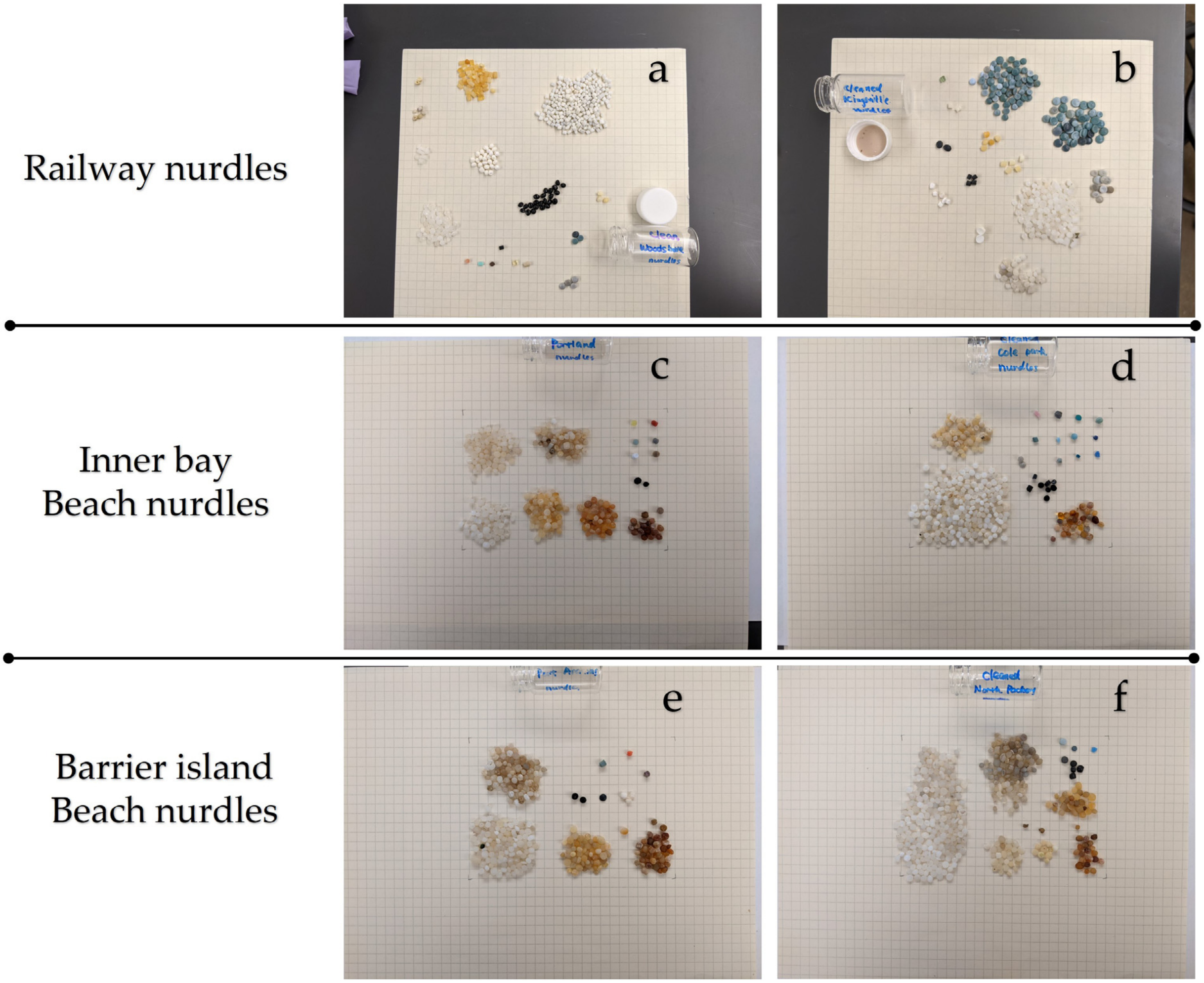

Fig. 3. Sorted nurdles at representative sites based on their colors and shapes, photographed on 6 mm*6 mm squared paper. The selected sampling sites are a. Site 1, Woodsboro; b. Site 5, Kingsville; c. Site 7, Portland; d. Site 9, Cole Park; e. Site 14, Port Aransas; f. Site 16, North Packery.

railway nurdles could be leaked during the transportation, loading and unloading, due to ineffective containers or transportation pipes, thus representing the diverse types of nurdles that are being produced and transported. However, nurdles collected from the bays and barrier island beaches were usually embedded within other floating debris, such as foamed plastic, string, wracks, and seagrass deposits, and the major component of beach nurdles were light density polymers, such as PE and PP, which could be floating more easily with runoff water and seawater. In other words, the drifting process may preferentially select light density nurdles, while the heavy ones may stay at the spots or sink to the sediment in streams, rivers and coastal oceans. This natural sorting process may further explain the floating plastic debris, dominated by PE or PP, in the open ocean garbage patch (Chen et al., 2018).

Beach nurdles were rarely discovered in sand dunes far from the shoreline, together with the fact that railway nurdles had never been found in the remote grasslands, suggested the negligible effect of wind on the nurdle transportation (Turner and Holmes, 2011). Instead, the beach nurdles likely have been transported and dispersed by runoffs and coastal currents. Similarly, Veerasingam et al. (2016) found that flood flushing played an important role in determining the seasonal variation of the most abundant PE and PP pellets along the Chennai coast. Hence, the source of beach nurdles is likely ocean-based, while the source of railway nurdles is land-based.

The drastic color differences between railway and beach nurdles may have been driven by the composition of the polymers and/or the residence time of nurdles in the environment. The small white nurdles that were only observed near the railways (i.e., sites 1 and 2) were made with PES (Fig. 3a), and some small grey nurdles were made with PS, and distinct green and blue nurdles which were also rarely found elsewhere. In addition to the color difference, PE and PP nurdles sampled in this study, similar in shape and size, had various extents of "darkening" or "cracking", suggesting that nurdles of similar manufacture origins may have undergone different degrees of weathering. Various degrees of "yellowing" or "darkening" of nurdles had been commonly observed in relevant studies (Endo et al., 2005; Jiang et al., 2021; Turner and Holmes, 2011; Veerasingam et al., 2016). For example, Turner and Holmes (Turner and Holmes, 2011) found plastic pellets with different degrees of yellowing on the island of Malta, and the yellowing degrees were associated with the increase of carbonyl index, which reflected the extent of aging. Natural weathering of nurdles usually occurs mainly due to photooxidation (Satoto et al., 1997), which leads to cross-linking or chain scissions of plastic backbone chains (Yamada et al., 2021), and produces oxygen-containing moieties such as carbonyl and ketone groups and cracks or grooves on the polymer surface (Jiang et al., 2021).

Furthermore, different indices have been used to evaluate the degradation/oxidation degree of polyolefin plastics, including hydroxyl, carbonyl, double bond, and carbon-oxygen index based on a FTIR spectrum (Brandon et al., 2016; Almond et al., 2020). In this study, no significant difference of oxidation indices was found between railway and beach nurdles, nor was the individual indices of hydroxyl, carbonyl, double bond, and carbon-oxygen among all the samples (Fig. 4). These indices

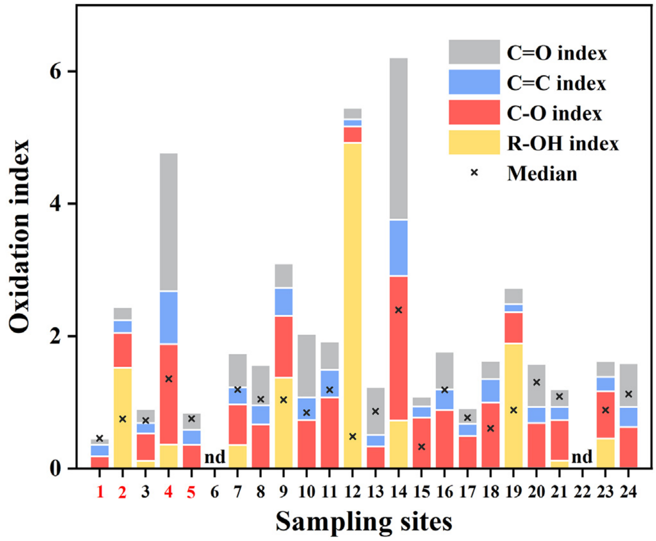

Fig. 4. Oxidation indices of PE nurdles from all sampling sites based on FTIR spectra, with median oxidation index of each site also plotted in corresponding column. The railway sites were marked in red (labels on the x-axis), and the shoreline beach sites were marked in black. The numbers of PE nurdles used for the index calculation were 1, 13, 24, 25, 22, 0, 23, 29, 23, 26, 27, 26, 25, 26, 17, 28, 23, 27, 30, 29, 23, 0, 28, 25 from site 1 to site 24, respectively.

varied greatly from site to site, indicating that individual nurdles might have experienced different weathering pathways and/or been spilled at different times at a given site (Tian et al., 2019).

Overall, nurdles collected in this study showed different yellowing status (Fig. 3) and different oxidation indices (Fig. 4), reflective of their different residence times in the environment, which all suggested that they were released to the environment from different times and likely different locations as well. In addition, based on the field observation (Gregory, 1983), fresh stranded plastic pellets had less embrittlement or degradation compared to the longer time stranded pellets, which indicated that the weathering/aging of beach stranded plastic pellets occurred mainly after settling down rather than during floating. Thus, given the fact that beach nurdles showed different oxidation indices, as well as "yellower" general colors than the railway ones, they could have been released into the environment for a much longer time period. Moreover, considering the different colors (white, yellow for beach nurdles vs. black, blue, and green for railway nurdles) and polymer compositions of both the railway and beach nurdles, it is clear that nurdles in this study were sourced from multiple spills/manufacturers, although the exact sources or the number of spills of the nurdles remained unknown. Further research is needed to identify how the nurdles are spilled and once spilled, how they are sorted and washed to the ocean. This information may be important for management and policy-making perspectives.

4.2. Chemical concentration levels

4.2.1. PCBs

Although PCBs have been commonly detected on beach plastic pellets (Endo et al., 2005; Hirai et al., 2011; Jiang et al., 2021; Ogata et al., 2009; Zhang et al., 2015), their concentrations are higher in heavily weathered nurdles with darker colors than those lightly weathered ones (Antunes et al., 2013; Endo et al., 2005). However, PCBs were not detected in this study. Jiang et al. (2021) measured PCBs on nurdles from Texas shorelines, which included some areas of this study, and their results showed that PCBs on nurdles were often undetectable, and the highest concentration was found at the Galveston Bay site, where the PCBs pollution is a known legacy problem (Oziolor et al., 2018; Sericano et al., 1995). In contrast, the sampling sites in this study focused on bays and estuaries in south Texas and did not include Galveston Bay. Although PCBs were legacy organic pollutants of the Gulf of Mexico, the detection of PCBs from the environmental matrices, such as water and sediment, in the ambient environment of this study region was rare, which suggests that PCBs concentrations are low in this region.

4.2.2. PAHs

Compared to other relevant studies, detection of PAHs on nurdles collected in this study were more erratic. For example, Jiang et al. (2021) investigated concentrations of PAHs on nurdles collected from the beaches of south Texas. They detected PAHs on all composite samples of nurdles at all sites, with concentrations ranging from 5 to 2000 ng/g-nurdle, in a similar concentration level to this work. Large piece-to-piece variability was also found for the total content of organic contaminants in previous studies (Hirai et al., 2011; León et al., 2019).

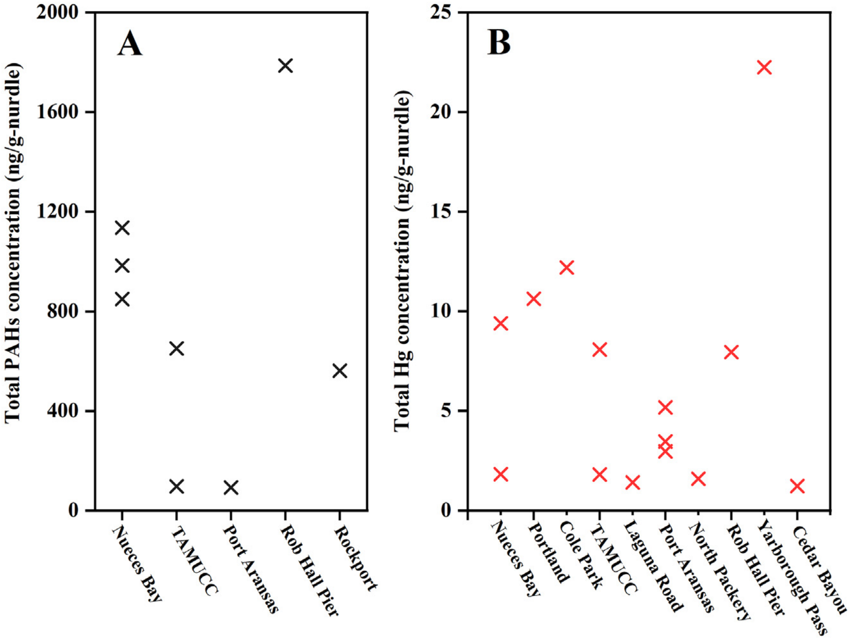

Fig. 5. Total concentration of PAHs (A) and Mercury (B) in nurdles from all the detected sampling sites (see Fig. 1 for the exact locations).

The composition of PAHs has been commonly used to indicate the source of PAHs, i.e., distinguishing petrogenic (e.g., oil spills) from pyrogenic sources (e.g., vehicle exhaust and incomplete combustion; Budzinski et al., 1997). Generally, PAHs with 2–3 rings (low molecular weight) are derived from petrogenic sources and those with 4–6 rings (high molecular weight) are from pyrogenic sources. In this study, high molecular weight PAHs (4–6 benzene rings) were dominant (65 % – 100 % of total PAHs) at all detected sites except for the Rockport site (site 21), suggesting that combustion of fossil fuel was the primary source of PAHs on these nurdles. This may also be related to the fact that high molecular weight PAHs are more hydrophobic, thus more likely being adsorbed to hydrophobic nurdles. At the Rockport site (site 21), however, only the 2-ring PAH, naphthalene, was detected, suggesting a different source of PAHs on nurdles at this site. It was commonly observed that beach plastic pellets were contaminated by tarry residues (Minchin, 1996; Shiber, 1987; Turner and Holmes, 2011), either embedded in tar deposits or stuck by small tar spheres. These tar residues are likely derived from the deposition and fragmentation of oil spills or natural seeps, and the discharge of bunker oil from ships (Herrera et al., 2018). Thus, nurdles from the Rockport site might also be contaminated by tarry residues, which might contain naphthalene.

All the detected PAHs were from beach nurdles, but not the railway ones. This result could be related to their weathering status or residence time mentioned previously. Weathering of nurdles increased their surface area due to the weathering-generated cracks or grooves, which enhance the sorption of organic pollutants, such as PAHs (Fisner et al., 2017; Jiang et al., 2021; Liu et al., 2018; Liu et al., 2019b). In addition, weathered nurdles are often fouled (Endo et al., 2005). Foulant such as biogenic organic matter or tarry residues could also contribute to the sorption of organic pollutants (Kuriyama et al., 2002). Furthermore, weathered nurdles tend to have longer residence times in the environment, which will increase the exposure to and chances of absorbing pollutants (Conkle et al., 2018; Jiang et al., 2021; Liu et al., 2019a), and thus might explain the higher concentrations of sorbed organic pollutants (Mato et al., 2001). In contrast, land-based railway nurdles were much less weathered than the ocean-based beach nurdles, and these nurdles did not have much contact with natural waters that always contain a certain level of PAHs. These factors might explain the results that PAHs were not detectable on the railway nurdles.

4.2.3. Mercury

Compared to other relevant studies, the concentration range of mercury on nurdles in this study was in the same magnitude as other studies, although the detection of mercury was erratic. For example, Conkle et al. (2018) found that the total mercury concentrations ranged from 2 to 14 ng/g-pellets for plastic from south Texas beaches. The total mercury concentration in sediment samples collected at the Nueces Bay during 2006 was 35 ng/g (Apeti et al., 2012), levels similar to this study. These results indicated that nurdles may accumulate mercury from the ambient environment. Coincidentally, relevant studies also revealed higher concentrations of mercury on weathered nurdles. For example, Acosta-Coley et al. (2019) tracked mercury concentration on plastic samples from the Caribbean and found greater mercury levels in white-degraded and black plastic pellets. Santos-Echeandía et al. (2020) also demonstrated the higher mercury concentrations on more degraded beach plastic from the Spanish Mediterranean coast. One possible explanation for the mercury accumulation is that photooxidation and attrition created more surface area and porosity on nurdles, which promote the attachment of organic matter or charged materials and thus facilitate a higher affinity for metals (Turner et al., 2015). In addition, black nurdles were also commonly found to contain various metals, especially mercury (Acosta-Coley et al., 2019; Turner et al., 2019), these black nurdles could be manufactured in special intention, for example, bio-beads (Turner et al., 2019), and mercury could be used as catalyzer or as an antimicrobial agent (Acosta-Coley et al., 2019). In our study, pure black nurdles were also sampled in several beach sites, which could affect the mercury detection on nurdles from these sites. Further work is needed to specifically focus on black nurdles for mercury analysis in order to elucidate the contamination source.

In general, neither the mercury nor the PAHs was constantly detected from all the nurdle samples, with ca. 20 % of total nurdles containing mercury, and ca. 12 % of the total nurdles containing PAHs. Since the nurdles were randomly chosen to form a composite sample for analysis and were not sorted by their weathering status, the highest concentration of pollutants detected could be contributed by heavily contaminated nurdles. It should also be noted that concentrations of contaminants on nurdles in the field could be higher than what were reported in this work. The nurdles were pretreated with ultrasonic rinsing in order to get rid of the attached natural organic matter or biofilm, which may also contain sorbed contaminants. Some of the samples with undetectable levels of PAHs could be due to the high detection limits of the EPA methods we adopted relative to relevant studies (e.g., Zhang et al., 2015; Chen et al., 2018; Jiang et al., 2021). Regardless, more than one composite samples from the Nueces Bay site, TAMUCC beach site, and Port Aransas site contained both PAHs and mercury, suggesting that the nurdles from these three sites could be commonly contaminated. Given the fact that the sorption of pollutants on plastics might be reversible (Endo et al., 2013; Jiang et al., 2021; Liu et al., 2019b; Liu et al., 2018), nurdles, especially the contaminated nurdles, could release pollutants to the ambient environments or be digested by marine organisms, which may lead to chronic toxicity or bioaccumulation in a long term.

5. Conclusion

In this study, we investigated the polymer type, morphology, and concentrations of associated PAHs, PCBs, and mercury on nurdles collected from the shorelines of the Coastal Bend. The polymer compositions of nurdles were dominated by PE and PP, but varied from site to site, and PES, PS, SA, and PVC nurdles were also identified. Nurdles stranded on the railways and beaches were sourced from multiple spills/manufacturers. The concentrations of pollutants on nurdles varied greatly between the sites. PAHs and mercury on nurdles were generally detectable except for a few sites, whereas PCBs were not detectable. Although the concentrations of pollutants were not in high toxic effect levels, the pollution status of the widespread nurdles and their associated nurdles is still of concern. There is a need to develop suitable and standardized protocols to investigate the risk of nurdles and to regulate the spillage of nurdles.

CRediT authorship contribution statement

Conceptualization, Xiangtao Jiang, Zhanfei Liu, and Jace W. Tunnell;
Methodology, Xiangtao Jiang;
Investigation, Xiangtao Jiang, Niki Conner, Kaijun Lu, Jace W. Tunnell, and Zhanfei Liu;
Resources, Zhanfei Liu, Jace W. Tunnell;
Data curation, Xiangtao Jiang, Niki Conner;
Writing-original draft preparation, Xiangtao Jiang.;
Writing-review and editing, Kaijun Lu, Jace W. Tunnell, and Zhanfei Liu;
Visualization, Xiangtao Jiang, Niki Conner, Jace W. Tunnell;
Supervision, Zhanfei Liu;
Project administration, Zhanfei Liu, Jace W. Tunnell;
Funding acquisition, Zhanfei Liu, Jace W. Tunnell.

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Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.156826.

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