

Marine Debris Polymers on Main Hawaiian Island Beaches, Sea Surface, and Seafloor

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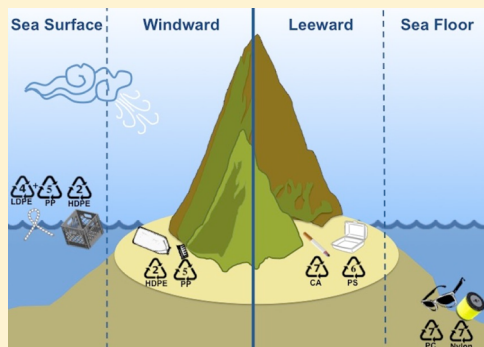
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Supporting Information

ABSTRACT: Polymeric differences of plastic debris were assessed across four compartments of the Main Hawaiian Islands (sea surface, windward beaches, leeward beaches, and seafloor) to better describe sources and fate. Plastic debris pieces ($n = 4671$) were collected from 11 beaches, three sea surface tows, and three seafloor dives. Fourier transform infrared spectroscopy identified the polymers of 3551 pieces. Significant differences ($p < 0.05$) in concentration, types, polymer composition, and weathering were found among four compartments. Windward beaches had 1–2 orders of magnitude more plastic pollution (g/m^2) than leeward beaches, despite smaller human populations on windward sides. Sea surface and windward beaches were dominated by severely weathered, less dense floating polymers (polyethylene and polypropylene comprised 92.7 and 93.5% on average, respectively, of the total debris mass), while leeward beaches and the seafloor debris consisted of less weathered and more dense sinking polymers (e.g., 41.0 and 44.7% of total mass consisted of the sum of polystyrene, nylon, cellulose acetate, polyethylene terephthalate, and additive-masked debris). These results are some of the first to provide evidence of polymeric stratification in the marine environment and emphasize that the majority of marine debris in Hawaii is floating in from distant sources rather than from Hawaii's residents or tourists.



INTRODUCTION

Plastic marine debris has received increased international attention.^{1–4} The Hawaiian Islands, one of the most remote archipelagos with high rates of endemism and endangered species,^{5,6} accumulate some of the highest reported amounts of marine debris.^{7–10} Hawaii is located south of the Subtropical Convergence Zone (STCZ) and southwest of the Eastern North Pacific Garbage Patch, where the highest concentration of floating plastic pollution on the planet accumulates because of wind-driven convergence.^{7,11,12} The Northeasterly trade winds are speculated to be the main driving force pushing floating marine debris from these accumulation areas to Hawaii.^{13,14}

Since Hawaii accumulates debris from a variety of sources, understanding the chemical composition of plastic marine debris is necessary.¹⁵ Seven standardized resin codes are assigned to the most commonly produced polymers:¹⁶ polyethylene terephthalate (PET, #1), high-density polyethylene (HDPE, #2), polyvinyl chloride (PVC, #3), low-density polyethylene [LDPE, #4, which includes linear low-

density polyethylene (LLDPE)], polypropylene (PP, #5), polystyrene (PS, #6), and other polymers (#7). Some consumer goods are stamped with their resin code, but weathered fragments are often missing these stamps, requiring chemical analyses for identification.

Polymer identification of plastic marine debris is crucial for understanding sources, fate, transport, and effects in the environment. Because different polymers have various chemical structures, their physical, chemical, and biological interactions within the environment will differ. Sorption rates and concentrations of organic and heavy metal pollutants vary among polymers, making certain polymers a greater threat of contaminant exposure to organisms.¹⁷ Chemical reactions during environmental degradation processes can lead to various polymeric degradation products that have not been

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widely studied.^{18–23} The release of additives, fillers, and greenhouse gases^{21,24} are highly variable among polymer type and in some cases even toxic.^{25,26} Polymer identification tools also provide indicators of the extent of the debris weathering, a sign of aging or possibly a time estimate since littering.^{20,27} Each polymer has a different chemical density, which is hypothesized to be a major (but not the only) influence in vertical stratification and fate of plastic debris in the ocean (Table 1).^{28,29} For instance, polymers less dense than seawater

Table 1. Chemical Densities of Pure Polymers^a

polymers	abbreviations	resin code	density (g/mL)
polypropylene	PP	5	0.85–0.92
low-density polyethylene	LDPE	4	0.89–0.93
ethylene-vinyl acetate copolymer	EVA	7	0.92–0.95
high-density polyethylene	HDPE	2	0.94–0.98
polystyrene	PS	6	1.04–1.06
acrylonitrile butadiene styrene	ABS	7	1.04–1.08
nylon (polyamide)	PA	7	1.12–1.24
poly(methyl methacrylate)	PMMA	7	1.14–1.20
polycarbonate	PC	7	1.20–1.22
cellulose acetate	CA	7	1.28–1.31
polyethylene terephthalate ^b	PET	1	1.37–1.41
poly(vinyl chloride)	PVC	3	1.38–1.45

^aWithout modification from additives, production, and degradation, plastic debris composed of polymers denser than seawater (1.020–1.029 g/mL) would sink, while less dense polymers will float on the ocean, indicated by the line. Densities were compiled from scientific literature or manufacturer web sites.^{38–42} ^bPolyester materials are often PET.

(e.g., PE and PP) float and are commonly found at the sea surface,^{30–34} while denser polymers predominantly sink to the seafloor.^{29,35,36} In addition, polymer identification can confirm that debris samples are in fact plastic and other material is not visually mistaken as plastic.³⁷ These reasons, plus the need to understand which polymers may affect different marine habitats, provided justification for the present study.

The goals of this study were to identify the polymers of plastic marine debris throughout the Main Hawaiian Islands (MHI), determine if vertical stratification was influenced by polymer density, and determine if the polymer composition was different among multiple environmental compartments: sea surface waters, beach surface sediment, and seafloor surface sediment in the photic zone. Beaches were subclassified into those with direct exposure to on-shore winds (windward) and those without (leeward). Five studies in Hawaii have been conducted for polymer identification of debris, but they had smaller sample sizes ($n \leq 248$ pieces) and focused on up to two beaches or one near-shore region.^{34,43–46} This study is the first to assess sites on six of the eight MHI, plus sea surface and seafloor sites in surrounding waters. Additionally, few empirical studies elsewhere have investigated vertical stratification of polymers in the ocean,^{47–49} and these focused on microplastics rather than the larger debris measured in the current study.

MATERIALS AND METHODS

Sample Collection. Beach sites ($n = 11$) were sampled between April and August 2017. Both windward and leeward beaches on Oahu were selected. Beaches on other islands were

opportunistic or commonly targeted for beach cleanups (Figure 1, Table S1).

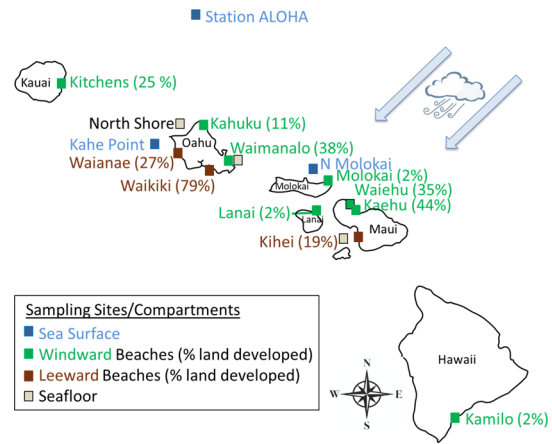


Figure 1. Sampling sites in the MHI ($n = 17$). Percentages after beach names indicate the percent of land development.

Beaches were sampled in triplicate transects parallel to the shoreline along the drift line. All three transects per beach were sampled on the same day at predetermined locations at $\approx 1/4$, half-way, and $3/4$ along the length of each beach. The rectangular-shaped transects varied in size. The transects were at least $1 \text{ m} \times 1 \text{ m}$ or covered an area where ≥ 50 pieces of plastic could be expected for sufficient polymer composition data. All plastic pieces lying on the sand surface that had a dimension $\geq 1 \text{ cm}$ inside the transect were collected. If < 50 pieces were obtained, the transect dimensions were increased to provide ample data for polymer composition. Rock boundaries prevented this for Waianae transect 1. The GPS locations were recorded. The transect length and width were measured to calculate the surface area sampled. Our sampling design did not follow the NOAA standardized 100 m protocol,⁵⁰ but we believe that it provided representative samples of plastic debris to determine polymer composition on the beaches.

Three sea surface trawls were performed on the R/V Kilo Moana using a $333 \mu\text{m}$ mesh size manta net with a 90 cm opening and an attached flow meter. The net was deployed with a Dynacon U-Frame designed to handle 13 608 kg towing load, 9072 kg of luffing load, and reach 3.7 m beyond the transom. The trawling speed was maintained between 0.514 and 1.02 m/s and the tow duration was typically 25 min, but varied because of environmental conditions. Dates and coordinates for Station ALOHA $\approx 100 \text{ km}$ north of Oahu, Station Kahe $\approx 10 \text{ km}$ west of Oahu, and a site $\approx 10 \text{ km}$ north of Molokai are provided in Table S1 and Figure 1. All visible plastic pieces from the tow, including those $< 1 \text{ cm}$, were collected using metal forceps.

SCUBA divers collected visible seafloor debris $> 1 \text{ cm}$ from three locations between April and June 2018. Dive sites were near-shore Oahu and Maui with depths less than 30 m (Table S1, Figure 1).

Sample Preparation. Plastic pieces from beaches ($n = 3280$ total; 292 pieces from leeward beaches and 2988 from windward beaches), sea surface ($n = 55$), and seafloor ($n = 216$) were processed without a microscope. To minimize chemical manipulation, solvents were not used for cleaning.¹⁵ Instead, sand and biological matter were brushed off gently to

avoid fragmentation. Samples were air-dried and weighed to the nearest 0.00001 g. Debris items that were too large to be transported to the laboratory were weighed to the nearest 0.1 g, or estimated from dimensions, and subsampled for analysis.

Each plastic piece was categorized by type (fragment, sheet, foam, line, pellet, other, or whole),²⁸ color, longest measurable dimension, and a weathering intensity rank (1 = mild, 2 = moderate, and 3 = severe). These physical characteristics for each debris piece are provided in [Supporting Information Table S2](#). Photos of cataloged transects from each compartment are shown in [Figure S1](#). “Whole” pieces were recognizable consumer goods that did not fit into the other type categories (e.g., cigarette filters, toothbrushes, and bottle caps); monofilament fishing line, rope, or net materials were classified as “line”; “sheets” were food wrappers, bags, and films; “foams” were expanded cellular plastics (blown with air); “other” was a category primarily for fabrics; “pellets” were preproduction polymers (e.g., nurdles); and “fragments” were unidentifiable pieces that did not fit in the other categories. Multicomponent pieces (e.g., sunglasses) were disassembled; each component was counted, weighed, and analyzed separately. The weathering intensity rank was based on the degree of visual square fracturing and white oxidation on the surface of the plastic ([Figure S2](#)). Mild was no square fracturing. Moderate was minimal square fracturing and/or a thin layer of white oxidation on the surface. Severe was deeply embedded square fracturing and/or a thick layer of white oxidation. The weathering rank focused on chemical weathering from photo-oxidative degradation as opposed to mechanical weathering (e.g., abrasions and bite marks). This method was used as an alternative to carbonyl index (CI) analysis because CI analysis has only been applied to polyolefins.^{19,43,51}

Fourier Transform Infrared Spectroscopy (FTIR) Analysis. Plastic pieces ($n = 3551$) from all beach, sea surface, and seafloor sites were analyzed for polymer identification using a PerkinElmer attenuated total reflectance (ATR) FTIR Spectrometer Spectrum Two (Waltham, MA) with 4 cm^{-1} resolution and 40 accumulations. Spectra were collected in transmission mode in a spectral range of $4000\text{--}450\text{ cm}^{-1}$. The ATR FTIR diamond crystal was cleaned with isopropanol and a background spectrum was collected prior to every measurement. A clean, less weathered inner surface was revealed by a razor blade and applied to the crystal with a force between 80 and 100 N. If pieces crumbled, the powder was analyzed. For a braided line, inner threads were analyzed. All spectral data are included in [Table S3](#).

Polymer Identification. Polymers were identified manually from spectra as described previously¹⁵ for 17 polymers in our in-house spectral library. If confirmation was required or the sample could not be manually identified, spectral libraries installed with the PerkinElmer software were used only if the search score was ≥ 0.90 . For LDPE and HDPE differentiation, the presence/absence of a band at 1377 cm^{-1} was used;¹⁵ however, undifferentiated samples were classified as “Unknown PE” without using a float/sink test. “PE/PP mixture” were samples that produced spectra with both PE and PP transmittance bands as previously described.¹⁵ “Other PE” were samples that produced high-intensity PE transmittance bands along with low-intensity bands associated with other functional groups, such as chlorinated PE. “Other” was a grouping of rare polymers [latex, petroleum wax, acrylic, PP/PET mixture, polycarbonate, and poly(vinylidene fluoride)].

“Unidentifiable” spectra were too noisy to interpret or were suspected copolymers. Samples were categorized as “additive-masked” when spectra produced bands characteristic of additives, mostly phthalic acid esters, which masked the underlying base polymer. These samples were typically elastomers, which consist of large percentages of phthalate plasticizer mixed with a base polymer.⁵² All “additive-masked” samples were searched with spectral libraries and will be the subject of a forthcoming manuscript.

Polymer Composition Calculations. Three transects, from Kamilo, Kahuku, and Kitchens, had too many pieces to analyze. From each transect, 100 pieces were randomly selected for analysis. One seafloor sample, Makai Pier on Oahu, had too many pieces of tangled monofilament line to count or analyze. The total mass of bundled line was divided by the average mass of 10 entire individual line pieces separated from the bundle to estimate the total number of lines included. Fifty representative pieces were cut from the bundle to analyze using FTIR. The count and mass of nonselected pieces ([Table S4](#)) were accounted for in all the following calculations.

To calculate the polymer percentages at each transect, dive, or tow, the sum of mass or number of pieces of each polymer type was divided by the total mass or total number of pieces, respectively, and multiplied by 100. The percentage of each polymer at each of the three transects (dive or tow) per site was used to calculate the mean and one standard deviation (SD) per beach (or seafloor or sea surface compartments).

Land Use Calculations. The percent of land developed, as a proxy for population, was used to estimate the influence of human population at each beach ($n = 11$) on marine debris accumulation. Census numbers are not available at the scale of each beach. The percent of land developed was estimated by overlaying a clear transparency with 160 dots (10 rows of 16 0.6 cm diameter zero symbols) onto Google Earth aerial photographs printed at an altitude of 6.1 km to include approximately 27.4 km^2 of Earth’s surface and the whole beach ([Figure S3](#)). The middle transect at each site was located in the center of the printed map. The percent of land developed was calculated as the number of dots of developed land (e.g., man-made impervious surfaces such as roads and buildings) divided by the total number of dots that were land and multiplied by 100. Dots that were counted as water were excluded. If a dot included both developed land and another category, it was counted as developed land.

Statistical Analysis. To determine if plastic debris abundance differed among beach sites, JMP 12.1.0 (SAS Institute Inc., Cary, NC) software was used. All abundance data, by pieces/ m^2 and by grams/ m^2 , were log-transformed, tested for normality using a Shapiro–Wilk test, and tested for unequal variances using an O’Brien test. When assumptions were met, an analysis of variance (ANOVA) was used followed by Tukey’s Kramer post hoc tests. When assumptions were violated, Kruskal–Wallis and Wilcoxon rank-sum post hoc tests were used. Pearson correlations examined if percent land development was related to plastic debris abundance, weathering rank, debris types, and polymer composition (residuals were normal based on Shapiro–Wilk tests). Differences in debris type (in pieces only), weathering rank (in pieces only), and polymer composition (in pieces and mass) among all sampling sites ($n = 17$) were determined via multiresponse permutation procedures (MRPP) coupled with indicator species analyses (ISA) using PC-ORD 6.08 (Gleneden

Beach, Oregon) software. The same procedure was followed to analyze the differences across the four compartments: seafloor, leeward beaches, windward beaches, and sea surface. All statistical output files are included in the [Supporting Information](#) as appendices.

RESULTS AND DISCUSSION

Debris Abundance Greater on Windward than Leeward Beaches. Across 11 beaches, a total of 3931 plastic pieces were collected with a total mass of 20552.3 g. Mean ($\pm$ SD) plastic abundance levels ranged from 0.404 ($\pm$ 0.549) to 68.3 ($\pm$ 41.5) pieces per square meter and from 0.320 ($\pm$ 0.280) to 188 ($\pm$ 234) g/m². The overall averages were 18.1 ($\pm$ 22.9) piece/m² and 48.8 ($\pm$ 59.8) g/m². Kahuku, located on the northern windward coast of Oahu, had the highest plastic marine debris abundance (Figure 2). Kamilo, known to be one

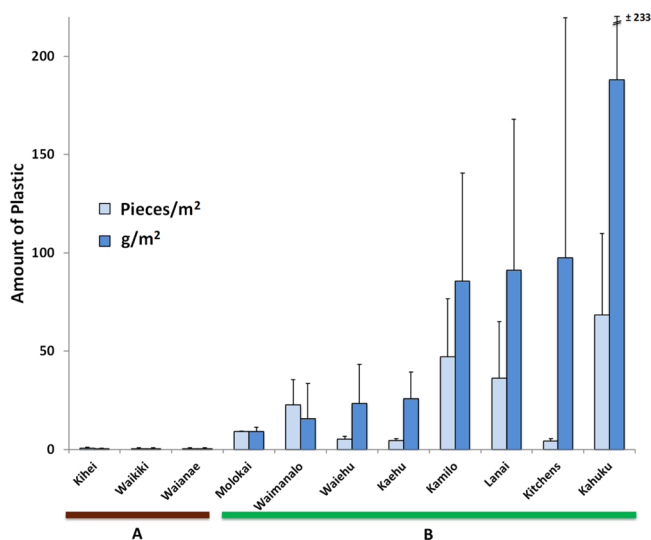


Figure 2. Plastic debris abundance (pieces $\geq$ 1 cm) on three leeward MHI beaches (brown line) is significantly less than that on eight windward beaches (green line) ($p < 0.0001$). Values are mean $\pm$ one SD.

of the worst plastic polluted beaches in the MHI, had lesser amounts than Kahuku. This unexpected trend was likely because of the predetermined sampling locations not overlapping with the most polluted portion of Kamilo beach (Figure S3a). All three leeward beaches had concentrations, <1 piece/m² or 1 g/m², 1–2 orders of magnitude lesser than windward beaches (Figure 2; ANOVA, $p < 0.0001$, [Supporting Information](#) Appendix 1). These field surveys on smaller-sized debris corroborate aerial surveys that found greater abundance of macro- to mega-sized debris on windward versus leeward beaches in the MHI.^{13,14}

Debris amounts are higher in the MHI than many other places. Ribic et al.¹⁰ reported that Oahu has higher debris loads than the US Pacific coast. MHI beaches sampled in the current study were more plastic polluted than South Korean beaches (means = 13.2 items/m² and 1.5 g/m² of 0.5–2.5 cm each)⁵³ even though they sampled additional particles in smaller size classes (<1 cm), which inflates their abundances compared to the current study. The current results are also 2 orders of magnitude greater than the North Atlantic Azores (0.62 pieces/m² of >2 cm) of a similar size range.⁵⁴ It is challenging to compare the present data with published debris abundances

on beaches because of the differences in particle sizes targeted. This emphasizes the need to report multiple measurements (piece counts, size distributions, and mass) to understand the type of debris in a region.

Physical Characteristics of Debris Differs Across Compartments. Spatial differences were found for debris types (MRPP, $p < 0.0001$). The differences among individual sites were numerous and complex but emphasized substantial windward versus leeward differences (Figure S4, Appendix 2). Interestingly, the debris types on windward Kaehu Beach was significantly different from all other windward beaches but similar to leeward beaches. We suspect that Kaehu debris is influenced by local littering. Abundant nylon stringtrimmer line may come from the adjacent golf course, while numerous expanded polystyrene foam food containers are trapped by the unique large quantities of logs on this beach.

When sites were grouped into the four compartments, whole items and line dominated the seafloor, while fragments were the most common debris type in the other three compartments (Figure 3). Windward beaches and the sea surface had similar

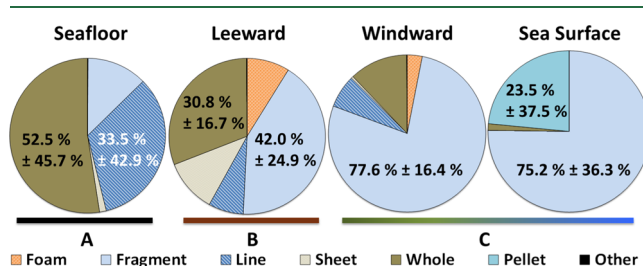


Figure 3. Types of MHI plastic marine debris sampled across compartments, percentages of pieces. The seafloor (A) and leeward beaches (B) are different from windward beaches and the sea surface (C) (MRPP, $p < 0.0001$). Values are mean $\pm$ one SD.

debris types and were different from leeward beaches and the seafloor (MRPP, $p < 0.0001$, Figure 3, Appendix 3). Using ISA, whole items significantly indicated (or influenced the compartment differences in) the composition of debris types on the seafloor, foam, and sheets on leeward beaches, fragments on windward beaches, and pellets on the sea surface ($p \leq 0.044$, Appendix 3). The most common whole items were sunglasses or dive masks (45%) and fishing lures (18%) on the seafloor, cigarette filters (48%) on leeward beaches, and oyster spacers (42%) on windward beaches. The sea surface had one whole item, a bottle cap. Because the debris types on windward beaches and the sea surface were similar, they likely receive inputs from a similar source (e.g., Eastern North Pacific Garbage Patch), whereas leeward beaches and the seafloor are receiving debris from local inputs and maritime activities, respectively.

All debris pieces were combined; the most abundant colors were white (42.3%), blue (14.0%), and black (12.0%) (Figure S5), similar to previous findings.^{51,55} The debris particle sizes were greater on the seafloor compared to beaches (Figure S6). The predominance of smaller plastic particles (1–1.9 cm) found on MHI beaches is consistent with global data¹¹ and is explained by fragmentation of larger items.

Greater Weathering on Sea Surface and Windward Beaches. Plastic fragmentation is of concern because of the release of potentially harmful substances and greenhouse gases, generation of nanoplastics that can permeate cell membranes, and increased affinity for contaminants because of increased

surface area.^{18,24,56} Debris on windward beaches and sea surface were more weathered than debris on leeward beaches and the seafloor (MRPP, $p < 0.0001$, Figure 4, Appendix 4).

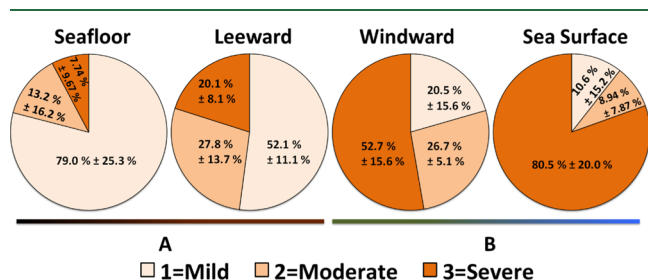


Figure 4. Weathering rank of MHI plastic marine debris across compartments, percentages of pieces. Debris on the seafloor and leeward beaches (A) are less weathered than windward beaches and the sea surface (B) (MRPP, $p < 0.0001$). Values are mean $\pm$ one SD.

Significant differences were also found among many of the individual sites (MRPP, $p < 0.0001$, Figure S7, Appendix 5). Weathering progressed from mild on the seafloor to severe on the sea surface. These results suggest that windward beaches and the sea surface may be at higher risk of negative effects resulting from fragmentation than the leeward beaches and seafloor. These data add insight into environmental exposure time and further support the hypothesis that most debris on windward beaches drift in from distant sources, whereas leeward beaches are presumably littered by local sources.

Polymer Composition Gradient from Sea Surface to Seafloor. Among individual sites, polymer composition differences were too complex to display but emphasized a substantial leeward versus windward divide (MRPP, $p < 0.0001$ by pieces and mass, Appendices 6 and 7, Figure S8). All four compartments had different polymer compositions by pieces (MRPP, $p < 0.0001$, Figure 5; Appendix 8). By mass, the

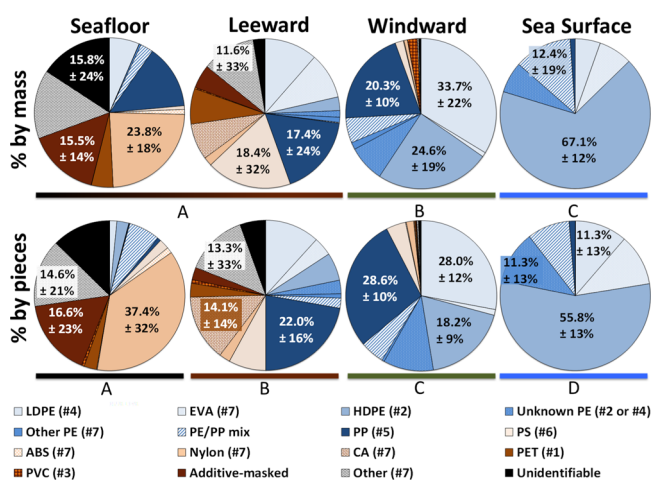


Figure 5. Comparison of MHI marine debris polymer composition across four compartments. Percentages are based on mass (top) and on the number of pieces (bottom). Compartments underlined with different letters are different from each other (MRPP $p < 0.0001$). Blue shades are floating polymers; brown and red shades are sinking polymers. Polymer abbreviations: low-density polyethylene (LDPE), ethylene vinyl acetate (EVA), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), acrylonitrile butadiene styrene (ABS), cellulose acetate (CA), polyethylene terephthalate (PET), and polyvinyl chloride (PVC). Values are mean $\pm$ one SD.

polymer composition on the seafloor and leeward beaches was similar, whereas the sea surface and windward beaches were different from all other compartments, including each other (MRPP, $p < 0.0001$, Figure 5, Appendix 9). The floating polymers (shown in blue shades) were in greater proportions on the sea surface and windward beaches, while the sinking polymers (shown in brown and red shades) were more frequent on leeward beaches and the seafloor. This trend supports our hypothesis, and the findings of a recent meta-analysis,²⁹ that the chemical density of the base polymer primarily drives the vertical stratification of plastic debris in the ocean. Specifically, HDPE dominated the sea surface, while LDPE by mass and PP by pieces dominated windward beaches. Leeward beaches were dominated by PS by mass and PP by pieces and the seafloor debris comprised mostly nylon. ISA revealed which polymers significantly indicated the compositional differences among compartments (Appendix 8 and 9). These were HDPE for sea surface; LDPE and PP for windward beaches; cellulose acetate (CA, cigarette filters) for leeward beaches; and nylon (e.g., monofilament fishing line), additive-masked (e.g., elastomeric fishing lures), and unidentifiable for seafloor. Only 1.4% of all pieces ($n = 48$) were unidentifiable, most found on the seafloor and leeward beaches.

Only polymers with chemical densities less than seawater (see Table 1) were found on the sea surface, consistent with global findings^{27,29–33,40,57–59} and with debris from the sea surface recently sampled near Maui County.³⁴ More than 90% of the samples found on windward beaches composed of floating polymers (LDPE, EVA, HDPE, Unknown PE, PE/PP mixture, and PP). Polymers denser than seawater comprised <10% (7.55% by pieces and 5.56% by mass) of windward samples. The most common sinking polymer by pieces was PS ($4.20 \pm 10.0\%$) and by mass was PVC ($2.02 \pm 7.77\%$), and most of these pieces contained air to keep the items floating. These results are consistent with Italian, Australian, Indian, and Portuguese beaches dominated by PE and PP.^{36,55,60,61} In previous MHI studies, PE (80%) and PP (20%) dominated Kauai's beaches,⁴³ and PE (85%), PP (14%), and PS or polyurethane (1%) were found at Kamilo.⁴⁵

More diverse polymers were found on leeward beaches. Less than half of the composition was composed of floating polymers (48.6% by pieces and 43.4% by mass), with PP being the most common floating polymer by pieces ($22.0 \pm 15.5\%$) and mass ($17.4 \pm 23.8\%$). The large proportion of PP was attributed to single-use consumer goods and food packaging items presumably left by beach-goers. Leeward beaches had a significantly greater proportion of sinking polymers compared to windward beaches (Wilcoxon, $p = 0.014$ by mass and $p = 0.025$ by pieces, Appendix 10). The most common sinking polymer by pieces was CA ($14.1 \pm 14.2\%$) as cigarette filters and by mass was PS ($18.4 \pm 32.4\%$) as expanded polystyrene (e.g., take-out containers). These results are not consistent with other studies, described above, that identified polymer composition on beaches. Only one other study in Singapore found a dominance of sinking polymers on beaches, with PS (40%) being the most abundant.³² Leeward beaches are less influenced by the Northeasterly trade winds that are speculated to push floating debris to the MHI. Thus, leeward beaches are not as susceptible to plastic pollution from the North Pacific Garbage Patch (e.g., floating polymers) as windward beaches. As a result, debris deposition on leeward beaches is explained by local littering on land, as there are limited physical

mechanisms to transport sinking plastics from the open ocean onto beaches.

Over 75% of the polymer composition on the seafloor, by both pieces and mass, was composed of sinking polymers. The less dense polymers found on the seafloor were heavily biofouled, which caused eventual deposition on the seafloor.⁶² The most common polymer found on the seafloor was nylon by both pieces ($37.4 \pm 31.9\%$) and mass ($23.8 \pm 17.8\%$) because of the large quantities of monofilament fishing line. Sinking polymers have dominated the seafloor in the Arctic (38% chlorinated PE and 22% polyamide)³⁵ and Antarctic (94.1% styrene–butadiene–styrene copolymers and nylon).³⁶ This emphasizes the need to fill the gap of missing plastic by analyzing the seafloor for plastic contamination.^{29,63} Many polymers are not being accounted for because most studies focus on the sea surface, which accumulates primarily PE and PP.

The seafloor had a greater portion of additive-masked items ($16.6 \pm 23.5\%$ by pieces; $15.5 \pm 14.0\%$ by mass) compared to other compartments. This category was the second most common category found on the seafloor. The spectra of these items were so dominated by phthalate bands that it was not possible to determine the base polymer with ATR FTIR alone (Figure S9). Additional testing is ongoing to better characterize these items, which will be the subject of a future manuscript. Additives, such as phthalates, have been reported in marine debris studies as a pollutant adsorbed onto the surface of plastic;⁶⁴ however, to our knowledge, there has never been a study that identified phthalates as the major structural component of plastic marine debris. Phthalates are commonly used as an additive in PVC comprising up to 30% mass fraction and can leach from plastic debris.⁶⁵ Most of the additive-masked samples were elastomers (e.g., fish lures) that are manufactured with >50% phthalates mass fraction.⁶⁶ More than 12 000 tons of fishing lures are deposited annually at the bottom of freshwater,⁶⁶ not counting lures lost in the ocean. Phthalates do not persist in the environment; however, they are well-known endocrine disruptors.⁶⁵ Additionally, there is evidence that concentrations in certain environments exceed concern thresholds.⁶⁵

Polymer Composition of Particular Types and Whole Items. The polymer composition of each debris type was examined (Figure 6). The dominant polymer(s) of fragments were PE and PP, foam was PS, line was nylon and PP, sheets were PP and LDPE, whole items were LDPE, pellets were HDPE, and the “other” category (e.g., fabric/fiber) was PET. Three of the seven types were dominated by PE, one of the most produced polymers in the United States.^{28,67} The overwhelming abundance of PE in the environment highlights the need to scrutinize the life cycle and improve the management of this polymer from production (most produced polymer⁶⁷), use (especially single-use products), and disposal (LDPE is not recycled in Hawaii).

Land Development Correlations with Marine Debris. Correlations between percent of coastal land developed and marine debris variables helped describe the potential influence of local population on debris found on the 11 beaches (Figure S10). The percent of land developed was weakly, insignificantly, and negatively correlated with the debris quantities by pieces/m² or g/m² (Figure S10A, Pearson $R^2 = 0.277$, $p = 0.096$ and $R^2 = 0.213$, $p = 0.153$, respectively). Regions with more land development had less debris. This contrasts from Brazil, where debris abundance decreased with distance from

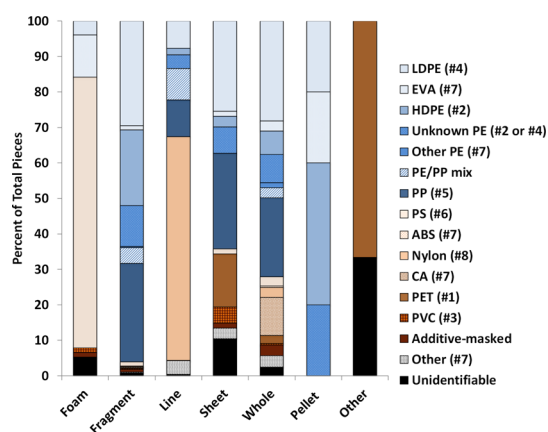


Figure 6. Polymer composition of different types of MHI plastic marine debris, calculated by pieces. Blue-shaded polymers float in seawater; brown-shaded polymers sink. Polymer abbreviations: low-density polyethylene (LDPE), ethylene vinyl acetate (EVA), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), acrylonitrile butadiene styrene (ABS), cellulose acetate (CA), polyethylene terephthalate (PET), and polyvinyl chloride (PVC).

urban centers.⁶⁸ These results further suggest that MHI beach debris, which is in largest abundance on the windward coasts, is primarily originating from nonlocal sources.

These correlations could be confounded by beach cleanups, but we believe that this possible confounder is a minor variable. Cleanup effort is undoubtedly higher on tourist beaches, such as Waikiki, but large-scale cleanup events are scheduled frequently for the less developed beaches. The exact timing of cleanup effort before our sampling was often unknown. Kahuku on windward Oahu has less land development, is located within the James Campbell National Wildlife Refuge, and received the largest debris amounts of all sampling sites.¹⁴ Portions of Kahuku are cleaned up approximately weekly to monthly. It was obvious that a recent cleanup had occurred at one of our three Kahuku transects. Still, Kahuku had the highest debris abundance, suggesting that recent cleanup had little impact on our overall findings.

Percent land development and weathering intensity showed a strong negative correlation (Figure S10B, Pearson $R^2 = 0.600$, $p = 0.0051$). Waikiki, the most developed, had the least weathered debris, suggesting that the small abundance of debris on this beach is from local sources with minimal exposure to environmental conditions. The least developed beaches (Kamilo, Lanai, and Molokai) had the most weathered debris. Weathering intensity for pieces exposed to sunlight could reflect environmental exposure time. The more weathered pieces on the sea surface and windward beaches were in the environment longer, arriving to Hawaii via wind and ocean currents from distant sources, compared to more recently littered debris on leeward beaches.

Types of debris were correlated with land development (Figure S10C). More fragments were found on less developed beaches (Pearson $R^2 = 0.362$, $p = 0.050$), while more sheets were found on more developed beaches (Pearson $R^2 = 0.443$, $p = 0.025$). Fragments are formed from mechanical and chemical weathering after extended environmental exposure. As such, the less developed windward beaches received debris dominated by fragments that were presumably washed ashore from older litter of distant sources, whereas sheets, mostly in

the form of food wrappers or bags, dominated leeward beaches likely as a result of recent local littering.

Polymer composition was correlated with percent land development (selected correlations are shown in Figure S10D,E, and all correlation statistics are given in Appendix 11). By mass, greater percentages of PET and PP were on more developed beaches (Pearson $R^2 = 0.577$, $p = 0.007$; and $R^2 = 0.441$, $p = 0.026$, respectively), while the opposite was found for HDPE (Pearson $R^2 = 0.458$, $p = 0.0227$). Proportionally by pieces, PVC and EVA were more dominant on more developed beaches (Pearson $R^2 = 0.637$, $p = 0.003$; and $R^2 = 0.578$, $p = 0.007$, respectively), while the opposite was found for PE/PP mixture (Pearson $R^2 = 0.515$, $p = 0.013$). PET and PVC are both sinking polymers from local sources, while HDPE and PE/PP mixtures are floating polymers more from nonlocal sources. EVA and PP, floating polymers, dominated more developed beaches because of single-use PP items (e.g., food wrappers/containers) and shoes composed of EVA left on the beach. This chemical methodology coupled with correlations adds a line of evidence for sourcing Hawaiian marine debris.

Globally, this is the largest known study to identify polymers of Hawaiian plastic marine debris with novel comparisons across space and habitat depths. Furthermore, this is the first known study to identify Hawaiian seafloor plastic debris and to identify additives, such as phthalates, as a major component of certain debris pieces. This is also the first study to create an efficient weathering rank. Floating, severely weathered polymers wash ashore from distant sources on windward beaches at a much greater abundance than denser, less weathered polymers found on leeward beaches and seafloor. These results support prior conclusions that the majority of marine debris in Hawaii is coming from distant sources,⁶⁹ often composed of maritime gear.¹⁰ Novel information suggests that the leeward beaches receive smaller quantities of litter, but from local activities (e.g., fishing, diving, boating, and picnicking). Stratification of polymers throughout the environment is evident because of the varying polymer densities that result in significantly different transport and fate of marine debris. Debris composed of denser polymers is more likely to sink near their source, while lighter polymers can travel great distances on the sea surface. This stratification leads to exposure of different debris types and chemicals in different habitats and associated biota. Thus, the chemical methodology of polymer identification is critical for understanding sources, fate, transport, and effects of this emerging global contaminant.

■ ASSOCIATED CONTENT

● Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b03561.

Individual plastic debris with raw data; debris representative; weathering intensity rank; aerial map of Kamilo Beach; aerial map of Waikiki Beach; types of plastic debris; colors of plastic debris; size distribution of MHI plastic marine debris; weathering rank of plastic debris; polymer composition of debris mass and pieces; ATR FTIR spectrum; relationships of percent land development; and statistical test outputs (PDF)

Sampling date sites; pieces analyzed; FTIR spectra; and pieces not analyzed (XLSX)

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Notes

The authors declare no competing financial interest. Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

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