Long-Term Field Measurement of Sorption of Organic Contaminants to Five Types of Plastic Pellets: Implications for Plastic Marine Debris

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S *Supporting Information*

ABSTRACT: Concerns regarding marine plastic pollution and its affinity for chemical pollutants led us to quantify relationships between different types of mass-produced plastic and organic contaminants in an urban bay. At five locations in San Diego Bay, CA, we measured sorption of polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) throughout a 12-month period to the five most common types of mass-produced plastic: polyethylene terephthalate (PET), high-density polyethylene (HDPE), polyvinyl chloride (PVC), low-density polyethylene (LDPE), and polypropylene (PP). During this long-term field experi-

ment, sorption rates and concentrations of PCBs and PAHs varied significantly among plastic types and among locations. Our data suggest that for PAHs and PCBs, PET and PVC reach equilibrium in the marine environment much faster than HDPE, LDPE, and PP. Most importantly, concentrations of PAHs and PCBs sorbed to HDPE, LDPE, and PP were consistently much greater than concentrations sorbed to PET and PVC. These data imply that products made from HDPE, LDPE, and PP pose a greater risk than products made from PET and PVC of concentrating these hazardous chemicals onto fragmented plastic debris ingested by marine animals.
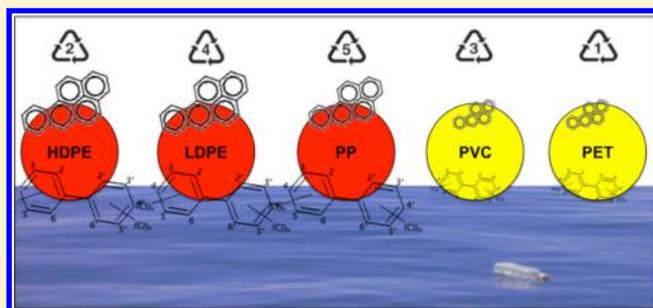
INTRODUCTION

Marine plastic pollution is recognized as a global environmental threat.[1] Plastic waste litters coastlines,[2,3] oceanic gyres,[4] and the sea floor.[5] For decades, plastic debris in the ocean has been of great concern as a physical hazard.[6,7] Because several plastics are composed of hazardous monomers and additives[8,9] and sorb chemical pollutants from aquatic environments,[8] plastic debris also poses a chemical hazard.[8,10] The ingestion of plastic debris by marine animals, including invertebrates,[11] fishes,[12,13] sea turtles,[14] seabirds,[15] and whales,[16] raises concerns that plastic is another mechanism for such chemicals to enter food webs.[8,10] This mixture of hazardous monomers, plastic additives, and sorbed pollutants, may impose a multiple stressor to marine organisms upon ingestion. This study focuses on the sorption of persistent organic pollutants (POPs), specifically polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), to the five most commonly mass-produced plastics. Our data suggest different types of plastic marine debris vary in their chemical hazards.

Early reports of PCBs on plastic debris collected from coastal and oceanic waters[12,17] and the use of plastics as passive samplers to quantify POPs in aquatic environments[18,19] led scientists to measure POPs on recovered plastic debris.[3,20] Plastic preproduction pellets, a recognizable component of marine debris, have been used to establish a global association between POPs and plastic discarded at sea.[20] The physical and

chemical properties of each type of plastic (e.g., surface area,[21] diffusivity,[22−24,27] and crystallinity[22,27]) influence the sorption of chemicals to plastic debris.[23,24] Moreover, sorption patterns are compound-specific (e.g., increasing in affinity with greater hydrophobicity).[25] For example, polyethylene has a larger surface area than polypropylene (PP) and polyvinyl chloride (PVC)[21] and an affinity for a wide range of organic contaminants varying in hydrophobicity.[26] For these reasons, polyethylene is commonly used as a passive-sampling device.[19,23] Here, we deployed different types of plastic preproduction pellets in an urban bay to measure how sorption of organic contaminants, ranging in hydrophobicity, varies among types of plastic debris in the environment.

Existing data on the sorption of organic contaminants to plastic preproduction pellets in laboratory studies are limited to three of the five most commonly produced polymers.[21,22] Those data, predictive modeling,[21] and comparisons of plastic debris of unknown origin collected from coastal and oceanic sites[20,27] suggest that polyethylene sorbs greater concentrations of PAHs and PCBs than does polypropylene (PP)[20,21,27] and polyvinyl chloride (PVC).[21] The only controlled field experi-
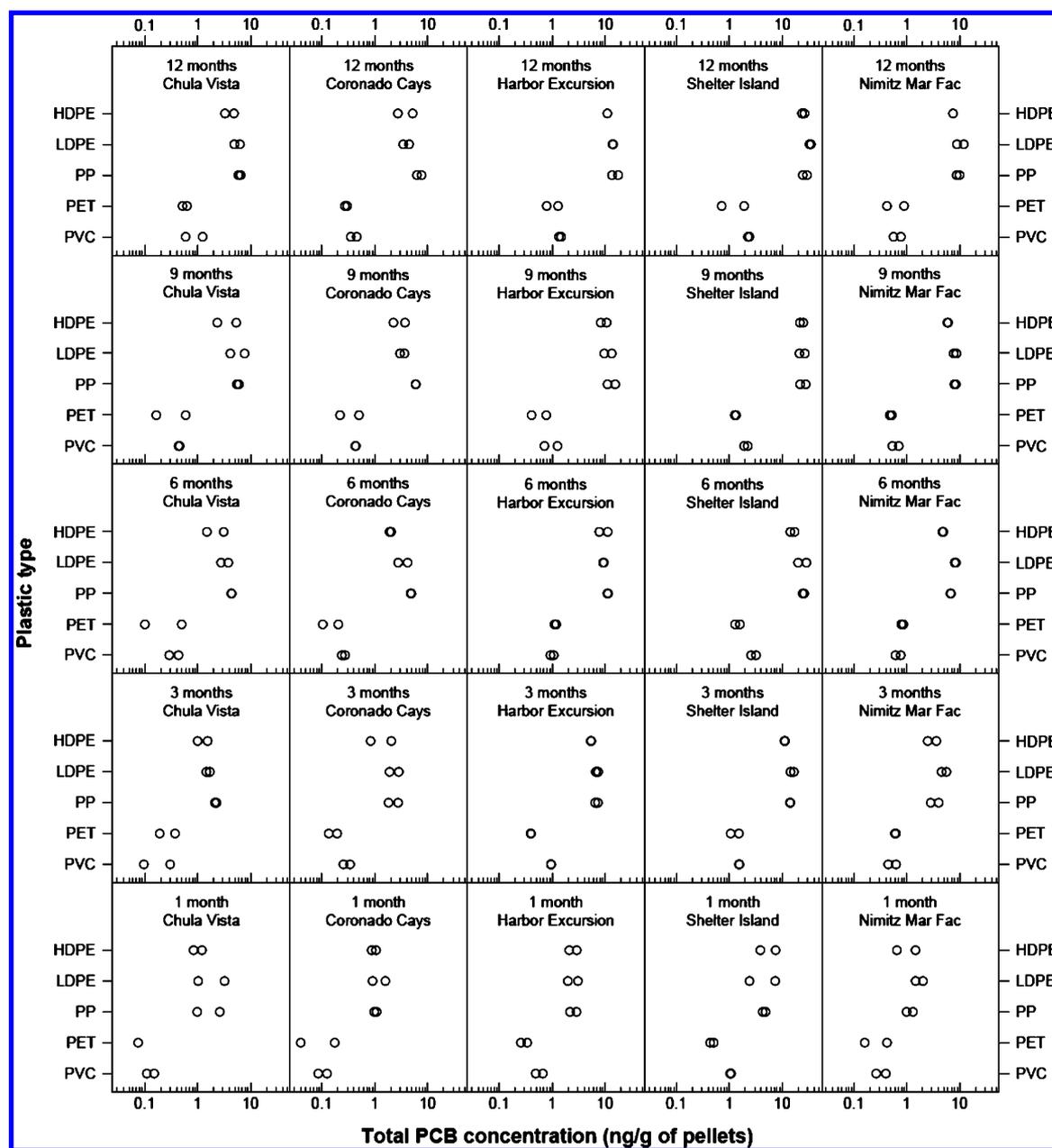
Figure 1. Concentrations of total PCBs (ng/g of pellets) sorbed to each plastic type at each location in San Diego Bay during each sampling period (*n* = 2 replicates). Each panel displays concentrations at one location during one sampling period. Concentrations are plotted on a log scale across the horizontal axis. Plastic types are listed along the vertical axis. Each row of five panels contains data from a single sampling period. Each column displays sites ordered from the back to front of the bay (Cornado Cays, Chula Vista, Harbor Excursion, Shelter Island, Nimitz Marine Facility). At each sampling period, 2-factor ANOVA showed significant differences among plastic types (*p* < 0.001) and locations (*p* < 0.001), and post hoc Tukey comparisons consistently distinguished a group of plastics with high PCB concentrations (HDPE, LDPE, and PP) from a group with low concentrations (PET and PVC).

ment measuring sorption of POPs (specifically PCBs) onto previously uncontaminated plastic preproduction pellets was limited to PP and a period of 6 days, which was too short to measure saturation.[27] Predictive modeling for low-density polyethylene (LDPE) passive sampling devices suggest that time to saturation in the field can vary from days to months depending on the chemical of interest and environmental conditions.[19] This study aimed to quantify sorption of organic contaminants to different types of plastic in an environment and period of time more relevant to plastic marine debris.

Our analyses targeted PCBs and PAHs (see Table S1 of the Supporting Information, SI, for list of targeted analytes).

Production of PCBs was banned in the U.S. in 1979, but measurable concentrations persist.[28] PAHs have continuous anthropogenic and natural sources into the environment.[29] We performed the first long-term field measurement of sorption of PCBs and PAHs to the five most common types of mass-produced plastic:[30] polyethylene terephthalate (PET), high-density polyethylene (HDPE), PVC, LDPE, and PP. Laboratory experiments precisely apply a dose of contaminants to polymers and hold many confounding variables constant (e.g., temperature). In contrast, our field experiment incorporated environmentally relevant factors, including in situ concentrations of contaminants, allowing the polymers to be
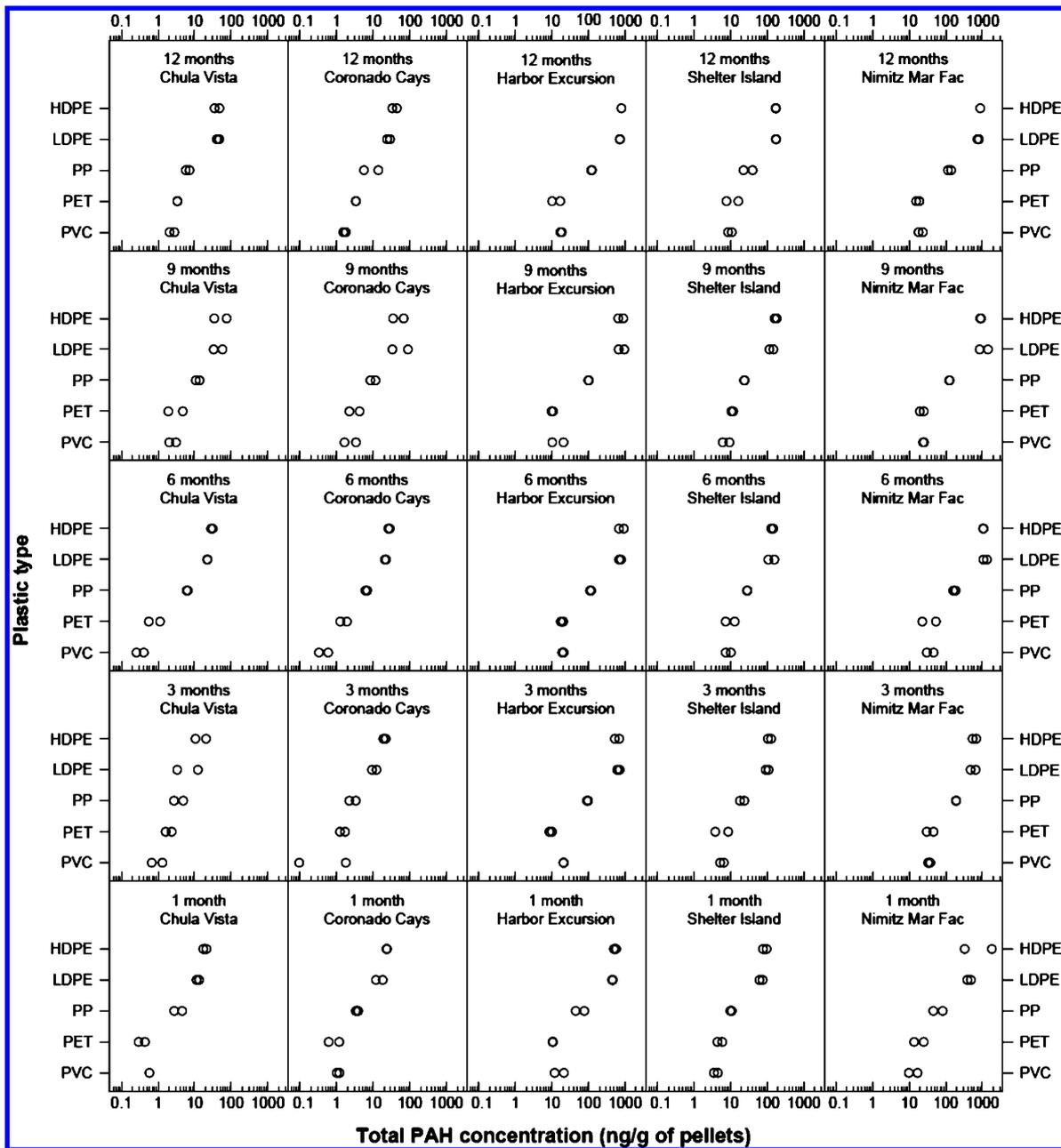
Figure 2. Concentrations of total PAHs (ng/g of pellets) sorbed to each plastic type at each location during each sampling period ($n = 2$ replicates). The format is the same as Figure 1. At each sampling period, 2-factor ANOVA showed significant differences among plastic types ($p < 0.001$) and locations ($p < 0.001$), and post hoc Tukey comparisons consistently distinguished a group of plastics with high PAH concentrations (HDPE and LDPE), a group with low concentrations (PET and PVC), and PP as sorbing an intermediate concentration.

exposed to in situ variations in all variables (including PAHs and PCBs) relevant at our field sites. We deployed two independent replicates of uncontaminated, preproduction pellets of each polymer at five locations in San Diego Bay (Figure S1 of the SI) and recovered them after 1, 3, 6, 9, or 12 months. We hypothesized that sorption of PAHs and PCBs would vary among plastic type and locations in the urban bay. On the basis of prior studies,[19,27] we expected to measure increasing concentrations during at least the first several months of this experiment.

■ EXPERIMENTAL SECTION

Experimental Design. On June 1st, 2009 five types of preproduction plastic pellets (PET, HDPE, PVC, LDPE, and

PP) were deployed from docks at five locations throughout San Diego Bay (Figure S1 of the SI): Chula Vista and Coronado Cays near the back of the bay, Harbor Excursions in the central bay, and Shelter Island and Nimitz Marine Facility near the mouth of the bay. At each location, two replicate samples (5 g) of each plastic type were deployed for future collection at the end of five time periods: 1, 3, 6, 9, and 12 months (250 total samples). See Supporting Information for additional methodological details.

Chemical Analyses. Methods for sample prep and instrumental parameters for chemical analyses were established previously in our laboratory.[3] Sample extracts were analyzed with an Agilent 6890 series gas chromatograph and Agilent 5973 mass spectrometer (Agilent, Santa Clara, CA, U.S.).
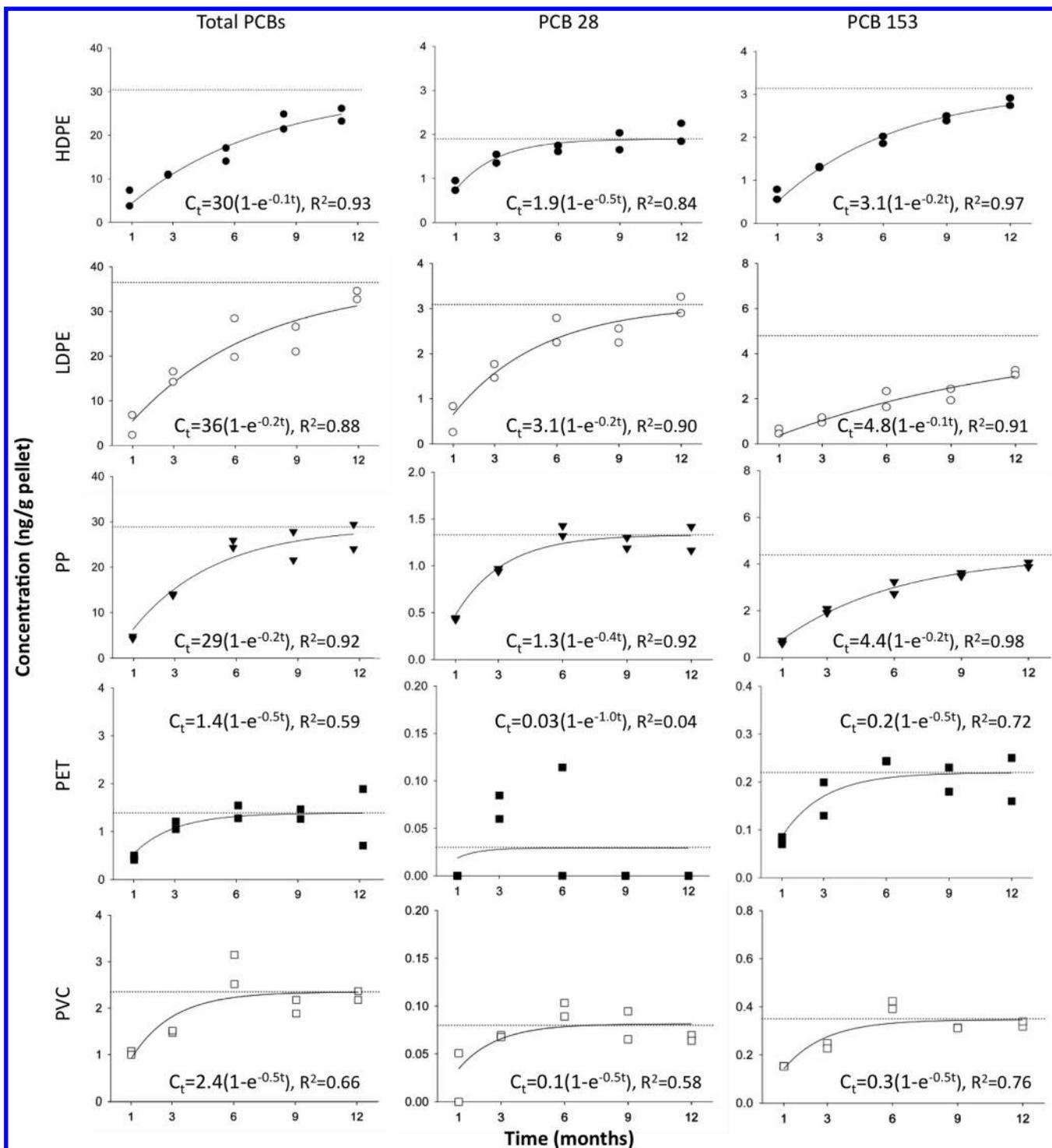
Figure 3. Concentrations of PCBs (ng/g of pellets) vs time for each type of plastic at Shelter Island. Rows represent plastic types HDPE, LDPE, PP, PET, and PVC (in order from top to bottom). Columns represent PCBs: Total PCBs, PCB28, PCB153 (in order from left to right). Please note that vertical axes differ among graphs. Data were fit to the first-order approach to equilibrium model[31] using the exponential rise to maximum equation $C_t = C_{eq}(1 - e^{-kt})$, where C_t is the concentration at time t, C_{eq} is the predicted equilibrium concentration, and k is the rate constant. The horizontal dotted line denotes the predicted C_{eq} for each plastic type.

Selected ion monitoring (SIM) was used to detect 15 PAHs (acenaphthalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(123-cd)pyrene, dibenzo(ah)anthracene, and benzo-(ghi)perylene), 27 PCBs (CB#8, 18, 28, 52, 44, 60, 101, 81, 77, 123, 118, 114, 153, 105, 138, 126, 187, 128, 167, 156, 180, 169, 170, 189, 196, 206, and 209), internal and recovery standards (see SI Table S1 for quantification and confirmation ions). For more detailed information regarding chemical standards and solvent materials, sample prep, chemical analyses, and QA/QC refer to the Supporting Information.

Statistical Methods. Statistical analyses were performed with SYSTAT 12 (SYSTAT Software, Chicago, IL). Concen-
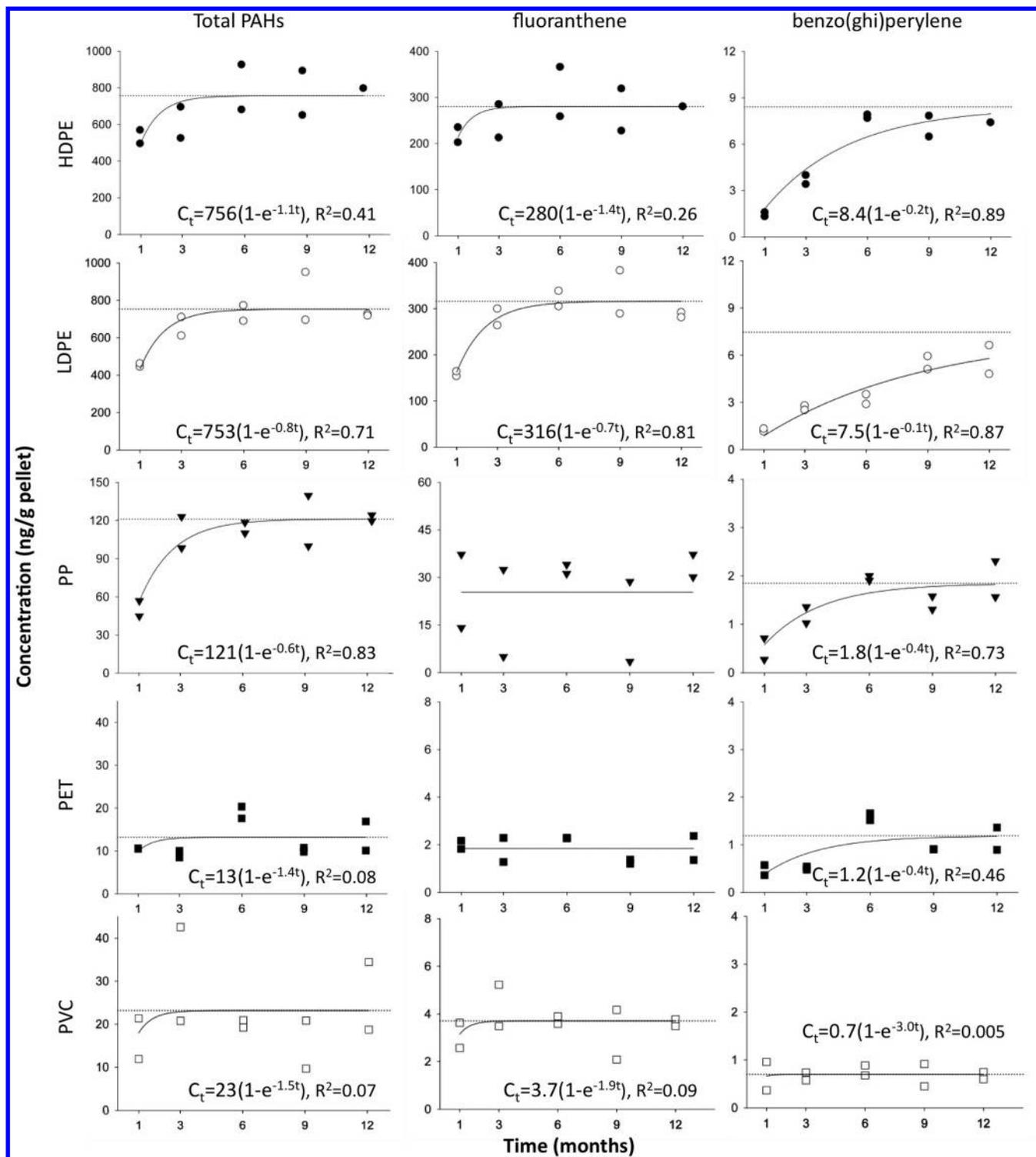
Figure 4. Concentrations of PAHs (ng/g of pellets) vs time for each type of plastic at Harbor Excursion. Rows represent plastic types HDPE, LDPE, PP, PET, and PVC (in order from top to bottom). Columns represent PAHs: Total PAHs, fluoranthene and benzo(ghi)perylene (in order from left to right). Please note that vertical axes differ among graphs. Data were fit to the first-order approach to equilibrium model[31] using the exponential rise to maximum equation $C_t = C_{eq}(1 - e^{-kt})$, where C_t is the concentration at time t, C_{eq} is the predicted equilibrium concentration, and k is the rate constant. The horizontal dotted line denotes the predicted C_{eq} for each plastic type. Where no equation is given, the model could not be fit to the data.

trations of sorbed PCBs and PAHs were log-transformed to achieve normality. We decided a priori to test for differences among plastic types and locations by performing a 2-factor ANOVA on data from each sampling period individually. Homogeneity of variance was verified by Levene's test. Post hoc

Tukey's tests were used to distinguish significantly different treatment means. We decided a priori to quantify temporal patterns by fitting a first-order approach to equilibrium model[31] to the concentrations sorbed to each type of plastic at each location in the bay. We expected sorption to follow first-order

kinetics as they do for polyethylene devices.[26] SigmaPlot 10 (SYSTAT Software, Chicago, IL) was used to fit the exponential rise to maximum equation $C_t = C_{eq}(1 - e^{-kt})$, where C_t is the concentration at time t, C_{eq} is the predicted equilibrium concentration, and k is the rate constant.

■ RESULTS AND DISCUSSION

Concentrations of PCBs and PAHs Vary by Plastic Type. Despite significant variability among locations in San Diego Bay, relative differences in sorption of total PCBs and total PAHs among the five polymers were consistent over space and time (Figures 1 and 2). Most importantly, HDPE, LDPE, and PP consistently sorbed greater concentrations of PAHs and PCBs than did PET and PVC. Rubbery polymers such as HDPE, LDPE, and PP are expected to demonstrate greater diffusion (e.g., permeability and greater free volume) than the glassy polymers PET and PVC,[23] which may explain trends observed here.

Throughout the 12-month period, concentrations of total PCBs sorbed to HDPE, LDPE and PP were consistently an order of magnitude greater than those sorbed to PET and PVC (Figure 1). Concentrations of PCBs sorbed to plastics varied significantly among polymers ($p < 0.001$) on each sampling date (Figure 1). There were not significant interactions between the main effects of location and polymer type ($p > 0.174$) except during the 3-month sampling ($p = 0.013$). At this sampling, sorbed PCBs at Coronado Cays, Harbor Excursion, Shelter Island, and Nimitz Marine Facility were greater on PVC than on PET, but this pattern between these two polymers was reversed at Chula Vista (Figure 1). By 12 months at Shelter Island, where PCB concentrations were greatest, mean concentrations of total sorbed PCBs were 25 ng/g on HDPE, 34 ng/g on LDPE, and 27 ng/g on PP compared to only 1 ng/g on PET and 2 ng/g on PVC.

Like PCBs, concentrations of PAHs sorbed to HDPE and LDPE were consistently an order of magnitude greater than those sorbed to PET and PVC (Figure 2). Furthermore, PP consistently sorbed an intermediate concentration (Figure 2). Concentrations of sorbed PAHs varied significantly among polymers ($p < 0.001$) on each sampling date (Figure 2). Concentrations of PAHs sorbed to PP were significantly less than those sorbed to HDPE and LDPE ($p < 0.001$) and significantly greater than those sorbed to PET and PVC ($p < 0.01$). Similar to PCBs, there were not significant interactions between location and plastic type ($p > 0.06$) except during the 3-month sampling ($p = 0.002$). At this sampling, sorbed PAHs were greater on PVC than on PET at Harbor Excursion, but concentrations on these two polymers were indistinguishable at the other locations (Figure 2). By 12 months at Harbor Excursion, where PAH concentrations were relatively large, mean concentrations of total sorbed PAHs were 797 ng/g on HDPE, 722 ng/g on LDPE, 122 ng/g on PP, 14 ng/g on PET, and 27 ng/g on PVC.

Trends among the five plastics in our field experiment reinforce previous laboratory studies in which polyethylene sorbed greater concentrations of PAHs than PP[21,22] and an order of magnitude greater concentrations of PCBs[23] and PAHs[21] than did PVC (to our knowledge, sorption of PCBs and PAHs to PET has not been measured and compared to other polymers in the laboratory). Polyethylene has a greater sorptive capacity than PP and PVC,[21] probably due to polyethylene's greater surface area[21] and free volume.[23] Moreover, diffusion into the polymer has been observed in

polyethylene pellets but not in PP.[22] Greater sorptive capacity is expected for polyethylene because its structure has a greater distance between polymeric chains than PP.[22] Thus, the greater sorptive capacity for PAHs that we observed in polyethylene compared to PP should be expected.[22] However, we caution that these similar results for PCBs and PAHs do not necessarily extend to all classes of contaminants, and compound-specific interactions in the polymer phase are also important.[25] For example, PVC has a greater affinity for alkylbenzenes than does HDPE.[32] While our study found that HDPE, LDPE, and PP sorb much greater PCBs and PAHs than do PET and PVC, future field experiments should measure sorption of additional priority pollutants to plastics to quantify the chemical mixtures associated with different types of plastic marine debris.

Sorption Rate of PCBs and PAHs among Plastic Types. Due to differences in surface area[26] and diffusion properties,[22,26] we expected to measure different temporal patterns among the five polymers. Fitting the first-order approach to equilibrium model[31] assumes a relatively constant background concentration of PAHs or PCBs. Although this assumption likely does not hold for chemical pollutants during a field deployment, the equation fits our data reasonably well over the long time scales of our experiment. We quantified temporal patterns for total PCBs and total PAHs for each type of plastic at each location in San Diego Bay (Figures 3 and 4). Because sorption is related to the hydrophobicity of each specific congener,[19,25,33] we also quantified temporal patterns for individual PCBs and PAHs at locations where concentrations were greatest (Figures S3–S9 of the SI).

At all locations, we observed increasing concentrations of PCBs and PAHs sorbed to HDPE, LDPE, and PP over time, and where PCB concentrations were relatively large, HDPE and LDPE did not always reach equilibrium within the 12-month experiment. For example, at Shelter Island, total PCBs sorbed to PET and PVC reached equilibrium by 6 months, concentrations sorbed to PP did not reach equilibrium until 12 months, and concentrations sorbed to HDPE and LDPE did not reach equilibrium (Figure 3). Using the predicted concentration at equilibrium (C_{eq}; Figure 3), and the assumption that total PCB sorption will follow the exponential rise to maximum equation, HDPE will achieve equilibrium within 44 months and LDPE within 19 months at Shelter Island. At Harbor Excursion, the relatively small concentrations of total PAHs sorbed to PET and PVC changed little over time, and PAH concentrations on HDPE, LDPE, and PP reached equilibrium by 6 months (Figure 4). Faster saturation on PET and PVC is expected based upon a smaller surface area[21] and a glassy polymeric structure,[23] which does not facilitate diffusion into the material as we would expect for HDPE,[19,23] LDPE,[19,23] and PP.[23] Diffusion into the polymeric matrix may be one reason we observe rapidly increasing concentrations during the first month (i.e., adsorption) followed by slower sorption over the following months (i.e., absorption).

Temporal patterns among individual congeners of PAHs and PCBs were also not equivalent (see Figures S3–S9 of the SI). They are often driven by hydrophobicity (octanol–water partition coefficient—log K_{ow}) and/or molecular weight (MW) of the chemical (See Table S1 of the SI for log K_{ow} and MW of targeted analytes).[25,33] Chemicals with less hydrophobicity and lighter MW are expected to reach saturation faster than those with greater hydrophobicity and heavier MW.[26] Here, PCB congeners with lighter MW and smaller log K_{ow} achieved saturation quicker than heavier congeners (Figures S3–S6 of

the SI). For example, PCB153 (log K_{ow} = 6.9; MW = 360.9) did not reach equilibrium within the 12-month period on HDPE, LDPE, or PP; however, PCB28 (log K_{ow} = 5.6; MW = 257.5) did (Figure 3). Similarly, PAHs with lighter MW and smaller K_{ow} achieved saturation faster (Figures S7–S9 of the SI). Benzo(ghi)perylene (log K_{ow} = 6.63; MW = 276.4) did not reach equilibrium within the 12-month period on HDPE and LPDE; however, fluoranthene (log K_{ow} = 5.0; MW = 202.3) reached equilibrium by the 6-month period (Figure 4). Furthermore, when concentrations of individual congeners were quantifiable, congeners with a log K_{ow} > 5 generally fit the first-order approach to equilibrium model, whereas those with a log K_{ow} < 5 generally did not. For example, acenaphthene, fluorene, phenanthrene, and anthracene sorption does not fit the model and instead appears to be decreasing in concentration over time (Figure S7 of the SI). This may be explained by a loss of lighter MW PAHs due to degradation.

Our finding that HDPE, LDPE, and PP required at least 6 months to reach equilibrium concentrations of PCBs or PAHs in San Diego Bay is much longer than estimated in laboratory studies with various plastics.[19,21,23] This may be due to weathering of the pellets over time, increasing the surface area and pore size of the material allowing greater diffusivity.[27] Comparing plastic waste discarded into the marine environment to plastic in controlled laboratory settings or used as short-term passive samplers has limited applicability to quantifying chemical hazards of plastic marine debris. In the marine environment, concentrations of chemical pollutants change over time depending on sources and environmental differences (e.g., temperature). Furthermore, plastic debris remains in the marine environment for an unknown period of time, drifts with the currents, and undergoes slow photo-degradation. Degradation increases surface area,[30,34] probably leading to greater sorption of contaminants over time,[30,34] suggesting that plastic debris may become more hazardous the longer it remains at sea.

International Pellet Watch (IPW) analyzes pellets collected globally that are fouled and/or weathered and yellowing.[20] They find greater concentrations of organic contaminants on pellets that are fouled and darker in color.[35] Discoloration is caused primarily by phenolic antioxidant additives to the plastics,[35] used in several polymers including those discussed here. It reflects a longer period of time in the marine environment.[20,34] Consistent with IPW, we observed discoloration of plastic pellets during our long-term field experiment (Figure S10 of the SI), further confirming that darker pellets reflect a longer period of time at sea.

Spatial Differences. While sample sites were relatively close to each other in a single bay (measuring approximately 25 × 3 km), concentrations of total PCBs and total PAHs sorbed to plastics varied significantly among locations in San Diego Bay (p < 0.001) on each sampling date (Figures 1 and 2). These differences were expected based on local sources of contamination and prior research showing that concentrations of sorbed pollutants on plastic reflect regional differences.[3,20,36–40] See Supporting Information for more details regarding differences among locations.

Small-scale variation observed in this study and larger-scale variation observed in other studies[41] has implications for management of plastic marine debris. Within San Diego Bay, we measured concentrations of PCBs on LDPE pellets that varied by an order of magnitude (3.4–35 ng/g). In our previous study, recovering pellets from eight beaches

throughout San Diego County, concentrations of the same suite of PCBs also varied by an order of magnitude (3.8–42 ng/g total PCBs).[3] Current data from International Pellet Watch show that PCBs on polyethylene pellets collected from beaches spanning over 1000 km of the California coastline also vary by an order of magnitude (23–605 ng/g), and this variation can be larger among locations within the United States (0.1–605 ng/g).[41] Discarded plastics may pose a greater hazard and thus a greater management priority in locations with greater point sources and greater concentrations of chemical pollutants. In addition, plastic debris drifts with currents; hypotheses regarding transport of hydrophobic chemicals from plastics, which may be released upon degradation of the material,[36] to remote areas should be tested.

Implications of This Study to Marine Plastic Debris. With escalating plastic production[10] and poor policy enforcement,[42] quantities of plastic debris in the marine environment will likely increase. In 2007, HDPE, LDPE, and PP accounted for 62% of all plastics produced globally, while PVC and PET represented only 19% and 7%, respectively.[30] In accord with production trends, HDPE, LDPE, and PP are the most common plastic debris collected at sea.[4,30] Our conclusion that HDPE, LDPE, and PP consistently sorbed greater concentrations of PCBs and PAHs than did PET and PVC implies that PET and PVC pose a reduced risk of concentrating these hazardous chemicals onto plastic debris ingested by animals. Although PVC sorbs relatively small concentrations of PCBs and PAHs, its vinyl chloride monomer is classified as carcinogenic and toxic.[9] In addition, PVC contains more hazardous additives than HDPE, LDPE, PP, and PET.[9] Because PET requires fewer additives and degrades faster than other polymers, it has been suggested as one of the least hazardous plastics.[9] These inherent properties in combination with the relatively low sorption of PCBs and PAHs we measured suggest that plastic products made from PET, especially items commonly encountered in marine debris such as plastic packaging and fishing gear,[30] might have fewer chemical impacts on marine life than products made from other types of plastic. Testing this hypothesis will require measuring bioaccumulation and toxicity from different types of plastic marine debris, all containing a unique cocktail of chemicals which may act as a multiple stressor when ingested by marine animals.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information
Table S1 shows Quantification and Confirmation Ions for each analyte targeted by GC/MS analysis. Figure S1 shows a map of our study sites in SD Bay and Figure S2 a schematic diagram of the experimental design. Figures S3–S6 show time trends for targeted PCBs at Shelter Island and Figures S7–S9 shows time trends for targeted PAHs at Harbor Excursion. Figure S10 shows the discoloration of pellets over time. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

■ REFERENCES

(1) *The Honolulu Strategy: A global framework for prevention and management of marine debris*. Proceedings of the 5th International Marine Debris Conference. National Oceanic and Atmospheric Administration and United Nations Environmental Programme, 2011; http://marinedebris.noaa.gov/projects/pdfs/HonoluluStrategy.pdf

(2) Browne, M. A.; Crump, P.; Niven, S. J.; Teuten, E. L.; Tonkin, A.; Galloway, T.; Thompson, R. C. Accumulations of microplastic on shorelines worldwide: Sources and sinks. *Environ. Sci. Technol.* **2011**, *45*, 9175−9179.

(3) Van, A.; Rochman, C. M.; Flores, E. M.; Hill, K. L.; Vargas, E.; Vargas, S. A.; Hoh, E. Persistent organic pollutants in plastic marine debris found on beaches in San Diego, California. *Chemosphere* **2011**, *86*, 258−263.

(4) Law, K. L.; Moret-Ferguson, S.; Maximenko, N. A.; Proskurowski, G.; Peacock, E. E.; Hafner, J.; Reddy, C. M. Plastic Accumulation in the North Atlantic Subtropical Gyre. *Science* **2010**, *329*, 1185−1188.

(5) Goldberg, E. D. Plasticizing the seafloor: An overview. *Environ. Technol.* **1997**, *18*, 195−201.

(6) Colton, J. B.; Knapp, F. D.; Burnes, B. R. Plastic particles in surface waters of Northwestern Atlantic. *Science* **1974**, *185*, 491−497.

(7) Thompson, R.; Olsen, Y.; Mitchell, R. P.; Davis, A.; Rowland, S. J.; John, A. W. G.; McGonigle, D.; Russell, A. Lost at sea: Where is all the plastic? *Science* **2004**, *304*, 838.

(8) Teuten, E. L.; Saquing, J. M.; Knappe, D. R. U.; Barlaz, M. A.; Jonsson, S.; Bjorn, A.; Rowland, S. J.; Thompson, R. C.; Galloway, T. S.; Yamashita, R.; Ochi, D.; Watanuki, Y.; Moore, C.; Viet, P. H.; Tana, T. S.; Prudente, M.; Boonyatumanond, R.; Zakaria, M. P.; Akkhavong, K.; Ogata, Y.; Hirai, H.; Iwasa, S.; Mizukawa, K.; Hagino, Y.; Imamura, A.; Saha, M.; Takada, H. Transport and release of chemicals from plastics to the environment and to wildlife. *Philos. Trans. R. Soc. B* **2009**, *364*, 2027−2045.

(9) Lithner, D.; Larsson, A.; Dave, G. Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. *Sci. Total Environ.* **2011**, *409*, 3309−3324.

(10) Thompson, R.; Moore, C.; vom Saal, F.; Swan, S. Plastics, the environment and human health: Current consensus and future trends. *Philos. Trans. R. Soc. B* **2009**, *364*, 2153−2166.

(11) Murray, F.; Cowie, P. R. Plastic contamination in the decapod crustacean *Nephrops norvegicus* (Linnaeus, 1758). *Mar. Pollut. Bull.* **2011**, *62*, 1207−1217.

(12) Carpenter, E.; Smith, K. Plastics on the Sargasso Sea surface. *Science* **1972**, *175*, 1240−1241.

(13) Davison, P.; Asch, R. Plastic ingestion by mesopelagic fishes in the North Pacific Subtropical Gyre. *Mar. Ecol.: Prog. Ser.* **2011**, *432*, 173−180.

(14) Lazar, B.; Gracan, R. Ingestion of marine debris by loggerhead sea turtles, *Caretta caretta*, in the Adriatic Sea. *Mar. Pollut. Bull.* **2011**, *62*, 43−47.

(15) Spear, L. B.; Ainley, D. G.; Ribic, C. A. Incidence of plastic in seabirds from the tropical pacific, 1984−91- Relation with distribution of species, sex, age, season, year and body-weight. *Mar. Environ. Res.* **1995**, *40*, 123−146.

(16) Tarpley, R. J.; Marwitz, S. Plastic debris ingestion by cetaceans along the Texas coast: Two case reports. *Aquat. Mammals Vol.* **1993**, *19*, 93−98.

(17) Carpenter, E.; Anderson, S.; Harvey, G.; Miklas, H.; Peck, B. Polystyrene spherules in coastal waters. *Science* **1972**, *178*, 749−750.

(18) Huckins, J.; Manuweera, G.; Petty, J.; Mackay, D.; Lebo, J. Lipid-containing semipermeable membrane devices for monitoring organic contaminants in water. *Environ. Sci. Technol.* **1993**, *27*, 2489−2496.

(19) Lohmann, R. Critical review of low-density polyethylene's partitioning and diffusion coefficients for trace organic contaminants and implications for its use as a passive sampler. *Environ. Sci. Technol.* **2012**, *46*, 606−618.

(20) Ogata, Y.; Takada, H.; Mizukawa, K.; Hirai, H.; Iwasa, S.; Endo, S.; Mato, Y.; Saha, M.; Okuda, K.; Nakashima, A.; Murakami, M.; Zurcher, N.; Booyatumanondo, R.; Zakaria, M. P.; Dung, L. Q.; Gordon, M.; Miguez, C.; Suzuki, S.; Moore, C.; Karapanagioti, H.; Weerts, S.; McClurg, T.; Burres, E.; Smith, W.; Van Velkenburg, M.; Lang, J. S.; Lang, R. C.; Laursen, D.; Danner, B.; Stewardson, N.; Thompson, R. C. International pellet watch: Global monitoring of persistent organic pollutants (POPs) in coastal waters. 1. Initial phase data on PCBs, DDTs, and HCHs. *Mar. Pollut. Bull.* **2009**, *58*, 1437−1446.

(21) Teuten, E. L.; Rowland, S. J.; Galloway, T. S.; Thompson, R. C. Potential for plastics to transport hydrophobic contaminants. *Environ. Sci. Technol.* **2007**, *41*, 7759−7764.

(22) Karapanagioti, H. K.; Klontza, I. Testing phenanthrene distribution properties of virgin plastic pellets and plastic eroded pellets found on Lesvos island beaches (Greece). *Mar. Environ. Res.* **2008**, *65*, 283−290.

(23) Pascall, M. A.; Zabik, M. E.; Zabik, M. J.; Hernandez, R. J. Uptake of polychlorinated biphenyls (PCBs) from an aqueous medium by polyethylene, polyvinyl chloride, and polystyrene films. *J. Agr. Food Chem.* **2005**, *53*, 164−169.

(24) Rusina, T.; Smedes, F.; Klanova, J.; Booij, K.; Holoubek, I. Polymer selection for passive sampling: A comparison of critical properties. *Chemosphere* **2007**, *68*, 1344−1351.

(25) Smedes, F.; Geertsma, R. W.; Van der Zande, T.; Booij, K. Polymer-water partition coefficients of hydrophobic compounds for passive samplilng: Application for cosolvent models for validation. *Environ. Sci. Technol.* **2009**, *43*, 7047−7054.

(26) Muller, J. F.; Manomanii, K.; Mortimer, M. R.; McLachlan, M. S. Partitioning of polycyclic aromatic hydrocarbons in the polyethylene/ water system. *Fresenius J. Anal. Chem.* **2001**, *371*, 816−822.

(27) Mato, Y.; Isobe, T.; Takada, H.; Kanehiro, H.; Ohtake, C.; Kaminuma, T. Plastic resin pellets as a transport medium for toxic chemicals in the marine environment. *Environ. Sci. Technol.* **2001**, *35*, 318−324.

(28) Wirgin, I.; Roy, N. K.; Loftus, M.; Chambers, R. C.; Franks, D. G.; Hahn, M. E. Mechanistic basis of resistance to PCBs in Atlantic Tomcod from the Hudson River. *Science* **2011**, *331*, 1322−1325.

(29) Ravindra, K.; Sokhi, R.; Van Grieken, R. Atmospheric polycyclic aromatic hydrocarbons: Source attribution, emission factors and regulation. *Atmos. Environ.* **2008**, *42*, 2895−2921.

(30) Andrady, A. Microplastics in the marine environment. *Mar. Pollut. Bull.* **2011**, *62*, 1596−1605.

(31) Allan, I. J.; Booij, K.; Paschke, A.; Vrana, B.; Mills, G. A.; Greenwood, R. Field performance of seven passive sampling devices for monitoring hydrophobic substances. *Environ. Sci. Technol.* **2009**, *43*, 5383−5390.

(32) Wu, B.; Taylor, C. M.; Knappe, D. R. U.; Nanny, M. A.; Barlaz, M. A. Factors controlling alkylbenzene sorption to municipal solid waste. *Environ. Sci. Technol.* **2001**, *35*, 4569−4576.

(33) Booij, K.; Hofmans, H. E.; Fischer, C. V.; van Weerlee, E. M. Temperature-dependent uptake rates of nonpolar organic compounds by semipermeable membrane devices and low-density polyethylene membranes. *Environ. Sci. Technol.* **2003**, *37*, 361−366.

(34) Fotopoulou, K. N.; Karapanagioti, H. K. Surface properties of beached plastic pellets. *Mar. Environ. Res.* **2012**, *81*, 70−77.

(35) Endo, S.; Takizawa, R.; Okuda, K.; Takada, H.; Chiba, K.; Kanehiro, H.; Ogi, H.; Yamashita, R.; Date, T. Concentration of polychlorinated biphenyls (PCBs) in beached resin pellets: Variability among individual particles and regional differences. *Mar. Pollut. Bull.* **2005**, *50*, 1103−1114.

(36) Hirai, H.; Takada, H.; Ogata, Y.; Yamashita, R.; Mizukawa, K.; Saha, M.; Kwan, C.; Moore, C.; Gray, H.; Laursen, D.; Zettler, E.; Farrington, J.; Reddy, C.; Peacock, E.; Ward, M. Organic micropollutants in marine plastic debris from the open ocean and remote and urban beaches. *Mar. Pollut. Bull.* **2011**, *62*, 1683−1692.

(37) Boonyatumanond, R.; Wattayakorn, G.; Togo, A.; Takada, H. Distribution and origins of polycyclic aromatic hydrocarbons (PAHs) in riverine, estuarine, and marine sediments in Thailand. *Mar. Pollut. Bull.* **2006**, *52*, 942−956.

(38) Rios, L. M.; Jones, P. R.; Moore, C.; Narayan, U. V. Quantitation of persistent organic pollutants adsorbed on plastic debris from the Northern Pacific Gyre's "eastern garbage patch". *J. Environ. Monit.* **2010**, *12*, 2189−2312.

(39) Fairey, R.; Roberts, C.; Jacobi, M.; Lamerdin, S.; Clark, R.; Downing, J.; Long, E.; Hunt, J.; Anderson, B.; Newman, J.; Tjeerdema, R.; Stephenson, M.; Wilson, C. Assessment of sediment toxicity and chemical concentrations in the San Diego Bay region, California, USA. *Environ. Toxicol. Chem.* **1998**, *17*, 1570−1581.

(40) Goldberg, E. D.; Bertine, K. K. Beyond the mussel watch—New directions for monitoring marine pollution. *Sci. Total Environ.* **2000**, *247*, 165−174.

(41) *International Pellet Watch* Website; www.pelletwatch.org.

(42) Shaxson, L. Structuring policy problems for plastics, the environment and human health: reflections from the UK. *Philos. Trans. R. Soc. B* **2009**, *364*, 2141−2151.