

Prepared in cooperation with the U.S. Environmental Protection Agency and the State of Hawai'i Department of Health

Pharmaceuticals and Personal Care Products in Passive Samplers at Seven Coastal Sites off West Maui, Hawai'i



Open-File Report 2022–1065

Cover: Passive membrane sampler (foreground) and a sediment tube trap deployed on the nearshore coral reef of Wahikuli, West Maui, Hawai'i.

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By Pamela L. Campbell, Nancy G. Prouty, and Curt D. Storlazzi

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U.S. Geological Survey**

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Conversion Factors

International System of Units to U.S. customary units

| Multiply | By | To obtain |
|--------------------------------------|----------|--------------------------------|
| Length | | |
| millimeter (mm) | 0.03937 | inch (in.) |
| meter (m) | 3.281 | foot (ft) |
| meter (m) | 1.094 | yard (yd) |
| kilometer (km) | 0.6214 | mile (mi) |
| kilometer (km) | 0.5400 | mile, nautical (nmi) |
| Area | | |
| square centimeter (cm ²) | 0.1550 | square inch (in ²) |
| square centimeter (cm ²) | 0.001076 | square foot (ft ²) |
| Volume | | |
| liter (L) | 33.81402 | ounce, fluid (fl. oz) |
| liter (L) | 2.113 | pint (pt) |
| liter (L) | 1.057 | quart (qt) |
| liter (L) | 0.2642 | gallon (gal) |
| Mass | | |
| gram (g) | 0.03527 | ounce, avoirdupois (oz) |

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$^{\circ}\text{F} = (1.8 \times ^{\circ}\text{C}) + 32.$$

Datum

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Supplemental Information

When discussing carbon numbers, they are denoted with a C and the appropriate numerical suffix; for example, C₂₀ is a carbon number of 20.

Contaminant concentrations are measured in nanograms per sampler (ng/sampler).

Minimum reporting level is measured in nanograms per milliliter (ng/mL).

Abbreviations

| | |
|------------------|---|
| DDM | degrees decimal minutes |
| DDT | dichlorodiphenyltrichloroethane |
| DNA | deoxyribonucleic acid |
| EST Lab | Environmental Sampling Technologies, Incorporated |
| HLB | hydrophilic-lipophilic-balanced |
| HMW | high molecular weight |
| LMW | low molecular weight |
| log | logarithmic |
| MDL | minimum detection level |
| MRL | minimum reporting level |
| <i>n</i> -alkane | normal alkane |
| ng | nanogram |
| PAH | polycyclic aromatic hydrocarbons |
| PCB | polychlorinated biphenyls |
| POCIS | polar organic chemical integrative sampler |
| PPCP | pharmaceutical and personal care products |
| SPMD | semipermeable membrane device |
| TCEP | tris(2-chloroethyl) phosphate |
| TCPP | tris(1-chloro-2-propyl) phosphate |
| USCRTF | U.S. Coral Reef Task Force |
| USGS | U.S. Geological Survey |

Pharmaceuticals and Personal Care Products in Passive Samplers at Seven Coastal Sites off West Maui, Hawai‘i

By Pamela L. Campbell, Nancy G. Prouty, and Curt D. Storlazzi

Abstract

Passive membrane samplers—semipermeable membrane devices and polar organic chemical integrative samplers—were deployed for 22 continuous days at 7 sites along the West Maui, Hawai‘i, coastline in February and March 2017 to assess organic contaminants at shallow coral reef ecosystems from diverse upstream inputs. The distribution of organic compounds observed at these coastal sites showed considerable variability; high concentrations of microbially sourced organic compounds observed at all sites, with pentadecane as the predominant normal alkane, showed the relative importance of marine and microbial organic matter to the coastal carbon pool. Pharmaceuticals and personal care products, as well as flame retardants, were also detected at all sites. Of the seven sites sampled, the Kahekili Beach Park site had the highest number of unique contaminants and the Honokōwai Stream site had the highest concentrations of compounds. Two individual compounds, a flame retardant and a fragrance, were ubiquitous across the studied West Maui reefs, including at the least-developed site. A direct correlation to upstream land-use practices or legacy agricultural inputs was not readily observed since polychlorinated biphenyls, pesticides, herbicides, or insecticides were not detected. Results provide a snapshot of relative contaminant abundances as well as inputs to select nearshore environments along the West Maui coastline captured during the 2017 wet season, which was drier than expected. These data can be useful for understanding the range of stressors potentially affecting nearshore ecosystems, such as groundwater inputs and watershed runoff.

Introduction

Coral reefs of the main Hawaiian Islands have been in decline since the 1990s owing to a multitude of factors, including excess nutrient runoff, shoreline development, and widespread agricultural and municipal runoff (Friedlander and others, 2008; Dailer and others, 2012). Long-term monitoring of coral reefs along the West Maui coastline has shown that the coral cover has declined by as much as 50 percent in some highly impacted sites (Bruno and Selig, 2007). The combined surface runoff and groundwater discharge

of terrestrially sourced organic and inorganic contaminants has affected nearshore ecosystems, including coral reefs, for much longer than previously suggested (Anthony and others, 2004; Fabricius, 2005; Brodie and others, 2012; Morrison and others, 2013).

Land-derived inputs to the reef system may include pesticides, petroleum hydrocarbons, trace metals, and wastewater contaminants such as pharmaceuticals and personal care products (PPCP) (Knee and others, 2010; Kitada and others, 2008). Petroleum hydrocarbons (found in asphalt sealer, boat bilge water, engine exhaust, runoff, and atmospheric deposition) are associated with paved coastal roads as well as vehicle and boat activity.

Long-range atmospheric transport of contaminants that include legacy and current-use pesticides, such as dichlorodiphenyltrichloroethane (DDT), polycyclic aromatic hydrocarbons (PAH), and polychlorinated biphenyls (PCB), is hypothesized to be a source of pollutants to coral reefs of the Caribbean Sea and Pacific Ocean (Bargar and others, 2013). Such long-distance transport mechanisms may be responsible for low-level legacy pesticide concentrations detected in coral samples from remote reefs, such as those of the Marshall Islands and the Hawaiian Islands National Wildlife Refuge (Wang and others, 2011).

In contrast to legacy contaminants, PPCP may be continuously discharged to the nearshore in stream discharge, surface runoff, and from submarine springs that are linked to upland watersheds. Nearshore hydrodynamic conditions can contribute to substantial dilution, and diffusion processes can make detection and ecological exposure difficult to quantify. The effects of chronic, low-level contamination on marine ecosystems such as coral reefs are not well characterized.

Over the past decade, there has been a notable change in bottom type off Kahekili Beach Park in West Maui; areas once populated with abundant corals are now mostly covered by turf algae or macro algae, suggesting eutrophication (Vermeij and others, 2010). In response to these findings, the site has been studied extensively to assess ecosystem effects from upstream municipal wastewater discharges, which are flushed into coastal waters by groundwater discharge (Hunt and Rosa, 2009; Dailer and others, 2012; Swarzenski and others, 2013; Miller-Pierce and Rhoads, 2016; Prouty and others, 2017; Prouty and others, 2018).

To assess surface-water and groundwater-derived organic loading rates along the U.S. Coral Reef Task Force (USCRTF) West Maui priority watersheds study area, a study was conducted using passive membrane samplers at seven sites along the West Maui coast (fig. 1; table 1) during the 2017 wet season (February and March) to determine the influence of surface water runoff from intermittent/seasonal streams to coral reef ecosystems.

The main objectives of this study were to (1) identify suites of pollutant classes in these different watersheds and (2) determine wet-season levels of organic contaminants per site. This study provides a unique opportunity to examine interactions between fragile coastal ecosystems and their adjacent upstream watersheds, and to examine the possible deleterious effects of land-based contaminants on the coral reef system of West Maui.

Study Sites

Passive samplers were deployed in the USCRTF West Maui priority watersheds study area off the Wahikuli, Honokeana, Kahana, Honolua, and Honokōwai stream mouths to capture the effects of surface water runoff. Another sampler was deployed in Kahekili Beach Park to capture the effects of submarine groundwater discharge (Swarzenski and others, 2017). The final sampler was deployed off Olowalu Beach to assess the possible effects of active coastal development via surface runoff, as well as to assess legacy contaminants. The drainages for the current sample sites are categorized by a series of small watersheds with historical agricultural use, consisting primarily of sugarcane and pineapple production. In addition, the coastal perimeter is heavily influenced by residential and tourist development and activity. Engott and

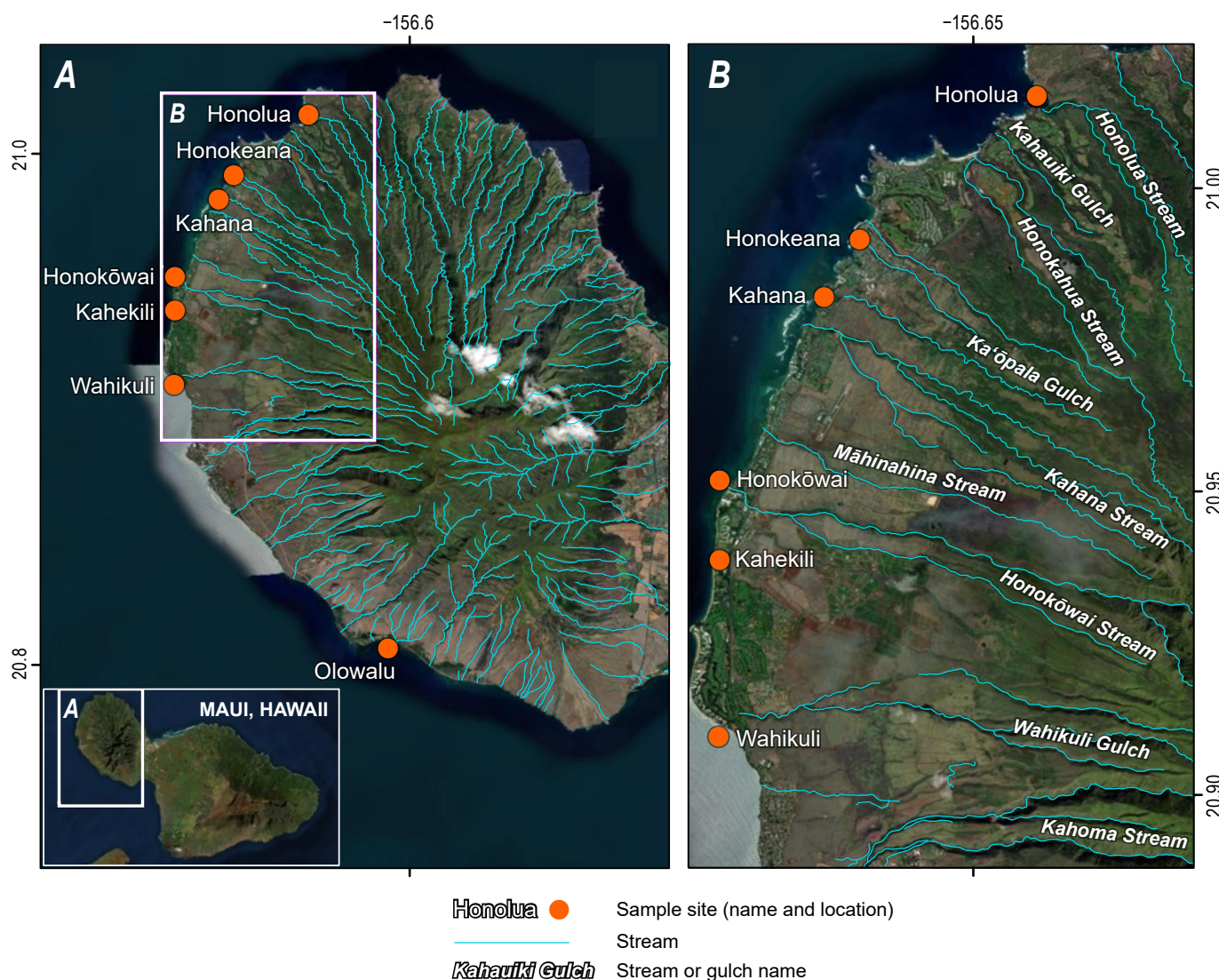


Figure 1. Maps of West Maui, Hawai'i, showing study area and coastal sample sites. A, Map of West Maui annotated with sample site locations and names. B, Inset of map A, covering the extent of all study sites excluding Olowalu.

Table 1. Sampling site locations (latitude and longitude), site watershed type, and deployment depth of samplers (semipermeable membrane devices and polar organic chemical integrative samplers).

[DDM, degrees decimal minutes; m, meters; °, degrees; ', minutes]

| Sample site | Latitude (DDM) | Longitude (DDM) | Watershed type | Water depth (m) of sampler |
|-------------|----------------|-----------------|----------------|----------------------------|
| Honolua | 21° 00.908' | 156° 38.373' | Stream | 26 |
| Honokeana | 20° 59.490' | 156° 40.127' | Stream | 6 |
| Kahana | 20° 58.918' | 156° 40.477' | Stream | 7 |
| Honokōwai | 20° 57.109' | 156° 41.509' | Stream | 26 |
| Kahekili | 20° 56.262' | 156° 41.600' | Stream | 5 |
| Wahikuli | 20° 54.573' | 156° 41.521' | Stream | 19 |
| Olowalu | 20° 48.388' | 156° 36.502' | Stream | 20 |

Vana (2007) estimated the historical land-use changes in West Maui to investigate the effects of rainfall and agricultural land-use changes on groundwater recharge in West Maui and the central Maui isthmus. From the early 1900s until about 1979, land use remained mostly unchanged except for some minor urbanization along the coast. As large-scale plantation agriculture practices declined after 1979, land use changes became more substantial. From 1979 to 2004, agricultural land use declined about 21 percent, mainly from the cessation of large-scale sugarcane production.

The Pioneer Mill Company was the major sugarcane cultivator on the west side of the mountains of West Maui, operating during the late 1800s until 1999, when it ceased sugarcane production on approximately 6,000 acres. Some of the land was subsequently converted to pineapple cultivation, including the area north of Honokōwai Stream. The extent of pineapple agriculture in West Maui decreased starting in the late 1990s and stopped entirely in 2009 (Gingerich and Engott, 2012). Today, large parts of the former sugarcane and pineapple fields remain fallow, whereas other parcels have been converted to low-density housing and diversified agriculture.

Sites chosen for this study were off the coast of Honolua, Honokōwai Beach Park, Kahekili Beach Park, Wahikuli Wayside Park, Honokeana, Olowalu Beach, and Kahana, all of which are in the USCRTF West Maui priority watersheds study area (table 1). The Olowalu Beach site is located approximately 13 km south of the main study area and represents a coastal site with minimal anthropogenic effects, owing to a lack of substantial recent development. The possibility of legacy contaminants at Olowalu (following the cessation of sugarcane operations in the late 1990s) may still represent a source of land-based pollutants, although this was chosen as the “pristine” site (Crites, 2006). Watersheds that drain into these seven sites contain a variety of land use types, including marinas, resorts, agricultural, and residential areas. At the Kahekili Beach Park site, there are multiple active groundwater seeps close to shore (at water depths of usually less than 2 meters), where lower salinity water (salinity less than 10) can be readily observed discharging into the nearshore water column (Swarzenski and others, 2013).

Approach

Chlorinated pesticides, PCB, hormones, pharmaceuticals, flame retardants, and *n*-alkanes were analyzed using passive water samplers placed at stream mouths and areas of known coral reef degradation (for example, Kahekili Beach Park coral reef) to assess the levels and environmental effects of contamination to the coastal waters of West Maui. The composition of organic matter, determined through the analysis of *n*-alkane lipid biomarkers, is frequently used to track terrestrial, marine, and environmental processes occurring in nearshore ecosystems (Waterson and Canuel, 2008). The carbon number and the distribution of straight chain normal alkanes found in organic matter are diagnostic signatures based on biosynthetic pathways, which vary with different vegetation types (terrestrial versus marine) and the microbial community present in the environment.

Passive membrane samplers, such as SPMDs and POCISs, overcome the limitations of sampling either small volumes of water or monitoring with sessile bivalve organisms (Prest and others, 1997; Alvarez and others, 2004; Adams and others, 2007) and have been deployed in a variety of settings (Booij and others, 2014). The diffusion of compounds onto the lipid-containing sampler membranes mimics natural biomembranes in their ability to take up bioavailable organic contaminants (Marrucci and others, 2013). In SPMDs, organic compounds are concentrated on a triolein membrane by partitioning hydrophobic compounds from the dissolved phase in seawater onto the membrane. These compounds are subsequently removed in the laboratory quantitatively through dialysis methods (Alvarez et al, 2004). The POCISs contain a hydrophilic-lipophilic-balanced (HLB) sorbent, a strongly hydrophilic polymer encased between two polyether sulfone membranes.

Many chemical contaminants (for example, PAH, PCB, and organochlorine pesticides) are present in the marine environment in low concentrations (concentration levels of parts per billion [micrograms per liter], or parts per trillion [nanograms per liter]). They are still, however, deleterious to coral reefs at these low levels owing to the effects of sustained bioaccumulation in coral tissue (Knutson and others, 2012). Therefore, discrete water sampling may underestimate the presence of contaminants

because of concentrations that are below the minimum detection level (MDL) in the standard one-liter grab sample volume, or because of temporal variations in contaminant concentrations. In addition, sessile organisms, such as bivalves, may metabolize the compounds of interest and alter the actual concentrations of the parent compound in the water column (Alvarez and others, 2004; Alvarez, 2010), decreasing the calculated exposure amount. The concentration of organic compounds collected in discrete water samples can also be influenced by tidal cycles, changes in the seasonal delivery of terrestrial inputs, or both. In contrast, sample collection techniques using SPMDs and POCISs can integrate over user-defined, longer durations, thus eliminating some sample bias (Alvarez and others, 2004; Arditoglou and Voutsas, 2008). Factors that may influence contaminant uptake rates into SPMDs or POCISs include biofouling, water turbulence, salinity, temperature, and flow rate (Petty and others, 2000).

Relating SPMD and POCIS uptake to coral tissue uptake has limitations that are site-specific and dependent on environmental and physicochemical parameters. Passive samplers do not always adequately measure the bioavailable concentrations of an organic compound (Bourgeault and Gourlay-Francé, 2013) owing to variable uptake rates between organisms and the sampler. For example, Richardson and others (2003) measured consistently higher concentrations of PAH and petroleum hydrocarbons in mussel tissue compared to the value calculated for the passive samplers. The uptake of organic compounds and contaminants by corals, and by their associated zooxanthellae, is dependent on physical factors including temperature, salinity, and pH, which can affect the solubility and particle adsorption rates of compounds.

Analytical Methods

For this study, SPMDs and POCISs were deployed near the seafloor at seven shallow (less than 30 meters deep) coastal sites in West Maui, Hawai'i, from February 21 to March 15, 2017.

The preassembled sampling canisters were shipped to the study sites on ice in airtight metal cans. Each stainless steel canister housed 3 POCISs (with sampling surface area of 41 square centimeters [cm²] and 200 milligrams [mg] of HLB sorbent each) and 3 SPMDs (with 460 cm² of surface area per milliliter [mL] triolein). These were deployed just above the seafloor on rebar posts at each site, according to procedures outlined by Alvarez and others (2004). Performance recovery compounds were added to the membranes by Environmental Sampling Technologies, Incorporated (herein referred to as EST Lab) before deployment to correct for incomplete adsorption of contaminants at the field sites. Field and trip blanks were used at each site during deployment and retrieval by opening the blank canister to the atmosphere along with the collection canisters. Upon retrieval, the canisters were placed back in their respective metal cans and shipped to the laboratory in coolers on ice for processing.

The samples were prepared by dialysis prior to gas chromatography/mass spectrometry or liquid chromatography/mass spectrometry analysis, as described by Alvarez (2010). After

the frozen samplers arrived at EST Lab, any marine debris or biofouling was removed from the canisters with a soft bristle brush for all samplers. Each POCIS and SPMD sampler was carefully opened, and the sorbent was transferred with deionized water into clean solid-phase extraction cartridges. Samples were filtered through a glass-fiber filter (Grade 6, 1.6 micrometer [μ m] nominal retention). The sorbent was subsequently dried by pulling air (by vacuum) through the sorbent bed for 10 minutes.

POCISs were each extracted with 25 mL of methanol (Fisher Scientific, Inc., Optima grade) which was then evaporated to 2–3 mL by rotary evaporation, combined into a single sample for each site, and adjusted to a final volume in methanol of 1 mL. POCIS samples were analyzed by Weck Laboratories (City of Industry, California). A gas chromatograph equipped with an electron capture detector was used to analyze samples for 27 chlorinated pesticides and 7 Aroclor PCB following the procedure in EPA Method 608. Liquid chromatography/mass spectrometry/mass spectrometry electron spray ionization was used to analyze samples for 34 PPCP, and liquid chromatography/mass spectrometry/mass spectrometry atmospheric pressure chemical ionization was used to analyze for 9 hormones following procedures in EPA method 1694. The PCB results are not presented here because all values were below the MDL.

SPMD samples were analyzed for *n*-alkanes and sterols following a micro-pipette column chromatography separation. The fractions were analyzed by splitless injection on an Agilent 6890 gas chromatograph interfaced to an Agilent 5973 mass spectrometer at the U.S. Geological Survey Organic Geochemistry Laboratories in Santa Cruz, California. The gas chromatograph oven program had an initial temperature of 90 degrees Celsius (°C), which was held for 4 minutes before increasing 5 °C per minute to a final temperature of 310 °C. The final temperature was then maintained for 10 minutes. The capillary column (Agilent J&W, DB-5ms; 30-m length, 0.25-millimeter [mm] inner diameter with a 25- μ m phase thickness) was directly interfaced to the ion source of the mass spectrometer. Compound identifications were made by comparison with known standards, published reference spectra, or both (National Institute of Standards and Technology, 2005).

Results and Discussion

Results are presented as total mass in nanograms (ng) of a specific compound, obtained in triplicate analyses, per site. The triplicate samples were combined during the processing steps to ensure detectable concentrations. The results reported here are qualitative, as absolute concentrations cannot be calculated without flow-rate data. These results represent a 22-day integrated water sample for each site that can be compared within and between sites in West Maui to describe the presence or absence of specific organic contaminants. The uptake of contaminants is assumed to be linear throughout the deployment of each passive sampler (Alvarez and others, 2004).

n-alkanes

Terrestrial biomarkers (long-chain normal alkanes for this study) are reliable indicators of terrestrial organic matter inputs to nearshore waters. Total *n*-alkane concentrations (an average of 27.2 ng per sampler) from the SPMDs reflect a predominantly marine signature, with an average value per sampler for combined marine *n*-alkanes of 21.5 ng, while the terrestrial *n*-alkanes had an average value of 3.9 ng, a fivefold difference. The *n*-alkanes had a carbon range from C₁₁ to C₃₁ (where the subscript describes the number of carbon atoms in

the molecule), with most hydrocarbons less than C₂₅ (fig. 2; table 2) reflecting marine and (or) bacterial inputs. These data are published in a companion data release (Campbell and others, 2022). The molecule with the highest carbon number out of all sites was pentadecane, a microbially (cyanobacteria-) produced alkane. The presence of pentadecane may reflect the increase in aqueous solubility of the lower molecular weight *n*-alkanes, whereas the higher molecular weight *n*-alkanes, representing terrestrial inputs, may be associated with particles (for example, colloids), and would not contribute to the truly dissolved phase sampled by the SPMDs.

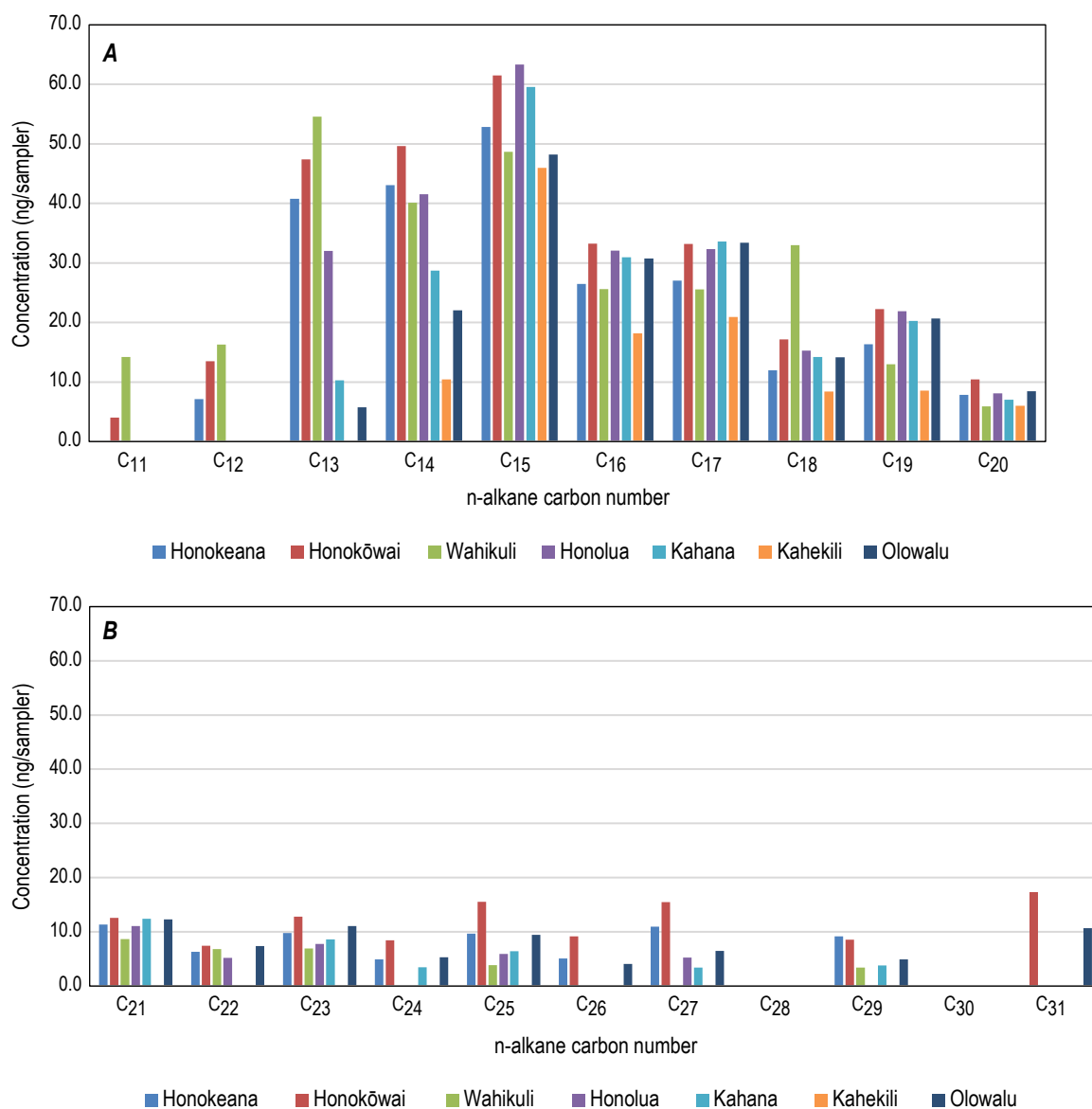


Figure 2. Bar graphs showing concentrations (in nanograms per sampler [ng/sampler]) of normal alkanes (*n*-alkanes) in semipermeable membrane devices after a 22-day deployment at the 7 coastal stream sites along the coast of West Maui. A, Concentrations of low molecular weight (LMW) *n*-alkanes at each site, where LMW is defined as less than 21 carbons. B, Concentrations of high molecular weight (HMW) *n*-alkanes at each site, where HMW is defined as more than 20 carbons.

Table 2. Normal alkane (carbon numbers C₁₁ through C₃₁) concentrations in nanograms per sampler from semipermeable membrane devices deployed at 7 sites for 22 continuous days.[Minimum detection level (MDL) is 2.0 nanograms per milliliter. *n*-alkane, normal alkane]

| <i>n</i> -alkane carbon number | Sample site | | | | | | |
|--------------------------------------|-------------|-----------|----------|---------|--------|----------|---------|
| | Honokeana | Honokōwai | Wahikuli | Honolua | Kahana | Kahekili | Olowalu |
| C ₁₁ | 0.0 | 4.1 | 14 | 0.0 | 0.0 | 0.0 | 0.0 |
| C ₁₂ | 7.2 | 14 | 16 | 0.0 | 0.0 | 0.0 | 0.0 |
| C ₁₃ | 41 | 47 | 55 | 32 | 10 | 0.0 | 5.8 |
| C ₁₄ | 43 | 50 | 40 | 42 | 29 | 10 | 22 |
| C ₁₅ | 53 | 62 | 49 | 63 | 60 | 46 | 48 |
| C ₁₆ | 27 | 33 | 26 | 32 | 31 | 18 | 31 |
| C ₁₇ | 27 | 33 | 26 | 32 | 34 | 21 | 33 |
| C ₁₈ | 12 | 17 | 33 | 15 | 14 | 8.4 | 14 |
| C ₁₉ | 16 | 22 | 13 | 22 | 20 | 8.6 | 21 |
| C ₂₀ | 7.8 | 10 | 5.9 | 8.1 | 7.0 | 6.0 | 8.5 |
| C ₂₁ | 11 | 13 | 8.7 | 11 | 12 | 0.0 | 12 |
| C ₂₂ | 6.3 | 7.4 | 6.8 | 5.2 | 0.0 | 0.0 | 7.4 |
| C ₂₃ | 9.8 | 13 | 7.0 | 7.8 | 8.6 | 0.0 | 11 |
| C ₂₄ | 4.9 | 8.5 | 0.0 | 0.0 | 3.4 | 0.0 | 5.3 |
| C ₂₅ | 9.7 | 16 | 3.8 | 5.9 | 6.4 | 0.0 | 9.4 |
| C ₂₆ | 5.1 | 9.1 | 0.0 | 0.0 | 0.0 | 0.0 | 4.1 |
| C ₂₇ | 11 | 15 | 0.0 | 5.3 | 3.4 | 0.0 | 6.5 |
| C ₂₈ | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| C ₂₉ | 9.2 | 8.6 | 3.4 | 0.0 | 3.8 | 0.0 | 4.9 |
| C ₃₀ | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| C ₃₁ | 0.0 | 17 | 0.0 | 0.0 | 0.0 | 0.0 | 11 |

Sterols

The total sterol concentrations per SPMD sampler are not reported—the concentrations were low and field blank concentrations exceeded the stream site concentrations. This may not reflect the actual concentrations of sterols in these samples because the gas chromatograph mass spectrometer was inoperable, and therefore there was a delay in analysis and injection following derivatization of the sterol compounds. Analysis of derivatized sterols after long term storage (greater than 6 months) may lead to degradation of the trimethylsilyl ether sterols.

Insecticides, Pesticides, and PCB

No PCB, insecticides, or pesticides were detected in the POCISs. See appendix 1 for a list of nondetected compounds and their minimum reporting levels, or limits of quantitation. The absence (or lack of detection) of PCB, insecticides or pesticides may reflect the low level of current agricultural practices in areas near the POCIS sites, a lack of long-range deposition during the sampling period, and (or) the rapid dilution of compounds entering the marine environment during the study period. The complete absence of legacy pesticides during the sample interval is also worth noting, though sampling took place during a

drier-than-expected wet season, and the flow rates of the seasonal streams were low due to drought conditions prevalent at the time the samplers were in the water.

Pharmaceuticals and Personal Care Products (PPCP)

In the United States, there are over 4,000 approved pharmaceutical products, including prescription and over-the-counter medications, which represent a wide range of chemical classes (U.S. Food and Drug Administration, 2015). For many of these products, there is likely a seasonality in concentration levels off the coast of West Maui, influenced by peak tourism periods and usage rates per person.

Fourteen polar organic compounds (PPCP and flame retardants) were identified in POCISs at the sites along the coast of West Maui (fig. 3; table 3). These data are published in the companion data release (Campbell and others, 2022). The highest concentrations of these compounds were found at the Honokōwai Stream site and the highest number of detects (13) occurred at the Kahekili Beach Park site (table 3). The two most abundant compounds per sampler were galaxolide (320 ng per sampler) and the flame retardant tris(1-chloro-2-propyl) phosphate (TCPP) (160 ng per sampler), which were found at the Honokōwai Stream

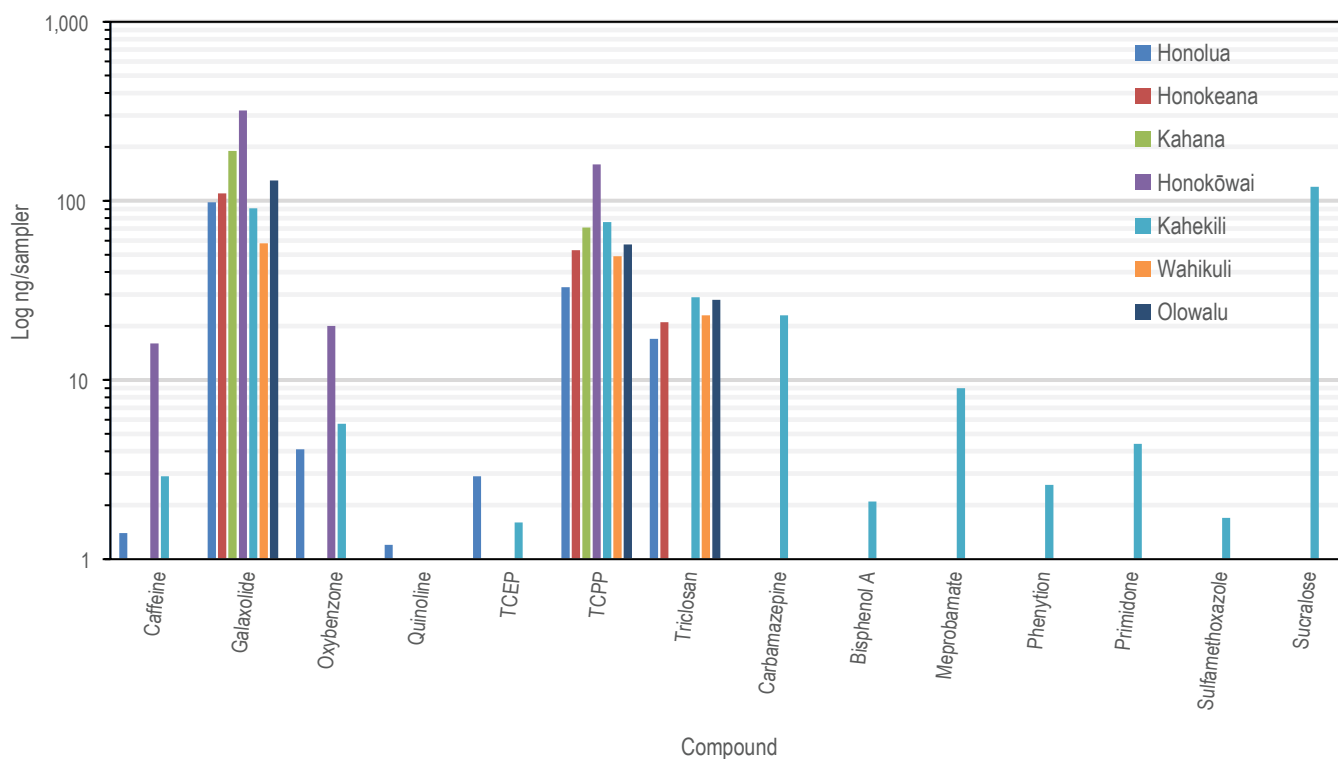


Figure 3. Logarithmic (log) concentrations in nanograms per sampler (ng/sampler) of pharmaceuticals and personal care products as well as flame retardants, measured by polar organic chemical integrative samplers. Abbreviations: TCEP, tris(2-chloroethyl) phosphate; TCP, tris(1-chloro-2-propyl) phosphate.

Table 3. Concentrations per sampler of polar organic contaminants (pharmaceuticals, personal care products, and pesticides) detected in polar organic chemical integrative samplers during the 22-day deployment.

[Concentrations in nanograms per sampler. MRL, minimum reporting level or limit of quantitation; ng/mL, nanograms per milliliter; —, non-detect; TCEP, tris(2-chloroethyl) phosphate; TCP, tris(1-chloro-2-propyl) phosphate]

| Analyte | Sample site | | | | | | | MRL (ng/ml) |
|---------------------|-------------|-----------|----------|---------|--------|----------|---------|-------------|
| | Honokeana | Honokōwai | Wahikuli | Honolua | Kahana | Kahekili | Olowalu | |
| Bisphenol A | — | — | — | — | — | 2.1 | — | 1.0 |
| Triclosan | 21 | — | 23 | 17 | — | 29 | 28 | 2.0 |
| Caffeine | — | 16 | — | 1.4 | — | 2.9 | — | 1.0 |
| Carbamazepine | — | — | — | — | — | 23 | — | 1.0 |
| Galaxolide | 110 | 320 | 58 | 98 | 190 | 91 | 130 | 10 |
| Meprobamate | — | — | — | — | — | 9 | — | 1.0 |
| Oxybenzone | — | 20 | — | 4.1 | — | 5.7 | — | 1.0 |
| Phenyton (Dilantin) | — | — | — | — | — | 2.6 | — | 1.0 |
| Primidone | — | — | — | — | — | 4.4 | — | 1.0 |
| Quinoline | — | — | — | 1.2 | — | — | — | 1.0 |
| Sucralose | — | — | — | — | — | 120 | — | 5.0 |
| Sulfamethoxazole | — | — | — | — | — | 1.7 | — | 1.0 |
| TCEP | — | — | — | 2.9 | — | 1.6 | — | 1.0 |
| TCP | 53 | 160 | 49 | 33 | 71 | 76 | 57 | 1.0 |

sample site. Oxybenzone was also detected at the sites off the coast of Honokōwai Stream (a master-planned town) and Kahekili Beach Park, two popular tourism destinations with many hotels and resorts. Oxybenzone is an active ingredient in many sunscreens and is found to cause coral bleaching, damage coral deoxyribonucleic acid (DNA) and harm coral larvae. Hawai'i became the first state in the nation to ban the sale and distribution of sunscreen products containing oxybenzone in January 2018, following the current study.

TCPP, which was detected at all sites, is the most widely distributed chlorinated organophosphorus flame retardant in the marine environment (Mercurio and others, 2021) and is an emerging environmental contaminant of concern. In addition, galaxolide was found at all sites in abundance. It is a heterocyclic lipophilic emerging contaminant of concern in the aquatic environment that is used as a fragrance (synthetic musk) in many cosmetics and other personal care products. It has been detected in remote areas far from its source and is considered the most abundant musk fragrance found in the marine environment. Because of its lipophilicity, it has been found in a variety of marine organisms (Ramirez and others, 2009; Shek and others, 2008), including in apex predators as a bioaccumulator (Tumová and others, 2019). Parolini and others (2015) found that galaxolide exposures in *Dreissena polymorpha* (zebra mussels) can induce genetic and oxidative damage. Owing to its presence at all stream sites and its possible deleterious effects to the coral reef environment, galaxolide is a contaminant, along with TCPP, in which continued monitoring would offer resource managers critical information to understand.

Summary

Passive membrane samplers (semipermeable membrane devices and polar organic chemical integrative samplers) were deployed for 22 continuous days at 7 coastal sites in West Maui to examine inputs of organic compounds (pesticides, polychlorinated biphenyls, pharmaceutical and personal care products, and flame retardants) to shallow coral reef ecosystems. The use of passive samplers allowed for almost one month of continuous, integrated sampling to accommodate fluctuations in daily and weekly inputs that would not be detected by grab water samples. The distribution of organic compounds observed at these sites was variable and indicated a predominance of marine-sourced organic matter; however, direct correlations to upstream land-use practices were not obvious because many compounds were below quantitation limits. There were distinct differences between the strongly groundwater-influenced Kahekili Beach Park site and the other coastal sites, which were influenced more by seasonal surface and stream water runoff. Tris(1-chloro-2-propyl) phosphate, tris(2-chloroethyl) phosphate, and a wide range of PPCP were detected at the Kahekili Beach Park site. The results presented here provide a snapshot of contaminants and inputs to the nearshore during a drier-than-expected wet season and can be useful for future evaluations and comparisons. At the time of sampling

(February and March), West Maui was under drought conditions, although these conditions ended with large rain events at the end of April 2017. Sampling during and after a wet season to evaluate runoff containing legacy and emerging contamination would be a next logical step in characterizing inputs in West Maui.

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Appendix 1. Supplemental Data of Polar Organic Contaminants

Table 1.1. List of polar organic contaminants including polychlorinated biphenyls, insecticides/pesticides, pharmaceutical and personal care products, and flame retardants that were not detected in polar organic chemical integrative samplers at the minimum reporting level or limit of quantitation.

[PCBs, polychlorinated biphenyls; PPCP, pharmaceutical and personal care products; DDD, dichlorodiphenyldichloroethane; DDE, dichlorodiphenyldichloroethylene; DDT, dichlorodiphenyltrichloroethane; BHC, benzene hexachloride; DEET, n,n-diethyl-meta-toluamide; TDCPP, tris(1,3-dichloroisopropyl)phosphate; MRL, minimum reporting level; ng/mL, nanograms per milliliter; —, not detected.

| Analyte | MRL (ng/mL) |
|-------------------------|-------------|
| PCBs | |
| Aroclor 1016 | 100 |
| Aroclor 1221 | — |
| Aroclor 1232 | — |
| Aroclor 124 | — |
| Aroclor 1248 | — |
| Aroclor 1254 | — |
| Aroclor 1260 | — |
| Insecticides/pesticides | |
| 2,4'-DDD | 5.0 |
| 2,4'-DDE | 5.0 |
| 2,4'-DDT | 5.0 |
| 4,4'-DDD | 5.0 |
| 4,4'-DDE N | 5.0 |
| 4,4'-DDT | 5.0 |
| Aldrin | 5.0 |
| alpha-BHC | 5.0 |
| alpha-Chlordane | 5.0 |
| beta-BHC | 5.0 |
| Chlordane (tech) | 100 |
| cis-Nonachlor | 5.0 |
| delta-BHC | 5.0 |
| Dieldrin | 5.0 |
| Endosulfan I | 5.0 |
| Endosulfan II | 5.0 |
| Endosulfan sulfate | 5.0 |
| Endrin | 5.0 |
| Endrin aldehyde | 5.0 |
| gamma-BHC (Lindane) | 5.0 |
| gamma-Chlordane | 5.0 |
| Heptachlor | 5.0 |
| Heptachlor epoxide | 5.0 |
| Methoxychlor | 5.0 |
| Mirex | 5.0 |
| Toxaphene | 500 |
| PPCP and Hormones | |
| 17-a-Estradiol | 10 |
| 17-a-Ethinylestradiol | 10 |
| 17-b-Estradiol | 10 |
| Diethylstilbestrol | 50 |

Table 1.1. List of polar organic contaminants including polychlorinated biphenyls, insecticides/pesticides, pharmaceutical and personal care products, and flame retardants that were not detected in polar organic chemical integrative samplers at the minimum reporting level or limit of quantitation.—Continued

[PCBs, polychlorinated biphenyls; PPCP, pharmaceutical and personal care products; DDD, dichlorodiphenyldichloroethane; DDE, dichlorodiphenyldichloroethylene; DDT, dichlorodiphenyltrichloroethane; BHC, benzene hexachloride; DEET, n,n-diethyl-meta-toluamide; TDCPP, tris(1,3-dichloroisopropyl)phosphate; MRL, minimum reporting level; ng/mL, nanograms per milliliter; —, not detected.

| Analyte | MRL (ng/mL) |
|-----------------------------|-------------|
| PPCP and Hormones—Continued | |
| Epitestosterone | 10 |
| Estriol | 10 |
| Estrone | 10 |
| Progesterone | 10 |
| Testosterone | 10 |
| PPCP and Pharmaceuticals | |
| Diclofenac | 1.0 |
| Gemfibrozil | 1.0 |
| Ibuprofen | 1.0 |
| Iopromide | 5.0 |
| Naproxen | 1.0 |
| Salicylic Acid | 50 |
| Acetaminophen | 20 |
| Amoxicillin | 10 |
| Atenolol | 1.0 |
| Atorvastatin | 1.0 |
| Azithromycin | 10 |
| Ciprofloxacin | 5.0 |
| Cotinine | 2.0 |
| DEET | 1.0 |
| Diazepam | 1.0 |
| Fluoxetine | 1.0 |
| Methadone | 1.0 |
| Praziquantel | 1.0 |
| Trimethoprim | 1.0 |
| Flame Retardants | |
| TDCPP | 1.0 |

