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

The Use of Passive Membrane Samplers to Assess Organic Contaminant Inputs at Five Coastal Sites in West Maui, Hawaii

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

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

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The Use of Passive Membrane Samplers to Assess Organic Contaminant Inputs at Select Coastal Sites in West Maui, Hawaii

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

Abstract

Five passive membrane samplers were deployed for 28 continuous days at select sites along and near the west Maui coastline to assess organic compounds and contaminant inputs to diverse, shallow coral reef ecosystems. Daily and weekly fluctuations in such inputs were captured on the membranes using integrative sampling. The distribution of organic compounds observed at these five coastal sites showed considerable variation; with high concentrations of terrestrially sourced organic compounds such as C29 sterols and high molecular weight *n*-alkanes at the strongly groundwater-influenced Kahekili vent site. In comparison, the coastal sites were presumably influenced more by seasonal surface and stream water runoff and therefore had marine-sourced organic compounds and fewer pharmaceuticals and personal care products. The direct correlation to upstream land-use practices was not obvious and may require additional wet-season sampling. Pharmaceuticals and personal care products as well as flame retardants were detected at all sites, and the Kahekili vent site had the highest number of detections. Planned future work must also determine the organic compound and contaminant concentrations adsorbed onto water column particulate matter, because it may also be an important vector for contaminant transport to coral reef ecosystems. The impact of contaminants per individual (such as fecundity and metabolism) as well as per community (such as species abundance and diversity) is necessary for an accurate assessment of environmental stress. Results presented herein provide current contaminant inputs to select nearshore environments along the west Maui coastline captured during the dry season, and they can be useful to aid potential future evaluations and (or) comparisons.

Introduction

Many coral reefs offshore of the main Hawaiian Islands have been in decline since the 1990s, primarily related to excess nutrient loading, algal bloom, shoreline development, and agricultural runoff (Friedlander and others, 2008; Dailer and others, 2012). Long-term monitoring of coral reefs along the west Maui coastline, Hawaii, has shown that the coral cover has declined by as much as 50 percent in some highly impacted areas (Bruno and Selig, 2007). Runoff and associated transport of terrestrial inorganic constituents and organic compounds has impacted nearshore ecosystems, including coral reefs, and the deleterious effects may last longer than previously suggested (Morrison and others, 2013; Brodie and others, 2012; Fabricius, 2005; Anthony and others, 2004). To assess surface- and groundwater-derived organic loading rates along the U.S. Coral Reef Task Force (USCRTF) west Maui priority watersheds study area, a passive membrane sampling study was conducted at five sites along the west Maui coast, some of which showed sustained and widespread groundwater discharge (Glenn and others, 2013; Swarzenski and others, 2013, 2016) and others that are more influenced by surface water runoff from intermittent streams. Passive samplers were deployed off the Wahikuli and Honokowai watersheds in the USCRTF west Maui priority watersheds study area to capture the effects of surface water runoff, another sampler was deployed in Kahekili Beach Park to capture the effects of submarine groundwater discharge, and the remaining sampler was deployed off Olowalu to capture the possible effects of active coastal development. At the Kahekili site there are multiple spring vents close to shore (water depths are usually less than 2 meters [m]), where much lower salinity water (<10 practical salinity units [psu]) can readily be observed discharging into the nearshore water column (Swarzenski and others, 2013). The main objectives of this study were to (1) determine the feasibility of using two types of passive samplers: semi-permeable membrane device, (SPMD) and a polar organic chemical integrated sampler, (POCIS), in the shallow waters of west Maui, Hawaii, where the predominant transport pathways of terrestrial material are submarine groundwater discharge or surface water runoff; (2) identify suites of pollutant classes in these different watersheds; and (3) determine ambient, dry-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, as well as the possible deleterious effects of land-based contaminants on the coral reef system of west Maui.

Study Sites

Some nearshore coral reefs off west Maui have recently undergone a dramatic and well-documented ecosystem shift from what was once a healthy coral reef to one that is currently dominated by turf algae (Vermeij and others, 2010). Of note, reduced coral cover, increased coral mortality, and decreased recruitment observed off Kahekili Beach Park in the Hawaii Department of Land and Natural Resources, Division of Aquatic Resources (DLNR/DAR) Herbivore Fisheries Management Area (FMA) are of particular concern, especially with the recent evidence of a hydraulic link to the Lahaina Wastewater Reclamation Facility (WWRF) (Glenn and others, 2013). This facility serves the municipal wastewater needs for the community and several major resorts located along the Maui coast. In 2014, the Lahaina WWRF was found in violation of the Clean Water Act for discharging millions of liters of treated wastewater into injection wells that delivered tainted groundwater to nearshore waters. This facility uses as many as four gravity-fed EPA Class V injection wells (wells that are used to inject non-hazardous fluids underground) to dispose of 11–19 million liters (3–5 million gallons) of wastewater effluent daily. The series of wells are located approximately 460 to 580 m from the shoreline of west Maui at about 10 m above sea level (https://www3.epa.gov/region09/water/groundwater/uic-pdfs/lahaina/Lahaina-renewal-SOB-final.pdf). The facility produces treated wastewater (tertiary treated with filtration) and, since October 2011, has been disinfected with chlorine to an R-2 standard, which is disposed of through four on-site injections wells. Prior research at this site based on nitrate-nitrogen isotopes (Dailer and others, 2010, 2012), as well as a suite of diagnostic organic municipal wastewater pollutants (Hunt and Rosa, 2009), has documented that wastewater effluent was carried by groundwater onto the shallow reef through a series of submarine vents (submarine springs) (Dailer and others, 2012, Swarzenski and others, 2013).

The sampling sites chosen for this study represent a variety of environments along the west Maui coast (fig. 1). The land can be categorized as a series of small watersheds with agricultural use as well as legacy use for sugarcane and pineapple production. Today, the coastal perimeter is heavily influenced by residential and tourist activity. Engott and Vana (2007)

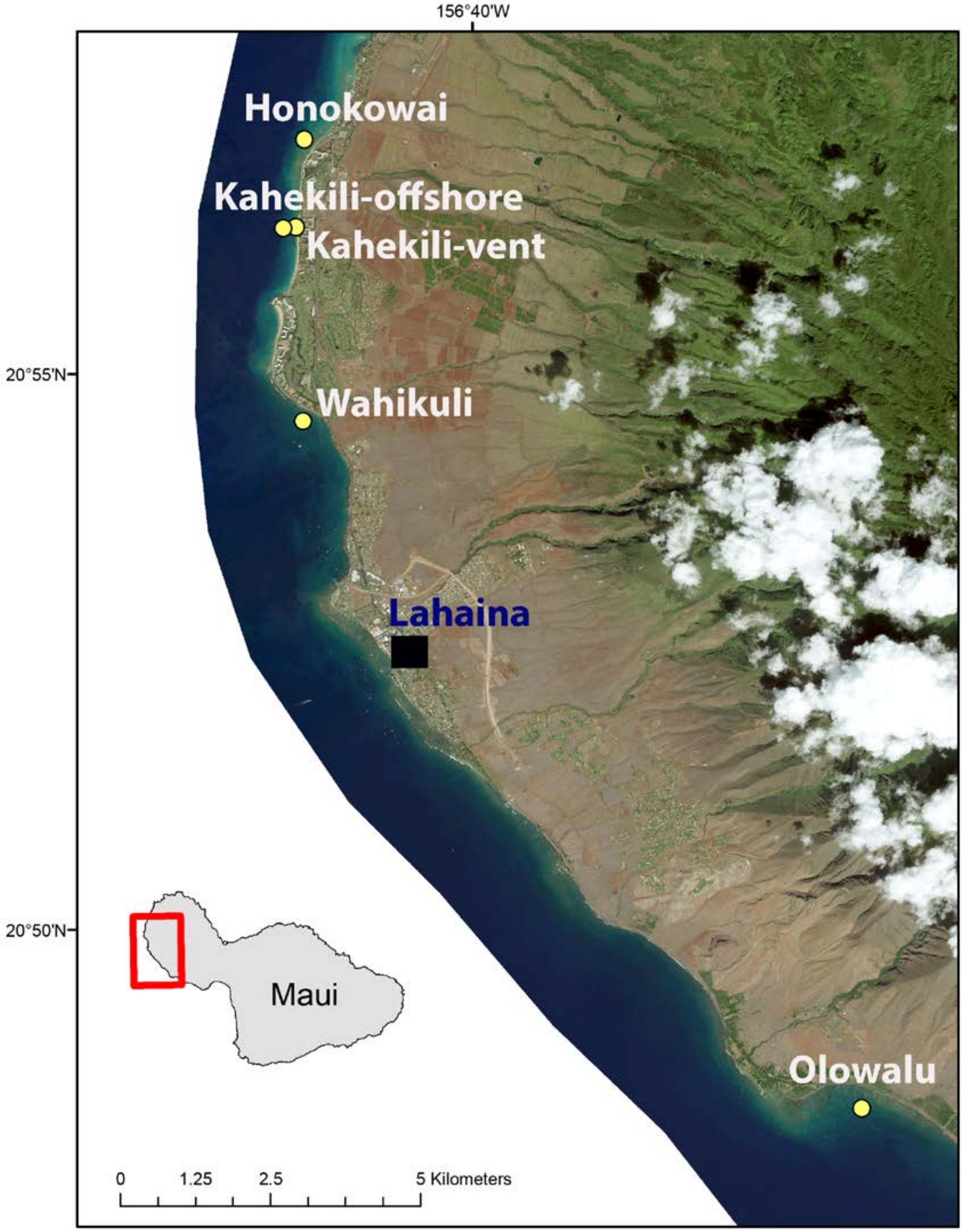

Figure 1. Location map of the five coastal sites along west Maui, Hawaii where passive membrane samplers were deployed for 28 continuous days.

estimated the historical land-use changes in west Maui to estimate the effects of rainfall and agricultural land-use changes on west and central Maui groundwater recharge. During the early 1900s until about 1979, land use was mostly unchanged except for some minor urbanization along the coast. As large-scale plantation agriculture declined after 1979, land-use changes became more significant. From 1979 to 2004, agricultural land use declined about 21 percent, mainly from the large-scale cessation of sugarcane production. The Pioneer Mill Company was the major sugarcane cultivator on the west side of the West Maui Mountains, 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 Honokowai Stream. The extent of pineapple agriculture in west Maui decreased extensively since the late 1990s, and it 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.

Passive membrane samplers were also deployed off Honokowai Beach Park, Kahekili Beach Park (one in shallow water adjacent to the primary submarine groundwater vents, and one offshore in deeper water), Wahikuli Wayside Park in the USCRTF west Maui priority watersheds study area, and off Olowalu approximately 10 kilometers (km) south of Lahaina where coastal development is beginning. The Olowalu site is located ~13 km south of the main study area and represents water with minimal anthropogenic impact owing to lack of development, although the possibility of legacy contaminants following the cessation of sugarcane operations in the late 1990s may still represent a source of land-based pollutants (Crites, 2006). Watersheds that drain these sites contain a variety of land-use types, including marinas, resorts, agriculture, and residential areas (table 1).

Table 1. Sampling dates, site locations (latitude and longitude), watersheds, and description (SPMD/POCIS or discrete water sample).

Sample Site	Latitude [d.deg]	Longitude [d.deg]	Watershed	Description	Sample Date
Honokowai	20.9518	156.6918	stream	SPMD/POCIS 1	12-Oct-2014
Kahekili - Vent	20.9451	156.6932	vent/groundwater	SPMD/POCIS 2	12-Oct-2014
Kahekili-offshore	20.9373	156.6935	offshore	SPMD/POCIS 3	12-Oct-2014
Olowalu	20.8065	156.6083	surface	SPMD/POCIS 4	12-Oct-2014
Wahikuli	20.9096	156.6920	stream	SPMD/POCIS 5	12-Oct-2014
Kahekili - Vent water 1	20.9451	156.6932	vent/groundwater	Vent 1	10-Jul-2013
Kahekili - Vent water 2	20.9451	156.6932	vent/groundwater	Vent 2	10-Jul-2013

Approach

Passive membrane samplers such as SPMD, POCIS, and other types of polyethylene membranes overcome the limitation 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 here in a variety of sites (Booij and others, 2014). The diffusion of compounds onto the lipid-containing sampler membranes mimic biomembranes in their ability to take up bioavailable organic contaminants (Marrucci and others, 2013). Compounds of interest are concentrated on the SPMDs triolein membrane, which are subsequently removed quantitatively through dialysis. The POCIS are another type of passive samplers containing Oasis hydrophilic-lipophilic-balanced (HLB) sorbent, a strongly hydrophilic polymer encased between two polyether sulfone membranes. Performance recovery compounds (PRC) were added by Environmental Sampling Technologies (EST) before deployment to correct for incomplete adsorption of contaminants at the field sites. Many chemical contaminants (for example, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), organochlorine pesticides) are present in the marine environment in very low concentrations (parts per billion (µg/L), or even parts per trillion (ng/L)), but they may still be deleterious to coral reefs owing to the effects of sustained bioaccumulation in coral tissue (Knutson and others, 2012). Therefore, discrete water sampling may underestimate the presence of contaminants due to concentrations being below the minimum detection level in the standard one-liter grab sample volume or temporal variations. Sessile organisms, such as bivalves, metabolize the compounds of interest and cause difficulty quantifying the concentrations of the parent compounds in the water column (Alvarez and others, 2004; Alvarez, 2010). The concentration of organic compounds collected in discrete water samples can also be influenced by tidal cycles and (or) changes in the seasonal delivery of terrestrial inputs. In contrast, SPMD and POCIS techniques may integrate, over user-defined, longer durations, thus eliminating some sample bias (Alvarez and others, 2004; Arditsoglou and Voutsa, 2008). Factors that may influence contaminant uptake rates into SPMD or POCIS include biofouling, water turbulence, salinity, temperature, and flow rate (Petty and others, 2000). Relating SPMD and POCIS uptake to coral tissue uptake is site specific and dependent on environmental and physicochemical parameters. Passive samplers do not always adequately measure the amount of bioavailable concentrations of an organic compound (Bourgeault and Gourlay-Francé, 2013) owing to differences in their uptake rates. For example, Richardson and others (2003) reported consistently higher concentrations in mussel tissue of PAHs and petroleum hydrocarbons compared to the value calculated for the passive samplers. Uptake of organic compounds and contaminants by corals, and their associated zooxanthellae, is dependent on physical factors including temperature, salinity, and pH, which can affect the solubility and particle adsorption rates of compounds.

The composition of the organic matter through the analysis of lipid biomarkers is often used to track terrestrial, marine, and environmental processes occurring in nearshore ecosystems (Waterson and Canuel, 2008). The carbon number and distribution of straight chain normal-alkanes (*n*-alkanes) show diagnostic signatures based on biosynthetic pathways that vary based on vegetation type (terrestrial versus marine). The analysis of sterols and *n*-alkanes can provide insight into the transport of compounds from the terrestrial environment to nearshore waters.

Methods

For this study, SPMD and POCIS were deployed near the seafloor at five unique, shallow (<20m), coastal sites along west Maui, Hawaii, from October 12, 2014 to November 10, 2014. SPMDs are passive samplers containing a triolein membrane that partitions hydrophobic compounds from the dissolved phase in seawater onto the membrane.

The preassembled canisters were shipped to the study sites on ice in airtight metal cans. A stainless steel canister, housing three POCISs (41 square centimeters [cm^2] sampling surface area, 200 milligrams [mg] of Oasis HLB each) and three SPMDs (460 cm^2 surface area per milliliter [mL] triolein), was deployed just above the seafloor on rebar posts at each of the five study sites, according to procedures outlined by Alvarez and others (2004). Deployment and retrieval time was less than 3 minutes for all but one of the canisters; at Wahikuli Wayside Park, the metal wing nut was difficult to remove, which exposed this sampler to ambient air for less than 10 minutes. Upon retrieval, the canisters were placed back in their respective metal cans and shipped to the laboratory, in coolers on ice, for processing.

The three POCISs and SPMDs from each site were combined following extraction into one SPMD and one POCIS sample per site. POCISs and SPMDs were removed from the metal cans at EST laboratories and rinsed with deionized water to remove any particles that may have fallen into the extraction cartridges. The samples were prepared by dialysis prior to Gas Chromatography/Mass Spectrometry (GC/MS) or Liquid Chromatography/Mass Spectrometry (LC/MS) as described by EST standard operating procedures (http://est-spmd.com/spmd.php).

Any marine debris or biofouling was first removed with a soft bristle brush, each POCIS and SPMD sampler was carefully opened, and then the sorbent was transferred with DI water into clean solid-phase extraction (SPE) cartridges (25 mL, Biotage, Charlotte, NC). Samples were then filtered through a glass-fiber filter (GFF) (Fisher, G-6 1.6 micrometer [µm] nominal retention). The sorbent was subsequently dried by pulling air (by vacuum) through the sorbent bed for 10 minutes. POCISs for the pharmaceuticals and pesticides were each extracted with 25 mL of methanol (Optima grade, Fisher Scientific Inc.), which was subsequently evaporated to 2–3 mL by rotary evaporation, combined into a single sample, and then adjusted to a final volume in methanol of 1 mL.

In an effort to further characterize the organic matter being discharged onto the reef flat from the submarine vents at Kahekili, two discrete 4 liter (L) vent water particulate and dissolved samples were collected during 2013 using Teflon tubing connected to a stainless steel piezometer tip, which was inserted into the main active vent. Vent water was pumped, using a peristaltic pump, through precombusted GFF filters. Although these discrete vent water samples were not collected during the SPMD/POCIS deployment, they do provide a snapshot of the types of organic matter present in the vent water. Filter particulates were extracted using a Dionex Accelerated Solvent Extractor (ASE, Dionex Corp, USA), Optima grade solvents, and a two phase extraction: 1. Hexane: Acetone–1:1; 2. Dichloromethane: methanol– 2:1. Samples were concentrated using a Biotage TurboVap system with high-purity N_2 to a final volume of 1.0 mL. Samples were analyzed for *n*-alkanes and sterols by splitless injection onto an Agilent 6890 gas chromatograph interfaced to a mass spectrometer at the USGS Organic Geochemistry Laboratories in Santa Cruz, California. The gas chromatograph oven program had an initial temperature of 90°C, which was held for 4 minutes then ramped at 5°C/minute to a final temperature of 310°C and maintained for 10 minutes. The capillary column (DB-5MS: 30 m length, 0.25 mm ID with a 25 µm phase thickness) was directly interfaced to the ion source of the mass spectrometer. Compound identifications were made by comparison with known standards and (or) published reference spectra.

POCIS samples were analyzed by Weck Laboratories (City of Industry, Calif.). The GC/MS was used to analyze samples for 27 chlorinated pesticides and 7 Aroclor PCBs (EPA Method 6080). Liquid chromatography/mass spectrometry/mass spectrometry electron spray ionization (LC/MS/MS-ESI) was used to analyze samples for 34 pharmaceuticals and personal care products (PPCPs) (EPA method 1694). The PCB results are not presented here.

Results and Discussion

Surface-water runoff and groundwater discharge can deliver a variety of contaminants to nearshore environments, including sensitive tropical coral reef ecosystems (Loos and others, 2010). Land-derived inputs may include pesticides, petroleum hydrocarbons, trace metals, and wastewater compounds such as PPCP (Focazio and others, 2008). Petroleum hydrocarbons (from asphalt sealer, boat bilge water, engine exhaust, runoff, and atmospheric deposition) are associated with paved coastal roads, and vehicle and boat activity (Latimer and others, 1990).

Long-range atmospheric transport of contaminants, including legacy and current-use pesticides (such as dichlorodiphenyltrichloroethane (DDT)), PAHs, and PCBs, 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). No insecticides or pesticides were detected in the POCISs. This may reflect the low level of current agriculture practices in areas near the POCIS sites, 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 the dry season when the seasonal streams were not flowing.

In contrast to legacy contaminants, PPCP may be continuously discharged to the nearshore in runoff via streams and rivers, as well as from submarine springs that are hydraulically linked to uplands watersheds that may house, for example, sewage treatment plants. Vigorous hydrodynamics in nearshore environments, significant dilution, and diffusion of runoff make detection and ecological exposure difficult to quantify. The affects of low level chronic contamination on vulnerable marine ecosystems is still mostly unknown.

Over the past decade, there has been a notable change in bottom type off Kahekili Beach Park in west Maui; areas once covered by abundant corals are now mostly covered by turf algae or macro algae, suggesting a prevailing nutrient imbalance (Vermeij and others, 2010). Owing to these findings, the site has been studied extensively to assess ecosystem impacts from upstream municipal wastewater discharges, which may be conveyed to the coastal waters by groundwater (Hunt and Rosa, 2009; Dailer and others, 2012; Miller-Pierce and Rhoads, 2016).

Results are presented as total mass in nanograms (ng) of a specific compound, obtained in triplicate analyses, per site. The results reported here are qualitative, as absolute concentrations cannot be calculated without flow-rate data. The results represent a 28-day integrated water sample that can be compared within and between sites in west Maui to describe the presence and (or) absence of specific organic contaminants. The uptake of contaminants is assumed to be linear throughout the deployment of the passive sampler (Alvarez and others, 2004).

Sterols and n-Alkanes

Terrestrial biomarkers (such as C29 sterols and long-chain normal alkanes) are the most reliable indicators of terrestrial organic matter inputs to nearshore waters. Among the five sites, the shallow Kahekili vent site had the highest relative concentrations of the terrestrial sterol biomarker β-sitosterol and terrestrial *n*-alkane biomarker C27. Total *n*-alkane concentrations from SPMDs reflect a predominantly marine signature, with average values per sampler for combined marine *n*-alkanes of 21.8 ng, while the terrestrial *n*-alkanes had an average value of 4.2

ng, a five-fold difference. The *n*-alkanes had a carbon range from C12 to C27, with the majority of hydrocarbons <C25 (fig. 2, table 2) reflecting a profile of marine/bacterial inputs. This may reflect the increase in aqueous solubility of the lower molecular weight alkanes, whereas the higher molecular weight *n*-alkanes representing terrestrial inputs may be associated with particles (for example, colloids), which were present in the vent water/particle samples, and not present in the truly dissolved phase sampled by the SPMDs.

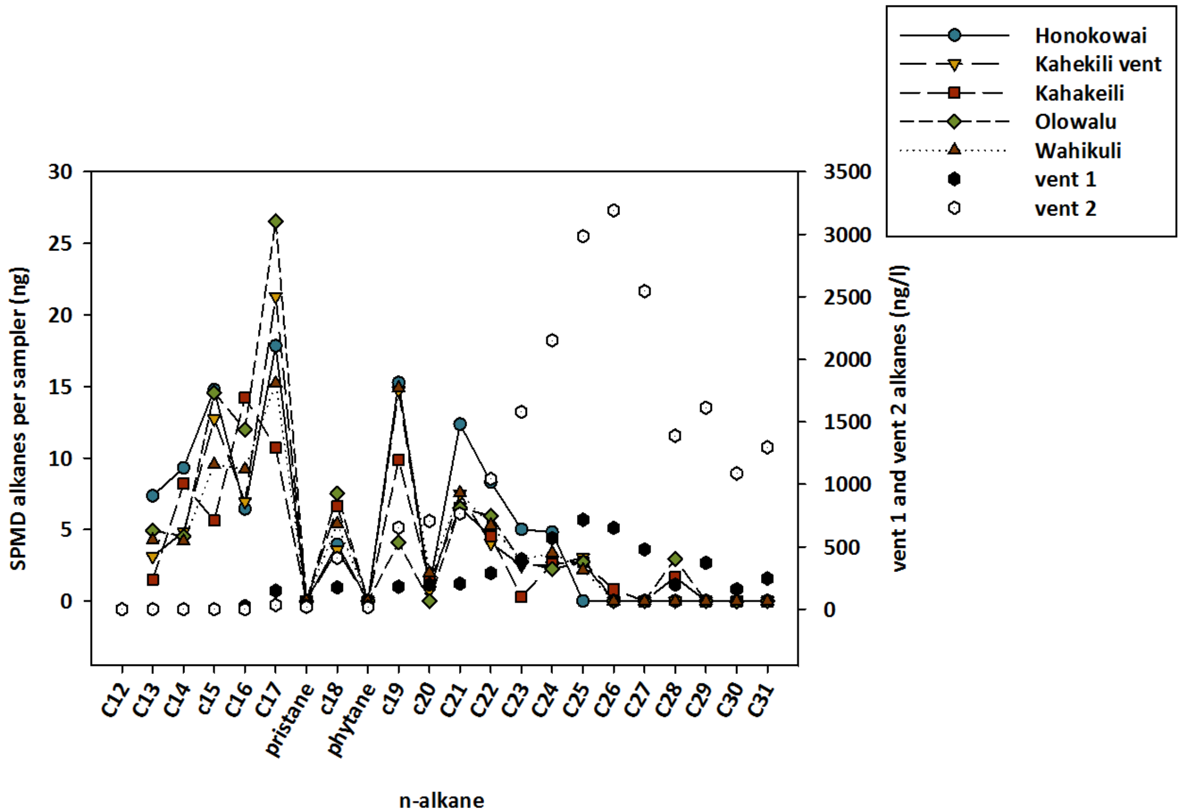

Figure 2. Concentration of *n*-alkanes (ng per sampler) in SPMDs and vent water filter samples after 28-day deployment, for the five coastal sites along west Maui.

Table 2. *N*-alkane (C11 through C31, including pristane and phytane) concentrations per sampler (ng) for five SPMD sites deployed for 28 continuous days and vent water samples collected at the Kahekili vent site

Sample Site	C11	C12	C13	C14	C15	C16	C17	pristane
Honokowai	23.68	7.35	9.31	14.78	6.43	17.85	3.96	0.00
Kahekili -Vent	8.97	3.12	4.84	12.73	6.94	21.29	3.58	0.00
Kahekeili	15.53	1.48	8.19	5.62	14.23	10.71	6.62	0.00
Olowalu	52.04	4.91	4.54	14.57	12.00	26.56	7.51	0.00
Wahikuli	31.48	4.25	4.18	9.54	9.20	15.23	5.40	0.00
Vent 1	0.00	0.00	0.00	0.00	0.00	25.91	148.08	27.13
Vent 2	0.00	0.00	0.00	0.00	0.00	0.00	33.90	19.38

Sample Site	C18	phytane	C19	C20	C21	C22	C23	C24
Honokowai	15.28	0.00	1.00	12.37	270.00	8.32	5.00	4.82
Kahekili -Vent	14.72	0.00	0.73	7.50	290.77	4.03	2.48	2.49
Kahekeili	9.86	0.00	1.39	6.51	266.79	4.54	0.28	2.59
Olowalu	4.11	0.00	0.00	6.50	272.54	5.96	2.75	2.22
Wahikuli	14.90	0.00	1.97	7.55	320.40	5.30	2.86	3.37
Vent 1	175.05	21.55	180.83	197.46	270.00	205.14	288.92	399.13
Vent 2	409.98	15.99	653.03	706.38	458.39	764.66	1,042.04	1,579.86

Sample Site	C25	C26	C27	C28	C29	C30	C31
Honokowai	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Kahekili -Vent	3.02	0.00	0.00	0.00	0.00	0.00	0.00
Kahekeili	2.75	0.80	0.00	1.67	0.00	0.00	0.00
Olowalu	2.72	0.00	0.00	2.96	0.00	0.00	0.00
Wahikuli	2.17	0.00	0.00	0.00	0.00	0.00	0.00
Vent 1	569.87	715.12	650.39	478.31	195.76	371.24	159.49
Vent 2	2,152.43	2,984.56	3,191.56	2,545.55	1,388.71	1,613.33	1,087.04

The SPMD total sterol concentrations per sampler ranged from 236.6 ng at Olowalu to a high of 2,513.8 ng at the Kahekili vent site (fig. 3, table 3). The sterol cholesterol, inferred to represent a marine source (Volkman, 1986), and the terrestrial sterol β-sitosterol explained 46 – 97 percent of the total sterol composition at all sites. Cholesterol, a sterol biomarker, originates from marine organism cell membranes whereas the β-sitosterol is derived from the epicuticular waxes of terrestrial plants (Volkman, 1986). Cholesterol and β-sitosterol were the most abundant compounds identified among the fatty alcohols, with the highest concentrations per sampler measured at the Kahekili vent site of 1,115.5 and 1,036.14 ng respectively. Cholesterol (cholest-5-en-3β-ol) is the major C27 sterol present in the marine environment and is used as a general indicator of zooplankton and other marine (for example, phytoplankton and fish), since it is the major sterol of marine organisms (Volkman, 1986). The major C29 sterol, β-sitosterol (24-ethylcholest-5-en-3β-ol), is indicative of terrestrial inputs since it is the most prevalent sterol in the epicuticular waxes of vascular plants (Santos and others, 2008), although it may also be derived from marine algae (Volkman, 1986). The values of the terrestrial-sourced sterol, β-sitosterol at the Kahekili vent site are close to an order of magnitude greater than the values at the other four coastal sites, indicating increased terrestrial inputs. The smallest contribution to the organic matter pool was calculated for other marine- and terrestrial-sourced sterols (brassicasterol, campesterol and stigmasterol). Coprostanol and epicoprostanol are produced by

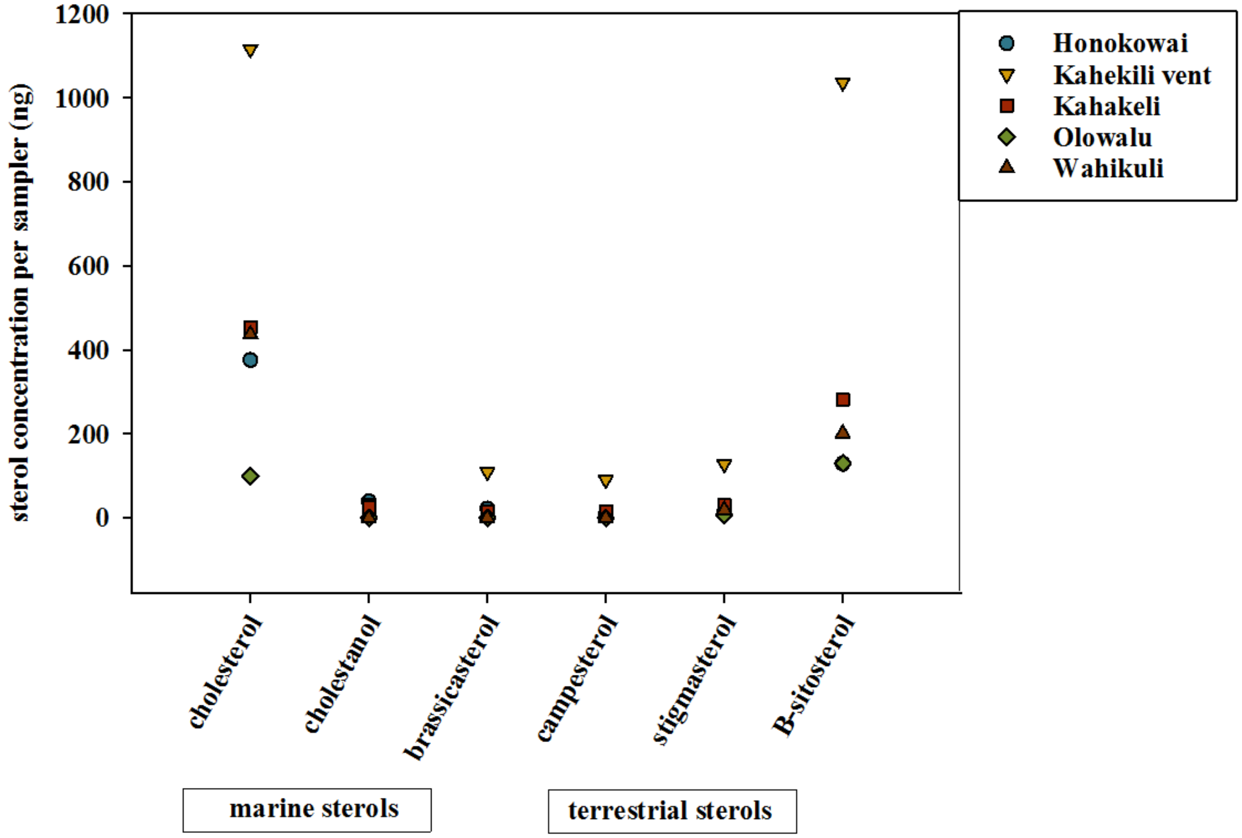

Figure 3. Concentration of sterols (ng per sampler) in SPMDs after 28-day deployment, for the five coastal sites along west Maui.

Table 3. Sterol concentrations per sampler (ng), total sterols and C27/C29 sterol ratios for five coastal sites along west Maui where the SPMDs were deployed for 28 continuous days.

Sample site	coprostanol	epicoprostanol	5-B-coprostanone	22-dehydrocholesterol	cholesterol
Honokowai	0.00	0.00	0.00	0.00	375.61
Kahekili	0.00	0.00	0.00	0.00	1,115.47
Kahekili	0.00	0.00	0.00	0.00	453.94
Olowalu	0.00	0.00	0.00	0.00	98.99
Wahikuli	0.00	0.00	0.00	0.00	436.97

Sample site	cholestanol	brassicasterol	campesterol	stigmasterol	B-sitosterol
Honokowai	38.93	21.82	7.36	14.22	128.78
Kahekili	35.30	109.08	90.24	127.57	1,036.14
Kahekili	24.39	15.64	15.50	30.88	281.13
Olowalu	0.00	0.00	0.00	7.96	129.68
Wahikuli	0.00	0.00	0.00	19.48	200.95

Sample site	stigmastanol	C29/C27	sterol sum
Honokowai	0.00	0.34	587.06
Kahekili	0.00	0.93	2,513.80
Kahekili	0.00	0.62	821.47
Olowalu	0.00	1.31	236.63
Wahikuli	0.00	0.46	657.40

the microbial reduction of cholesterol as well as through the intestinal microflora of higher animals, including marine mammals and birds, but are frequently used as sewage tracers correlated with anthropogenic inputs (Takada and others, 1994). Coprostanol (5β-cholest-3β-ol), a human sewage biomarker, was not detected at any of the sites. The absence of coprostanol at all sites suggests that efficient degradation and (or) dispersion of certain biomarkers/organic compounds must have occurred.

The C29/C27 sterols ratio (such as β-sitosterol/cholesterol and stigmasterol/cholesterol) is used as an indicator to differentiate terrestrial-sourced input of organic material from marine-sourced material (Mudge and Norris, 1997). In general, the C29/C27 sterols ratios calculated for all stations, except for the Olowalu site, were less than 1 (table 3). These results illustrate the dominance of marine biomass in the coastal waters of west Maui relative to terrestrial plant sources. The relatively higher ratio at Olowalu indicates the dominance of terrestrial inputs of organic matter from that watershed, presumably through surface runoff. The C29/C27 ratio at the Kahekili vent site is close to unity, indicating a higher degree of terrestrial organic matter input relative to marine organic matter input at this site. Overall, total sterol concentrations were greatest at the Kahekili vent site and lowest at the Olowalu site (table 3). The combination of sterol and n-alkane concentrations and ratios show that terrestrial-derived organic matter inputs were highest at Kahekili reef vent site. It seems likely that other contaminants may be also transported to the reef either in surface runoff and groundwater.

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 large number of chemical classes (U.S. Food and Drug Administration, 2015). For many of these products, there is likely a seasonality influenced by peak tourism and usage rates per person. Since the majority of pharmaceutical products are more polar and less hydrophobic than other contaminants, POCIS were chosen for sampling of possible pharmaceutical contaminants.

Seventeen polar organic compounds (PPCPs and flame retardants) were identified in POCIS at the five sites along west Maui (fig. 4, table 4). The highest concentrations of the compounds, and the highest number of detects (12), occurred at the Kahekili vent site (table 4). The compounds with the highest concentrations per sampler were acetaminophen at Wahikuli (1,800 ng), followed by the flame retardant Tris (1-chloro-2-propyl) phosphate (TCPP) at the Kahekili vent site (550 ng per sampler), and antimicrobial/antifungal triclosan at Olowalu (700 ng per sampler). The anti-epileptic pharmaceutical drug carbamazepine was detected at high levels (350 ng per sampler) in the Kahekili vent site utilizing the POCIS, this pharmaceutical has been previously detected in groundwater and wastewater from numerous sites worldwide (Khetan and Collins, 2007). There are no data on its effects to corals, but it is classified as a hazardous contaminant to the aquatic environment based on ecotoxicity bioassays performed on fish, micro-crustaceans, algae, and bacteria (Ferrari and others, 2003).

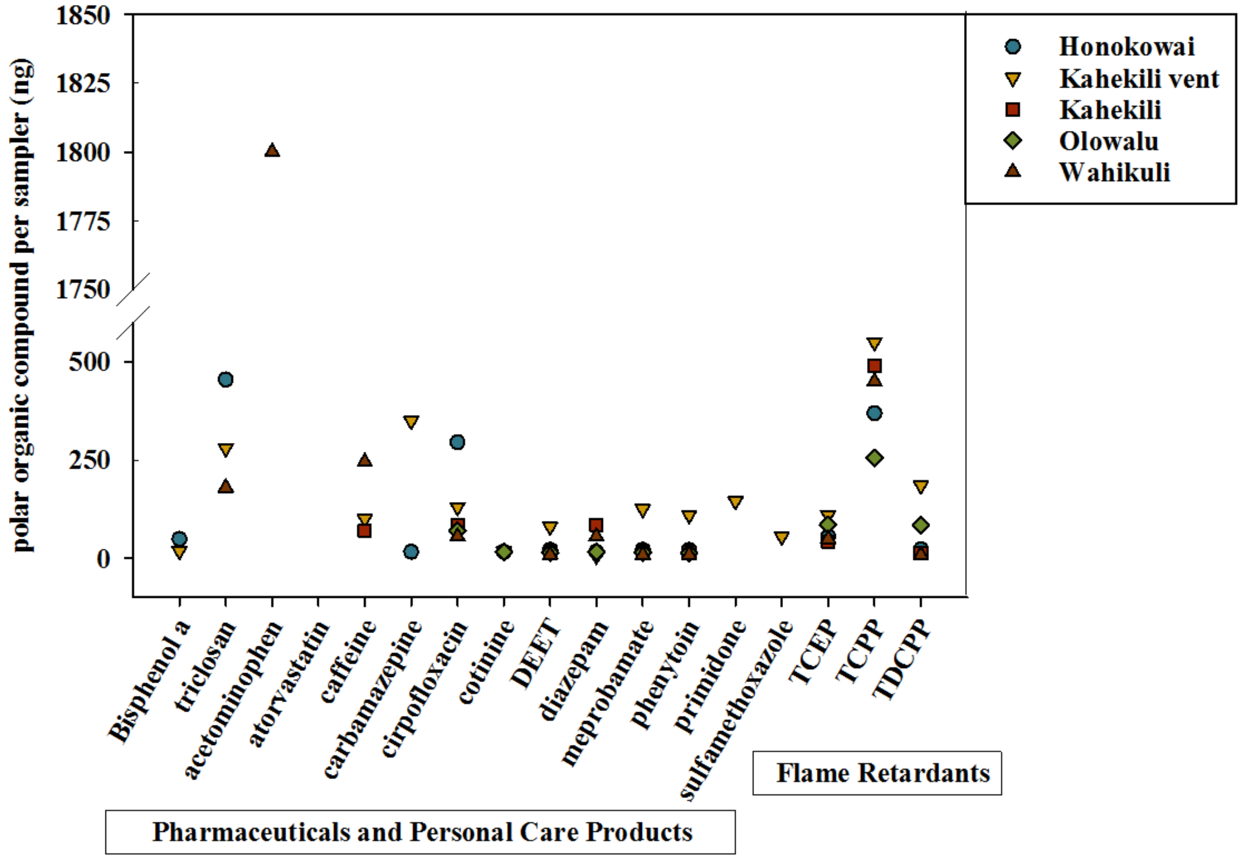

Figure 4. Concentrations (ng per sampler) of pharmaceuticals, PPCPs, and flame retardants in POCIS samples.

Table 4. Polar organic contaminants (pharmaceuticals, PPCP, and pesticides) detected in the POCIS during the 28-day deployment. Concentrations in (ng).
[ND = non detect]

Analyte	Honokowai	Kahekili - vent	Kahekili	Olowalu	Wahikuli
2,4'-DDD	ND	ND	ND	ND	ND
2,4'-DDE	ND	ND	ND	ND	ND
2,4'-DDT	ND	ND	ND	ND	ND
4,4´-DDD	ND	ND	ND	ND	ND
4,4´-DDE	ND	ND	ND	ND	ND
4,4´-DDT	ND	ND	ND	ND	ND
Aldrin	ND	ND	ND	ND	ND
alpha-BHC	ND	ND	ND	ND	ND
alpha-Chlordane	ND	ND	ND	ND	ND
Aroclor 1016	ND	ND	ND	ND	ND
Aroclor 1221	ND	ND	ND	ND	ND
Aroclor 1232	ND	ND	ND	ND	ND
Aroclor 1242	ND	ND	ND	ND	ND
Aroclor 1248	ND	ND	ND	ND	ND
Aroclor 1254	ND	ND	ND	ND	ND
Aroclor 1260	ND	ND	ND	ND	ND
beta-BHC	ND	ND	ND	ND	ND
Chlordane (tech)	ND	ND	ND	ND	ND
cis-Nonachlor	ND	ND	ND	ND	ND
delta-BHC	ND	ND	ND	ND	ND
Dieldrin	ND	ND	ND	ND	ND
Endosulfan I	ND	ND	ND	ND	ND
Endosulfan II	ND	ND	ND	ND	ND
Endosulfan sulfate	ND	ND	ND	ND	ND
Endrin	ND	ND	ND	ND	ND
Endrin aldehyde	ND	ND	ND	ND	ND

Analyte	Honokowai	Kahekili - vent	Kahekili	Olowalu	Wahikuli
gamma-BHC (Lindane)	ND	ND	ND	ND	ND
gamma-Chlordane	ND	ND	ND	ND	ND
Heptachlor	ND	ND	ND	ND	ND
Heptachlor epoxide	ND	ND	ND	ND	ND
Methoxychlor	ND	ND	ND	ND	ND
Mirex	ND	ND	ND	ND	ND
Toxaphene	ND	ND	ND	ND	ND
trans-Nonachlor	ND	ND	ND	ND	ND
17-a-Ethynylestradiol	ND	ND	ND	ND	ND
17-b-Estradiol	ND	ND	ND	ND	ND
Estrone	ND	ND	ND	ND	ND
Progesterone	ND	ND	ND	ND	ND
Testosterone	ND	ND	ND	ND	ND
Bisphenol A	49	18	ND	ND	ND
Diclofenac	ND	ND	ND	ND	ND
Gemfibrozil	ND	ND	ND	ND	ND
Ibuprofen	ND	ND	ND	ND	ND
Iopromide	ND	ND	ND	ND	ND
Naproxen	ND	ND	ND	ND	ND
Salicylic Acid	ND	ND	ND	ND	ND
Triclosan	455	280	365	700	180
Acetaminophen	ND	ND	ND	ND	1,800
Amoxicillin	ND	ND	ND	ND	ND
Atenolol	ND	ND	ND	ND	ND
Atorvastatin	16	ND	ND	ND	ND
Azithromycin	ND	ND	ND	ND	ND
Caffeine	175	100	70	75	245
Carbamazepine	ND	350	ND	ND	ND

Analyte	Honokowai	Kahekili - vent	Kahekili	Olowalu	Wahikuli
Ciprofloxacin	295	130	85	70	55
Cotinine	16	17	ND	16.5	ND
DEET	21	80	16	14.5	9
Diazepam	ND	6	ND	ND	ND
Fluoxetine	ND	ND	ND	ND	ND
Meprobamate	ND	125	ND	ND	ND
Methadone	ND	ND	ND	ND	ND
Phenytoin (Dilantin)	ND	110	ND	ND	ND
Primidone	ND	145	ND	ND	ND
Sulfamethoxazole	ND	55	ND	ND	ND
TCEP	55	110	42	85	49
TCPP	370	550	490	255	450
TDCPP	22.5	185	14	ND	8
Trimethoprim	ND	ND	ND	ND	ND

Bisphenol(a) (BPA) is classified as an endocrine disrupting chemical (EDC) that potentially interferes with reproduction and developmental processes. BPA is commonly released into the environment as a result of its use in epoxy resins and polycarbonate plastics (Kang and others, 2007; Kitada and others, 2008). BPA leaching from plastics is of concern in tropical environments such as Hawaii, as leaching rates are a function of time and ambient water and (or) air temperature (Sajiki and Yonekubo, 2003). The presence of low-level concentrations of BPA at Honokawai (49 ng per sampler) and at the Kahekili vent site (19 ng per sampler) merits increased monitoring at these and other nearby sites due to its endocrine disrupting capability. BPA has been shown to be deleterious to corals (Kitada and others, 2008).

Caffeine, a central nervous system stimulant and the most widely used psychoactive drug in the world, was present at all five sites. Caffeine has been proposed as a wastewater contaminant tracer because of its elimination through human urine (~3 percent of consumption is excreted) or household disposal. Pollack and others (2009) have suggested that coral bleaching occurrences may increase due to the presence of caffeine, which acts as a general environmental stressor. Knee and others (2010) reported concentrations of caffeine in coastal groundwater and surface water on the north shore of Kauai with seasonal fluctuations related to rainfall and tourism. The higher levels of caffeine observed at Wahikuli Park maybe be caused by proximity to upstream residential communities with onsite private septic systems. Trace organic contaminants may be detected in wastewater from septic systems in concentrations that are orders of magnitude higher than typical concentrations reported in treatment plant wastewater

(Conn and others, 2006; Godfrey and others, 2007). Caffeine was also present at the shallow Kahekili vent water site and in deeper water offshore close to large hotel developments implying offshore transport of contaminants.

Summary

Passive membrane samplers (SPMD and POCIS) were deployed for 28 continuous days at five coastal sites along west Maui to examine organic compound inputs to diverse, shallow coral reef ecosystems. The use of passive samplers allowed for one month of continuous, integrated sampling to accommodate fluctuations in daily and weekly inputs. The distribution of organic compounds observed at these five coastal sites along west Maui showed considerable variations; however, direct correlations to upstream land-use practices were not clearly obvious. There were distinct differences between the strongly groundwater-influenced Kahekili vent site and the other coastal sites, which were influenced more by seasonal surface/stream water runoff. High concentrations of terrestrial biomarkers as well as numerous PPCP and flame retardants were detected at the Kahekili vent site. Future work may determine the concentrations of organic compounds adsorbed onto water column particulate matter and coral algal symbionts, at the Kahekili vent site, and investigate offshore and alongshore contaminant dispersal from the vent site. The effect of contaminants at the individual level (such as fecundity and metabolism), as well as the community level (such as species abundance and diversity), in coral reefs is necessary for an accurate assessment. Results presented here provide current contaminant inputs to the nearshore during the dry season and can be useful for future evaluations and (or) comparisons.

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