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Case Report

Evaluation of stormwater as a potential source of polychlorinated biphenyls (PCBs) to Pearl Harbor, Hawaii

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ABSTRACT

A study was implemented to determine whether stormwater from Oscar 1 Pier outfall is a contributing source of polychlorinated biphenyls (PCBs) to Decision Unit (DU) N-2, Pearl Harbor Sediment Site, Honolulu, Hawaii USA. Results suggested that PCBs were discharged from the outfall, remained in suspension, and dispersed elsewhere before settling. Stormwater PCBs were characterized by heavier congeners, likely associated with particles. Surface water PCB samples collected throughout DU N-2 exhibited heavier congeners and lighter congeners that are typically associated with the dissolved phase. These lighter congeners could have originated from a different source(s) and/or partitioned from the suspended phase.

1. Introduction

The Pearl Harbor Sediment Site (the Site), included within Joint Base Pearl Harbor-Hickam (JBPHH), Honolulu (Oahu), Hawaii, USA, was placed on the National Priorities List by the United States Environmental Protection Agency (USEPA) in October 1992. Contaminant sources at the Site are attributed to naval activities and releases from commercial, industrial, residential, and agricultural sources in the surrounding watershed, and discharged from the tributary streams and storm drains that enter Pearl Harbor.

The Site has been the subject of many environmental investigations over the last three decades and summarized in the Site record of decision (ROD) [1]. Of relevance to this project is the Pearl Harbor Sediment Remedial Investigation Addendum [2]. This study began in 2009 and recommended seven Decision Units (DUs) for further consideration of remedial action, including DU N-2 (Oscar 1 and 2 Piers Shoreline). DU N-2 has a total area of approximately 10.8 ha and is located on the eastern bank of the Pearl Harbor navigation channel (Fig. 1). Water depths within DU N-2 range from less than 3 m along the shoreline to depths of 15–18 m in a channel that extends from Dry Dock 4 to the navigation channel.

DU N-2 was evaluated for sediments contaminated with cadmium, copper, lead, mercury, zinc, dieldrin, and polychlorinated biphenyls (PCBs). DU N-2 sediment data indicated elevated concentrations of PCBs in the immediate vicinity of Oscar 1 Pier, decreasing in concentration with increasing distance from the outfall. From these data, PCBs are hypothesized to be delivered during stormflow to surface water and ultimately, site sediment, via the Oscar 1 Pier outfall: *The COC [contaminant of concern] concentration profile for sediments near an outfall south of Dry Dock 4 and Oscar 1 Pier indicates that concentrations are stable or increase toward the sediment surface, indicating potential continuing input of contaminants localized to the area immediately off of the outfall* [1].

The remedy footprint for DU N-2, as reported in the Site ROD, includes areas of enhanced natural recovery, monitored natural recovery, activated carbon treatment, and no remediation [1]. To date, enhanced natural recovery remedy actions have been implemented and activated carbon has been placed under Oscar 1 Pier. Long-term fish monitoring for total PCBs (TPCBs) was also recommended for DU N-2.

A detailed PCB study, using OPTICS (OPTically-based In-situ Characterization System), was performed for evaluation and characterization of stormwater as a potential source of contaminants to DU N-2. OPTICS is a tool that combines robust, in-situ aquatic instrumentation, discrete analytical samples, and multi-parameter statistical modeling techniques to provide concentrations of chemical contaminants at high temporal or spatial resolution, at significantly reduced cost relative to traditional sampling methods [3,4]. OPTICS is particularly well-suited for

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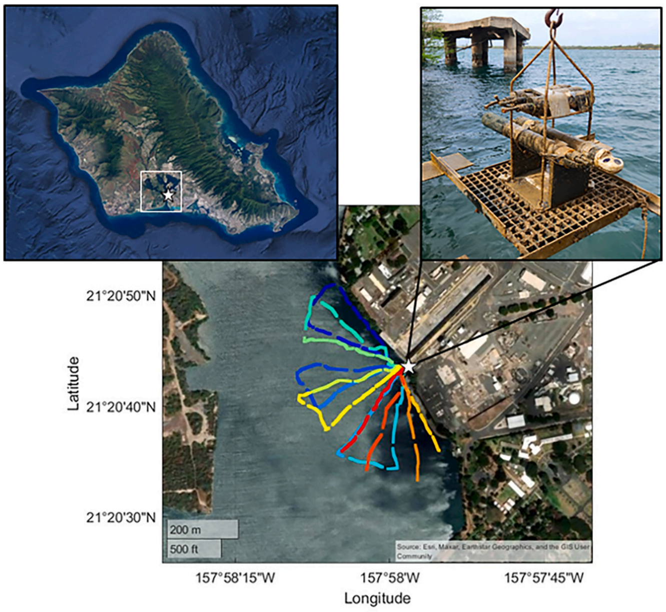

Fig. 1. Top left: Oahu, Hawaii USA with Pearl Harbor outlined in white. Bottom: Mobile monitoring transect lines and stationary monitoring platform (inset) in Decision Unit N-2 of the Pearl Harbor Sediment Site. The white stars indicate the location of Oscar 1 Pier outfall.

hydrophobic COCs, like PCBs, because of their tendency to sorb to materials in the water column. These materials have unique optical signatures that can be assessed using in-situ optical and water quality sensors (e.g., Boss and Pegau [5]; Boss et al. [6,7]; Babin et al. [8]; Boss et al. [9]; Coble et al. [10]; Sullivan et al. [11]; Twardowski et al. [12]; Chang et al. [13]; Briggs et al. [14]; Slade and Boss [15]), thereby providing an effective and robust means of quantifying high-resolution concentrations of chemical contaminants through statistical methods. Optically-based chemical contaminant characterization methods have been successfully applied at coastal, estuarine, riverine, and lacustrine sites for a variety of COCs, including PCBs [3,4,16–18].

The primary objectives of the Pearl Harbor PCB study are to.

- Determine whether stormwater from the Oscar 1 Pier outfall is contributing ongoing PCBs to DU N-2.
- If applicable, characterize the stormflow plume of PCBs generated by Oscar 1 Pier outfall in terms of plume extent (area affected) during a stormflow event.

2. Materials and methods

The PCB study was conducted during rainy season between November 2022 and March 2023, to determine the potential for Oscar 1 Pier outfall to deliver PCBs to DU N-2. The study involved mobile (vessel-mounted) OPTICS monitoring to characterize stormwater plume spatial variability. Two baseline events and one stormflow event were captured during mobile monitoring. In addition to mobile monitoring, near-continuous stationary (moored) OPTICS monitoring was conducted at the base of Oscar 1 Pier outfall over the four-month period to evaluate baseline conditions and stormwater discharge over time-varying conditions (tidal, dry-period, flood event, etc.).

2.1. In-situ measurements

A vessel-mounted, mobile OPTICS monitoring system was deployed in tow-yo mode (vertical profiling while transecting) following a predetermined “spoke” pattern (Fig. 1); vertical profiles were collected from 1 m of the surface to 1 m above the sediment bed. OPTICS instrumentation included sensors to measure water quality and optical parameters (conductivity-temperature-depth [CTD], optical turbidity, dissolved oxygen [DO], chlorophyll-a [Chl-a] fluorescence, and optical near-forward and backscattering). Near-forward scattering was measured with a Laser In-Situ Scattering and Transmissometry (LISST) sensor and used to derive particle size distribution [19]. Mobile OPTICS measurements were collected at 1 Hz continuously, thus providing high spatial resolution data vertically and horizontally throughout DU N-2. Mobile monitoring was conducted during baseline (dry) conditions November 16 and 17, 2022 and March 28, 2023, and repeatedly over two days of rainfall on February 17 and 18, 2023. Over these two days, rainfall was intermittent and varied in intensity, enabling stormflow mobile monitoring activities to capture the onset, peak, and cessation of stormflows with variable flowrate.

A stationary OPTICS monitoring platform with instrumentation similar to the vessel-mounted system was installed and maintained at the base of Oscar 1 Pier outfall (Fig. 1). In addition to water quality and optical parameters, an acoustic Doppler current profiler measured current velocity throughout the water column. Monitoring equipment was placed such that instrumentation sensing volumes were within 1 m of the sediment bed and sensors were programmed to collect data at 1 Hz for 30 s, every 20 minutes, continuously. The OPTICS stationary platform was serviced (recovered, cleaned, data offloaded, batteries changed, and recalibrated [if necessary]) twice over the deployment period at 4- to 6-week intervals to ensure high quality data.

2.2. Discrete water samples

Unfiltered discrete surface water samples were collected periodically, collocated with OPTICS instrumentation using a peristaltic pump with sterile sample tubing. A suite of 47 discrete surface water samples, including five field duplicates, were collected along six of the transect spokes, at the base of the outfall (1 m above the sediment bed) and at three distances from the outfall (15 m, 30 m, and 75 m) at 2 m above the sediment bed during stormflow conditions. Samples collected at 15 m or greater from the outfall were obtained along every other transect line to cover a wide spatial area. Surface water samples were collected to cover a range of flow conditions, from baseline to peak flow. Two additional samples were collected directly from Oscar 1 Pier outfall discharge waters during stormflow. Four more samples were obtained during baseline (dry) conditions at the base of the outfall (1 m above the sediment bed) and 300 m directly offshore of the outfall (2 m above the sediment bed), for a total of 51 samples. Samples were analyzed in the laboratory for PCBs (congeners) following USEPA Method 1668C [20] (high resolution mass spectrometry after separatory funnel extraction) for statistical correlation to in-situ mobile and stationary OPTICS measurements.

2.3. OPTICS statistical modeling

The response variable for the OPTICS model was TPCBs (sum of congeners) obtained through laboratory analysis of discrete surface water samples. These were statistically correlated with the predictor variables through partial least-squares (PLS) multi-parameter regression analysis [21,22]. OPTICS predictor variables were parameters that are likely to describe variability in TPCBs. These included primary and higher-order mechanisms for PCB mobility, transport, and partitioning (e.g., Adeyinka and Moodley [23]). OPTICS model predictor variables thus included in-situ measurements of depth, temperature, salinity, turbidity, optical backscattering, DO, and Chl-a. OPTICS PLS regression was performed on root-transformed predictor and response datasets with 10-fold cross-validation. The optimal number of model components was identified to maximize the variance explained between predictors and response, minimize the root mean square error of predictions, and avoid model over-constraint. OPTICS model results were vertically and horizontally resolved TPCBs along predetermined spoke transect lines throughout DU N-2 and high-resolution time series estimates of TPCBs at the base of Oscar 1 Pier outfall (1 m above the sediment bed).

3. Results and discussion

OPTICS model skill was assessed through computation of OPTICS modeled versus measured comparative statistics, and results were compared to the same comparative statistics between paired field duplicates (Table 1). Statistical metrics computed between modeled and measured TPCBs were not as strong as those determined during previous OPTICS studies [3,4]. A likely explanation for this was highly variable (patchy) solids consisting of silt-sized particles discharged from Oscar 1 Pier outfall (Fig. 2) and the relatively high standard deviation in discrete surface sample TPCBs (Table 2). Strong, small-scale variability in TPCBs created difficulties in direct correlations between in-situ measurements (collected at 1 Hz and averaged over 30 s) and discrete water samples (pumped over 60–90 s). However, despite these discrepancies, OPTICS results were very good, with model error metrics generally better (lower) than field duplicate errors and bias metrics that were better (closer to 1.0) for the OPTICS model as compared to paired field duplicates (Table 1).

Discrete surface water samples were collected primarily to serve as response variables for the OPTICS model and to assess model performance. However, these data also provided valuable data with which to evaluate Oscar 1 Pier outfall as a potential source of PCBs to DU N-2, and for characterizing PCBs across DU N-2. Discrete surface water sample TPCBs ranged from 52.2 pg/L to 573 pg/L across the surface water samples collected near the sediment bed in November 2022 and February and March 2023 (Table 2). TPCB concentrations of the two water samples that were collected directly from outfall discharge waters were 26,512 pg/L and 36,115 pg/L – two orders of magnitude higher than TPCBs measured at the base of Oscar 1 Pier outfall (1 m above the sediment bed) (Table 2; Fig. 3). Although pollutant transfer via industrial outfalls in Pearl Harbor has been significantly reduced since the 1980s [24–27], these results indicate that the outfall is likely a source of PCBs to DU N-2.

Further investigation of discrete surface water sample data revealed that the PCB fingerprints (congener fractions of TPCBs) of Oscar 1 Pier outfall discharge waters differed from those of samples collected within 2 m of the sediment bed throughout DU N-2 (Fig. 4) [28]. Discharge waters were largely characterized by heavier congeners between PCB-129 and PCB-193. These heavier congeners were also present in surface water samples collected elsewhere in DU N-2, but to a lesser extent relative to peaks observed for the lighter congeners: PCB-11, between PCB-44 and PCB-54, and PCB-65. A potential explanation for this was that during stormflow, Oscar 1 Pier outfall delivered primarily PCBs associated with silt-sized particles (e.g., Cao et al. [29]), whereas both particulate and dissolved PCBs were found in DU N-2 surface water. Lighter congener PCBs may have originated from another source or through partitioning processes to the dissolved phase. Balasubramani et al. [30] reported similar patterns in industrial effluents, where higher concentrations of lighter PCBs are in the dissolved phase and higher concentrations of heavier PCBs are in the suspended phase. Cao et al. [31] suggested that heavier PCBs are associated with stormwater particles because of increased hydrophobicity.

Table 1
Mobile OPTICS and field duplicate statistical performance metrics.

	R	Slope	Ratio	MAE	MPD	RMSE	CVRMSE
TPCBs (field duplicates)	0.60	1.46	0.92	57 pg/L	15.1%	75 pg/L	27%
TPCBs (modeled)	0.72	0.80	0.98	40 pg/L	13.2%	50 pg/L	20%

R = correlation coefficient, Slope = Model II slope, Ratio = median(modeled/measured), MAE = mean absolute error or mean(|modeled – measured|), MPD = mean absolute percent difference or 100*(|modeled – measured|)/measured, RMSE = root mean square error, CVRMSE = 100*coefficient of variation of RMSE, pg/L = picograms per liter.

Discrete surface water samples provided evidence to support the hypothesis that Oscar 1 Pier outfall is a source of PCBs to DU N-2. However, interestingly, TPCBs collected at the base of the outfall (1 m above the sediment bed) during stormflow events did not significantly exceed concentrations recorded during non-storm conditions in November 2022 or March 2023 and were at times, less than non-storm concentrations (Fig. 3). The temporal variability of OPTICS-derived TPCBs at the base of the outfall was not directly correlated to turbidity, which is a proxy for particle concentration (Fig. 5). Further, lower concentrations of TPCBs were observed at the base of the outfall during periods of lower salinity (i.e., rainfall), indicating a dilution effect of stormwater near the sediment bed (Fig. 5) [32]. The apparent discrepancy between outfall discharge water characteristics and observations at the base of the outfall can be explained through analysis of high spatial resolution OPTICS-derived TPCBs.

OPTICS enabled spatial mapping of TPCBs throughout DU N-2 during stormflow conditions (Fig. 6), and helped resolve transport patterns of TPCBs from the outfall to other areas within DU N-2. TPCB exceedance, or $\text{TPCBs}_{\text{plume}}$, was defined as the difference between spatially-resolved, OPTICS-derived TPCBs and the one-third quantile of TPCB concentrations measured in discrete surface water samples between November 2022 and March 2023 (Fig. 6). Quantile difference analysis indicated that TPCB exceedance was observed in approximately 10% of data within DU N-2 during stormflow conditions (February 2023), compared to 0% in November 2022 and approximately 2% during baseline mobile monitoring activities in March 2023. TPCB exceedance observed in March 2023 may have been due to residual PCBs following significant rainfall (2.2 cm) one day prior to mobile OPTICS monitoring.

Fig. 7 shows results from stormflow TPCB exceedance analysis for water depths between 1 m and 12 m, in 1-m depth bins. TPCB exceedances during stormflow were found throughout most of DU N-2 and were more dispersed at depths greater than 4 m. Near the surface, TPCB exceedances were found closer to the outfall than at depth (Fig. 7A). At 1 m below the surface, the maximum distance between the outfall and the location of TPCB exceedance was 18.6 m, as compared to 314 m maximum distance at a depth of 12 m below the surface (Fig. 7). These results suggest that PCBs were discharged from Oscar 1 Pier outfall, remained in suspension, and dispersed elsewhere in DU N-2 before settling.

These results are consistent with those presented by Chadwick et al. [33,34], who used innovative drifter technologies for tracking and sampling stormwater plumes in DU N-2. Chadwick et al. [33,34] released 19 sediment tracking and sampling drifters at the base of Oscar 1 Pier outfall over a full 25-h tidal cycle. Results indicated that due to tidal currents, silt-sized particles released from Oscar 1 Pier outfall could potentially deposit over 1–1.5 km to the north and south of the outfall. Analysis of sediment deposition data showed that particle settling rates were relatively low at the base of the outfall compared to away from the outfall (∼70 g and ∼330 g cumulative deposition over 42 days, respectively), and the timing of deposition near Oscar 1 Pier outfall was correlated with storm events. Similar to the findings reported here, Chadwick et al. [33,34] presented TPCB concentration near Oscar 1 Pier that was more than an order of magnitude higher than concentrations found up to 1.5 km away from the outfall. The combined results from Chadwick et al.'s [33,34] experiment and this PCB study has provided data showing that Oscar 1 Pier outfall is a source of PCBs to DU N-2 during stormflow, and that particle-bound PCBs remain in suspension, are advected away from the outfall by tidal currents, and settle elsewhere in DU N-2.

4. Summary and conclusions

A PCB study was conducted to evaluate Oscar 1 Pier outfall as a

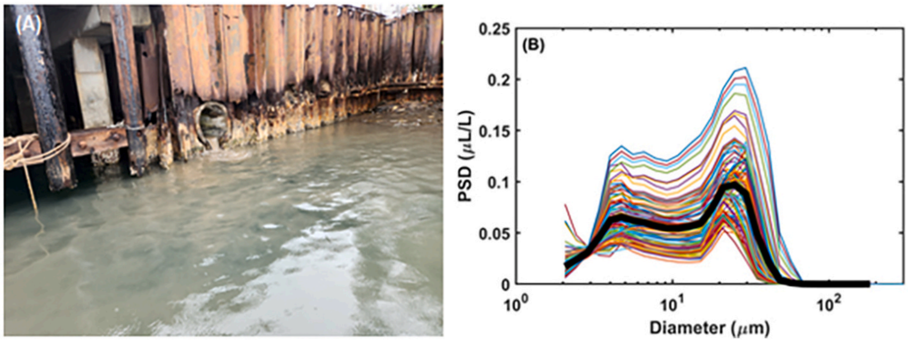

Fig. 2. (A) Small scale patchiness near Oscar 1 Pier outfall observed on February 17, 2023. (B) Particle size distributions (PSDs) measured throughout DU N-2; the thick black line is the mean (μm = micrometer).

Table 2
Discrete surface water sample basic statistics.

Analyte	N	Mean	Median	Minimum	Maximum	Standard Deviation
TPCBs (pg/L) (all)	51	1482	271	52.2	36,115	6163
TPCBs (pg/L) (no discharge)	49	264	271	52.2	573	104
TPCBs (pg/L) (discharge only)	2	31,314	31,314	26,512	36,115	6791

potential source of PCBs to DU N-2 of the Pearl Harbor Sediment Site, and to monitor and characterize the stormwater plume in the region. OPTICS monitoring provided robust, high spatial and temporal resolution TPCBs showing that during stormflow conditions, elevated TPCBs were observed near the surface, closest to the outfall (within 20 m), and higher concentrations of TPCBs were found at distances greater than 200 m from the outfall at depths greater than 4 m. These results suggested that PCBs were discharged from the outfall, remained in suspension, and dispersed elsewhere at DU N-2 before settling.

PCB data provided strong evidence that Oscar 1 Pier outfall is a source of contamination to DU N-2. Detailed PCB analysis showed that stormwater PCBs were characterized by heavier congeners and likely associated with fine-grained particles. Surface water PCB samples collected within 2 m of the sediment bed throughout DU N-2, on the other hand, exhibited both the heavier congeners similar to those present in stormwater, as well as lighter congeners that are typically associated with the dissolved phase. These lighter congeners could have originated from a different source (or sources) and/or were partitioned from the suspended phase.

This PCB study employed novel techniques (e.g., OPTICS) to assess and characterize stormwater delivery of PCBs to an industrial harbor. This study is of global interest due to the potential environmental, human health, and ecosystem impacts of persistent COCs delivered to surface waters. In particular, PCBs, once widely used in industrial applications, are known for their toxicity and ability to bioaccumulate in aquatic ecosystems. Understanding the mechanisms and pathways of

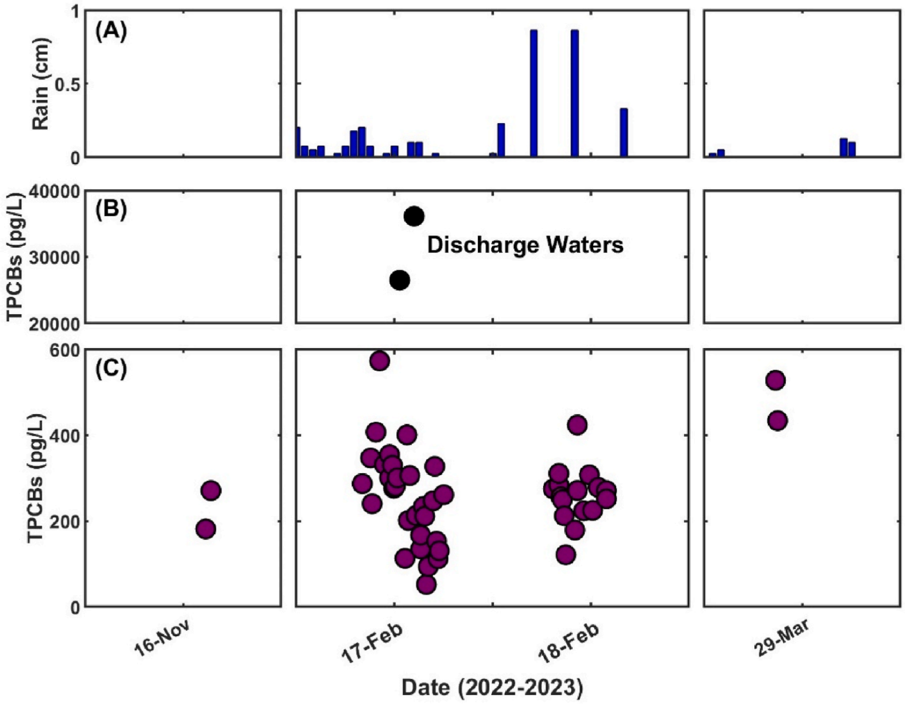

Fig. 3. (A) Hourly rainfall at Iroquois Point (Pearl Harbor), (B) Oscar 1 Pier outfall discharge water sample total PCBs (TPCBs; sum of congeners), and (C) discrete surface water TPCBs collected within 2 m of the sediment bed in Decision Unit N-2 (pg/L = picograms per liter).

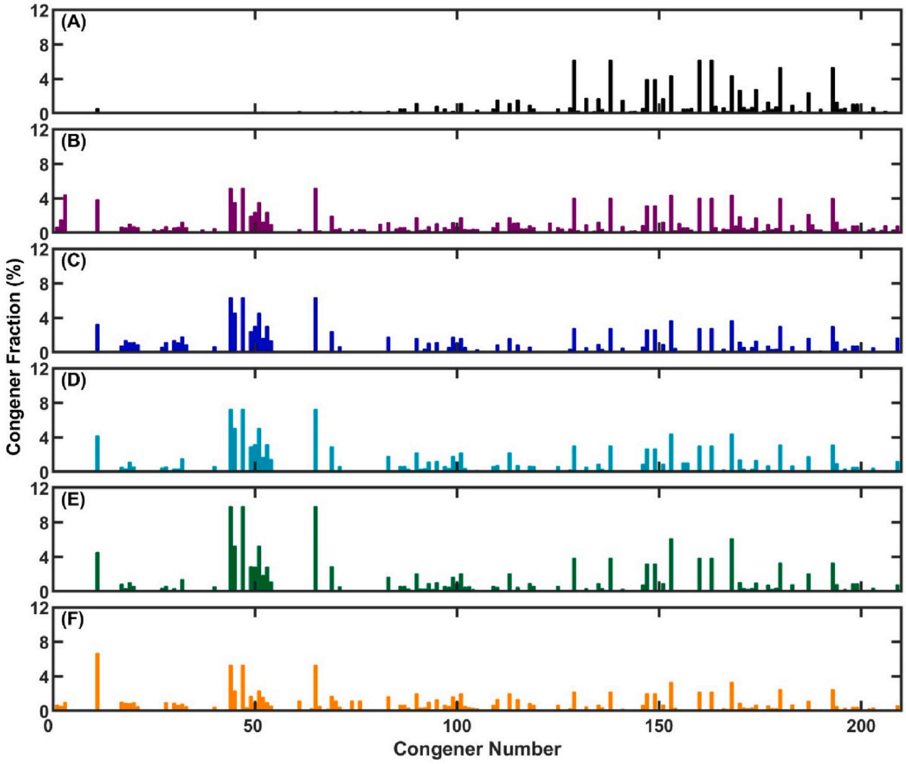

Fig. 4. PCB congener fraction to total PCBs averaged over samples collected (A) directly from Oscar 1 Pier outfall discharge waters; and within 2 m above the sediment bed at varying distances from the outfall: (B) 5 m, (C) 15 m, (D) 30 m, (E) 75 m, and (F) 300 m.

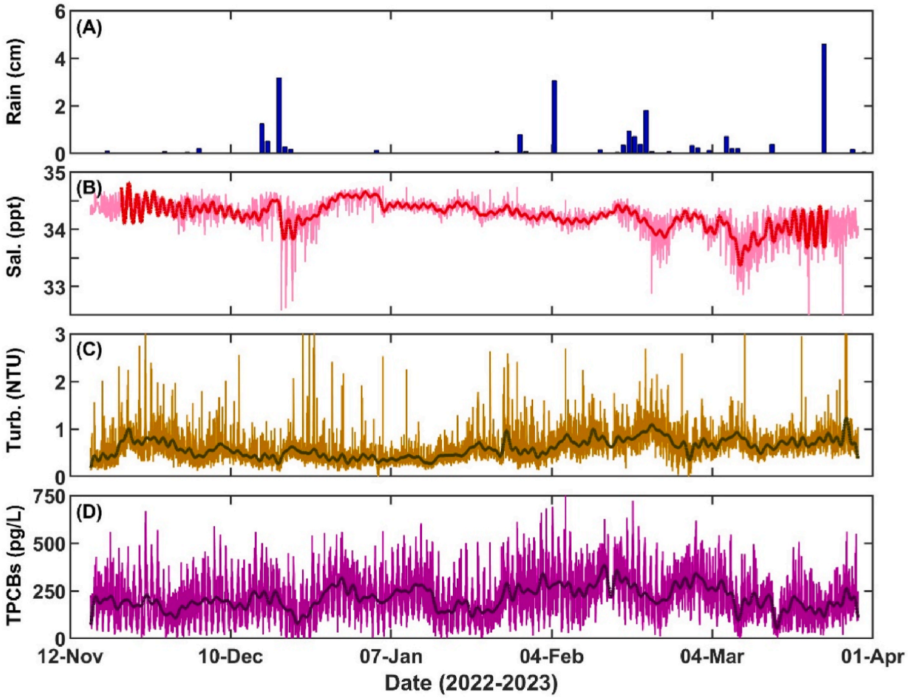

Fig. 5. (A) Daily rainfall at Iroquois Point (Pearl Harbor). Time series data from the base of Oscar 1 Pier outfall: (B) salinity, (C) optical turbidity, and (D) OPTICS-derived TPCBs. Lighter colors indicate data at 20-min intervals and 35-hr low-pass filtered data are shown as darker lines in (B)–(D). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

PCB transport via stormwater is crucial for assessing the extent of contamination, identifying sources, and implementing effective management strategies to mitigate environmental impacts.

Future studies in the region would benefit from OPTICS monitoring systems deployed within the outfall and/or at the sea surface just below Oscar 1 Pier outfall as stationary platforms, as well as on a vessel for additional stormflow mobile monitoring. Filtered and unfiltered sampling for PCBs would provide high temporal and spatial resolution of

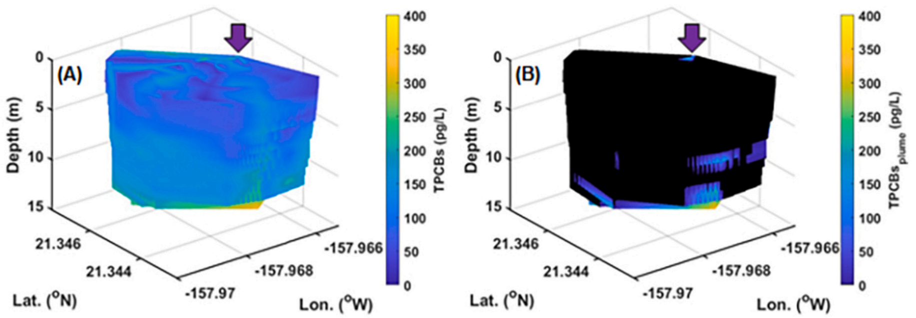

Fig. 6. Stormflow monitoring (February 2023) volume plots of (A) OPTICS-modeled TPCBs and (B) TPCB exceedance, where exceedance is defined as concentration greater than the one-third quantile. The purple arrow indicates the location of Oscar 1 Pier outfall, in the “back” of the figure. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

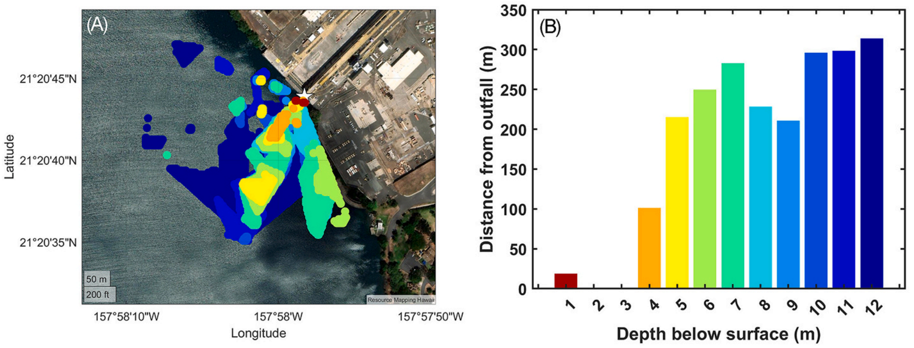

Fig. 7. (A) Map of TPCB exceedances as a function of water depth. The white star is the location of the Oscar 1 Pier outfall and color coding indicates depth of TPCB exceedance from 1 m (red) to 12 m (blue) in 1-m depth increments, as shown in (B). (B) Maximum distance between the location of TPCB exceedance and the outfall. Exceedance is defined as concentration greater than the one-third quantile. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

COCs in dissolved and suspended phase. Coupled with outfall flow meter data, the stationary OPTICS monitoring system(s) would enable computation of filtered, unfiltered, particulate (by difference), and total PCB flux and mass loading from the outfall as a function of time and/or storm event. Numerical hydrodynamic and sediment transport modeling of DU N-2, validated with e.g., particle tracing studies [35] would provide insights into fate and transport of particulate PCBs to inform effective remedial design.

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CRediT authorship contribution statement

Grace Chang: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Frank Spada:** Writing – review & editing, Project administration, Methodology, Investigation, Data curation. **Keith Brodock:** Writing – review & editing, Validation, Investigation, Conceptualization. **Craig Hutchings:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Kim Markillie:** Writing – review & editing, Validation, Supervision, Resources, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Grace Chang has patent #11,079,368 issued to Integral Consulting Inc. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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