



## Contaminant concentrations and risks associated with the Pacific oyster in the highly urbanized San Diego Bay

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### ABSTRACT

Contaminant concentrations in filter-feeding shellfish may indicate the health of coastal waters and consumption risks. Widespread expansion of the Pacific oyster (*Crassostrea gigas*) and its popularity as food make it a useful sentinel. We surveyed intertidal Pacific oysters in San Diego Bay, California for contaminants during summer 2018 and winter 2019. We compared contaminants in Pacific oyster to California mussel from California's State Mussel Watch Program (1993–2003) and human consumption thresholds. Contaminants such as neonicotinoid and chlorinated pesticides, selenium, and several metals were higher in Pacific oysters in summer, while PBDEs, benzylbutyl phthalate, and plastics were higher in winter. Contaminant levels were generally lower in Pacific oyster than mussel except for copper and zinc. Bay-wide PCB concentrations in oysters exceeded thresholds but individual samples (locations) also met or surpassed chlordane, PCB and PAH thresholds. Monitoring and risk assessments that consider species' biology, season, location, effects of multiple contaminants, and human consumption patterns will contribute to more effective consumption guidelines.

### 1. Introduction

Shellfish such as mussels and oysters absorb and accumulate contaminants making them both a risk to food webs, including humans, and excellent indicators of the health of coastal waters (e.g., Harris et al., 2009; EPA, 2011, 2013; Melwani et al., 2013). Mussels (*Mytilus* spp.) have been used nationally for monitoring but the popularity of oysters as farmed and human harvested food, the interest in aquaculture growth in the US and California in particular (NOAA, 2020; OPC, 2020; Noaa, 2021), and the recent expansion of feral populations, especially the Pacific oyster (*Crassostrea gigas*), throughout Southern California (Crooks et al., 2015; Langevin, 2019; Tronske et al., 2018) and worldwide (Ayres, 1991; Reise et al., 2017; Herbert et al., 2016) makes oysters an important focal organism for assessment of contamination levels and potential consumption risk. The feeding strategy and natural history of the oyster, including responses to environmental change, differ from that of the more commonly used mussels, with uncertain consequences

for oyster contaminant uptake and accumulation, and subsequent risks (e.g., Newall and Jordan, 1983; Cognie et al., 2003; Rosa et al., 2018). Furthermore, the target of monitoring efforts on the U.S. west coast, the California mussel (*M. californianus*), is predominantly an open-ocean species, not commonly found in inner bay and estuarine environments, with historic monitoring conducted using transplanted mussels (Melwani et al., 2013).

Understanding the drivers of contamination risk is particularly important for urbanized bays and estuaries, where relatively high contamination loads converge with dense human populations, including vulnerable populations. Our project focused on San Diego Bay (32.67°N, –117.15°W), a 12,000-acre urban bay with 5 public fishing piers, and 17 public shoreline parks. The San Diego Bay watershed includes flows from the Sweetwater River, Otay River, Chollas Creek, Paleta Creek, Paradise Creek, and Switzer Creek, all of which are heavily urbanized and serve as the stormwater system, shunting flows from San Diego and surrounding metropolitan areas downstream into San Diego Bay. An

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estimated 74% of San Diego Bay's shoreline is armored with riprap seawalls (Tierra Data, Inc., 2013), providing much publicly accessible settlement substrate for oysters. Most of the land surrounding San Diego Bay is a mix of commercial, recreational, and military uses (Henderson et al., 2015), with the highest urban development and population density, and often lowest incomes, along the eastern shore of San Diego Bay (Steinberg and Moore, 2017). These adjacent areas of higher density and lower incomes are also linked with the highest numbers of recreational and subsistence fishers in San Diego Bay (Steinberg and Moore, 2017).

The feral Pacific oyster predominantly occurs in San Diego Bay between tidal elevations of 0.3–1.0 m MLLW (Tronske et al., 2018). Higher density areas and/or patches with larger individuals may be at higher risk of predation by potential predators, such as octopus, shorebirds, and the commercially important spiny lobster (Ambrose, 1984; Robles et al., 1990; Herbert et al., 2018) and, when coupled with public access points, human harvest. With the large Pacific oyster being largely unmonitored, the risks to consumers are uncertain, yet some sort of risk is certain based on the well-documented presence of contaminants in sediment, water, and a suite of other organisms (e.g., endangered species, finfish of fishing interest, benthic invertebrates, zooplankton, bird eggs) in San Diego Bay (McLaughlin et al., 2020; Bay et al., 2016; Stransky et al., 2016; Loflen et al., 2018; Komoroske et al., 2011). These data have led to the publication of waterbody-specific State of California consumption advisories due to elevated levels of polychlorinated biphenyls (PCBs) and mercury in finfish and spiny lobsters (OEHHA, 2018), as well as further studies and cleanup activities in San Diego Bay.

Despite documented contaminant risks, however, the State of California ceased regular monitoring for contaminants in transplanted mussels (California's State Mussel Watch Program) in 2003 due to a lack of consistent funding. Therefore, recent information on contamination levels in sessile species likely predated by wildlife and harvested for food (e.g., feral Pacific oysters, Olympia oysters, Mediterranean mussels) is limited. The most recent data revealed that concentrations of PCBs in transplanted Mediterranean mussel closer to the mouth of San Diego Bay were higher than "no consumption" California Advisory Tissue Levels (Anderson et al., 2017), warranting further evaluation of shellfish. The age of these data, combined with the use of transplant organisms, prevents the State of California from developing waterbody-specific consumption advice for chemical contaminants in shellfish. Furthermore, most contaminants tested are the "usual suspects" of bioaccumulation and other major contaminants, such as mercury and PCBs (e.g., Bay et al., 2016; Stransky et al., 2016), with less emphasis on those of emerging concern (e.g., Busse and Nagoda, 2015). In particular, contaminants of emerging concern in San Diego Bay include small plastics and other anthropogenic debris, plasticizers (phthalates) (e.g., SDBDSW, 2016; Talley et al., 2020), newer-use pesticides, personal care products and pharmaceuticals (Busse, 2010; Busse and Nagoda, 2015; Bay et al., 2016). These compounds may have acute and long-term health effects on the organisms that consume shellfish, including humans, so the risks posed by consumption of shellfish, in particular the Pacific oyster which has high desirability but little information, need to be better understood.

### 1.1. Project goal and objectives

The goal of this study was to better understand contaminant dynamics in the Pacific oyster, in order to inform management decisions and collaboratively develop solutions for San Diego Bay and beyond. This goal was met by achieving the following four objectives:

1. Determine intertidal shellfish distributions;
2. Test for contaminants and anthropogenic debris, including small plastics, in the Pacific oyster;
3. Compare contaminants in California mussels and Pacific oysters; and

4. Compare contaminants in Pacific oysters with fish tissue and human health guidelines.

## 2. Materials and methods

In summer 2018 and winter 2019, we conducted intertidal surveys and collected Pacific oysters from eleven public access sites around San Diego Bay, California (Fig. 1). All sites had artificially armored shorelines and all overlapped with sites included in previous contaminant surveys (Bay et al., 2016, Stransky et al., 2016).

### 2.1. Intertidal shellfish distributions (Obj. 1)

In summer 2018 and winter 2019, surveys of sessile and epifaunal shellfish (Mollusca, Crustacea, Echinodermata) were conducted at each of 11 armored shoreline sites (between -0.3 and 1 m above MLLW; Fig. 1) during low tide ( $\leq 0.3$  m MLLW). At each site, a 50 m long  $\times$  3 m wide transect was established parallel to the shoreline and through the highest density oyster zone (between ca. 0.3–1 m above MLLW). The highest density zone was targeted (vs. random surveys; Sagarin and Gaines, 2002) because this zone of productivity is likely where people focus their collections. Mobile epifauna were identified and counted from above within the whole transect area by stepping slowly and in a zig zag fashion along the transect line. Counts of sessile organisms then were made within a 50  $\times$  50 cm quadrat that was placed every 5 m along the transect alternating between a haphazard placement within 1.5 m above and 1.5 m below the transect line. Within each quadrat, identification and counts of live organisms and clean oyster scars (indicating recent removal) were visually assessed from directly above (the sides of boulders and riprap were not assessed). Differences in shellfish abundance and diversity, as well as Pacific oyster abundance, length, weight and percent lipid content between season and sites were tested using paired *t*-tests (season) and one-way ANOVA (sites) in JMP® Pro 15.

### 2.2. Pacific oyster contaminants and plastics (Obj. 2)

Between 20 and 23 Pacific oyster individuals were collected in summer 2018 and 32 in winter 2019 from each of the 11 armored shoreline sites. Pacific oyster individuals that were in the mid- to large size range within each site, as assessed visually, were selected since this was assumed to be the size range most likely to be harvested. Three of the oysters per site were placed whole in individual zip top bags for debris analysis. The rest were measured (longest length of shell), wet weighed and shucked, and the meat was combined in acid washed, sterile glass jars for contaminant analysis. All samples were placed on ice until frozen in the laboratory.

#### 2.2.1. Plastics analysis

In the laboratory, the whole Pacific oyster collected for plastics analysis were thawed, measured (longest length of shell), shucked, and the meat was wet weighed. Oyster meat was digested in 10% potassium hydroxide solution at room temperature for one week (Dehaut et al., 2016; Kühn et al., 2017). Potassium hydroxide has been generally effective at digesting biological tissues and other natural materials (e.g., wool fibers) with little to no degradation of plastic polymers especially at low temperatures (Dehaut et al., 2016, Kühn et al., 2017). After digestion, oyster samples were rinsed with tap water through a 53  $\mu$ m mesh sieve, the smallest size easily visible using a dissecting microscope, to remove potassium hydroxide and reduce the sample volume, and carefully returned to each jar for sorting in water. Samples were examined under a dissecting microscope and, when needed, particles were examined under a compound microscope. Proper microplastic laboratory contaminant control measures were taken, including pre-cleaning work areas, keeping clear lids on or over petri dishes as much as possible while sorting, and using control dishes to measure numbers of ambient fibers, which were then subtracted from sample

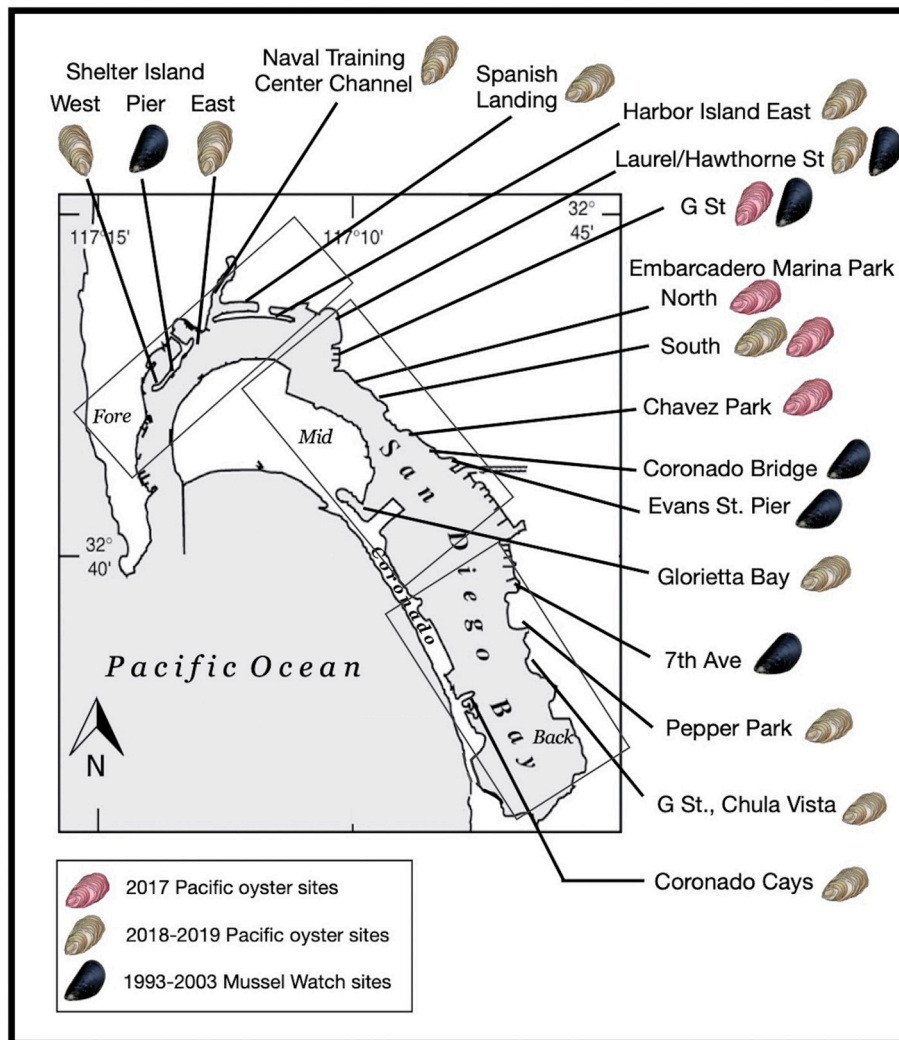


Fig. 1. Publicly accessible, armored shoreline (riprap) sites in San Diego Bay at which 2018–2019 surveys and Pacific oyster samples were collected, a 2017 composite Pacific oyster sample was collected, and 1993–2003 California’s State Mussel Watch Program mussels were deployed and monitored.

data (e.g., Rochman et al., 2015). Differences in abundance and composition of plastics between sites and season were tested using two-way ANOVA.

### 2.2.2. Contaminant analysis

The composite samples for contaminant analysis were kept frozen until analysis at Physis Environmental Laboratory, Inc., Vista Analytical Laboratory, San Diego State University (SDSU), CalScience Environmental Laboratory, or the Marine Pollution Studies Laboratory at Moss Landing Marine Lab (each lab ran different analyses). Tissue samples were analyzed for 69 to 165 compounds across 14 major contaminant classes (Table 1), which included major urban coastal contaminants, and contaminants of emerging concern from the Los Angeles Regional Water Quality Control Board Monitoring List. Traditional bioaccumulation parameters (PCBs, PAHs, PBDEs, OC Pesticides, Metals) were analyzed in accordance with the State of California’s Quality Assurance Program Plans for coastal waters (“QAPP”, SWAMP, 2009, 2018), which requires data results to meet minimum method quality objectives (MQOs) for quality assurance for usability, in addition to specifying required method detection limits (MDLs) and reporting levels (RLs). Quality assurance data, including compliance information for laboratory results, are available to the public on the State of California’s California Environmental Data Exchange (CEDEN.org). All laboratories used in the study, with the exception of SDSU, are certified as accredited under the

California Environmental Laboratory Accreditation Program. Oysters sent to SDSU for individual total mercury analysis used a Milestone direct mercury analyzer (DMA-080) in accordance with USEPA method 7473. Data that did not meet State of California MQOs were not included in the dataset for analysis. For the remaining contaminant classes and compounds, samples were analyzed using best available methods and were run by the same laboratories across samples and seasons.

Contaminant data were grouped by region in San Diego Bay—fore bay ( $n = 5$  sites), mid bay ( $n = 3$  sites), and back bay ( $n = 3$  sites). Differences in percent lipid content of samples between season and site were tested using paired *t*-test and ANOVA, respectively, in JMP® Pro 15. Differences in the suites of contaminants between region and season were visualized using non-metric multidimensional scaling (nMDS; see Clarke, 1993) on Euclidean distance similarity indices of fourth root transformed normalized data in Primer-e v7 (Clarke and Gorley, 2016). Six different random starting points with up to 1000 steps were used. The stress values from the six runs were examined for stability to determine whether a global solution had been found. Only analyses with stress values of  $<0.2$  were used; stress is a measure of how well the solution (in this case the two-dimensional MDS plots) represents the distances between the data. Clarke (1993) suggests values  $<0.1$  are good and  $<0.2$  are useful. Significance testing for differences in contaminant composition among regions of San Diego Bay and seasons was completed using an analysis of similarity (ANOSIM) procedure (Clarke,

**Table 1**

Collection dates, numbers of individuals per composite sample, and contaminant analyses run on Pacific oysters collected from San Diego Bay. The 11 sites sampled in 2018–2019 are shown in Fig. 1. The four east San Diego Bay sites sampled in 2017 include G St., Embarcadero Marina Park North, Embarcadero Marina Park South, and Chavez Park.

Sample information		Sample date				
		Winter 2017	Summer 2018		Winter 2019	
		06 March 2017	16 July 2018	31 July–01 Aug 2018	18 January 2019	14–15 March 2019
# Sites per sample		4	1	1	1	1
# Samples collected		1	11	11	11	11
# Individuals per composite sample		18 (4–5 from each of 4 sites)	5	12–15	5	24
# Analytes tested per sample		159	69	165	69	165
Contaminant classes	Analytical method					
Acid extractable compounds (phenols)	EPA 8270D	x	x		x	
Base/neutral extractable compounds (phthalates, caffeine)	EPA 8270D	x	x		x	
Chlorinated pesticides & degradates (chlordane, DDT, dieldrin), PCBs	EPA 8270D	x		x		x (except PCBs <sup>c</sup> )
Fipronil & degradates	EPA 8270D-NCI	x	x		x	
Metals	EPA 3052M, EPA 245.7 (Hg)	x		x <sup>b</sup>		x
Neonicotinoid compounds	EPA 8270D-NCI	x	x		x	
Organophosphorus Pesticides (chlorpyrifos, DEET, etc.)	EPA 8270D	x	x	x	x	x
Organotins	Krone et al., 1989	x	x		x	
Perfluorooctanesulfonic acid (PFOS)	Modified EPA 537	x	x <sup>a</sup>		x	
Pharmaceuticals and Personal Care Products	EPA 1694	x	x <sup>a</sup>		x	
Phosphate Flame Retardants	EPA 8270D	x	x		x	
Polybrominated diphenyl ethers (PBDEs)	EPA 8270D-NCI	x	x	x	x	x
Polynuclear aromatic hydrocarbons	EPA 8270D	x		x <sup>c</sup>		x
Pyrethroid pesticides	EPA 8270D-NCI	x	x		x	

<sup>a</sup> Only one composite sample from Embarcadero Marina Park South was collected.

<sup>b</sup> Supplemental mercury sampling conducted by San Diego State University at each site ( $n = 88$  total individual oysters).

<sup>c</sup> Laboratory errors prevented the production of usable summer 2018 PAH data and winter 2019 PCB data.

1993) in Primer-e v7. Analyses of contaminant dissimilarities between region and season groups, and the particular contaminants contributing to the dissimilarity, were carried out using SIMPER (Clarke, 1993) in Primer-e v7. The SIMPER results specify which contaminants are responsible for the ANOSIM results by comparing the average normalized concentrations of contaminants between groups. The average dissimilarity between samples from the groups is computed and then broken down into contributions from each contaminant. Those contaminants with high average terms relative to the standard deviation are important in the differentiation of groups.

### 2.3. Comparison with mussel analyses (Obj 3)

Results from contaminant analyses run on a pilot composite oyster sample collected from four sites along the eastern shore of San Diego Bay in March 2017 (Table 1) and the 2018–2019 samples were compared with mussel contaminant data from the same or nearby sites collected during California's State Mussel Watch Program between 1993 and 2003. This was the last decade that mussel monitoring was conducted around San Diego Bay. Data from three State Mussel Watch sites that were nearby the four sites used for the 2017 composite sample were averaged to create comparable contaminant values. Data from all sites were then averaged across sampling dates. Although the sites used in the State Mussel Watch Program and this study aligned well (Table 2), the timeframes did not overlap, with mussel data from 1993 to 2003 and oyster data from 2017 to 2019. We used descriptive statistics to assess contaminant patterns across sites and to indicate large differences between mussels and oysters. Prior to comparison, contaminant analysis methods were evaluated for comparability given the extended time period separating sampling (15–25 years). Samples were largely deemed appropriate for comparison purposes, though PCB analysis methods differed between the two studies. Mussels were historically analyzed for

**Table 2**

Locations of San Diego Bay sites used to compare contaminants in mussel from California's State Mussel Watch Program (1993–2003) and Pacific oysters from this study (2017–2019). The composite sample from 2017 used in this study was compared to an average of three similarly located sites used from 1993 to 2003 in the State Mussel Watch Program.

Bay region	State Mussel Watch		This study	
	Site name (1993–2003)	Lat/Long	Site name (2017–2019)	Lat/Long
Fore bay	Shelter Island Pier	32.712°, –117.228°	2018–2019 Shelter Island West	32.708°, –117.234°
Central east shore-north	Laurel St	32.728°, –117.179°	2018–2019 Laurel/Hawthorne	32.727°, –117.179°
Back bay	7th Ave	32.670°, –117.123°	2018–2019 Pepper Park	32.650°, –117.112°
	1993–2003 Averaged data		2017 Composite sample	
Central east shore-south	G St	32.712°, –117.176°	G St	32.712°, –117.175°
	Evans St	32.693°, –117.149°	Embarcadero	32.708°, –117.169°
	Pier	32.692°, –117.151°	Marina Park North	32.703°, –117.164°
	Coronado		Embarcadero	32.703°, –117.164°
	Bridge		Marina Park South	32.696°, –117.151°
			Chavez Park	32.696°, –117.151°

PCBs as various Aroclors, while Pacific oyster monitoring used a State of California list of PCB congeners (SWAMP, 2009). Thus, PCB comparisons were done for mussels and Pacific oysters using Aroclor 1254 and summed total PCB congener concentrations, respectively. While mussels were typically tested for more than one Aroclor, Aroclor 1254 was selected due to its consistency in analysis across samples and years, and to avoid any potential inherent bias with summation of Aroclors.

2.4. Comparison with fish tissue limits and human health guidelines (Obj 4)

Contaminant concentrations in Pacific oysters from this study and mussels from California’s State Mussel Watch Program were both compared to human consumption thresholds published by the State of

California as California Fish Contaminant Goals and California Advisory Tissue Levels (Klasing and Brodberg, 2008, 2011), as well as European Union commercial bivalve import criteria (EU, 2006). California’s fish contaminant goals represent “a starting point for OEHHA [Office of Environmental Health Hazard Assessment] to assist other agencies that

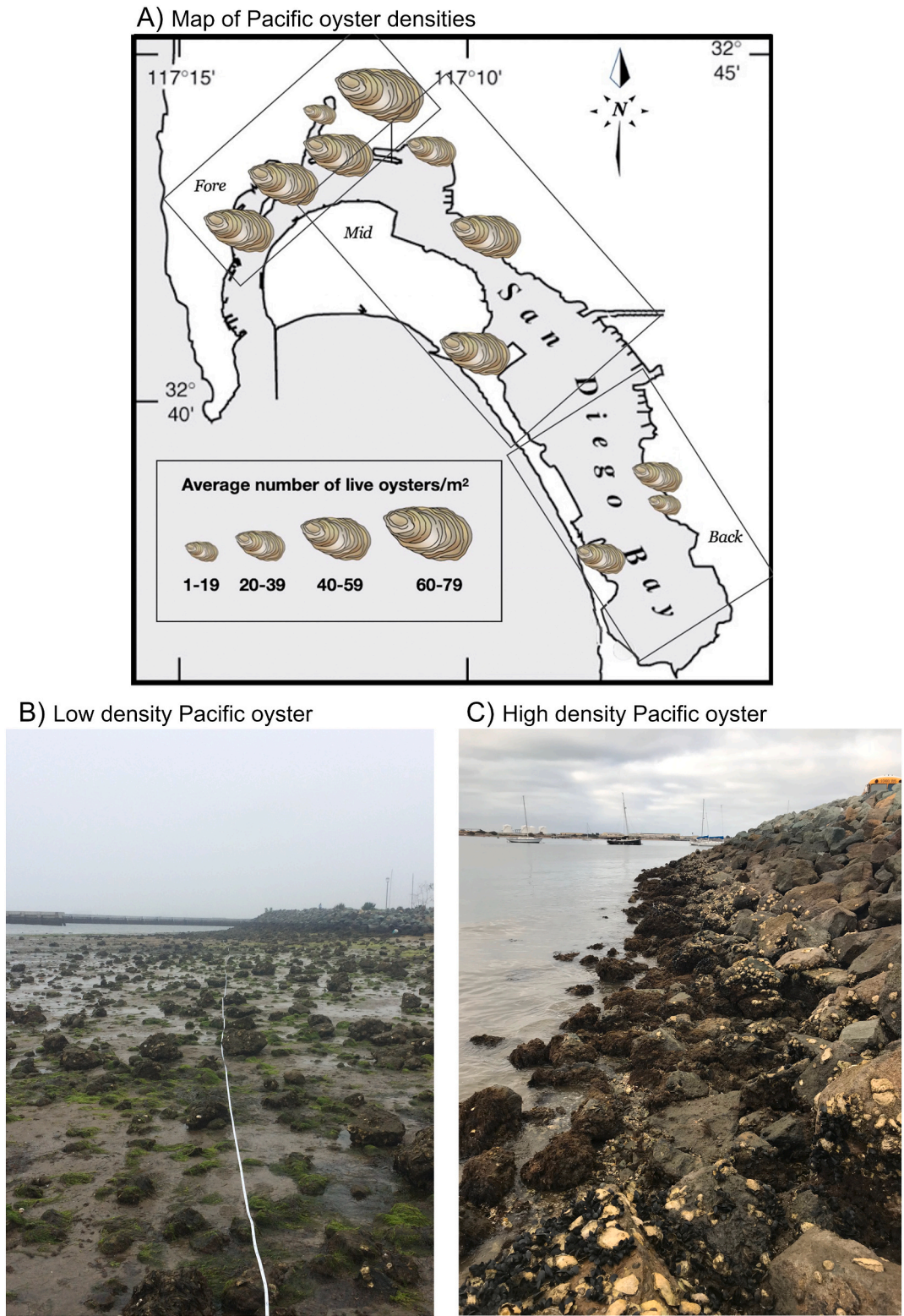


Fig. 2. Pacific oyster densities on San Diego Bay. (A.) Map of study locations and average ( $\pm 1SE$ ) Pacific oyster densities around San Diego Bay during summer 2018 and winter 2019. Densities were averaged across dates,  $N = 20$  quadrats per site (10 quadrats per site  $\times$  2 dates). Photos are of (B.) low density (G. Street, Chula Vista) and (C.) high density (Shelter Island East) Pacific oyster distributions in sites (photos taken: January 2019).

wish to develop fish tissue-based criteria with a goal toward pollution mitigation or elimination,” while advisory tissue levels “provide a number of recommended fish servings that correspond to the range of contaminant concentrations found in fish and are designed to prevent consumers from being exposed to more than the average daily reference dose for non-carcinogens or to a risk level greater than  $1 \times 10^{-4}$  for carcinogens” (Klasing and Brodberg, 2008). The same comparisons were outlined for raw plankton samples collected during the same year as the summer oyster collection (2018) during a separate study (Bay and Parks, 2020) that analyzed plankton as tissue for a subset of pollutants (PCBs, DDTs, Chlordane).

Differences between the Pacific oyster contaminant concentrations found in this study and the published consumption thresholds were tested using *t*-tests or, when assumptions of normality were not met, non-parametric Wilcoxon tests in R (R Core Team, 2020).

### 3. Results

#### 3.1. Shellfish distributions

Shellfish species richness (number of species) on the riprap tended to be highest at both Shelter Island sites, Harbor Island East, and Embarcadero (ANOVA  $p < 0.001$ ,  $F_{10,99} \geq 14.5$ ) during both sample dates with 10–17 species per site (Avg  $\pm$  1SE:  $3.4 \pm 0.4$ – $7.1 \pm 0.7$  species per  $0.25 \text{ m}^2$ ). The other 7 sites were each populated by 3–9 species per site ( $1.4 \pm 0.4$ – $4.4 \pm 0.3$  species per  $0.25 \text{ m}^2$ ). Species richness did not differ between summer and winter (Paired *t*-test:  $p = 0.61$ ,  $t_{109} = 0.52$ ). Most of these species would likely not be of interest to harvesters for food (e.g., chitons, limpets), however some are of interest for other uses, such as bait (e.g., lined shore crabs; Pedersen and Talley, 2021).

Of the species found in our sites, the Pacific oyster and Olympia oyster, and Mediterranean mussel are the three species that would likely be of most interest for food harvest. The Pacific oyster was the predominant oyster in all sites except for Shelter Island west where Olympia oyster dominated the survey area ( $47 \pm 9$  individuals/ $\text{m}^2$  in winter to  $63 \pm 18$  individuals/ $\text{m}^2$  in summer, but Pacific oyster was prevalent nearby in an area with more difficult access). Average ( $\pm$ 1SE) Pacific oyster densities across sites ranged from  $8 \pm 2$ – $56 \pm 7$  individuals/ $\text{m}^2$  in summer and  $1 \pm 1$ – $53 \pm 9$  individuals/ $\text{m}^2$  in winter, with the highest densities ( $\geq 40$  oysters/ $\text{m}^2$ ) found in the northern half of San Diego Bay on both dates (ANOVA  $p \leq 0.001$ ,  $F_{10,99} \geq 3.2$ ; Fig. 2). There was a bay-wide average of  $7.8 \pm 3.0/\text{m}^2$  fewer Pacific oysters in winter than summer across sites (paired *t*-test:  $p = 0.010$ ,  $t_{109} = 1.98$ ).

The lengths and weights of Pacific oysters collected did not differ with date (overall average:  $111 \pm 2$  cm length,  $27.6 \pm 1.5$  g wet wt; Paired *t*-test:  $t_{43} \leq -0.9$ ,  $P \geq 0.37$ ), but did differ across sites (ANOVA:  $F_{10,77} = 4.6$ ,  $P < 0.001$  for both length and weight), with smaller oysters in the fore bay ( $81.6 \pm 4.8$ – $84.1 \pm 4.7$  cm long and  $14.5 \pm 2.6$ – $18.4 \pm 1.5$  g at Shelter Island West and East) as compared to sites in the mid and back bay ( $111.8 \pm 10.0$ – $116.7 \pm 5.9$  cm and  $29.5 \pm 2.2$ – $37.5 \pm 4.9$  g at Spanish Landing, Laurel/Hawthorne, Glorietta, and Pepper Park). Percent lipid content of samples did not differ with season (paired *t*-test,  $p = 0.76$ ,  $t_{10} = 0.31$ ) or site (ANOVA:  $F_{10,11} = 0.91$ ,  $P = 0.55$ ), with an overall average ( $\pm$ 1SE) of  $8.6 \pm 0.3\%$  lipid.

The presence of clean attached oyster shells, evidence of recent oyster removals, was most obvious at the sites at the northern part of San Diego Bay in summer (both Shelter Island sites, Harbor Island, NTC Channel) and at all sites in winter. In fact, density of clean shell increased, on average across sites, by  $2.5 \pm 0.27$  shells/ $\text{m}^2$  between summer and winter (paired *t*-test:  $p < 0.001$ ,  $t_{109} = 9.11$ ).

The Mediterranean mussel had a patchy distribution between and within sites. The mussel was found in only six sites around San Diego Bay on each date. When present, the average density of the mussel per site ranged from  $0.4 \pm 0.4$ – $74 \pm 30$  individuals/ $\text{m}^2$  in summer and  $0.4 \pm 0.4$ – $151 \pm 49$  individuals/ $\text{m}^2$  in winter, with the highest densities at Shelter Island East and West (ANOVA  $p < 0.001$ ,  $F_{10,99} \geq 5.5$ ), the

closest sites to the ocean. Density of mussels did not vary between summer and winter (paired *t*-test:  $p = 0.27$ ,  $t_{109} = 1.10$ ).

#### 3.2. Pacific oyster contaminants

Pacific oysters from all sites contained between 7–11 classes of contaminants in summer and 9–10 classes of contaminants in winter (Table 3). Many contaminants were present in Pacific oysters from all 11 sites around San Diego Bay on both dates, including benzylbutyl phthalate, tributyltin, most metals (zinc, aluminum, arsenic, cadmium, copper, lead, manganese, nickel, silver), selenium, and mercury (Table 3). Both season and location were associated with differences in the composition (types and concentrations) of contaminants in Pacific oyster (Table 4; Fig. 3). In the summer across San Diego Bay, Pacific oysters contained higher concentrations of pesticides, including neonicotinoid and chlorinated compounds, selenium, and several metals (aluminum, cadmium, copper, nickel, silver and zinc) (Table 3). PCB concentrations were high, too, but a laboratory error prevented the production of usable winter PCB data and therefore comparison across season. In winter, the oysters across San Diego Bay had higher concentrations of PBDEs, pyrethroids, benzylbutyl phthalate, chromium and manganese (SIMPER, analytes explaining up to 75% of the differences between seasons across regions). PAH concentrations were also relatively high in winter, but a second laboratory error prevented the production of usable summer PAH data thereby limiting comparisons across season. There were also differences in Pacific oyster contaminants between the three regions of San Diego Bay within each season (Table 4; Fig. 3). During the summer, contaminant composition of Pacific oyster from the back bay had higher concentrations of chromium, nickel and silver, than those in both the fore bay, which had higher concentrations of arsenic, cadmium, lead and manganese, and in the mid bay, which were higher in chlorpyrifos, PBDEs, lead and mercury. In the winter, Pacific oyster contaminant composition differed between the back and fore bay with higher concentrations of aluminum and manganese in the back bay, and higher concentrations of mercury, lead, tributyltin and copper in the fore bay (SIMPER, analytes explaining up to 75% of the differences between seasons across regions).

Two sites had unique contaminants: Embarcadero Marina Park South was the only site in which the organophosphate pesticide chlorpyrifos was detected, and Harbor Island East was the only site to detect the organochlorine pesticide dieldrin (both only in summer). Other contaminants were detected in Pacific oysters across all sites in one season, but then limited to particular sites during the other seasons (Table 3). Pyrethroids and Galaxolide were detected in oysters from all sites in winter, but only at Spanish Landing (pyrethroids) and Embarcadero (Galaxolide) in summer (Table 3). Similarly, the chlorinated pesticide DDT and its degradates were detected in Pacific oysters from all sites in summer, but only at Shelter Island West in winter (Table 3).

#### 3.3. Pacific oyster plastics

On both dates, all 11 sites around San Diego Bay had Pacific oysters that contained small plastics with average ( $\pm$ 1SE) abundance per individual ranging from  $0.7 \pm 0.7$ – $9.3 \pm 2.0$  in summer 2018 to  $3.3 \pm 0.7$ – $14.3 \pm 0.9$  in winter 2019 (Fig. 4). Of the 33 individual oysters tested on each date (3 individuals per site  $\times$  11 sites), 88% (29) contained plastics in summer 2018 and 97% (32) had plastics in winter 2019.

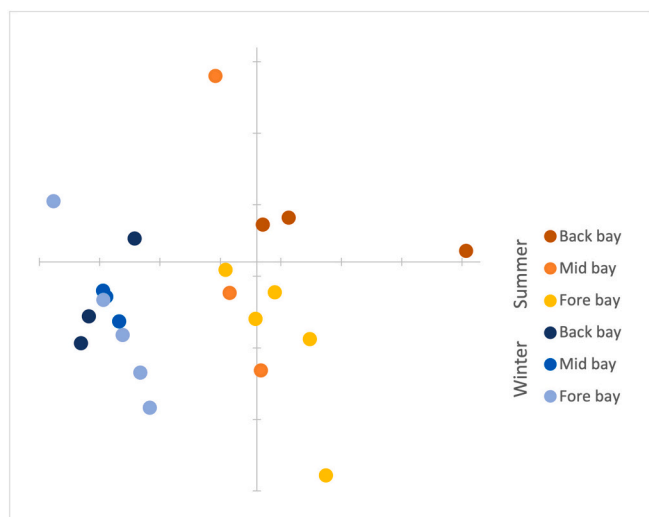
The number of plastics per individual varied with date and site (2-Way ANOVA  $P = 0.0137$ ,  $F_{21,44} = 2.2$ ,  $n = 66$ , Site  $p = 0.24$ , Date  $p = 0.0015$ , Date  $\times$  Site  $p = 0.0415$ ; Fig. 4). Averaged across San Diego Bay, 64% more plastics were found in Pacific oysters in winter ( $6.7 \pm 0.8$  pieces per individual) than summer ( $4.1 \pm 0.6$  pieces per individual). The winter increase in plastics was particularly prominent at Spanish Landing, Laurel/Hawthorne, Embarcadero, G St, and Coronado Cays, where the number of particles per individual more than doubled (Fig. 4).



**Table 4**

Differences in the suites of contaminants observed between the three regions of San Diego Bay (fore, mid and back) and between seasons (summer, winter). Data are from summer 2018 and winter 2019,  $n = 4$  fore, 3 mid, and 3 back bay sites. Differences in contaminants between sites are measured as average summed Euclidean distances. ANOSIM Global  $P = 0.007$ ,  $R = 0.28$ ; pairwise  $p$  values are shown below the diagonal, within-site and season differences (distances) are along the diagonal, and pairwise differences (distances) are above the diagonal. Pairwise tests with  $p$  values of  $\geq 0.10$  are shown in bold.

Season	Bay regions	Summer			Winter		
		Fore	Mid	Back	Fore	Mid	Back
Summer	Fore	11	30	<b>36</b>	<b>43</b>	<b>41</b>	<b>45</b>
	Mid	0.36	18	<b>41</b>	<b>43</b>	<b>41</b>	<b>44</b>
	Back	<b>0.05</b>	<b>0.10</b>	15	<b>57</b>	<b>52</b>	<b>49</b>
Winter	Fore	<b>0.01</b>	<b>0.02</b>	<b>0.02</b>	12	14	25
	Mid	<b>0.02</b>	<b>0.10</b>	<b>0.10</b>	0.84	3	14
	Back	<b>0.02</b>	<b>0.10</b>	<b>0.10</b>	<b>0.09</b>	0.20	8



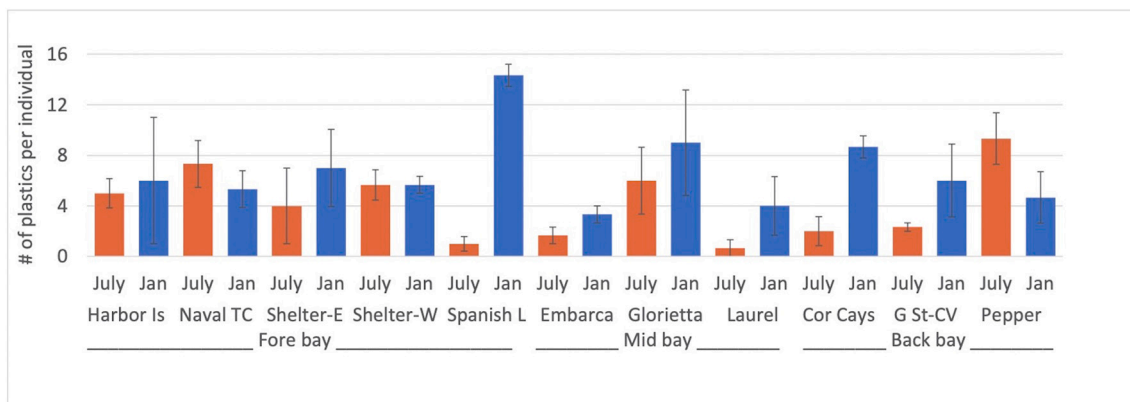
**Fig. 3.** Non-metric multi-dimensional scaling (nMDS) of the suite of contaminants found in three regions of San Diego Bay over two seasons, summer and winter. Data are from summer 2018 and winter 2019,  $n = 3$  back bay sites, 3 mid bay sites, and 5 fore bay sites. Stress = 0.15.

**4. Discussion**

The lack of recent bivalve contaminant data for San Diego Bay, coupled with both a dearth of publicly available data for Pacific oysters and recent evidence of contamination in higher-trophic level species, revealed a key gap in our knowledge about the suitability of Pacific oysters for human and wildlife consumption. Filling this gap is especially important given the recent proliferation of feral non-native Pacific oysters throughout southern California bays and estuaries – including on publicly accessible points throughout San Diego Bay (Tronske et al., 2018, this study) – which has provided an easily accessible food source to natural predators and to those in the community interested in recreational and subsistence consumption. Risk to consumers can be difficult to predict because there are many factors influencing contaminant availability and shellfish uptake and accumulation, including seasonal weather and bay circulation patterns, location-specific factors (e.g., local hydrology and sediment characteristics, adjacent and upstream land and water uses), and the biology of the organisms (e.g., local population genetics, phenology, life-style, and feeding strategies) all of which can influence an organism’s contaminant concentrations (e.g., Schiff et al., 2000; Rowe, 2008; Katagi, 2010; Walkinshaw et al., 2020).

**4.1. Seasonal risks**

In general, season strongly influenced contaminant composition in Pacific oysters from San Diego Bay. This seasonality of risk is well known for algal toxins, such as paralytic shellfish and domoic acid poisoning, for which seasonal harvest restrictions are common around the world, including the May 1 through October 31 annual bivalve quarantine in San Diego (CDPH, 2021). For anthropogenic chemicals, understanding seasonality in contamination provides managers with important information as to potential pollutant sources (e.g. stormwater) in addition to consumer risk and may be especially important in semi-arid regions such as Southern California, where contaminant transport, accumulation and availability may vastly differ across the hot dry summers and cool wet winters (Stein et al., 2006; Tiefenthaler et al., 2008; SDBDSW, 2016). Although unintended, protection from some chemical contaminants may be conferred to consumers by the five-month long annual shellfish closure for harmful algae. During the summer as compared to winter in San Diego Bay, there were elevated concentrations of neonicotinoid pesticides, a relatively new class of water-soluble pesticides that are



**Fig. 4.** Average ( $\pm 1SE$ ) number of plastic pieces per individual Pacific oyster collected in summer (July) 2018 and winter (January) 2019 from sites around San Diego Bay.  $N = 3$  individuals per site.



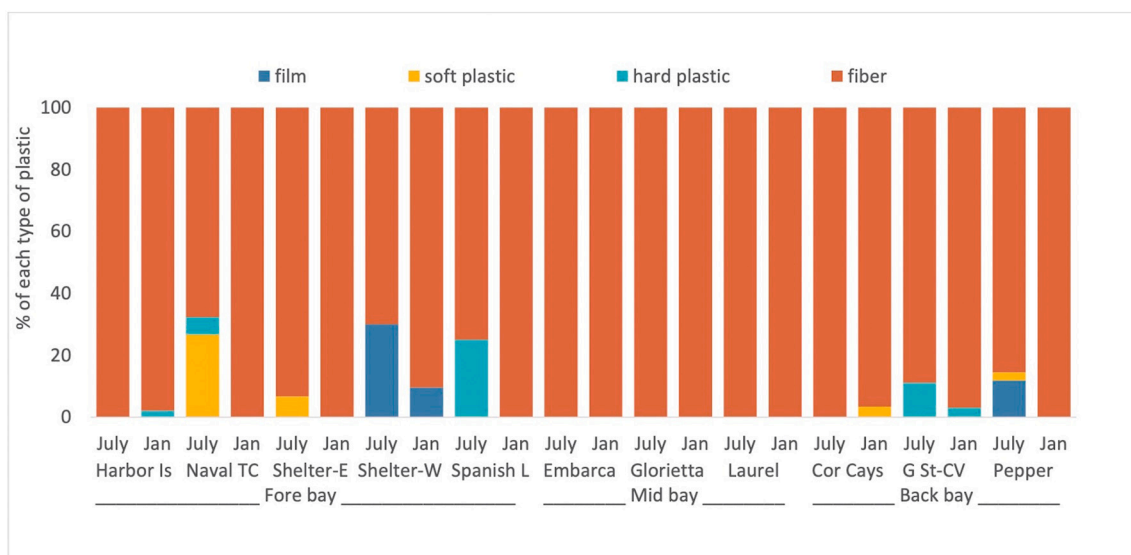


Fig. 5. Average proportions of microplastics in Pacific oysters from San Diego Bay in summer (July) 2018 and winter (January) 2019. N = 3 individuals per site per date.

Table 5

Comparison of pollutant concentrations (ng/g wet weight) in bay mussels (*Mytilus* spp.) and Pacific oysters (*Crassostrea gigas*) collected from the same areas around San Diego Bay. Shown are the contaminants for which concentrations in the two organisms did not overlap and/or that have experienced changes in use across the study period (i.e., are of management interest). Mussels were analyzed as part of the California State Mussel Watch program between 1993 and 2003 (the last decade of available data); Pacific oysters were analyzed as part of this study (2017–2019). Data are average ± 1 standard error; n = 2–10 mussel samples (one per year for as many years as the data were available), and for Pacific oyster samples: n = 1 (central east south), n = 1 (other sites for PCBs and PAHs), and n = 2 (other sites for all other analytes). Sites with the same or similar locations were paired between the two studies for comparison: Back bay = 7th Street (mussel) and Pepper Park (oyster); Central-east-south = averaged Evans St and Coronado Bridge (mussel) and a composite from G St, Embarcadero Marina Park North and South, and Chavez Park (oyster); Central-east-north = Laurel St (mussel and oyster); Fore bay = Shelter Island pier (mussel) and Shelter Island west (oyster). Contaminants that had overlapping concentration ranges across the two taxa and thus are not shown here include chlordane, Chlorpyrifos, organochlorine pesticides, arsenic, selenium, and manganese. – = no data.

	Biological properties				Organics					
	Lipid (% weight weight)		Moisture (% wet weight)		PAHs		PCBs			
	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster		
Site	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	
Back bay	1.1 ± 0.2	7.0 ± 0.6	95 ± 10	88 ± 2	7850 ± 4649	364	85 ± 5	9		
Central east south	1.1 ± 0.2	10.6	85 ± 1	86	2133 ± 1829	919	94 ± 10	44		
Central east north	0.6 ± 0.1	8.1 ± 1.7	88 ± 0	88 ± 2	179 ± 61	442	110 ± 10	60		
Fore bay	1.1 ± 0.0	7.6 ± 1.0	84 ± 1	83 ± 5	194 ± 74	425	73 ± 9	36		
	Metals									
	Chromium		Copper		Lead		Nickel		Zinc	
	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster
Site	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE
Back bay	1159 ± 524	182 ± 21	6923 ± 825	144,111 ± 1969	841 ± 135	90 ± 8	210 ± 37	87 ± 28	62,368 ± 5924	640,534 ± 239,246
Central east south	891 ± 321	–	3518 ± 434	283,900	1083 ± 115	254	269 ± 32	–	43,114 ± 4327	1,302,020
Central east north	1239 ± 695	96 ± 96	2667 ± 445	162,054 ± ###	1295 ± 195	148 ± 16	311 ± 89	66 ± 6	35,882 ± 4287	539,649 ± 12,304
Fore bay	1088 ± 526	134 ± 134	2893 ± 259	163,743 ± 4743	865 ± 117	178 ± 21	410 ± 107	91 ± 31	49,146 ± 2656	422,971 ± 85,289
	Trace or light metals, & related									
	Aluminum		Cadmium		Mercury		Tributyltin			
	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster
Site	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE	Avg ± SE
Back bay	147,114 ± 28,033	29,837 ± 1579	777 ± 116	424 ± 32	29 ± 2	9 ± 4	115 ± 9	13 ± 0		
Central east south	86,849 ± 15,834	–	997 ± 188	–	39 ± 6	19	101	19		
Central east north	53,977 ± 8234	17,830 ± 5036	1395 ± 350	426 ± 29	30 ± 5	13 ± 1	127 ± 32	5 ± 1		
Fore bay	153,892 ± 11,612	29,707 ± 3047	1005 ± 175	611 ± 50	42 ± 9	17 ± 1	30 ± 29	14 ± 7		

**Table 6**

Threshold concentrations of organic and metal contaminants put forth by various groups as guidelines for wildlife tissue limits and human consumption, compared to concentrations found in Pacific oyster from this 2017–2019 study, mussels from 1993 to 2003 of California's State Mussel Watch Program, and plankton in the same bay from 2018 (Bay and Parks, 2020).

Organism	Pollutant	Threshold (ppb wet weight)					Results					
		FCG	ATL (2)	ATL (1)	ATL (0)	EU	Min	Max	Mean	SD	p-Value <sup>a</sup>	
Oyster, <i>C. gigas</i> (2017–2019)	Chlordane	5.6	190	280	560	–	ND	5.69	0.27	1.21	NA	
	DDTs	21	520	1000	2100	–	ND	2.68	0.67	1.52	NA	
	Dieldrin	0.46	15	23	46	–	ND	0.01	0.00005	0.002182	NA	
	Mercury	220	70	150	440	–	5	47.65	26.82	8.04	NA	
	PCBs	3.6	21	42	120 <sup>a</sup>	–	8.33	132.57	36.27	33.64	<0.001	
	Selenium	7400	2500	4900	15,000	–	324.45	589.57	444.52	80.64	NA	
	PBDEs	310	100	210	630	–	ND	16.17	2.22	2.85	NA	
	Benzo[a]pyrene	–	–	–	–	5	ND	26.69	2.85	7.94	<0.05	
	PAHs	–	–	–	–	30	ND	516.88	47.13	148.19	<0.05	
	Cadmium	–	–	–	–	1000	320.15	829.3	474.16	128.52	NA	
	Lead	–	–	–	–	1500	68.327	302.4	147.73	63.00	NA	
	Mussel, <i>Mytilus</i> (1993–2003, n = 28–38)	Chlordane	5.6 <sup>a</sup>	190	280	560	–	0.6	12.9	2.91	2.17	<0.001
		DDTs	21	520	1000	2100	–	2.42	18.9	6.34	3.50	NA
		Dieldrin	0.46	15 <sup>a</sup>	23	46	–	ND	4.4	0.98	0.69	<0.001
Mercury		220	70 <sup>a</sup>	150	440	–	19	94.5	34.54	13.70	<0.001	
PCBs		3.6	21	42	120 <sup>a</sup>	–	26.7	142.2	84.35	31.81	<0.001	
Selenium		7400	2500	4900	15,000	–	ND	781.77	355.86	184.75	NA	
PBDEs		310	100	210	630	–	nm	nm	nm	nm	nm	
Benzo[a]pyrene		–	–	–	–	5	0	369.1	71.5	123.9	NSL	
PAHs		–	–	–	–	30	6.5	3518	658.9	1040	NSL	
Cadmium		–	–	–	–	1000	500	1933.34	1000.05	419.22	NSL	
Lead		–	–	–	–	1500	420	2300	1006.05	406.57	<0.001	
Plankton (2018, n = 12)		DDTs	21	520	1000	2100	–	1.02	8.85	3.82	2.56	NA
		PCBs	3.6	21	42	120 <sup>a</sup>	–	12.80	176.00	57.88	44.40	<0.001
		Chlordane	5.6 <sup>a</sup>	190	280	560	–	ND	15.20	2.40	4.39	<0.001

FCG = California Fish Contaminant Goal.

ATL() = California Advisory Tissue Level, with () indicating number of weekly servings.

EU = European Union Import Criteria.

NA = maximum value at or below lowest threshold.

ND = non-detect.

PAHs = Sum of benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene.

NSL = not significantly lower than threshold.

nm = not measured.

<sup>a</sup> Indicates the maximum criteria level for which the mean values were significantly less.

widely used in residential, public, and agricultural settings (Craddock et al., 2019; Bakker et al., 2020), but for which there are not yet fish tissue or consumption thresholds. The high summer concentrations of neonicotinoid pesticides may have been due to increased use to combat summer insect pests throughout the urban watersheds upstream of and in parks surrounding San Diego Bay, where localized irrigation runoff was observed during sampling for this project.

Summer was also associated with relatively high concentrations of banned hydrophobic pollutants, in particular chlorinated pesticides and PCBs, known to be persistent in sediment (e.g., surface and suspended sediments), where they can be taken up and accumulated by organisms (Honeycutt and Shirley, 2014; Stransky et al., 2016). Although a laboratory error prevented the production of useable winter PCB data for seasonal comparison, mean and maximum summer PCB levels were similar to and sometimes higher than those for fish listed in the State of California advisory (OEHHA, 2018). Several metals and selenium, although abundant in both seasons, were also elevated in Pacific oysters during the summer. While high metal concentrations are expected during winter in association with storm water runoff from developed areas (Tiefenthaler et al., 2008; Chiba et al., 2011), some metals are predominantly used in summer, such as copper and zinc, which are commonly involved in boat hull anti-fouling (e.g. Niera et al., 2009; Briggs and D'Anna, 2012).

Other extrinsic drivers of the high summer metal and persistent organic pollutant burden are also likely, especially considering the similar size and lipid content of Pacific oysters between seasons. Summer is associated with lower rates of bay circulation and less turn-over with “clean” ocean waters, as well as a higher average inundation time of bay water, all primarily driven by summer thermal stratification

(Chadwick and Largier, 1999; Chadwick et al., 2004), which could increase exposure of mid-intertidal organisms, like the Pacific oyster, to contaminants. The average higher summer temperatures (+10 °C) may influence biological processes, such as increasing activity of benthic bioturbators and blooms of plankton and bacteria that can increase suspension and/or availability of sediment-bound hydrophobic contaminants (e.g., Baines et al., 2004; Peng et al., 2001; Banta and Andersen, 2003; Cabrita et al., 2020). When coupled with greater oyster feeding rates in the summer (Brown, 1988), summer increases in bioavailability can translate to increased contamination levels.

In winter, when the shellfish quarantine is not in effect, several classes of pollutants were found in Pacific oysters in higher concentrations than in summer, including PBDEs, pyrethroids, benzylbutyl phthalate, chromium, manganese, and plastics. Although we had no summer data, PAHs were also found in Pacific oyster in winter which was expected given their association with urban and industrial stormwater runoff and sediment resuspension (Stein et al., 2006; Niera et al., 2009; Koudryashova et al., 2019). In some samples, PAH concentrations exceeded human health thresholds (EPA, 2000a; EU, 2006). Pyrethroids, a group of hydrophobic pesticides commonly bound to sediment, were more widespread and in higher concentrations in winter, likely due to stormwater flows (Hayman et al., 2019; Méjanelle et al., 2020). No human consumption thresholds exist for pyrethroids, which were initially considered safe for humans, but for which human health effects have recently emerged (Chrutek et al., 2018), making risk uncertain. Like pyrethroids, winter stormwater (and sediment) flows also likely contributed to the highest Pacific oyster concentrations of PBDE flame retardants, the plasticizer butylbenzyl phthalate, and both chromium and manganese (e.g., Mi et al., 2019; Wu et al., 2019). PBDE

concentrations did not exceed human consumption thresholds (Klasing and Brodberg, 2011). While chromium and manganese occur naturally in the environment and in food, there are forms of these elements that are toxic by-products of manufacturing processes (EPA, 2000b; ATSDR, 2012). Since the toxic forms were not distinguished in this study, consumption risks are uncertain. No food consumption limits exist for butylbenzyl phthalate, although a risk evaluation for butylbenzyl phthalate is underway (EPA, 2020) and there is a daily oral dose limit of 1200 mg/day for a 58 kg woman (OEHHA, 2012). This daily limit is five orders of magnitude higher than the estimated amount that would be in an average sized Pacific oyster (28 g wet wt) if it contained the highest levels of this phthalate (0.0005 mg/g wet wt) found in this study.

Winter also brought with it greater abundances of plastics in Pacific oysters, especially fibers and film particles. This seasonal increase is consistent with prior plastic plankton surveys in San Diego Bay, which documented increased densities after winter storm events (SDBDSW, 2016). Small plastics and other anthropogenic debris have been found in wetland and bay fishes and crustaceans from San Diego Bay (SDBDSW, 2016; Talley et al., 2020; Pedersen and Talley, 2021), and in Pacific oysters from the U.S. west coast and around the world (Danopoulos et al., 2020). The frequency of plastics presence (88–97% of all individuals tested) and/or average ( $\pm 1$ SE) density ( $0.25 \pm 0.29$  pieces per g ww or  $5.4 \pm 1.6$  pieces per ind.) in Pacific oysters found in this study were similar to or higher than those reported in studies from Europe (80–93%,  $\leq 0.47 \pm 0.16$  pieces per g ww,  $2.1 \pm 1.7$  pieces per ind.) and South Korea ( $\sim 95\%$ ,  $0.07 \pm 0.06$  pieces per g ww,  $0.77 \pm 0.74$  pieces per ind.); and within the range of those reported from China (84–100%,  $0.26 \pm 0.29$ – $0.80 \pm 0.20$  pieces per g ww,  $1.5 \pm 1.06$ – $4.7 \pm 0.3$  pieces per ind.) and elsewhere on the U.S. west coast (33% of individuals;  $0.35 \pm 0.13$  pieces per g ww;  $0.69 \pm 0.26$ – $10.95 \pm 2.43$  pieces per ind.) (Van Cauwenbergh and Janssen, 2014; Rochman et al., 2015; Phuong et al., 2018; Baechler et al., 2019; Cho et al., 2019; Teng et al., 2019; Zhu et al., 2019). The effects of plastics consumption on individual organisms and food webs are still being evaluated, but so far include the transfer of environmental contaminants, which are adsorbed to the plastics, to the consumer (Teuten et al., 2009; Rochman et al., 2014), accumulation of plastics in the gut and gills of consumers (Murray and Cowie, 2011; Watts et al., 2014; Browne et al., 2008), and physical or chemical damage to consumers' internal organs and cellular function (Rochman et al., 2013; Browne and Thompson, 2013). Knowledge about human health risks from seafood containing small plastics is in its infancy (SCCWRP, 2020) so that, for now, consumption limits and associated risks remain uncertain.

#### 4.2. Locational risks

Location-specific information on contaminant dynamics—as compared to bay- or region-wide information—is especially important for informing and encouraging safe consumption of sessile species, whose contamination levels may be strongly tied to local conditions, as well as informing regulatory management for pollutant control and remediation purposes. This study revealed differences in Pacific oyster contaminant composition across broad regions of San Diego Bay, in particular, higher concentrations of several metals associated with manufacturing (aluminum, chromium, nickel, silver) in the back bay, relative to the mid- and fore bay which were higher in lead and mercury despite decades-long bans on uses of these elements, although neither exceeded consumption thresholds. Compared to the back bay, mid bay Pacific oysters also had higher levels of PBDEs and chlorpyrifos, which we hypothesize is due to stormwater inputs from the adjacent urbanized watersheds and industrial areas, although neither exceeded consumption thresholds. Pacific oysters in the fore bay also had higher levels of arsenic, and both copper and tributyltin likely associated at least in part with past and present boatyard activities. Higher concentrations of total arsenic may be partially explained by seepage from groundwater (Ford et al., 2008), but consumption risks are uncertain since levels of the toxic

inorganic form of arsenic were not distinguished.

Some contaminants were unique to particular sites. The pesticides chlorpyrifos and dieldrin, banned in the U.S. since 1987, were found in only one site each. Dieldrin concentrations were lower than human consumption thresholds (Klasing and Brodberg, 2008). Consumption guidelines do not exist for chlorpyrifos but there is a daily oral dose limit of  $0.58 \mu\text{g}/\text{day}$  (OEHHA, 2020). This daily limit is 10 times higher than the estimated amount that would be in one average sized Pacific oyster (28 g wet wt) if it contained the highest levels of chlorpyrifos ( $0.060 \mu\text{g}/\text{g}$  wet wt) found in this study. In other instances, location interacted with season. Contaminants such as pyrethroids and Galaxolide were found throughout San Diego Bay in winter, but found in only one site each during the summer. Further, despite the ubiquity of pyrethroids in winter, concentrations were particularly high at only one site. However, no guidelines for pyrethroids and Galaxolide exist. Similarly, levels of DDT and its degradates, which were below consumption thresholds (Klasing and Brodberg, 2008), were present in Pacific oysters from all sites in summer, but only one site in winter, indicating that locally-specific processes (e.g., localized irrigation runoff or stormdrain inputs, local hydrologic patterns) were at play, contributing to year-round availability and uptake at particular locations throughout San Diego Bay, and highlighting the need for location-specific information on contaminant dynamics.

#### 4.3. Species-specific risks

The differences in California mussels, a commonly used sentinel species and that used in California's State Mussel Watch Program, and Pacific oyster contaminant levels observed in this study were likely due in part to interspecific differences in biology and life history. Lipid content was counter-intuitively higher in the less-contaminated Pacific oysters, which does not explain observed differences. Lifestyle, such as benthic, demersal and pelagic dwelling, trophic level, and feeding strategy, such as filter feeding and deposit feeding, may influence organisms' exposure to and uptake of contaminants, including small plastics from sediment and water (SDBDSW, 2016; Stransky et al., 2016; Talley et al., 2020). Even within a group of organisms that share a general feeding strategy, such as filter feeders, there may still be inter- and intraspecific differences associated with the organisms' biology and interactions with the environment. In contrast to California mussels, Pacific oysters are estuarine species that exhibit differences in feeding behavior based on abiotic environmental factors (e.g., temperature, salinity; Comeau et al., 2008; Casas et al., 2018), as well as prey selectivity (Rosa et al., 2018). Further, the organisms' physiology, which can be linked to local population genetics, may influence the uptake, physiological responses, metabolic conversion, and/or elimination of contaminants (Katagi, 2010). These factors may be partially responsible for the differences in contamination levels observed across individual oysters and between oysters and mussels in this study. Further side-by-side studies of contaminant uptake for the two species, both of which are available for human harvest, are warranted.

#### 4.4. Temporal changes in risk

Changes in the environment, management actions and use of chemicals with time may also have influenced differences in contamination levels between Pacific oysters and California mussels. Environmental shifts such as those associated with climate change, including altered precipitation and hydrologic regimes, sediment dynamics, and water temperatures, can influence the modes of contamination, contaminant availability, and vulnerability of organisms (e.g., changes to lipid content), which can in turn influence contaminant uptake rates, accumulation, and elimination rates (e.g., Landrum and Fisher, 1999). Further, the environment may interfere with management actions to prolong contaminant availability, as is often the case with persistent organic pollutants, and as was exemplified by the high lead

concentrations that were seen in the 1993–2003 California mussel samples despite the 1992 California ban on lead in gasoline and paints (Melwani et al., 2013). The combination of pollutant bans (e.g., PCBs, PBDEs) and implementation of state and federal regulatory programs (e.g., Porter Cologne Water Quality Control Act, Clean Water Act) have, over time, resulted in the elimination of toxic discharges and the initiation of cleanup and remediation efforts, precipitating a consistent decline in mussel contamination, primarily from organic legacy pollutants, across multiple waterbodies (Melwani et al., 2013). Bans on tributyltin and the use of mercury in many products from the mid 1990s to early 2000s may have influenced the lower concentrations of these compounds found in the more recent Pacific oyster samples relative to the California mussel samples. Lower concentrations of PCBs, banned federally in 1979, found in Pacific oyster relative to the California mussel may similarly reflect larger-scale declines and cleanup actions. However, recent independent data from higher trophic levels and plankton reveal that PCB levels can still be high in San Diego Bay, reminding us of the need to consider both management actions and natural history when interpreting monitoring data.

Other discharge types such as stormwater from impervious surfaces have, however, increased since the early 1970s due to extensive upstream development, bringing with them contaminants of emerging concern (e.g., neonicotinoids, butylbenzyl phthalate, plastics) and upland sources of other pollutants of concern (e.g., PAHs, PBDE flame retardants, pyrethroid pesticides). Copper and zinc are common in stormwater flows, largely due to releases from motor vehicle tires and brake pads, and in bay waters because they replaced tributyltin in anti-fouling ship paint (although both are now the focus of reduction programs, e.g., CSQA, 2019, Jablon, 2021, PSD, 2021). Higher levels of copper and zinc found in the more recent Pacific oyster samples relative to the earlier mussel samples indicate that there may be more copper and zinc available now, or that species-specific uptake rates may influence contaminant levels, though concentrations in the oysters were similar to levels found in California spiny lobster, a potential predator, recently tested in San Diego Bay (Loflen et al., 2018).

Finally, contaminant concentration differences between California mussels and Pacific oysters may in part be an artifact of the oyster survey being conducted over a shorter period of time (<1 yr) than the 10-yr mussel data, resulting in a lower likelihood of seeing pulsed appearances or inputs of contaminants, especially for those tied to stormwater runoff (e.g., PAHs), and/or due to higher variability due to fewer Pacific oyster samples. Additional research is needed to explore potential species-specific differences for informed decisions regarding risk.

## 5. Considerations for future contaminant monitoring, research, thresholds and guidelines

The use of bivalve molluscs as a biosentinel species has been a part of water quality monitoring programs for over thirty years (Melwani et al., 2013), and the use of a consistent species across space and time, such as in the Mussel Watch Programs, has allowed for long-term comparisons within and among waterbodies across the USA. However, the use of transplanted mussels is not meant to be a substitute for site and species-specific evaluation using local species. Further, use of a single species for monitoring can often conflict with actual species availability and patterns of harvest and consumption on a waterbody-specific basis, as well as miss part of the contamination story if, as observed in this study, contamination loads differ across species. We recommend conducting studies using paired mussel and oyster sampling to create a more complete picture of contamination risks.

The variability in contaminant types and concentrations linked with seasonality, location, and the changing uses and legacy potential of pollutants illustrate a need for more regular and comprehensive contaminant monitoring efforts. In addition, species- or taxon-specific monitoring for pollutants that organisms of interest, such as Pacific oyster, cannot easily or quickly depurate (e.g., mercury and PCB) will

also be helpful in better understanding risks to consumers and crafting warnings. While monitoring that incorporates all these variables seems unfeasible, we in fact already consider each of these in different regulatory contexts, and the information can be important for regulatory management and informing guideline development efforts. For instance, guidelines for algal toxins are seasonal (summer paralytic shellfish poisoning) and targeted at select locations around San Diego Bay. Location can be important for chemical contaminants—and addressing sources, such as for those single samples that were at or exceeded limits for chlordane and PAHs. Chemical contaminant guidelines, although established on a bay-wide basis in California, are already focused on species, and we recommend adding Pacific oysters to the OEHHA guidelines for San Diego Bay because of high PCB concentrations.

Research is still needed to inform the risk determinations that are used to develop thresholds and consumption guidelines. Research needs include gaining a better understanding of the interactions between pollutants (e.g., Krishnan and Brodeur, 1994; Tang et al., 2009; Teuten et al., 2009), the additive effects of pollutants that on their own may be below thresholds but together pose increased risk, and the effects of many of the emerging contaminants found in this study, including plastics. Research to better understand the human dimensions underlying shellfish harvest is also needed. Human health risks are influenced by the timing and rates of consumption, preparation methods (e.g., raw or cooked, parts of organism used; Wong et al., 1981; Zabick et al., 1992; Wilson et al., 1998), a lack of awareness or understanding of risk (no or ineffective outreach; Tyson, 2012), and drivers of a disregard for risk (e.g., Torchetti, 1998; Burger, 2002; Harris et al., 2009; Pitchon and Norman, 2012; Steinberg and Moore, 2017; Pedersen and Talley, 2021). Although not observed directly by our research team, frequent visitors to Embarcadero pier reported occasionally witnessing individuals harvest and consume Pacific oysters during summer morning low tides, and a California Fish and Wildlife Warden reported observing people collecting for consumption in the back bay. These reports coupled with both year-round inquiries from the public about the safety of recreational oysters harvest from local bays (Talley and Loflen pers. obs.) and the common occurrence of oyster scars indicate that harvest likely occurs regularly and there may be particular ethnic, racial and/or socio-economic groups that are especially vulnerable to contaminant exposure via oyster consumption (e.g., Torchetti, 1998; Pitchon and Norman, 2012; Tyson, 2012). Guidelines and even laws like the annual bivalve quarantine are difficult to enforce, and the extent that they are followed by the public is uncertain. Socially informed and tailored outreach surrounding risks and solutions will also be crucial next steps.

## CRediT authorship contribution statement

**Theresa Sinicrope Talley:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Chad Loflen:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Rich Gossett:** Resources, Methodology. **David Pedersen:** Funding acquisition, Resources. **Nina Venuti:** Data curation, Investigation, Methodology, Supervision, Visualization, Writing – review & editing. **Julie Nguyen:** Resources. **Richard Gersberg:** Resources, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2021.113132>.

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