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Seabirds as regional biomonitors of legacy toxicants on an urbanized coastline

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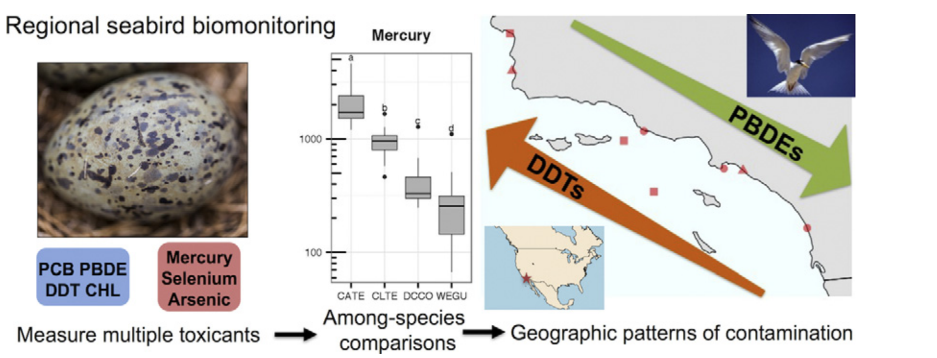
HIGHLIGHTS

- Coastal species are susceptible to mixtures of chemical pollution.
- We compared contaminant concentrations in seabird eggs across a regional water body.
- Legacy contaminants remain dispersed and persistent in seabirds in the SCB.
- Concentrations of contaminant classes and congeners displayed geographic patterns.
- Seabird contaminant monitoring informs remediation & management of coastal regions.

GRAPHICAL ABSTRACT



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ABSTRACT

Seabirds are often cited as sentinels of the marine environment, but are rarely used in traditional ocean and coastal contaminant monitoring. Four classes of persistent organic pollutants (POPs, n = 68) and three trace elements (mercury, selenium, and arsenic) were measured in the eggs of California least terns (*Sterna antillarum browni*), caspian terns (*Hydroprogne caspia*), double-crested cormorants (*Phalacrocorax auritus*), and western gulls (*Larus occidentalis*) that nest in the Southern California Bight. Building on a periodic five year regional monitoring program, we measured contaminant exposure and assessed the utility of seabirds as regional contaminant biomonitors. We found that the eggs of larger, more piscivorous species generally had the highest concentrations of POPs and trace elements while California least terns had the lowest concentrations, except for mercury which was higher in least terns. As expected, DDT concentrations were elevated near the Palos Verdes Superfund site. However, we also detected a previously unknown latitudinal pattern in PBDE concentrations in least terns. POP congener profiles also confirmed differences in contamination in urban least tern colonies closest to urban centers. Though toxicants were at detectable levels across species and sites, concentrations were below those known to cause adverse effects in avian taxa and are steady or declining compared to previous studies in this region. Our results suggest that regional seabird monitoring can inform site-specific remediation and support management and protection of regionally-threatened wildlife and coastal systems. Integration of seabird contaminant data with traditional sediment, water, bivalve and fish monitoring is needed to further our understanding of exposure pathways and food web contaminant transfer.

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Abbreviations

Birds

CATE	Caspian tern
CLTE	California least tern
DCCO	Double-crested cormorant
WEGU	Western gull

Toxicants

POPs	persistent organic pollutants
CHLs	chlordanes
DDTs	dichlorodiphenyltrychloroethanes
PBDEs	polybrominated diphenyl ethers
PCBs	polychlorinated biphenyls
SCB	Southern California Bight

1. Introduction

Human population density continues to increase in coastal areas worldwide, including coastal California (Crossett et al., 2004). Point source pollution, runoff, and atmospheric deposition associated with urban, suburban, agricultural, and industrial development has led to spikes in persistent organic pollutants (POPs) and trace elements in coastal environments (Elliott and Elliott, 2013; Schiff et al., 2001). While production of some toxicants is banned or closely regulated, persistent toxicants remain in coastal waters and sediments for decades and cycle through aquatic food webs. This is especially problematic for long-lived, top predators like seabirds, as many POPs and some trace elements are subject to bioaccumulation and biomagnification (Elliott, 2005; Rowe, 2008). At high concentrations, toxicants can reduce individual survival and reproduction, resulting in population decline (Bustnes et al., 2003; Hellou et al., 2013). Even at sub-lethal levels, these toxicants can impair physiological, immune, and reproductive function (Finkelstein et al., 2007; Tartu et al., 2013; Goutte et al., 2015) and in some species, combinations of toxicants even below effects thresholds have been linked to endocrine disruption (Silva et al., 2002; Bryan et al., 2005). Though effects vary by species, contaminant type, and concentration, the impacts have been observed in multiple taxa and are severe enough to warrant regular screening.

Despite their widespread distribution and ecological effects, multiple contaminant classes are rarely quantified among species or sites for regional analysis (but see Braune et al., 2002, Mallory and Braune, 2012). While single-site, single -species studies can provide data on species' vulnerability in one location, these analyses can overlook regional patterns of contaminant exposure, distant points of contamination, or fail to account for the mobility of marine taxa (Jarvis et al., 2007; Parnell et al., 2008). Given our nascent understanding of the synergistic or additive effects of multiple contaminant types (Finkelstein et al., 2007; Rowe, 2008; Goutte et al., 2015; Noyes and Lema, 2015), a multi-site and species approach can enhance our baseline knowledge of mixtures of toxicants present in impacted ecosystems. This information is particularly relevant along urbanized coastlines, where wildlife have higher exposure to a wide range of anthropogenic toxicants (Phillips et al., 1997; Schiff and Allen, 2000; Jarvis et al., 2007; Millow et al., 2015).

The Southern California Bight (SCB), which extends from Point Conception, CA to Cabo Colnett, Baja California, Mexico, is a seabird biodiversity hotspot that is home to many species of conservation concern, including the California least tern (*Sterna antillarum browni*; Gray, 1997). As high trophic level consumers, seabirds in the SCB are exposed to high concentrations of toxicants and declines in seabird populations in the SCB have been linked to exposure to several compounds, including DDT (dichlorodiphenyltrichloroethane) dispersal from the Palos Verdes Superfund site (Risebrough et al., 1967; Ohlendorf et al., 1985; Fry, 1994). Numerous other toxicants, including mercury (Hotham and Powell, 2000; Komoroske et al., 2012), selenium (Ohlendorf et al., 1985; Hotham and Powell, 2000), arsenic (Komoroske et al., 2011), PCBs (polychlorinated biphenyls, e.g. industrial and electrical byproducts, Fry, 1995, Schiff and Allen, 2000, Brown et al., 2006, Jarvis et al., 2007), PBDEs (polybrominated diphenyl ethers, e.g., flame retardants, Brown et al., 2006) and CHLs (chlordanes, Ohlendorf et al., 1985, Schiff and Allen, 2000), have also been detected in wildlife, sediments, and waters (Zeng et al., 2005; Dodder et al., 2012) in the SCB.

Although seabirds have been recognized as sentinels of marine systems (e.g., Burger and Gochfeld, 2002; Elliott and Elliott, 2013), most contaminant monitoring efforts have yet to include seabirds as part of the typically studied samples, a list that often includes water, sediment, bivalves, and fish (e.g., Zeng et al., 2005; Parnell et al., 2008; Dodder et al., 2012). Here, we assess the loads of the four classes of POPs and three trace elements in four seabird species nesting in the SCB to compare differences in toxicant concentrations within and among species, look for spatial trends in exposure levels within species, and consider the link between contaminant exposure and biological responses. Our research highlights the utility of seabird tissues and ecology in examining spatial, temporal, and biologically-relevant trends in regional contaminant biomonitoring.

2. Methods

2.1. Study species

Salvaged seabird eggs, i.e. eggs left at the end of a breeding season, have been demonstrated to serve as a robust tissue type for toxicant analyses (Hickey and Anderson, 1968; Braune et al., 2002; Burger, 2002; Mallory and Braune, 2012; Millow et al., 2015). Using salvaged eggs, we analyzed the egg contents of four seabird species: California least tern (*Sterna antillarum browni*), Caspian tern (*Hydroprogne caspia*), double-crested cormorant (*Phalacrocorax auritus*), and western gull (*Larus occidentalis*). The selected species differ in foraging strategies and ranges, which are known to influence toxicant load (Mallory and Braune, 2012; Lavoie et al., 2015). For instance, California least terns and Caspian terns are both plunge diving, piscivorous birds, but may consume different prey species (Ohlendorf et al., 1985; Lewison and Deutschman, 2014). Double-crested cormorants are also piscivorous and forage by diving at depth. Western gulls are generalists that forage on the ocean surface as well as on marine, coastal, and terrestrial subsidies. These differences in foraging strategies and prey items may result in varying contamination levels in the eggs of each species.

2.2. Egg collection, processing, and chemical analysis

Salvaged eggs were collected from the nests of the study species from 16 sites in the Southern California Bight (Fig. 1, Table A.1) during spring and summer 2013. Egg collection was executed by permitted individuals at each site in accordance with State, Federal and IACUC guidelines. All collected eggs were determined to be fail-to-hatch eggs due to nest abandonment or were taken as part of a depredation effort. Eggs were placed in cardboard cartons and transported to the US Fish and Wildlife Office in Carlsbad, CA for subsequent morphometric analysis, and other laboratories as described in the Supporting Information for chemical analysis. Eggs were processed using standard protocols for avian egg harvest for chemical analysis, embryo examination, and shell thickness determination. Because a single least tern egg does not contain enough material for all chemical analyses, we combined the contents of multiple least tern eggs into composite samples until sufficient matrix was present for subsequent analyses. Least tern composite samples comprised the egg contents of 2–4 eggs collected from the

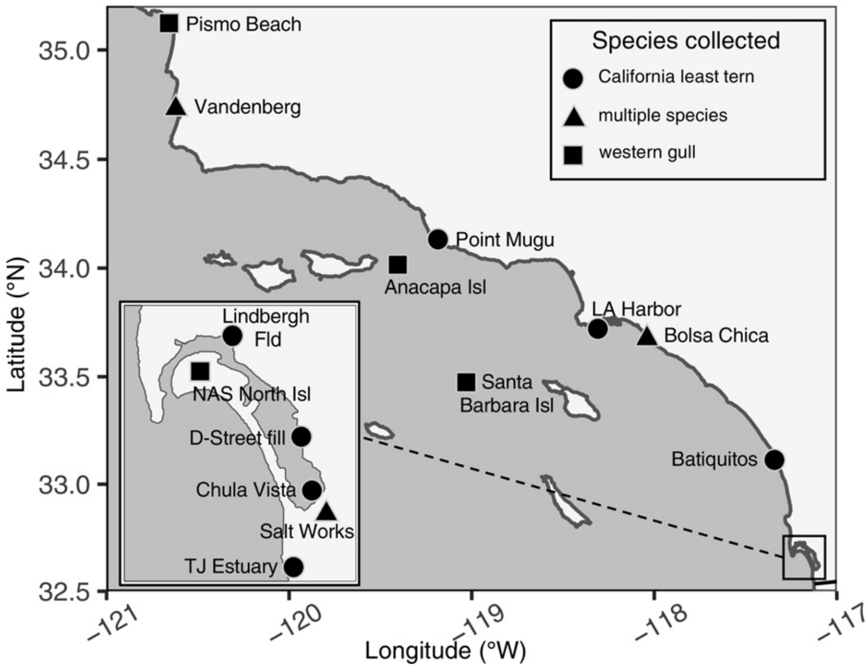

Fig. 1. Egg collection locations in the Southern California Bight.

same site. Egg morphometrics of each egg in the composite sample were averaged to obtain mean measurements per composite sample. The majority of samples (92/102) were either not fertilized or in the early stages of development. The analytical methods and quality assurance/quality control (QA/QC) protocols closely followed those of the Southern California Bight Program (Dodder et al., 2016). The analytes included 41 polychlorinated biphenyl (PCB) congeners, 15 polybrominated biphenyl ether (PBDE) congeners, 7 dichlorodiphenyltrichloroethane (DDT) related compounds, 5 chlordanes (CHLs), mercury, selenium, and arsenic. Additional information on egg processing, analysis, and quality assurance is available in Appendix B: Supplementary methods.

2.3. Statistical analysis

All statistical analysis was performed in R (R Core Team, 2015). Results from Physis Environmental Labs were reported on a wet weight basis; the percent lipid was also determined. All concentrations were standardized to unadjusted dry weight, ng/g (ppb), to account for desiccation based on differences in egg collection dates. Summed concentrations by contaminant class were $\log_{10}$-normalized to fit test and model assumptions of normality. For non-detect samples, we set values to zero for statistical analysis and summary statistics (Table A.2). For log-scaled graphing purposes only, we added 1 ng/g dry weight to all CHL values.

Based on results from Wilk-Shapiro and Levene's test which showed that data among species were nonparametric ($p < 0.05$), we used Kruskal-Wallis ANOVAs with post-hoc Dunn's test and Holm's correction to compare differences in toxicant concentrations among species across all sites, among species at a single site, and within a species across multiple sites. We used Welch's *t*-tests to assess differences in contaminant concentrations between two species at a single site.

We conducted spatial analyses for California least terns and western gulls as sample size and egg collection distribution were sufficient to allow for spatial comparison. To assess spatial relationships with toxicant concentrations within species, we used linear mixed models with latitude, distance to urban areas, and the type of collection site (e.g., designated marine protected area) as fixed effects and site as a random effect. We compared models using Akaike's Information Criterion (AIC) and described significant predictors using likelihood ratio tests and the importance function in the R package "AICcmodavg" (Mazerolle, 2016). The ratio test yields the relative importance of the predictor variables in each model set based on the sum of Akaike weights that include the variable of interest (w+). We set a w+ critical value of 0.75 for high relative importance for each predictor. We performed two additional analyses to further evaluate geographic toxicant patterns: Mantel tests and principal components analysis (PCA). Mantel tests examine the relationship between distances between sites and mean toxicant concentrations by site; only CLTE had sufficient data to perform these tests. PCAs were used to examine loadings of individual POP congeners and the resulting groupings of individual samples based on the similarity in individual congener profiles. For this analysis, congener concentrations were converted to a percent of the summed POP concentration per sample to normalize abundance that would otherwise obscure variation among congener profiles. For CLTE, we performed PCA on samples from urban sites only, as these are the areas of greatest potential for contaminant exposure (Dodder et al., 2012).

2.4. Biological response

To consider potential biological responses to toxicant exposure we compared toxicant concentrations to eggshell thickness measurements and published toxicant concentrations associated with adverse effects in other avifauna. Because both PBDEs and DDTs have been associated with decreased eggshell thickness in avifauna, we ran linear regressions to compare eggshell thickness and Ratcliffe's index to log-normalized PBDE and DDT concentrations (Ratcliffe, 1970; Harris and Elliott, 2011). Because eggshell thickness is species-specific, we did not compare eggshell thicknesses between species. Effect levels can be used to delineate the toxicant concentrations at which adverse effects may occur. To put our results in this context, we compare our detected toxicant levels to previously published contaminant effect levels associated with adverse effects in other avifauna. Although effect levels vary by species and toxicant, and there are limited data available on effect levels for particular species or toxicants, the selected thresholds are ones that have been established by published research on toxicant levels in avian eggs. Two sets of thresholds were used in this analysis: No Observed Adverse Effect Concentration (NOAEC) and Lowest Observed Adverse Effect Concentration (LOAEC). NOAEC indicates a concentration threshold where there is no concern of adverse effects and LOAEC indicates the lowest level at which adverse effects may occur. Levels between NOAEC and LOAEC suggest the toxicant merits additional consideration. We compared the range and mean for our focal species to estimates from other avian species (Table 1).

3. Results

3.1. POPs: levels detected among species

We detected all targeted toxicants by class in every egg sample except CHLs, although toxicant concentrations varied among species. Percent lipid was not related to organic contaminant concentrations. In general, Caspian terns (CATE) had the highest concentrations of all targeted toxicants while California least terns (CLTE) had the lowest, and PCBs and DDTs had the highest concentrations among species (Fig. 2). CATE and double-crested cormorants (DCCO) had similar ($p = 0.983$) and greater amounts of PCBs ($\chi^2(3) = 35.252$, $p \ll 0.001$) compared to western gulls (WEGU) and CLTE ($p = 0.983$). There was a similar pattern in DDTs ($\chi^2(3) = 51.813$, $p \ll 0.001$), where DCCO and CATE had the highest concentrations of DDTs ($p < 0.772$), but WEGU differed from CATE ($p \ll 0.001$), DCCO ($p < 0.001$), and CLTE ($p < 0.001$). DCCO had similar concentrations of PBDEs as CATE ($p < 0.084$), WEGU ($p < 0.879$), and CLTE ($p < 0.084$), but all other species were different from each other ($\chi^2(3) = 40.485$, $p \ll 0.001$). CHLs also differed among species ($\chi^2(2) = 37.329$, $p \ll 0.001$), with CHL concentrations higher in CATE than CLTE ($p < 0.006$) and WEGU ($p < 0.001$), and CHL concentrations higher in CLTE ($p < 0.001$) than WEGU. We did not include DCCO in CHL analyses because a high proportion (3/8) of samples were non-detects.

Two sites had sufficient sample size to examine differences in contaminant concentrations among species: Bolsa Chica and Salt Works. We sampled CATE and CLTE eggs at Bolsa Chica and CATE, CLTE, and DCCO eggs at Salt Works. At Bolsa Chica, PCB (Welch's *t*-test: $t_{6.66} = 10.474$, $p < 0.001$), PBDE ($t_{5.20} = 9.366$, $p < 0.001$), DDT ($t_{5.98} = 8.724$, $p < 0.001$), and CHL ($t_{6.11} = -5.278$, $p < 0.002$) concentrations were higher in CATE than CLTE (Fig. 3).

At Salt Works, DDT concentrations differed ($\chi^2(2) = 8.07$, $p < 0.018$) among species, where CATE ($p = 0.043$) and DCCO ($p = 0.043$) had higher concentrations than CLTE, but CATE and DCCO concentrations were similar ($p = 0.351$; Fig. 3). There were no observed differences in PCB ($\chi^2(2) = 5.66$, $p = 0.059$), PBDE ($\chi^2(2) = 4.17$, $p > 0.124$), or CHL ($t_{2.42} = 0.264$, p > 0.812) concentrations between species at Salt Works.

3.2. Trace elements: levels detected among species

We found some evidence of differences in trace element levels among species. Mercury concentrations significantly differed ($\chi^2(3) = 71.05$, $p \ll 0.001$) among species in a repeated pattern of concentrations ($p < 0.05$), with greatest to smallest found in CATE, CLTE, DCCO and WEGU in that order (Fig. 2). For other elements there were fewer obvious patterns, although DCCO samples were not analyzed for selenium or arsenic. Selenium concentrations were significantly ($\chi^2(2) = 26.412$, $p \ll 0.001$) greater in CLTE than WEGU, but CATE and WEGU ($p = 0.086$) and CATE and CLTE ($p = 0.884$) had similar selenium concentrations. CATE and WEGU had similar arsenic concentrations ($p = 0.075$), and both CATE ($p < 0.004$) and CLTE ($p \ll 0.001$) had higher arsenic concentrations than WEGU ($\chi^2(2) = 27.733$, $p \ll 0.001$). DCCO samples were not analyzed for selenium or arsenic.

There was also evidence for differences in element concentrations among species nesting at the same site that was similar to the overall patterns among species. At Bolsa Chica, CATE harbored significantly more mercury than CLTE ($t_{4.80} = 4.680$, $p < 0.006$; Fig. 3), but the two species had similar concentrations of selenium ($t_{4.54} = 0.656$ $p > 0.543$) and arsenic ($t_{6.62} = -0.928$, $p > 0.386$). At Salt Works, mercury concentrations differed ($\chi^2(2) = 27.733$, $p \ll 0.001$) and were higher in CATE than CLTE ($p < 0.029$) and DCCO ($p < 0.002$), whereas mercury concentrations were similar between CLTE and DCCO ($p > 0.125$, Fig. 3). Samples at Salt Works were not analyzed for selenium or arsenic.

3.3. Spatial patterns in toxicant concentration

To look for spatial patterns in toxicant concentrations, we evaluated toxicant levels across the region by class, investigated whether any of the available landscape predictors explained the detected variability, and looked for spatial differences in the concentrations of single POP compounds among the most urban sites. We had sufficient sample size and resolution to assess CLTE and WEGU toxicant levels across the region, and differences in concentrations of single POP compounds in CLTE eggs. AIC scores of regional comparisons and toxicants are in Table A.5. For CLTE, we found marine protected area status ($\chi^2(1) = 4.622$, $p < 0.032$) and latitude ($\chi^2(1) = 4.898$, $p < 0.005$) were significant and independent predictors of PBDE exposure; PBDE concentrations in CLTE samples decreased about 36% per degree of latitude and were 26% lower in sites located in MPAs (Fig. 4). Conversely, DDT concentrations in CLTE samples increased with latitude ($\chi^2(1) = 11.553$, $p < 0.001$) by about 45% per degree of latitude (Fig. 4).

Latitude was the strongest predictor for concentrations of PBDEs and DDTs (Table 2). DDT concentrations in CLTE were also significantly

Table 1
Screening values (NOAEC and LOAEC) for analyzed toxicants in ng/g fresh weight. No Observed Adverse Effect Concentration (NOAEC) are values below which no adverse effects are predicted. Lowest Observed Adverse Effect Concentration (LOAEC) are values at which eggshell thinning and/or reproductive success are impaired. NO and LO indicate the number of samples above NOAEC and LOAEC, respectively. Values between LOAEC and NOAEC may be of concern. NOAEC thresholds for DDTs are conservative estimates for all birds. No thresholds are available for CHL data.

Toxicant	NOAEC	LOAEC	Species (sample size)								Reference
			CATE (15)		CLTE (55)		DCCO (8)		WEGU (24)		
			NO	LO	NO	LO	NO	LO	NO	LO	
PCB	2600	23,000	0	0	0	0	0	0	0	0	Harris and Elliott, 2011
PBDE	200	1000	10	0	0	0	1	0	8	0	Rattner et al., 2011; Harris and Elliott, 2011
DDT[a]	200	10,000	15	0	21	0	8	0	19	0	DOI 1998
DDT[b]	1000	5000	12	2	1	0	2	0	3	0	DOI 1998
Mercury	500	800	4	2	0[c]	0[c]	0	0	0	0	Burger and Gochfeld, 1997; Henny et al., 2002
Selenium	900	3000	1[d]	0[d]	0[e]	0[e]	–	–	0[f]	0[f]	Ohlendorf and Heinz, 2011
Arsenic	910	>910	0[d]	0[d]	0[e]	0[e]	–	–	0[f]	0[f]	DOI 1998

[a] Thresholds for observed eggshell thinning in seabird species.
[b] Thresholds for reduced reproductive activity in seabird species.
[c] Sample size is 52.
[d] Sample size is 5.
[e] Sample size is 29.
[f] Sample size is 15.

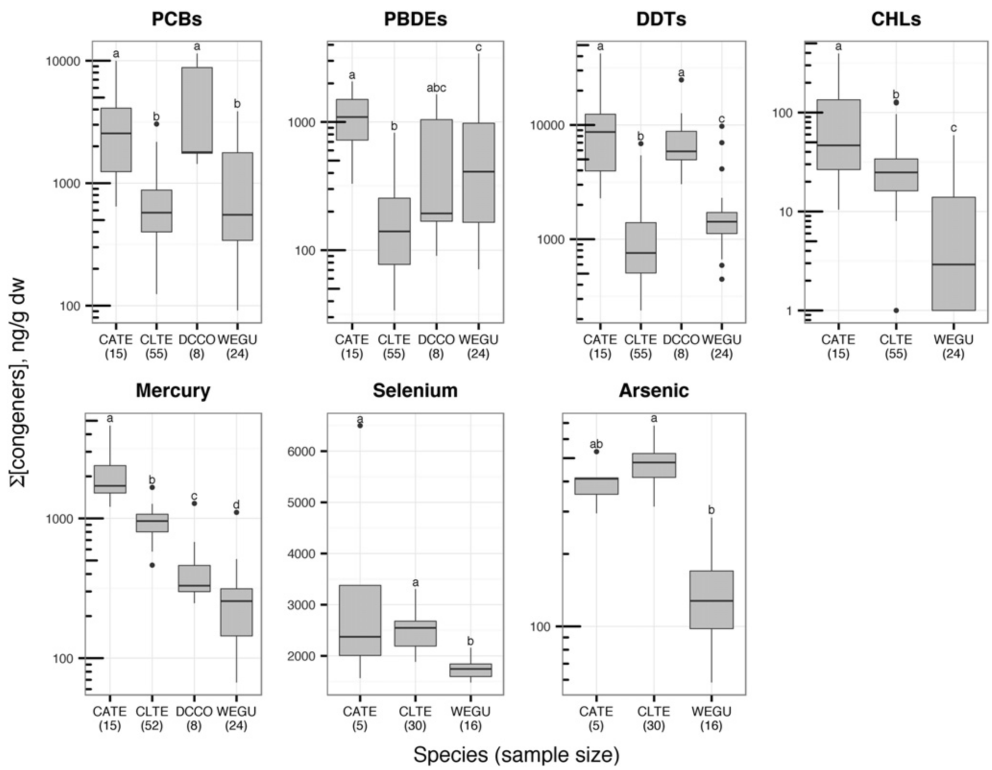

Fig. 2. Sum toxicant concentrations by species. The concentrations of congeners within organic contaminant classes are summed by sample. Each boxplot indicates the median (horizontal line), 25%–75% interquartile range (box), and 1.5 times the interquartile range (error bars). Letters represent similarities in sum toxicant concentration within each toxicant class among species.

related to distance between sites (Mantel test: $r = 0.764$, $p = 0.004$), with a similar, but less robust pattern for PBDE ($r = 0.329$, $p = 0.055$). No model adequately explained the spatial variation of PCB or CHL concentrations in CLTE. Additionally, no toxicant groups other than DDTs were significantly related by distance between sites (Mantel test, $p > 0.05$). Principle Components Analysis demonstrated spatial differences in congener profiles among CLTE in urban regions (Fig. 5). CLTE nesting in LA Harbor clustered negatively on PC1, which was dominated by DDT congener *p,p*-DDE followed by *p,p*-DDMU. Congener profiles of CLTE in San Diego Bay were dominated by PCB-138, −153, and −187, which loaded positively on PC1, whereas Tijuana River Estuary CLTE clustered positively on PC2 which indicated these samples have proportionally more PBDE-47 and, to a lesser degree, PBDE-99 and -100.

In WEGU, we found a significant relationship between PCB concentrations and marine protected area status where PCB concentrations were significantly lower ($\chi^2(1) = 5.106$, $p < 0.024$) by about 250% for WEGU nesting in the protected Channel Islands (Fig. A.1), although relative importance of MPAs was equivocal ($w+ = 0.66$, Table 2). Similarly, PCA showed the POP loads of WEGU from the Channel Islands contained proportionally less PCB-138 and -153 compared to WEGU nesting at NAS North Island in San Diego Bay (Fig. A.2). No predictors or their interactions significantly predicted PBDE, DDT, or CHL concentrations in WEGU, and no predictor was relatively more important than others (Table 2).

Though spatial patterns were evident for some POPs, likelihood ratio tests showed that no fixed effect significantly predicted mercury, selenium, or arsenic concentrations in regional comparisons of CLTE or WEGU samples. Similarly, no predictor had high relative importance (Table 2) and trace element concentrations were not related by distance (Mantel test, $p > 0.05$) in the CLTE model set. We did not conduct regional comparisons of selenium and arsenic in WEGU because samples from NAS North Island were not tested for these toxicants.

3.4. Potential biological responses

Regressions between CLTE eggshell thickness and PBDE and DDT concentrations explained very little of the observed variability in the eggshell data (PBDEs: $F_{1,52} = 2.02$, $R^2 = 0.037$, $p = 0.16$; DDTs: $F_{1,52} = 3.40$, $R^2 = 0.06$, $p = 0.07$, Fig. A.3). For WEGU, PBDE concentrations were not significantly related to eggshell thickness ($F_{1,21} < 0.003$, $R^2 < 0.0002$, $p = 0.961$, Fig. A.4). There was a significant but weak relationship between WEGU DDT concentrations and eggshell thickness ($F_{1,52} = 5.11$, $R^2 = 0.20$, $p = 0.034$, Fig. S2), which suggests DDT concentration may be one of many factors contributing to variation in WEGU eggshell thickness. The relationship between PBDE and DDT concentrations and Ratcliffe's index also explained little variability in the data for CLTE (PBDE: $F_{1,51} = 1.16$, $R^2 = 0.02$, $p = 0.29$; DDT: $F_{1,21} = 2.53$, $R^2 = 0.05$, $p = 0.12$, Fig. A.3) and WEGU (PBDE: $F_{1,21} = 0.10$, $R^2 = 0.004$, $p = 0.75$; DDT: $F_{1,21} = 0.45$, $R^2 = 0.02$, $p = 0.51$, Fig. A.4).

Across the region, no species exceeded the LOAEC-based thresholds for the legacy toxicants measured on a fresh weight basis (Table 1). However, DDT concentrations were above the NOAEC threshold for eggshell thinning for the majority of individuals in all species except CLTE (Table 1). Of all species, CATE had the highest proportion of individuals above NOAEC thresholds for multiple toxicants (Table 1). Effect thresholds were not available for CHLs.

4. Discussion

Regional contaminant monitoring in the SCB has been ongoing since 1994 and represents coordinated agency efforts to enhance the understanding of local and non-local pollutants in a regional marine environment (Cross and Weisberg, 1995). Environmental monitoring efforts of Southern California's coastal ocean typically focus on environmental

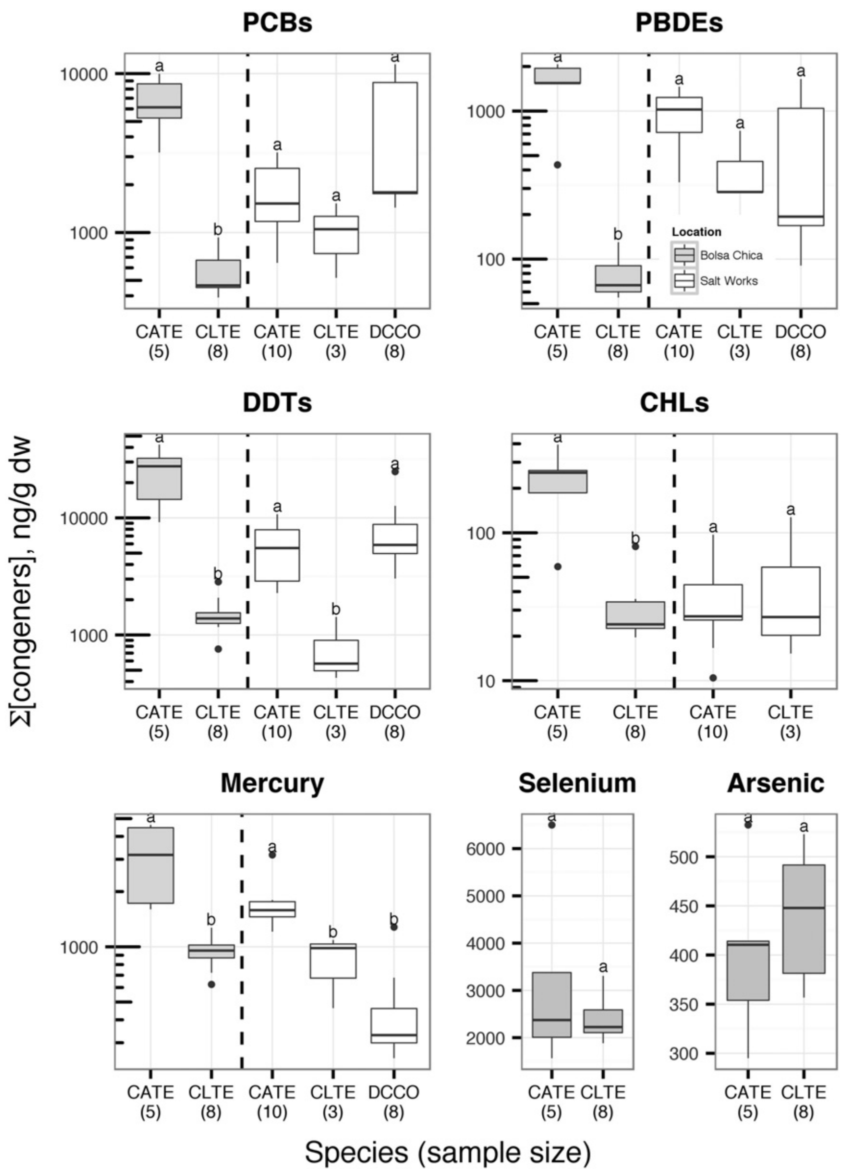

Fig. 3. Sum toxicant concentrations in Bolsa Chica (grey) and Salt Works (white) by species. Letters represent similarities in sum toxicant concentration within each toxicant class among species, but within site.

(water, sediment) or lower order taxa (bivalves) monitoring and are not designed to describe large-scale changes or to assess cumulative impacts from multiple compounds or monitor upper trophic level species. Results from this study, and other published research (Braune et al., 2002; Blasius and Goodmanlowe, 2008; Maruya and Schiff, 2009; Mallory and Braune, 2012), confirm the importance of coordinated regional monitoring efforts and demonstrate that levels of banned or highly regulated toxicants of interest are present but decreasing in the animals at the top of the SCB food webs. Our research also highlights exposure patterns of toxicants of interest among seabird species and across sites within the SCB and confirms that salvaged seabird eggs can be used to monitor larger regions (>100 km) of the coastal and marine environment, in support of restoration and protection of vulnerable species in this region (Braune et al., 2002; Elliott and Elliott, 2013).

4.1. Seabird toxicant exposure: differences among species

Every sample across each of 13 sites (Fig. 1) contained congeners from each class of pollutants assessed with the exception of CHLs. Among species, we found clear differences, i.e. up to an order of magnitude difference, in toxicant concentrations (Fig. 2). In general, we found larger, piscivorous species (CATE and DCCO) had higher organic contaminant levels than the generalist (WEGU) and smaller (CLTE) species (Figs. 2–3), a finding common with previous published research (Burger and Gochfeld, 1997; Braune et al., 2005). While all species in this study are piscivorous, there are likely differences in the trophic position and size of prey among the species we sampled. DCCO and CATE diets likely comprise larger and older fish due to a larger gape size and may consume higher proportions of higher trophic level fish in their diet versus other marine species like krill. The differences in contaminant levels we detected may also be driven by the extent or range of movement during breeding and non-breeding periods.

In contrast to the patterns in POP exposure among species, we found that CLTE had higher mercury concentrations (Fig. 2) than expected given their size and trophic level (Burger, 2002). Like POPs, mercury is both a point and non-point source pollutant, with mercury levels in top predators varying based on local anthropogenic activity at smaller temporal and spatial scales and the amount of sulfate and sulfate-reducing bacteria at the base of the food web that methylates elemental mercury (Elliott and Elliott, 2016). Increased mercury concentrations in

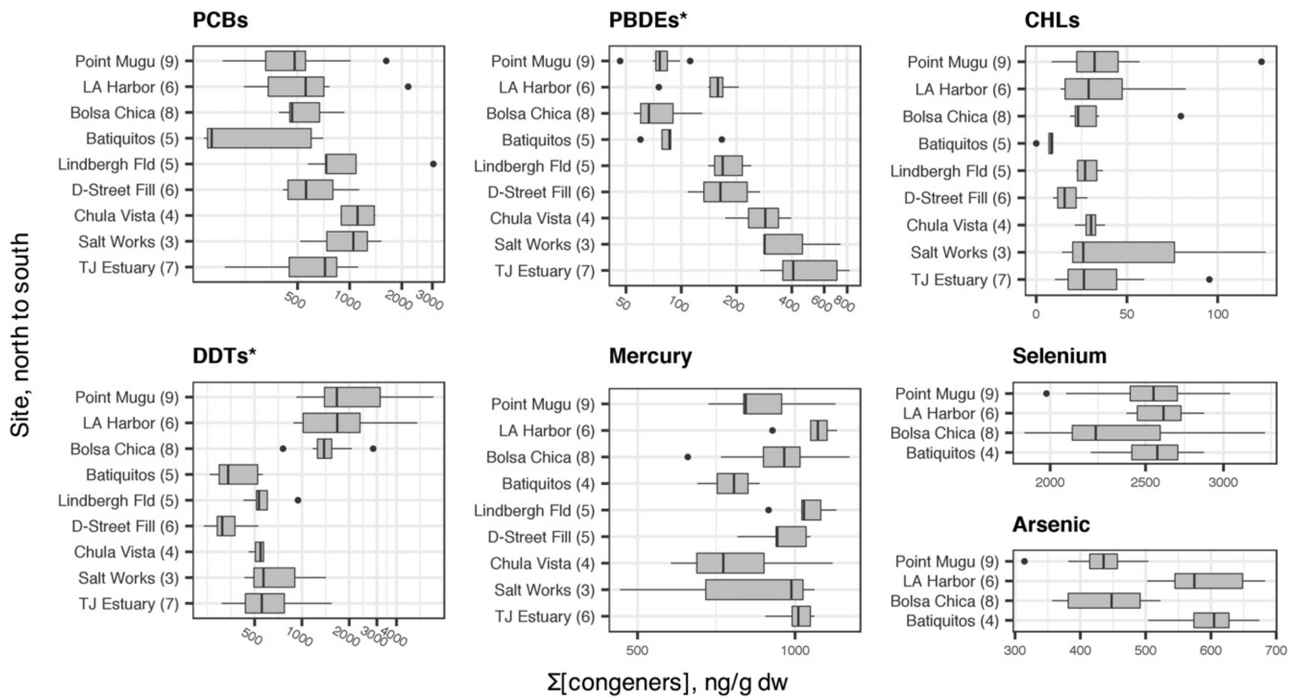

Fig. 4. Latitudinal comparisons of sum toxicant concentrations in California least terns. Parentheses indicate sample size by site. Asterisks represent plots where a significant latitudinal trend is present.

CLTE versus the larger species in this study, CATE and DCCO, may be due to differences in diet or foraging location. Other studies have also found higher mercury concentration in smaller seabirds (auklets and murrelets) versus piscivorous species that feed at a higher trophic level (e.g., herons, Elliott and Elliott, 2016). The relatively high mercury concentrations in CLTE could also reflect conditions at their overwintering area, as has been shown in some migratory populations of CATE and DCCO in the central US and Canada (Lavoie et al., 2015). Because mercury is not lipophilic like POPs (Ackerman et al., 2013), seabirds may have limited capacity to excrete body-bound mercury via burning adipose tissue, a decretion pathway that has been suggested for POPs.

4.2. Detecting toxicant trends in space and time

Based on the data from the two species for which samples were available across the study area, CLTE and WEGU, we also found evidence of significant distribution patterns of organic contaminant exposure. For CLTE, DDTs were highest near Los Angeles (Pt. Mugu south to Bolsa Chica) and PBDEs were highest in sites in and near San Diego Bay (Lindbergh Field south to Tijuana River Estuary, Fig. 4). The observed pattern for DDTs is likely explained by the location of the Palos Verdes Shelf Superfund Site, which lies ~23 km west of Bolsa Chica in the northern area of the SCB (Fry, 1994; Schiff and Allen, 2000; Zeng et al., 2005). While many seabird populations have recovered as contaminant exposure has declined, DDT levels remain detectable in coastal wildlife in the SCB (Macintosh et al., 2016). Although we found that across all colonies, DDT exposure was most similar at colonies in close proximity, the highest DDT concentrations were found in CLTE nesting north of Batiquitos (Fig. 4).

The spatial pattern in PBDEs also appears to be largely a geographic pattern rather than site-specific differences as the highest levels of PBDEs were detected in the CLTE colonies in and near San Diego Bay, a regional finding that has not been documented previously in seabirds. However, sediments in San Diego Bay and Los Angeles Harbor contain the highest concentrations of PBDEs in the SCB, likely from stormwater runoff (Dodder et al., 2012). Additionally, PCA revealed that the composition of POP congeners was significantly different among CLTE nesting sites in Los Angeles Harbor, San Diego Bay and Tijuana River Estuary (Fig. 5). These patterns suggest regional differences in contamination among contaminant class and individual congener profiles. The observed toxicant patterns also suggest CLTE may be a strong candidate for future regional monitoring in this area. The lack of spatial patterns for other toxicants (e.g., CHLs, trace elements) in this study suggests that exposure to these toxicants does not vary in the seabird species we sampled substantially across the region.

In WEGU, PCB concentrations increased from north to south and WEGU from the southern site, NAS North Island, had congener profiles containing greater proportions of PCBs than other WEGU (Fig. A.1). This finding reflects known patterns of PCB contamination in the SCB, where sediments in embayments harbored greater PCB concentrations than offshore areas (Maruya and Schiff, 2009). However, interpretation of spatial differences in WEGU contamination should be approached with caution because gulls feed omnivorously and opportunistically on marine and terrestrial resources.

Because there has been contaminant monitoring at specific sites and species within the SCB, we can also consider trends in toxicant levels

Table 2
Relative importance (w+) and rank of each variable for CLTE and WEGU model sets (Table A.5). Bold indicates w+ > 0.75.

	Latitude		MPA		UrbanDist	
	w+	Rank	w+	Rank	w+	Rank
CLTE						
PCB	0.31	2	0.70	1	0.09	3
PBDE	**0.88**	1	0.43	2	0.04	3
DDT	**0.98**	1	0.20	2	0.03	3
CHL	0.24	2	**0.81**	1	0.09	3
Mercury	0.30	2	0.60	2	0.12	3
Selenium	0.46	2	0.49	1	0.08	3
Arsenic	0.65	1	0.29	2	0.18	3
WEGU						
PCB	0.51	2	0.66	1	0.01	3
PBDE	0.52	2	**0.75**	1	0.01	3
DDT	0.54	2	0.58	1	0.01	3
CHL	0.57	2	0.59	1	0.01	3
Mercury	0.49	2	0.59	1	0.01	3

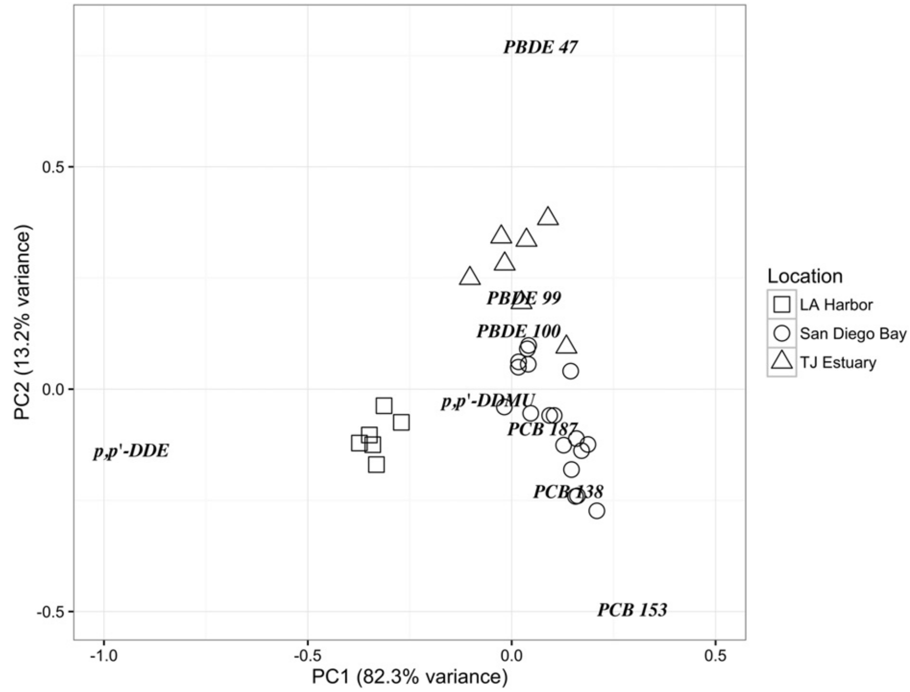

Fig. 5. PCA biplot of organic contaminant congeners among California least terns nesting at urban sites. Single congener concentrations within individual samples were converted to a percentage of the total organic contaminant concentration prior to analysis. Congeners are plotted according to their loadings.

detected over time. Our findings confirm that there is a continued decline in many POPs in the SCB (Maruya et al., 2015), yet many legacy toxicants persist in the SCB. On average, POPs were detected in lower concentrations in this study than those found in the recent past in seabird eggs in the SCB, including DDTs in WEGU nesting at NAS North Island (Jimenez-Castro et al., 1995), PCBs, PBDEs, and DDTs in nesting CATE at Salt Works (Zeeman et al., 2008), PBDEs in nesting CLTE at Salt Works (Zeeman et al., 2008), and PCBs and DDTs in nesting CLTE at the Tijuana River Estuary (Hotham and Powell, 2000). However, mean DDT concentrations in CLTE (764 ng/g ww) nesting at Salt Works were higher by about 400 ng/g ww on average, and above the maximum value of DDT concentrations in 2008 (Zeeman et al., 2008).

For trace elements, there are fewer data points to identify temporal trends as selenium and arsenic exposure were not available for our focal species. Identification of a temporal trend is also complicated because of temporal variability among studies, particularly in mercury concentrations. For example, mean mercury concentrations in our study are lower than those reported at the D-Street Fill CLTE colony in the 1980's (Hotham and Zador, 1995) but higher than mean concentration values reported from CLTE nesting at Tijuana River Estuary from 1994 to 1996 by ~300 ng/g dw (Hotham and Powell, 2000). The mechanism causing this variation merits further investigation (see Section 4.1, Elliott and Elliott, 2016).

Overall, these spatial and temporal trends suggest that concentrations of many legacy toxicants are steady or decreasing in seabirds in the SCB. Though the spatial trends in contamination we identified confirm findings from previous research on contamination in the SCB, we also identified important differences in contaminant profiles among seabird colonies that can inform local and regional management of SCB waters.

4.3. Biological response to toxicants

Regulated environmental monitoring is typically required to examine the potential biological effects of toxicant exposure, based on known thresholds which can help contextualize how toxins detected compare with known levels at which adverse effects take place. Eggshell thinning, which can lead to non-viable eggs and reproductive failure, is another commonly used metric in combination with identified thresholds to contextualize potential biological responses of toxicant exposure. All four monitored species exceeded the DDT NOAEC threshold for eggshell thinning. While there is historical precedence of eggshell thinning in the SCB associated with exposure to *p,p*-DDE, we did not find a relationship between DDT or PBDE contamination and eggshell thickness or Ratcliffe's index in CLTE or WEGU (Hickey and Anderson, 1968, Fig. A.3 and A.4). Although shell thickness in these species is approaching pre-1945 levels, neither CLTE or WEGU shell thickness has returned to values observed before DDT was in widespread use (Kiff, 1994; Jimenez-Castro et al., 1995; Zeeman et al., 2008).

When considering the adverse effects thresholds (NOAEC, LOAEC) independently, the evidence was equivocal. No species, on average, exceeded the adverse effects threshold, though a few individuals harbored contaminants at or above the LOAEC (Table 1). We found that some species exceeded the NOAEC for a compound class, but information on the effects of toxicants at these low concentrations and among species with varying sensitivities to toxicants is limited. Even larger data gaps exist regarding the additive or synergistic effects of contaminants and their interaction with other stressors, such as low food availability or changes in ocean climatic regimes (e.g., Noyes and Lema, 2015).

4.4. Seabirds as regional biomonitors

Monitoring contaminants at the regional scale is essential to aid in early detection of contaminant trends and adverse effects, and also to inform marine environmental policy with important implications for species and ocean health. Seabirds are considered effective monitors of marine ecosystem health (Mallory et al., 2006; Elliott and Elliott, 2013), but few large-scale toxicant monitoring efforts include seabirds as biomonitors. Here, we demonstrated that seabird biomonitoring can detect not only expected spatial and temporal patterns of contamination, but also reveal undescribed patterns in contaminant exposure both among species and across a nesting region (see Section 4.2).

There are advantages to using seabird tissues to examine regional contamination patterns. Abandoned and fail-to-hatch eggs are easily sampled at low cost on seabird colonies, compared to effort needed for sampling marine sediments, macrofauna, and fish. Seabird eggs are often large enough to test for multiple contaminant classes, or can be combined within site to give site-specific parameters. Seabird tissues are also easily archived and are used to describe temporal differences in toxicant values among species, sites, and regions (Braune et al., 2002; Mallory and Braune, 2012; Bond et al., 2015). However, tissues from migratory seabirds may have toxicants incorporated from both breeding and overwintering foraging areas, which hinders tracing the source of toxicants (Braune et al., 2002; Bond and Diamond, 2010). Additional samples from tissues formed at different times within the life cycle, such as feathers or otoliths formed overwinter outside of the breeding season, can further clarify geographic sources of contamination (Ramos and González-Solís, 2012; Lavoie et al., 2015).

Another important comparison to contextualize detected contaminant levels in seabirds at the regional scale is to analyze concentrations among sample types, e.g. sediment, bivalves, invertebrates, fish, and water to provide greater understanding of the pathway by which seabirds are exposed to toxicants in a food web. Identification of the exposure pathways may be supported using seabird diet, stable isotope, and telemetry data (Braune et al., 2002; Ramos and González-Solís, 2012). Additional efforts are needed to compare the contaminant levels in the SCB across these sample types. Nevertheless, the detected values in our study can be used to address region-wide questions of pollutant sources and potential impacts, and have conservation relevance, as one of our study species, California least tern, is a federally and state-listed Endangered species. The findings from this study serve as a baseline for regional contaminant assessment, and can be used to direct future studies of contamination sources to support research on biomagnification, and food web ecology in coastal and marine regions, as well as inform management efforts for vulnerable species in the SCB.

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Appendix A & B

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