Contaminant Bioaccumulation in Seabird Eggs

BIGHT '13

Southern California Bight
2013 Regional Monitoring
Program
Volume V

SCCWRP Technical Report 944

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Contaminant Bioaccumulation in Seabird Eggs of the Southern California Bight

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September 2016
Technical Report 944

ACKNOWLEDGEMENTS

We thank the USFWS Analytical Control Facility for providing the egg reference material. We thank Physis Environmental Laboratories, the Sanitation Districts of Los Angeles County, and the City of San Diego for providing analytical services. Thanks to Dovi Kacev, Chris Tyson, and Matthew Savoca for input on statistical analysis. Thanks to members of the Bight committee for reviews. And thanks to Amec Foster Wheeler, Andrew Yamagiwa, and Hal Beral for the cover page photos.

DISCLAIMER

The finding and conclusions in this report are those of the author(s). As such, they do not necessarily represent the views of the U.S. Fish and Wildlife Service.

EXECUTIVE SUMMARY

Local and global processes concentrate anthropogenic toxicants in aquatic systems. There, toxicants enter biotic food webs and can potentially adversely affect organismal physiology and immune function at lethal and sub-lethal concentrations. Natural resource and water quality stakeholders are often tasked with monitoring the extent and magnitude of contamination in aquatic food webs. However, most monitoring efforts are limited by spatial scale, numbers of species, and number of toxicants assessed, thus are unable to consider bioaccumulation at regional scales. Here, we show the utility of seabird eggs in a regional bioaccumulation monitoring program across an urbanized region of coastline, the Southern California Bight. We assessed the egg contents from four seabird species for four organic contaminant classes (polychlorinated biphenyls, polybrominated diphenyl ethers, DDTs, and chlordanes) and three elements (mercury, selenium, and arsenic). Results indicate toxicants are detectable across species throughout the region, and levels are steady or declining based on comparison to results from historic site-specific monitoring. While some individual eggs were found to have toxicants at levels above those known to cause adverse effects, on average no species met or exceeded lowest-observed adverse effect concentrations (LOAECs) and eggshell thicknesses were not related to PBDE or DDT concentrations. Our results suggest that continued monitoring of legacy and more recent contaminants in seabird eggs is useful in the Southern California Bight to inform site remediation, management, and protection of threatened wildlife in coastal systems.

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INTRODUCTION

Human population density continues to increase in coastal areas of California (Crossett et al. 2004). This growth has led to increased urban, suburban, agricultural, and industrial development that introduces organic contaminants and heavy metals to coastal environments via point sources, runoff, and atmospheric deposition (Elliott and Elliott 2013, Schiff et al. 2001). While production of many toxicants is banned or closely regulated, many persistent toxicants remain in coastal waters and sediments for decades and are biomagnified in aquatic food webs. At high concentrations, contaminants can reduce individual survival and reproduction, resulting in population decline, particularly for top predators (Bustnes et al. 2003, Hellou et al. 2013). Even at sublethal levels, these toxicants can impair physiological, immune, and reproductive function (Finkelstein et al. 2007, Tartu et al. 2013, Goutte et al. 2015). Many stakeholders, including ocean coastal communities, fisheries, ports, and wildlife managers, are concerned with water quality and tasked with biomonitoring in coastal food webs. These monitoring efforts are typically local in spatial extent, with a single organization or agency monitoring a single site. While site-specific monitoring is mandated and provides useful information on toxin exposure, regional monitoring is also essential to provide comparable data among sites within a geographic area and to previous studies. Regional monitoring maximizes the ability to use biomonitoring efforts to meet mandated monitoring objectives, prioritize site remediation, and trace the dispersal and uptake of toxicants in marine food webs.

The Southern California Bight (SCB), which extends from Point Conception, CA to Cabo Colnett, Baja California, Mexico is oceanographically complex and has high biodiversity (Gray 1997). The SCB abuts a densely populated coastline that houses an estimated 22 million people. Numerous natural and anthropogenic 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 1994, Schiff and Allen 2000, Brown et al. 2006, Jarvis et al. 2007), PBDEs (polybrominated diphenyl ethers, e.g., flame retardants, Brown et al. 2006) CHLs (chlordanes, Ohlendorf et al. 1985, Schiff and Allen 2000), and DDTs (e.g. pesticides, Risebrough et al. 1967, Ohlendorf et al. 1985, Fry 1994, Schiff and Allen 2000) have been identified in wildlife in the SCB. Many of these toxicants have also been detected in SCB sediments and coastal waters (Zeng et al. 2005, Dodder et al. 2012). For some avian species, these toxicants have been directly linked to population declines in this region (Hickey and Anderson 1968, Blus et al. 1972, Gress et al. 1973).

Seabirds have been identified as effective and efficient biomonitors of coastal ecosystem health (Elliott and Elliott 2013). As top predators, they generally feed at high trophic levels, which are reflected in the biomagnification of toxicants in their body tissues (Burger and Gochfeld 2002). During egg formation, birds can maternally transfer toxicants into egg contents; thus, salvaged egg (eggs left on a colony at the end of a nesting season) collection can be a low cost and non-invasive method to assess toxicant concentrations in coastal environments. Many seabird contaminant studies, as with general biomonitoring, are limited by the cost of chemical analyses to measure multiple toxicant classes, the number of species studied, or the spatial region where samples were obtained. In this study, we assessed contamination levels of six toxicant groups in four different species of seabirds that nest across the Southern California Bight. Building on decades of contaminant research in the SCB, our study objectives were to assess the extent and magnitude of contamination in the SCB, characterize the risks to seabirds from contaminants in this region, and highlight the utility of non-invasive seabird tissues in regional contaminant biomonitoring and assessments.

METHODS

Study species

We examined 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 traits, which is known to influence contaminant load. For instance, California least terns and Caspian terns are both plunge diving, piscivorous birds, but California least terns consume a variety of marine taxa and have been observed foraging within 2 km of shore during the breeding season, when this species is found in California (Fournier et al. 2016 *in review*). Double-crested cormorants are piscivorous and forage by diving. Western gulls are generalists that forage on the ocean surface as well as on marine, coastal, and terrestrial subsidies. Though the foraging ranges and diet of these species is not well-defined in this region, all chosen species are constrained by body and gape size, where smaller species (California least tern) cannot consume larger prey items, unlike larger species. Thus, each species provides a unique signal in contamination differences in the prey available from each foraging strategy.

Egg collection and processing

Eggs were collected from the nests of these 4 species from 16 sites in the Southern California Bight (Fig 1, Table 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 processing.

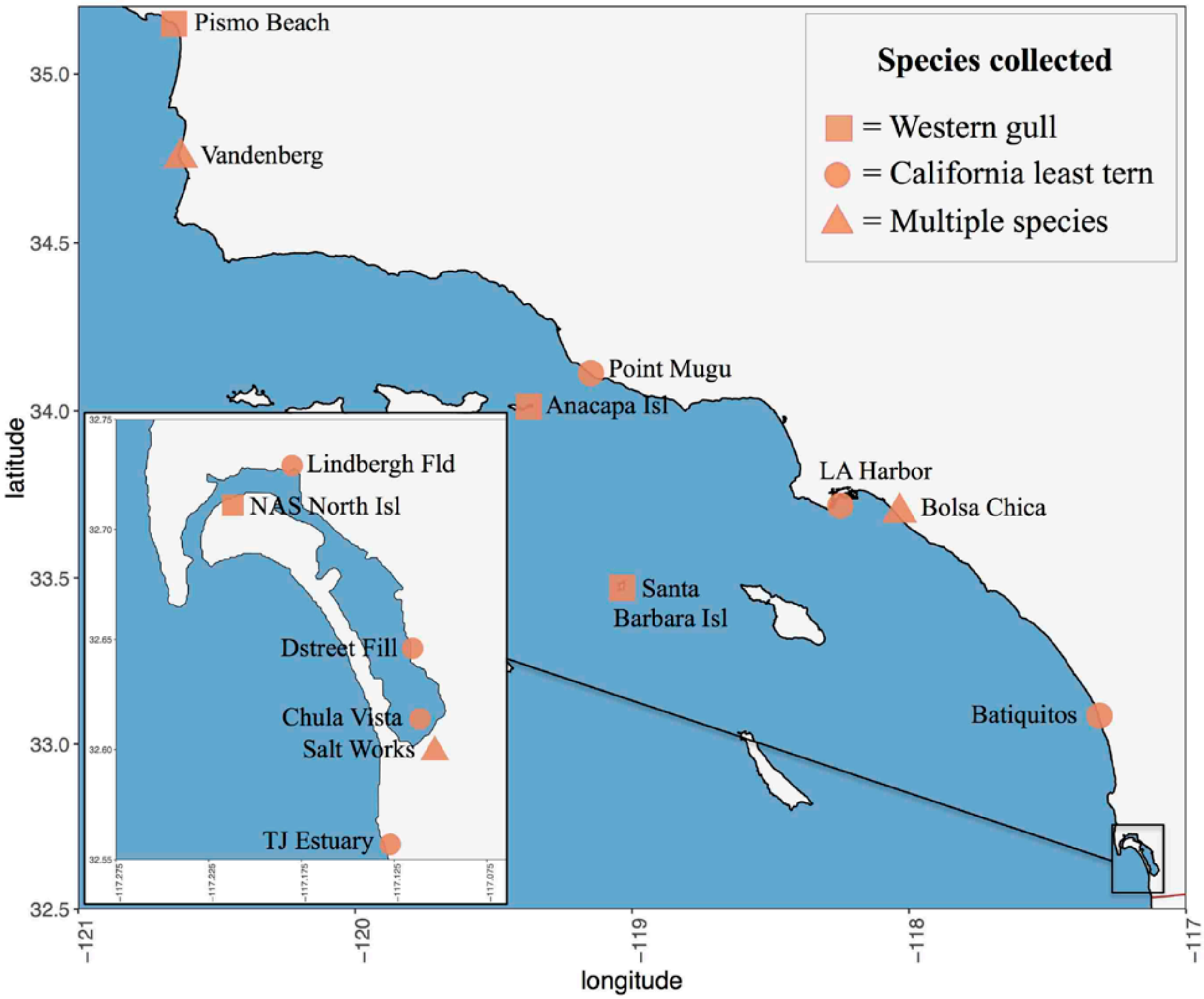

Figure 1. Egg collection locations.

Table 1. Number of egg samples collected from each species by site. Asterisks indicate sites within a Marine Protected Area (MPA).

	California least tern	Caspian tern	Double-crested cormorant	Western gull
Anacapa Island*				5
Batiquitos Lagoon*	5			
Bolsa Chica Reserve*	8	5		
Chula Vista Reserve	4			
D-Street Fill	6			
LA Harbor	6			
Lindbergh Field	5			
NAS North Island				8
Pismo Beach				1
Point Mugu	9			
Salt Works	3	10	8	
Santa Barbara Island*				9
Tijuana Estuary	7			
Vandenberg	2			1
SUM:	**55**	**15**	**8**	**24**

Eggs were processed using standard protocols for avian egg harvest for chemical analysis, embryo examination, and shell thickness determination. Eggs were cleaned with distilled water, weighed, and measured for maximum length and width to the nearest 0.1mm using an analog dial caliper. We measured volume as the weight of water displaced by the egg. For cracked eggs, we estimated volume using the generic approach by Hoyt (1979). Afterwards, we sliced eggs through the equator using a scalpel pre-rinsed with dilute nitric acid, distilled water, reagent grade acetone, and reagent grade hexane. We examined egg contents for approximate embryo age and malposition, placed contents into a kilned glass jar, and stored in a -20°C freezer until subsequent chemical analysis.

We let eggshells dry at room temperature for 30 days before measuring eggshell thickness. The thickness of each eggshell (shell + shell membrane) was measured at four points around the girth using a Starrett Model 1010M dial micrometer, which is accurate to 0.01 mm, and capable of estimatoin to 0.001 mm. Although a dial micrometer is commonly used, this instrument is affected by eggshell curvature and may overestimate measurements for small eggs (i.e., California least tern). We averaged the 4 eggshell thickness measurements for each sample to derive one thickness measurement per sample. To account for errors in measuring thinner eggshells, we also calculated Ratcliffe's index, $RI = \frac{S}{L \cdot W}$, where L is the maximum shell length (mm), W is the maximum shell width (mm), and S is the weight of the dry shell (g) (Ratcliffe 1970). 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 same site, and we averaged resulting least tern egg morphometrics by sample.

Chemical analysis and quality assurance

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. The individual analytes and reporting levels are provided in Table S1. Organic contaminants and selenium were measured by Physis Environmental Laboratories (PEL; in Anaheim, CA). Mercury was measured by the Sanitation Districts of Los Angeles County (LACSD; in Whittier, CA), and the City of San Diego, CA (CSD). Selenium and arsenic were measured by LACSD only.

An elemental inter-laboratory comparison was performed prior to the analysis of field samples. A single lab performed organic contaminant analyses, so no inter-laboratory comparisons took place. Two reference materials were used: National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 1946: Lake Superior Fish Tissue, and a chicken egg homogenate containing spiked concentrations of the target elements. For both materials, all laboratories were within ± 30% of the mean value for each element.

Each lab used established EPA methods or machinery to perform toxicant and egg content analysis (Table S2). Laboratories ran a set of QC materials with the field samples, including method blanks, spiked blanks (elements only), reference materials, matrix spikes, and laboratory sample duplicates. Each QC material had associated criteria for analytical frequency and accuracy (Table S3). The success of meeting these criteria was evaluated for each contaminant class (Table S4). In all cases, the frequency success was 100%. The accuracy success was generally between 84% and 100%, except as noted (Table S4).

Statistical analysis

All statistical analysis was performed in R (R Core Team, 2015). Results from PEL were reported on a wet-weight basis, in addition to percent lipid in each sample. All concentrations were standardized to unadjusted dry weight, ng/g (ppb), to account for desiccation based on differences in egg collection dates. Contaminant levels were log10-normalized to fit test and model assumptions of normality. All boxplots indicate the median (horizontal line), 1^{st} and 3^{rd} interquartile range (box), and 1.5 times the interquartile range (error bars).

After Wilk-Shapiro and Levene's test showed that data between species were nonparametric ($p<0.05$), we used Kruskal-Wallis ANOVAs with post-hoc 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 t-tests to assess differences in contaminant concentrations between 2 species at a single site. For samples which were non-detects for any contaminant class, we set values to 0 for statistical analysis and summary statistics and to ½ MDL for geometric means. 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.

We also considered how organic contaminant levels changed relative to eggshell thickness as both PBDEs and DDTs have been associated with decreased eggshell thickness in avifauna (Harris and Elliott 2011). Because eggshell thickness is species-specific, we did not compare eggshell thicknesses between species. Instead, we ran linear regressions to compare eggshell thickness and Ratcliffe's index to log-normalized toxicant concentrations.

Effect levels

Effect levels can be used to delineate the toxicant concentrations at which adverse effects may occur. To put our results in this context, we compared our detected contaminant levels to previously published contaminant effect levels associated with adverse effects in other avifauna (Table 4). Although effect levels vary by species and contaminant, and there are limited data available on effect levels for particular species or contaminants, the selected thresholds are ones that have been used in other studies on contaminant levels in avian eggs. Two sets of thresholds were used in this analysis: No Observed Adverse Effect Concentration (NOAEC) and LowestObserved 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. Additional information on selection of effect levels is available in the supplement. We compared the range and mean for our focal species to estimates from other avian species.

Due to the variety of reported contaminant concentrations in the literature, we used the R package "OrgMassSpecR" to convert contaminant concentrations to a standardized reporting metric, ng/g fresh weight. We reported both means and geometric means to ease comparisons among studies. Fresh weight concentrations are what the wet weight concentrations would be if the egg sample were fresh and before any moisture loss that occurs, especially in abandoned eggs. The adjustment eliminates an analytical artifact that significantly affects unadjusted wet weight-based concentrations. The extent of moisture loss from individual eggs was variable, such that unadjusted wet-weight-based concentrations in some eggs would be over-reported by as much as nearly four-fold. To obtain fresh weight-based values, wet-weight-based contaminant levels reported by the laboratory were adjusted according to methods by Stickel et al. (1973), using an adjustment factor equal to the ratio of the egg volume to the egg weight for each egg that was sampled. Mean adjustment factors were calculated for those samples that were composites of multiple eggs (i.e., least terns).

Current methods for PCB screening measures PCB congeners, whereas historic data and screening levels used Aroclor mixtures to examine sum PCB concentrations. More recent studies used the Aroclor approach in conjunction with the sums of homologs and/or 90 or more congeners. All three measures of total PCB concentrations were in close agreement in studies of seabird eggs for which total PCB concentrations were measured in all three ways (Zeeman et al. 2008). We used a method from Zeeman et al. (2008), who calculated a least squares linear regression formula to relate the total PCB contaminants of past studies (>90 PCB congeners) to the 41 congeners measured in this study:

Total PCBs (ng/g fw) = 75.2 ng/g fw + 1.17 (∑ lab-reported PCB concentrations ng/g fw)

RESULTS

Organic contaminants: levels detected

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

For CLTE and WEGU, we had sufficient sample size and spatial distribution of sampling to consider contaminant levels across the region by compound class to investigate whether any of the available predictor values explained the detected variability. AIC scores of regional comparisons and toxicant are in Table 2A and 2B. For CLTE, we found that marine protected area status ($\chi^2(1) = 4.622$, $p < 0.032$) and latitude ($\chi^2(1) = 4.898$, $p < 0.005$) were significant predictors of PBDE exposure, though there was no significant interaction between the two predictors ($\chi^2(1) = 0.532$, $p = 0.466$). PBDE concentrations in CLTE samples decreased about 36% per degree of latitude and were 26% lower in sites located in MPAs (Figure 3). Conversely, DDT concentrations in CLTE samples increased with latitude ($\chi^2(1) = 11.553$, $p < 0.001$) by about 45% per degree of latitude (Figure 3). No model adequately explained variation in CLTE PCB or CHL concentrations.

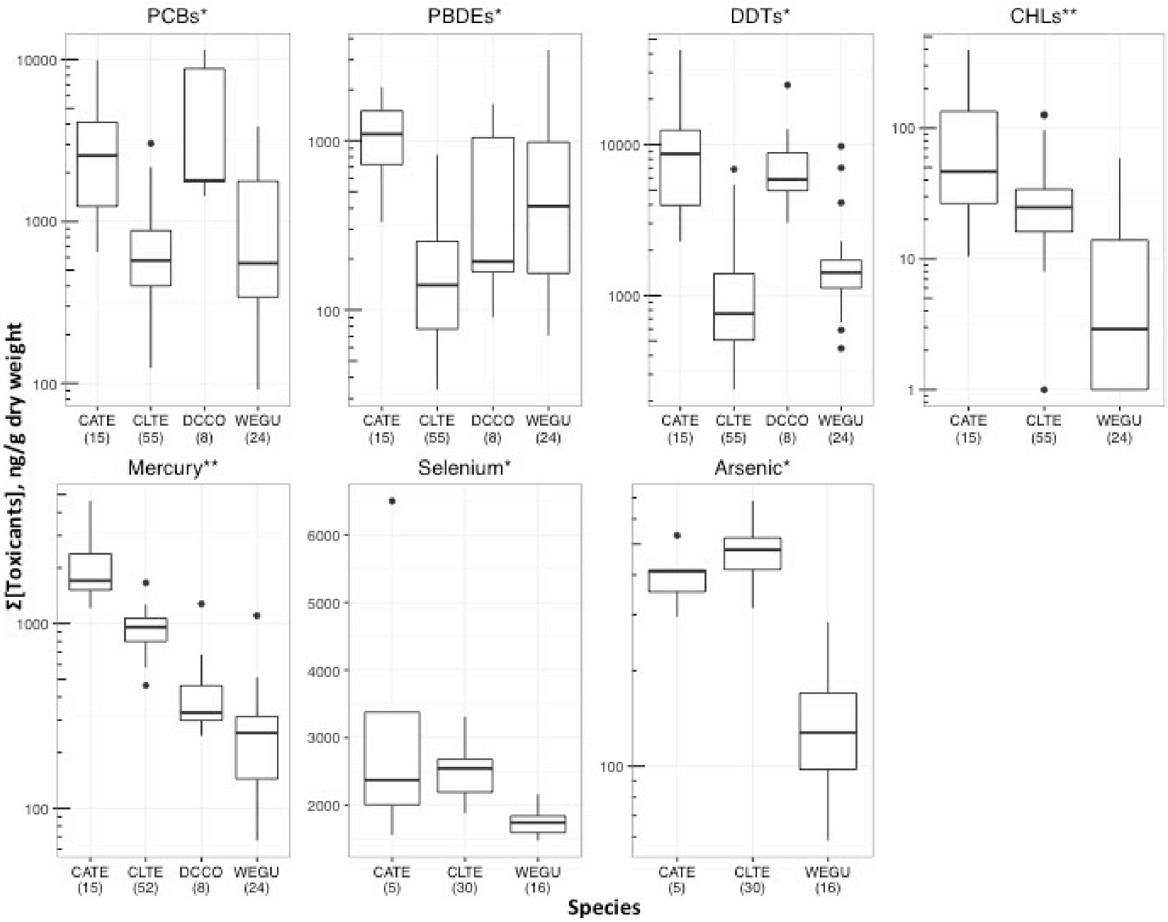

Figure 2. Toxicant concentrations by species. The concentrations of congeners within organic contaminant classes are summed by sample. Asterisks represent significant differences between species, and figures with two asterisks represent toxicant classes where all species have differing concentrations. Sample sizes are listed in parentheses below the 4-letter species ID for each toxicant class.

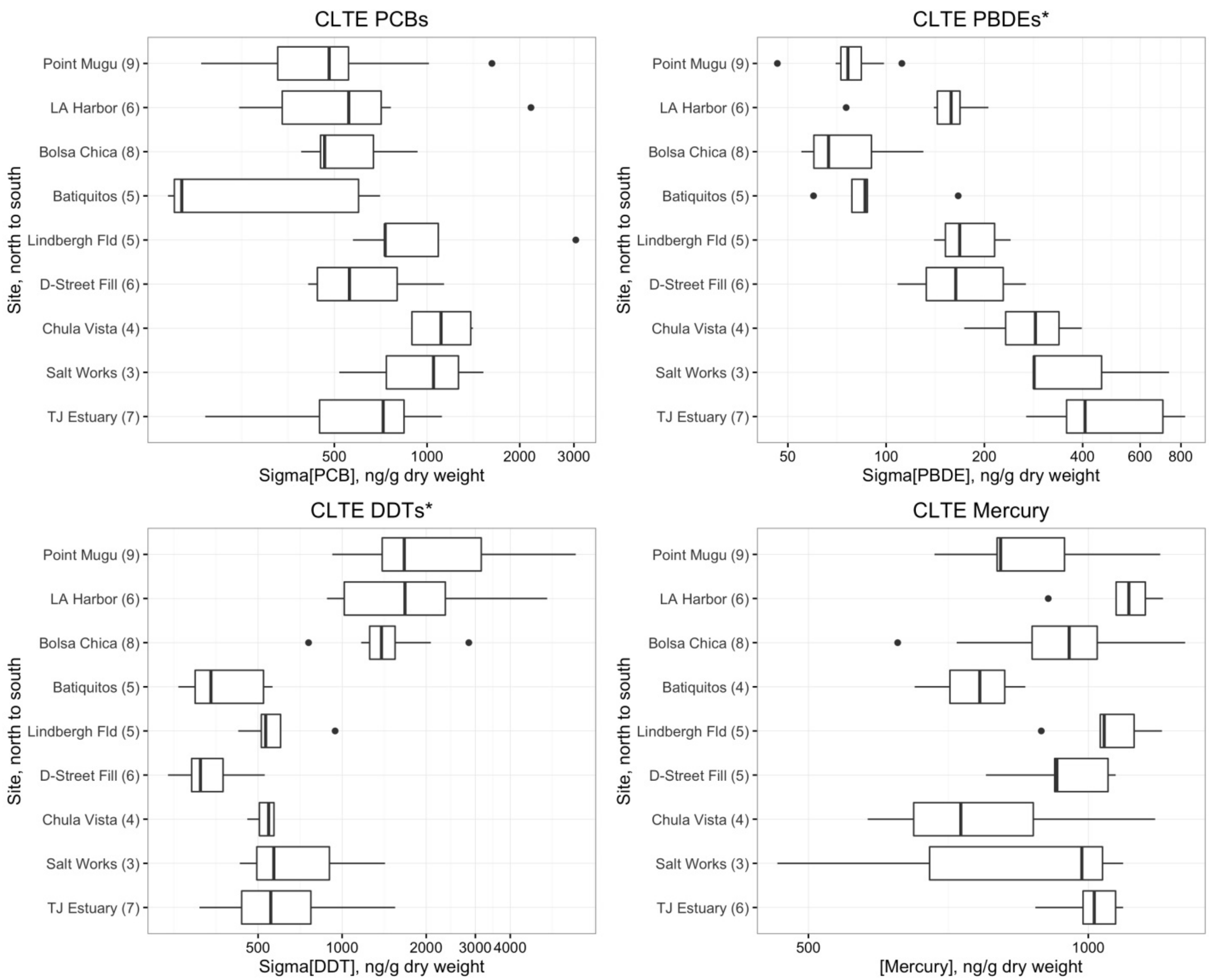

Figure 3. Latitudinal comparisons of toxicant concentrations in California least terns. Parentheses indicate sample size by site. Asterisks represent plots where a significant latitudinal trend is present.

Table 2A. Model selection tables for CLTE spatial data. LogLik is the likelihood of the model fit. AICc is a relative measure of quality of the model with the given data. Delta is the difference between the listed model and the model with the lowest AICc. Weight is a proportional estimate of how often a model will best predict new data.

Model (**Toxicant class**)	df	logLik	AICc	delta	weight
PCBs					
MPA + (1 | Site)	4	-9.38	27.59	0	0.603
Lat + (1 | Site)	4	-10.34	29.51	1.917	0.231
Lat + MPA + (1 | Site)	5	-10.28	31.84	4.251	0.072
UrbanDist + (1 | Site)	4	-11.75	32.34	4.742	0.056
MPA + UrbanDist + (1 | Site)	5	-11.3	33.87	6.277	0.026
Lat + UrbanDist + (1 | Site)	5	-12.46	36.19	8.599	0.008
Lat + MPA + UrbanDist + (1 | Site)	6	-12.37	38.56	10.965	0.003
PBDEs					
Lat + (1 | Site)	4	13.1	-17.36	0	0.532
Lat + MPA + (1 | Site)	5	13.79	-16.31	1.046	0.315
MPA + (1 | Site)	4	11.44	-14.05	3.312	0.101
Lat + UrbanDist + (1 | Site)	5	11.17	-11.07	6.292	0.023
UrbanDist + (1 | Site)	4	9.18	-9.54	7.823	0.011
Lat + MPA + UrbanDist + (1 | Site)	6	11.65	-9.47	7.888	0.01
MPA + UrbanDist + (1 | Site)	5	10.14	-9	8.356	0.008
DDTs					
Lat + (1 | Site)	4	-1.23	11.3	0	0.764
Lat + MPA + (1 | Site)	5	-1.43	14.13	2.829	0.186
Lat + UrbanDist + (1 | Site)	5	-3.46	18.2	6.901	0.024
MPA + (1 | Site)	4	-5.49	19.81	8.512	0.011
UrbanDist + (1 | Site)	4	-5.68	20.19	8.888	0.009
Lat + MPA + UrbanDist + (1 | Site)	6	-3.69	21.2	9.902	0.005
MPA + UrbanDist + (1 | Site)	5	-6.26	23.81	12.507	0.001

Model (**Toxicant class**)	df	logLik	AICc	delta	weight
Mercury					
MPA + (1 | Site)	4	44.58	-80.28	0	0.578
Lat + (1 | Site)	4	43.86	-78.84	1.438	0.281
UrbanDist + (1 | Site)	4	42.95	-77.02	3.263	0.113
Lat + MPA + (1 | Site)	5	42.3	-73.23	7.052	0.017
MPA + UrbanDist + (1 | Site)	5	41.2	-71.03	9.252	0.006
Lat + UrbanDist + (1 | Site)	5	41.05	-70.73	9.547	0.005
Lat + MPA + UrbanDist + (1 | Site)	6	39.63	-65.31	14.97	0
Selenium					
MPA + (1 | Site)	4	32.61	-55.41	0	0.465
Lat + (1 | Site)	4	32.54	-55.27	0.142	0.433
UrbanDist + (1 | Site)	4	30.81	-51.8	3.614	0.076
Lat + MPA + (1 | Site)	5	31.08	-49.29	6.117	0.022
Lat + UrbanDist + (1 | Site)	5	28.9	-44.95	10.459	0.002
MPA + UrbanDist + (1 | Site)	5	28.68	-44.5	10.91	0.002
Lat + MPA + UrbanDist + (1 | Site)	6	27.28	-38.36	17.054	0
Arsenic					
Lat + (1 | Site)	4	30.41	-51	0	0.544
MPA + (1 | Site)	4	29.35	-48.88	2.124	0.188
UrbanDist + (1 | Site)	4	29.18	-48.54	2.461	0.159
Lat + MPA + (1 | Site)	5	30.14	-47.43	3.573	0.091
Lat + UrbanDist + (1 | Site)	5	28.06	-43.25	7.749	0.011
MPA + UrbanDist + (1 | Site)	5	27.44	-42.02	8.986	0.006
Lat + MPA + UrbanDist + (1 | Site)	6	27.23	-38.26	12.746	0.001

Table 2B. Model selection tables for WEGU spatial data. CHLs are not included due to non-detects.

Model (**Toxicant class**)	df	logLik	AICc	delta	weight
PCBs					
Lat + (1 | Site)	4	-12.1	34.5	0	0.457
MPA + (1 | Site)	4	-12.3	34.9	0.43	0.368
Lat + MPA + (1 | Site)	5	-11.4	36.6	2.067	0.162
MPA + UrbanDist + (1 | Site)	5	-14.9	43.5	8.998	0.005
Lat + MPA + UrbanDist + (1 | Site)	6	-13.4	44.4	9.958	0.003
UrbanDist + (1 | Site)	4	-17.3	44.8	10.358	0.003
Lat + UrbanDist + (1 | Site)	5	-15.7	45.2	10.753	0.002
PBDEs					
Lat + MPA + (1 | Site)	5	-13.2	40.1	0	0.369
MPA + (1 | Site)	4	-14.9	40.1	0.086	0.353
Lat + (1 | Site)	4	-15.2	40.7	0.638	0.268
UrbanDist + (1 | Site)	4	-19	48.4	8.385	0.006
Lat + MPA + UrbanDist + (1 | Site)	6	-16.3	50.2	10.173	0.002
MPA + UrbanDist + (1 | Site)	5	-18.6	50.9	10.863	0.002
Lat + UrbanDist + (1 | Site)	5	-19	51.7	11.637	0.001

Model (**Toxicant class**)	df	logLik	AICc	delta	weight
DDTs					
Lat + (1 | Site)	4	-8.09	26.53	0	0.437
MPA + (1 | Site)	4	-8.16	26.68	0.145	0.406
Lat + MPA + (1 | Site)	5	-7.53	28.8	2.272	0.14
UrbanDist + (1 | Site)	4	-11.9	34.11	7.579	0.01
MPA + UrbanDist + (1 | Site)	5	-11.2	36.17	9.635	0.004
Lat + UrbanDist + (1 | Site)	5	-11.8	37.37	10.838	0.002
Lat + MPA + UrbanDist + (1 | Site)	6	-9.92	37.45	10.914	0.002
Mercury					
Lat + (1 | Site)	4	-2.05	14.45	0	0.421
MPA + (1 | Site)	4	-2.23	14.8	0.352	0.353
Lat + MPA + (1 | Site)	5	-1.18	16.1	1.653	0.184
UrbanDist + (1 | Site)	4	-5.2	20.76	6.306	0.018
MPA + UrbanDist + (1 | Site)	5	-3.64	21.04	6.589	0.016
Lat + UrbanDist + (1 | Site)	5	-4.61	22.97	8.515	0.006
Lat + MPA + UrbanDist + (1 | Site)	6	-3.44	24.48	10.024	0.003

In WEGU, we found a significant relationship between PCB concentrations and marine protected area status where PCB concentrations were significantly higher ($\chi^2(1) = 5.106$, $p < 0.024$) by about 250% for WEGU nesting outside of an MPA (e.g., NAS North Island, Figure 4). No fixed effects or their interactions significantly predicted PBDE, DDT, or CHL concentrations in WEGU.

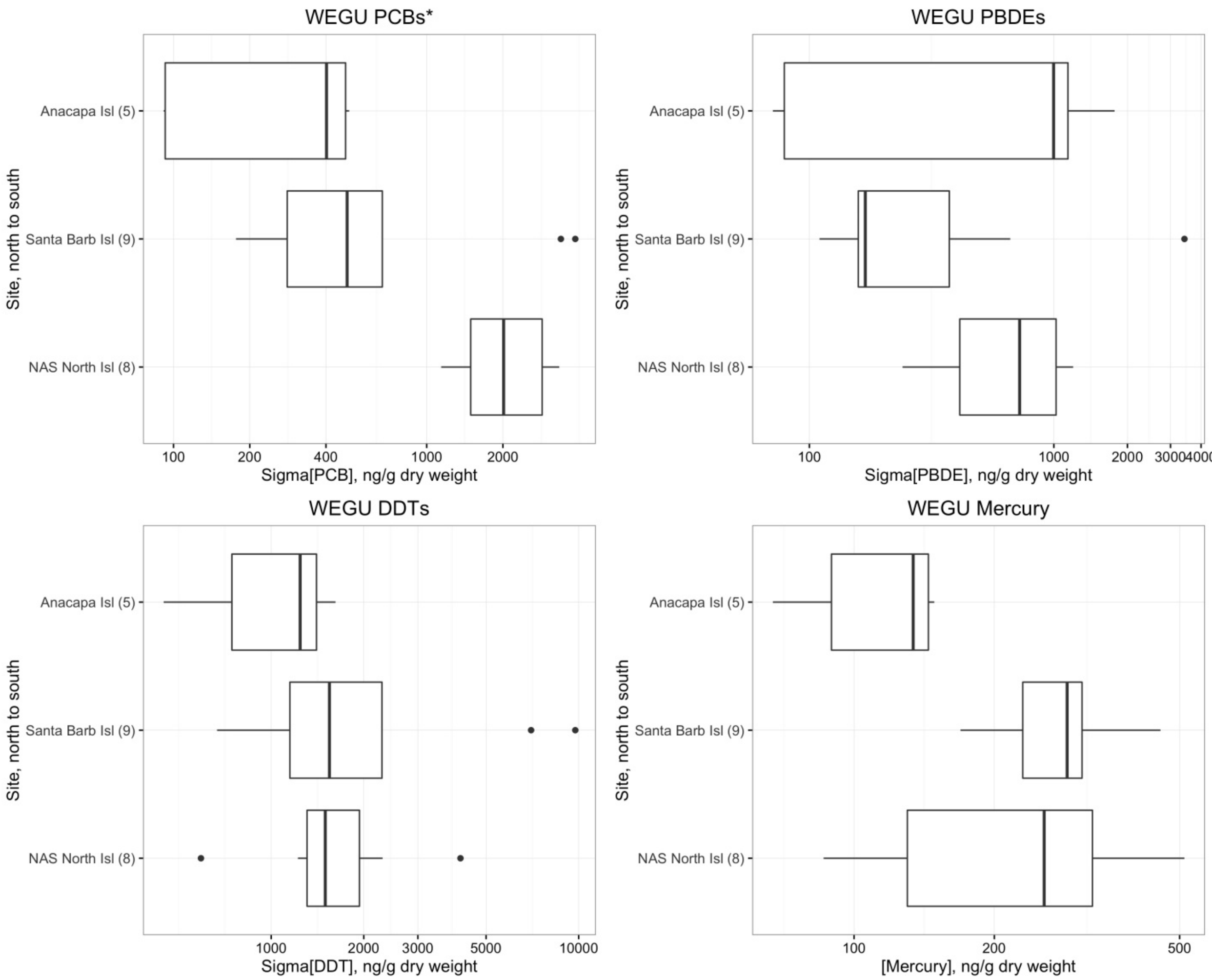

Figure 4. Latitudinal comparisons of toxicant concentrations in western gulls. Parentheses indicate sample size by site. Asterisks represent plots where a significant latitudinal trend is present.

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: $t=10.474$, $df = 6.66$, $p < 0.001$), PBDE ($t = 9.366$, $df = 5.20$, $p < 0.001$), DDT ($t = 8.724$, $df = 5.98$, $p < 0.001$), and CHL ($t = -5.278$, $df = 6.11$, $p < 0.002$) concentrations were higher in CATE than CLTE (Figure 5).

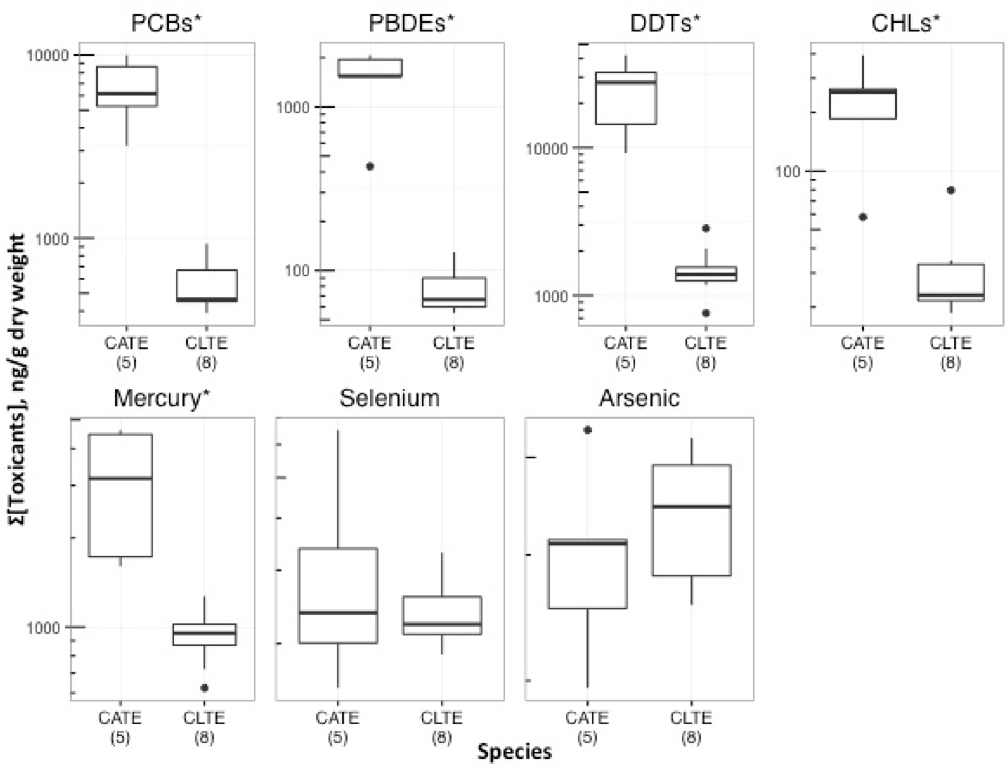

Figure 5. Toxicant concentrations in Bolsa Chica by species. The concentrations of congeners within organic contaminant classes are summed by sample. Asterisks represent significant differences between species. Sample sizes are listed in parentheses below the 4-letter species ID for each toxicant class.

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 ($p = 0.351$; Figure 6). 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 = =0.264, df = 2.42, p > 0.812) concentrations between species at Salt Works.

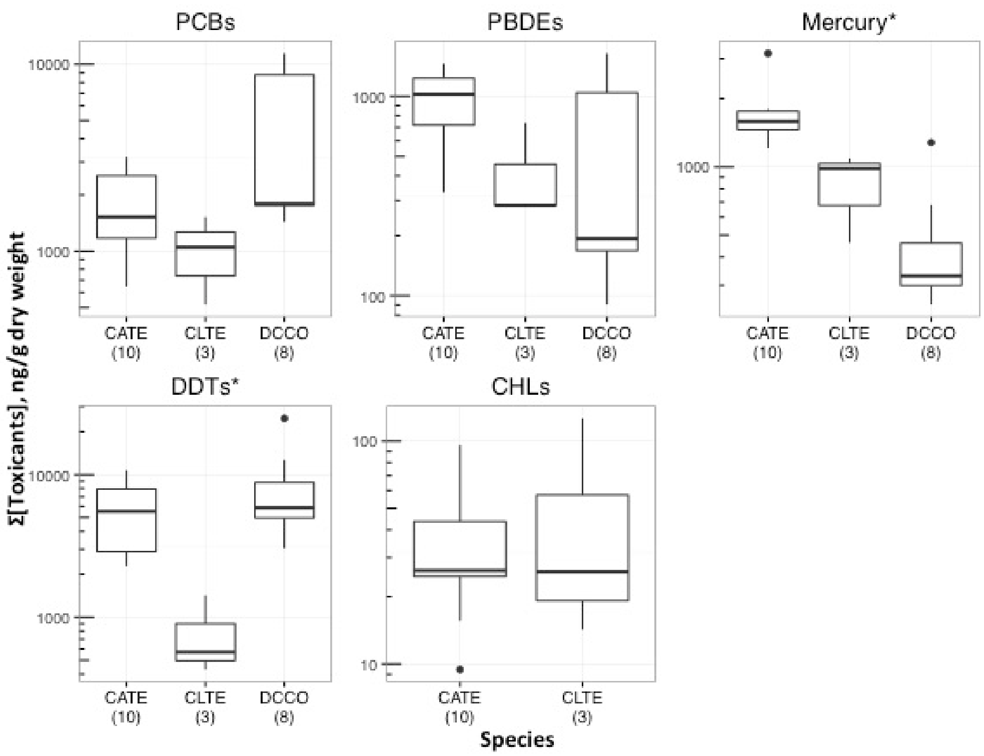

Figure 6. Toxicant concentrations in Salt Works by species. The concentrations of congeners within organic contaminant classes are summed by sample. Asterisks represent significant differences between species. Sample sizes are listed in parentheses below the 4-letter species ID for each toxicant class

Organic contaminants: effect on eggshell thickness

Regressions between CLTE eggshell thickness and PBDE and DDT concentrations explained very little of the observed variability in the data (PBDEs: $F(1,52) = 2.02$, $R^2 = 0.037$, $p = 0.16$; DDTs: $F(1,52) = 3.40$, $R^2 = 0.06$, $p = 0.07$, Figure S1). For WEGU, PBDE concentrations were not significantly related to eggshell thickness ($F(1,21) < 0.003$, $R^2 < 0.0002$, $p = 0.961$, Figure S2). There was a significant but weak relationship between WEGU DDT concentrations and eggshell thickness ($F(1,52) = 5.11$, $R^2 = 0.20$, $p = 0.034$, Figure 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: $F(1,51) = 2.53$, $R^2 = 0.05$, $p = 0.12$, Figure S1) and WEGU (PBDE: $F(1,21) = 0.10$, $R^2 = 0.004$, $p = 0.75$; DDT: $F(1,21) = 0.45$, $R^2 = 0.02$, $p = 0.51$, Figure S2).

Element contaminants: levels detected

We found some evidence of significant difference in element contamination among species. Mercury concentrations significantly differed ($\chi^2(3) = 71.05$, $p << 0.001$) 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 (Figure 2). For other elements there were fewer obvious patterns, although DCCO samples were not analyzed for selenium or arsenic. Selenium concentrations were significantly ($\chi^2(2) = 26.412$, $p << 0.001$) greater in CLTE than WEGU ($p << 0.001$), 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 ($p << 0.001$) had higher arsenic concentrations than WEGU ($\chi^2(2) = 27.733$, $p << 0.001$). DCCO samples were not analyzed for selenium or arsenic.

Likelihood ratio tests showed that no fixed effect significantly predicted mercury, selenium, or arsenic concentrations in regional comparisons of CLTE samples. No fixed effect significantly predicted mercury samples in WEGU. We did not conduct regional comparisons of selenium and arsenic in WEGU because samples from NAS North Island were not tested for these toxicants.

There was some evidence for differences in element concentrations between species nesting at the same site that was similar to the overall between species comparisons. At Bolsa Chica, CATE harbored significantly more mercury than CLTE ($t = 4.680$, $df = 4.80$, $p < 0.006$; Figure 5), but the two species had similar concentrations of selenium ($t = 0.656$, $df = 4.54$, $p > 0.543$) and arsenic ($t = -0.928$, $df = 6.62$, $p > 0.386$). At Salt Works, mercury concentrations differed ($\chi^2(2) = 27.733$, $p << 0.001$) and were higher in CATE than CLTE ($p < 0.029$) and DCCO ($p < 0.002$), whereas mercury concentrations were similar between CLTE and DCCO ($p > 0.125$). Samples at Salt Works were not analyzed for selenium or arsenic.

Screening levels

Across the region, no species exceeded the LOAEC-based thresholds for the legacy toxicants measured on a fresh weight basis (Table 4). However, DDT concentrations were above the NOAEC threshold for eggshell thinning for the majority of individuals in all species except CLTE (Table 4). In CLTE, only one sample was above the NOAEC threshold for reduced productivity associated with DDT. Of all species, CATE had the highest proportion of individuals above NOAEC thresholds for multiple toxicants (Table 4). The majority of CATE also had DDT concentrations above the NOAEC threshold for reduced productivity associated with DDT in most sensitive species, with two individuals above the LOAEC threshold for less sensitive seabirds. Four individual CATE were above the NOAEC threshold for mercury, and two individual CATE also were above the LOAEC threshold for mercury in sensitive species. In DCCO, one individual was above the NOAEC level for PBDE and two were above the NOAEC threshold for reduced reproductive activity associated with DDT. In WEGU, eight individuals were above the NOAEC threshold for PBDEs and three were above the NOAEC threshold for reduced productivity associated with DDT. No individuals in any species were above the NOAEC thresholds for PCBs or Arsenic. Effect thresholds were not available for CHLs.

Table 3. Summary statistics of egg contaminant data. All units are ng/g (ppb) basis. Geometric mean for CHL data was calculated by replacing NDs with ½ MDL, 0.025 ng/g.

		DRY WEIGHT				WET WEIGHT			
		CLTE	WEGU	CATE	DCCO	CLTE	WEGU	CATE	DCCO
PCBs	Range	124.3 - 3041.5	91.39 - 3863.44	645 - 9967	1436 - 11448	33.43 - 833.36	24.09 - 1089.49	140.6 - 2162.8	207.2 - 2255.2
	Mean	709.3	1235.45	3413	4795	186.83	314.68	802.4	848.8
	Geom Mean	562.5	712.8	2509.3	3284.2	142.7	187.5	626.9	527
PBDEs	Range	33.87 - 824.26	70.92 - 3420.65	330.1 - 2069.7	90.43 - 1644.82	9.11 - 274.48	18.44 - 2749.2	87.29 - 449.12	10.58 - 324.03
	Mean	198.02	675.98	1130.9	573.89	51.65	549.1	278.2	103.52
	Geom Mean	145.6	409.7	988.6	339.2	37	107.8	247	54.4
OCs	Range	248.6 - 6990.5	447 - 9749	2291 - 42493	3036 - 24834	56.67 - 1635.78	122.5 - 2749.2	536.1 - 9221	355.2 - 4395.6
	Mean	1216	1444	12251	8692	297.93	549.1	2819.3	1482.2
	Geom Mean	869.1	1533.6	8137.2	6975.8	220.5	403.5	2032.9	1119.3
Mercury	Range	462 - 1666.7	66.92 - 1104.84	1210 - 4617	247 - 1280	142 - 400.8	17.03 - 274	322 - 1100	35.09 - 227.84
	Mean	949.4	256	2184	482.4	243.1	73.6	537	82.18
	Geom Mean	927	223.9	1982.8	411.2	235.8	58.9	495.4	66
Selenium	Range	1883 - 3307	1480 - 2160	1566 - 6500	NA	460 - 774	367 - 656	368 - 1170	NA
	Mean	2495	1752	3165	NA	609.3	495.6	642	NA
	Geom Mean	2473.8	1741.6	2772.8	NA	605	490.3	586.8	NA
Arsenic	Range	314.5 - 682.9	58.46 - 565.69	295 - 532.3	NA	67.9 - 191	15.2 - 155	53.1 - 107	NA
	Mean	493.8	160.31	401.1	NA	122.3	45.25	85.7	NA
	Geom Mean	484.7	136	393.6	NA	118.5	38.3	83.3	NA

		LIPID WEIGHT				FRESH WEIGHT			
		CLTE	WEGU	CATE	DCCO	CLTE	WEGU	CATE	DCCO
PCBs	Range	172.3 - 7740.7	245.5 - 10894.9	1953 - 30987	4855 - 34274	25.84 - 685.02	22.69 - 1005.6	133.57 - 1959.5	171.78 - 1950.77
	Mean	1819.4	3368.7	9305	18266	138.06	315.1	751.44	727.83
	Geom Mean	1388	1905.8	6843.9	14033.6	109.08	183.35	592.45	461.76
PBDEs	Range	42.04 - 3147.55	192.5 - 8215.2	1063 - 6987	548.3 - 4924.5	7.04 - 165.51	17.37 - 981.2	82.93 - 413.86	8.77 - 280.29
	Mean	544.08	1777.8	3064	1983.6	37.49	171.1	261.21	88.86
	Geom Mean	359	1095.4	2696.2	1449.4	28.23	99.64	233.4	47.69
OCs	Range	166.3 - 18256.5	1201 - 27492	5691 - 117436	13158 - 68043	48.79 - 1406.77	118.31 - 2537.56	518.97 - 8354.25	293.44 - 3648.35
	Mean	3154.2	5580	33437	34981	231.52	529.26	2616.19	1280.75
	Geom Mean	2144	4100.2	22193.1	29807.9	168.53	386.74	1921.19	980.78
Mercury	Range	309 - 5388	181.6 - 2988	3381 - 16587	835.3 - 4938.3	107.62 - 279	16.39 - 245.5	295.27 - 1019.69	30.88 - 192.3
	Mean	2437	742.2	6083	2169	185.48	71.23	504.3	71.13
	Geom Mean	2267.6	598.6	5407.9	1757	180.44	57.61	468.15	57.8
Selenium	Range	3914 - 10813	3311 - 6090	3728 - 23353	NA	356.01 - 623.84	328.83 - 635.66	397.81 - 988.65	NA
	Mean	6321	4655	9243	NA	487.1	470.52	591.47	NA
	Geom Mean	6170.6	4596.8	7403.9	NA	483.1	464.38	557.02	NA
Arsenic	Range	821.4 - 1761	158.7 - 1519.6	711.1 - 1537.4	NA	54.86 - 146.69	14.32 - 149.73	44.87 - 105.18	NA
	Mean	1237.2	426.6	1083.3	NA	97.76	43.56	82.73	NA
	Geom Mean	1209	359.1	1051	NA	94.65	36.51	79.07	NA

		CLTE	WEGU	CATE	DCCO
Shell Thickness (mm)	Range	0.123 - 0.169	0.331 - 0.442	0.3 - 0.371	0.328 - 0.466
	Mean	0.145	0.371	0.3413	0.4096
Ratcliffe Index	Range	0.573-0.804	1.567-1.954	1.279-1.682	1.773-2.123
	Mean	0.666	1.804	1.529	1.986
Percent Lipid	Range	4.08 - 24.8	6.75 - 13.6	5.01 - 12.3	0.77 - 6.58
	Mean	10.33	10.01	9.42	4.525

Table 4. Screening values for analyzed toxicants. 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. Values between LOAEC may be of concern. NOAEC thresholds for DDTs are conservative estimates for all birds. No thresholds are available for CHL data.

			Number of samples above threshold (total sample size)				
Toxicant	**Site**	**NOAEC (ng/g fw)**	CATE	CLTE	DCCO	WEGU	**Reference**
PCB		2600	0 (15)	0 (55)	0 (8)	0 (23)	Harris and Elliott 2011
PBDE		200	10 (15)	0 (55)	1 (8)	8 (23)	Rattner et al. 2011
	Anacapa Isl					2 (4)	
	Bolsa Chica		4 (5)				
	NAS North Isl					4 (8)	
	Salt Works		6 (10)		1 (8)		
	Santa Barb Isl					2 (9)	
DDT[a]		200	15 (15)	21 (55)	8 (8)	19 (23)	DOI 1998
	Anacapa Isl					2 (4)	
	Bolsa Chica		5 (5)	6 (8)			
	LA Harbor			3 (6)			
	Lindbergh Fld			1 (5)			
	NAS North Isl					7 (8)	
	Pismo Beach					1 (1)	
	Point Mugu			8 (9)			
	Salt Works		10 (10)	1 (3)	8 (8)		
	Santa Barb Isl					8 (9)	
	TJ Estuary			1 (7)			
	Vandenberg			1 (2)		1 (1)	
DDT[b]		1000	12 (15)	1 (55)	2 (8)	3 (23)	DOI 1998
	Bolsa Chica		5 (5)				
	NAS North Isl					1 (8)	
	Point Mugu			1 (9)			
	Santa Barb Isl					2 (9)	
	Salt Works		7 (10)		2 (8)		
Mercury		500	4 (15)	0 (52)	0 (8)	0 (23)	Burger and Gochfeld 1997
	Bolsa Chica		3 (5)				
	Salt Works		1 (10)				
Selenium		900	1 (5)	0 (29)	-	0 (15)	Ohlendorf and Heinz 2011
	Bolsa Chica		1 (5)				
Arsenic		910	0 (5)	0 (29)	-	0 (15)	DOI 1998

Toxicant	Site	LOAEC (ng/g fw)	Number of samples above threshold (total sample size)				Reference
			CATE	CLTE	DCCO	WEGU	
PCB		23000	0 (15)	0 (55)	0 (8)	0 (23)	Harris and Elliott 2011
PBDE		1000	0 (15)	0 (55)	0 (8)	0 (23)	Harris and Elliott 2011
DDT[a]		10000	0 (15)	0 (55)	0 (8)	0 (23)	DOI 1998
DDT[b]		5000	2 (15)	0 (55)	0 (8)	0 (23)	DOI 1998
	Bolsa Chica		2 (5)				
Mercury		800	2 (15)	0 (52)	0 (8)	0 (23)	Henny et al. 2002
	Bolsa Chica		1 (5)				
	Salt Works		1 (10)				
Selenium		3000	0 (5)	0 (29)	-	0 (15)	Ohlendorf and Heinz 2011
Arsenic		>910	0 (5)	0 (29)	-	0 (15)	DOI 1998

[a]Thresholds for observed eggshell thinning in seabird species

[b]Thresholds for reduced reproductive activity in seabird species

DISCUSSION

Although seabirds have been established 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 species, a list that often includes sediment, bivalves, fish, and water quality (Dodder et al. 2012). This study confirms that salvaged eggs can be used to monitor both organic and element contaminants (Braune et al. 2002, Elliott and Elliott 2013).

Organic contaminants

Our findings confirm evidence of a continued decline in many organic contaminants (Dodder et al. 2012) in the SCB, yet these legacy toxicants persist in the SCB food web. Every sample across each of 13 sites contained congeners from each class of pollutants assessed with the exception of CHLs. In general, we found larger, piscivorous species (CATE and DCCO) had higher contaminant levels than the generalist (WEGU) and smaller (CLTE) species, a finding common with previous research (Burger and Gochfeld 1997, Braune et al. 2005). While all species in this study are piscivorous, DCCO and CATE diets likely comprise larger and older fish (due to a larger gape size) and may consume higher proportions of fish in their diet (versus other marine species like krill).

Using eggs from the two species for which samples were collected across the SCB, California least tern (CLTE) and WEGU, we found evidence for significant spatial patterns of organic contaminant exposure. For CLTE, PBDEs increased and DDTs decreased from north to south. The observed pattern for DDTs may be explained by the location of the Palos Verdes Shelf Superfund Site, which lies ~23 km west of the Bolsa Chica nesting site. The increase in PBDEs detected in the southern CLTE colonies has not been documented previously. However, sediments in San Diego Bay harbor higher concentrations of PBDEs than elsewhere in the SCB, followed by Los Angeles Harbor and Long Beach Harbor. Dodder et al (2012) showed that stormwater runoff is a primary source of PBDEs in the SCB. A previous study of CLTE in San Diego Bay found demonstrably higher mean levels of PBDEs (2,210 ng/g lipid weight) than those measured here (Zeeman et al. 2008). However, both this study and existing literature confirm that the highest levels of PBDEs in CLTE were found in sites near southern San Diego Bay. Increased PBDE monitoring in San Diego Bay will better inform this spatial pattern. In WEGU, PCBs decreased from north to south. 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).

Element contaminants

The results from the element analyses differ both in terms of contamination levels and spatial patterns of accumulation than the organic compounds. Although piscivorous seabirds like CLTE are not thought to be sensitive to mercury at the levels reported here (Shore et al. 2011), our data suggest that CLTE had higher mercury concentrations than other monitored species. Unlike many organic contaminants, mercury is both a point and non-point source pollutant, with mercury levels varying based on local anthropogenic activity at small temporal and spatial scales as well as from the global mercury cycle (Selin 2009). Additionally, mercury is not lipophilic and, in eggs, is concentrated in albumen predominantly in the form of bioavailable methylmercury (Ackerman et al. 2013). Thus, while mercury can biomagnify in food webs, the mechanisms by which biomagnification occurs differs from organic contaminants. The increased mercury concentrations in least terns versus higher trophic species (in this study, CATE and DCCO) may be due to differences in foraging location. Breeding least terns largely forage on resources in nearshore surface waters (Fournier et al. 2016 *in review*). Elevated levels of mercury have also been found in SCB sediments and fish (Maurer et al. 1994, Phillips et al. 1997). Like PBDEs and DDTs, it is possible that mercury levels in CLTE may mirror patterns of mercury levels in the nearshore environments in the SCB. A second explanation could be that CLTE mercury concentrations are associated with their overwintering area, as migratory CATE and DCCO in the central US and Canada have shown (Lavoie et al. 2015). This evidence suggests seabirds may have little capacity to excrete body-bound mercury via burning adipose tissue during migration, a decretion pathway that has been suggested for organic contaminants. Further study of mercury concentrations in different body tissues (e.g., feathers) and stable isotope analysis would inform possible sources of mercury in these species (Lavoie et al. 2015).

While site-level data on selenium and arsenic was not available for our focal species, detected mercury concentrations were temporally variable at local scales. For instance, mean mercury concentrations decreased in CLTE nesting at the D-Street fill (948 ng/g dw) in the 1980's (Hotham and Zador 1995) but increased in CLTE nesting at Tijuana River Estuary (1010 ng/g dw) by ~300 ng/g dw compared to mean concentrations measured in eggs from 1994-1996 (Hotham and Powell 2000). Mean mercury levels in CATE (541 ng/g fw) nesting at Salt Works in San Diego Bay were also similar to those analyzed in 2005 (Zeeman et al. 2008). Adverse reproductive effects from mercury exposure, including reduced clutch size, egg viability, and egg hatchability may occur at low concentrations (e.g., between 600 and 800 ng/g fw) in sensitive species (Shore et al. 2011). Mercury levels may not have fluctuated greatly in San Diego Bay over the last 10 – 20 years, but the increase in mercury levels at Tijuana River Estuary likely merits further investigation.

Potential for adverse effects and trends over time

Screening levels can help contextualize how detected toxins compare with toxicant concentrations at which adverse effects take place and may govern management of contaminant sources. In terms of potential for adverse effects, the evidence was mixed. No species on average was at risk of adverse effects from any toxicant class, though a few individuals harbored contaminants at or above the LOAEC. Results above the NOAEC levels show that species may potentially be adversely affected by toxicants, but the specificity of the effects of toxicants at these concentrations is low. Unfortunately, little is known about the toxicity of these chemicals at low, sub-lethal concentrations, and even less is known about the additive or synergistic effects of contaminants and other stressors, including interannual periods of low food availability and climate change (e.g., Noyes and Lema 2015).

Because there has been contaminant monitoring at specific sites within the SCB, we can also compare results from this study to previously monitored sites. On average, organic contaminants were detected in lower concentrations here than those found previously in seabird eggs in the SCB, including DDTs (424 ng/g wet weight) in WEGU nesting at NAS North Island (Jimenez-Castro et al. 1995), PCBs (512 ng/g ww), PBDEs (2550 ng/g lipid weight), and DDTs (1596 ng/g ww) in nesting CATE at Salt Works (Zeeman et al. 2008), PCBs (290 ng/g ww) and PBDEs (1320 ng/g lw) in nesting CLTE at Salt Works (Zeeman et al. 2008), and PCBs (165 ng/g ww) and DDTs (179 ng/g ww) 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 on average, and above the maximum value of DDT concentrations in 2008 (Zeeman et al. 2008). While these contaminants continue to decline below LOAEC thresholds, they still persist at detectable levels in coastal wildlife in the SCB.

Another important comparison to contextualize detected contaminant levels in seabirds at the regional scale is to compare concentrations among sample types, e.g. sediment, bivalves, fish, and water. While important, these comparisons are challenging without a clearer understanding of the pathway by which seabirds are exposed to toxicants in a food web. Clarity on this pathway may be supported using diet, stable isotope or additional contaminant data from water, sediment and prey invertebrates and fish (Hobson et al. 1997, Braune et al. 2002). Additional samples from seabird tissues formed at different times within the life cycle or annually, such as feathers or otoliths, can help pinpoint the geographic source of contamination (Lavoie et al. 2015). Telemetry and movement data for seabirds may provide greater explanatory power and links to observed toxicant concentrations throughout the SCB food web. Additional efforts are needed to compare the contaminant levels in the SCB across these sample types.

Contaminants and eggshell thinning

Although all four species exceeded the DDT NOAEC threshold for eggshell thinning in most sensitive species, we did not find a relationship between DDT or PBDE contamination and eggshell thickness or Ratcliffe's index in CLTE or WEGU. Values for eggshell thickness in CLTE and WEGU are similar to recent findings (Jimenez-Castro et al. 1995, Zeeman et al. 2008, Figure S1 and S2), and demonstrate that shell thickness for neither species have returned to thicknesses observed before DDT was in widespread use (Kiff 1994). Eggshell thinning is a concern for seabirds because it can lead to non-viable eggs and reproductive failure. There is historical evidence of eggshell thinning in the SCB associated with exposure to p,p-DDE, a metabolite of DDT converted in aquatic systems (Hickey and Anderson 1968). Avian species have varying sensitivities to DDE exposure (USDOI 1998). Many avian species nesting in the SCB now lay eggs with shell thicknesses approaching levels prior to the DDT discharge off the Palos Verdes shelf (e.g., 0.367 mm for WEGU in Jimenez-Castro et al. 1995). This study supports this trend, where eggshell thicknesses approached pre-1945 thicknesses (CLTE: 0.152mm in Blus & Prouty 1979, WEGU: 0.376 mm by L. Kiff in Jimenez-Castro et al. 1995, Figure S1 and S2 in this study) and many species laid eggs with mean DDT concentrations between NOAEC and LOAEC (USDOI 1998). Other factors, including laying order and egg age, also influence eggshell thickness (Hunt and Hunt 1973). While we did not have access to this in our study, our regressions on eggshell thickness suggest that other predictors may explain more variability in eggshell thickness than the compounds analyzed here.

Importance of regional monitoring

Monitoring contaminants at the regional scale across taxa and sample types is essential to tracking the health of marine systems. Seabirds are considered effective monitors of marine ecosystem health (Elliott and Elliott 2013), but few large-scale toxicant monitoring efforts include seabirds as indicator species (but see Braune et al. 2005). Here, we demonstrated the importance of including seabirds in a long-term biomonitoring program of the SCB, a 400 km urbanized coastline. 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 reliably combined within site to give site-specific parameters. Due to seabirds' position atop many aquatic food webs, recorded contaminant values are biologically relevant to other top predators in coastal and marine wildlife food webs.

The ability to compare contaminants regionally or among sites remains limited largely due to non-standard reporting for toxicant levels, sample type and inter-laboratory variation. The inability to transpose reporting metrics presents a substantial challenge to larger scale comparative research. To address this, we have reported toxicant concentrations in four different units – dry weight, wet weight, fresh weight, and lipid weight - to enable comparisons of toxicant concentrations with future studies. While many seabird tissues can be used to assess body burdens of toxicants, concentrations in each are not comparable to each other due to differences in how each toxicant may be stored or metabolized in the body. Inter-laboratory variation in quality assurance and quality control standards will also affect the accuracy with which contaminant levels are reported. We quantified this inter-laboratory variation by conducting round robin exercises with bird egg samples prior to the regional survey. Improved standardization within the monitoring community such as this will aid comparisons between local studies and scale-up to regional assessments.

CONCLUSIONS

- **Contaminant concentrations in seabird eggs were frequently detected throughout the SCB.**

Over 100 bird egg samples were analyzed as part of the Bight regional monitoring in 2013. Most contaminants analyzed were detected in virtually every sample from all four bird species, regardless of location.

- **Contaminant concentrations are comparable to or lower than previous studies in the SCB.**

While we cannot compare contaminant concentrations at regional scales because this is the first regional survey of bioaccumulation in bird eggs, we can compare the regional concentrations in 2013 to historical site-specific studies. DDT and PCB concentrations measured in historical studies, which occurred largely in San Diego Bay and date as far back as 20 years, were typically similar to or higher than what was observed in the 2013 regional monitoring.

- **Observed contaminant levels were generally lower than those that have potential for adverse effects, but synergistic or additive effects are unknown at this time.**

We used two thresholds from the literature for comparing the relative risk of regional contaminant concentration data in bird eggs: no observed apparent effects concentrations (NOAEC) and lowest observed apparent effects thresholds (LOAEC). Only 2 of 102 bird egg samples exceeded the LOAEC for any single contaminant, indicating that the probability of effects was likely low. However, many – and sometimes the majority – of the samples for single species exceeded NOAEC thresholds. The cumulative effects of multiple contaminants at these very low levels are uncertain.

- **There was no evidence of a relationship between eggshell thickness and PBDE or DDT levels in seabird eggs.**

Based on the regional distribution of DDT and PBDE in bird eggs from western gulls and California least terns, we did not see strong relationships with eggshell thickness. The lack of relationship may be a result of low concentrations relative to studies from the 1960's and 70's, when eggshell thinning was an important indicator of seabird population effects.

- **This study highlights the utility of seabirds as an indicator species for contaminant bioaccumulation in this region.**

The regional monitoring program was able to successfully sample, process, analyze, and assess contaminants in seabird eggs. The collaboration, coordination, and integration among sampling teams, laboratories, and managers proved that a regional monitoring program for bioaccumulation in seabirds is a viable and productive monitoring approach.

Recommendations

- **Compare contaminant concentrations in seabird eggs (this study) to contaminant concentrations in water, sediment, invertebrates, and fish from the SCB to track exposure pathways for wildlife.**

While we were able to assess the extent and magnitude of contaminants that bioaccumulate in seabird eggs, we do not yet know how or where the contaminants came from. Future studies should investigate trophic transfer through the coastal food web. This will be especially important for improving the State Water Board's Sediment Quality Objectives exposure modeling for wildlife risk.

- **Introduce additional monitoring tools, including stable isotope, telemetry, or GPS technology, to improve current understanding of exposure pathways in SCB wildlife.**

One mechanism for deciphering where contaminants come from is to use tools that either track where seabirds are feeding (i.e., near sediment contaminant hot spots) or geochemical tracers of contaminants. Tools currently exist to track feeding locations, including GPS transmitters that can be attached to individual birds nesting in the Bight. A tool currently available for tracking contaminants is stable isotope chemistry. Stable isotope chemistry has been used for other elements (i.e., lead), but is more difficult for organic contaminants.

- **Continue regional monitoring efforts to detect the occurrence of emerging contaminants in coastal and marine ecosystems.**

Since seabirds have proven to be a feasible and useful bioindicator for legacy (DDT, PCB) and more recent (PBDE) contaminants, seabirds can also be useful indicators for new and emerging contaminants. Bioaccumulation of new contaminants has been identified as a priority by the State Water Board's Expert Panel on constituents of emerging concern.

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SUPPLEMENTARY MATERIAL

Methods: Selecting screening levels

Contaminant levels measures in seabird eggs collected for this study were compared with levels associated with adverse effects in other studies of avian species. The amount of information on effect levels is variable depending on the contaminant and some may be field-based while others are laboratory-based. Effect levels may vary with species and effect, and often there are no data on effect levels for the species being studied. Consequently, contaminant levels reported for California least terns (CLTE), Caspian terns (CATE), double-crested cormorants (DCCO), and western gulls (WEGU) in this study were compared with estimated No Observed Adverse Effect Concentrations (NOAECs) and ranges of Lowest Observed Adverse Effect Concentrations (LOAECs) reported for sensitive adverse effects relating to maintenance of viable populations. Low ends of ranges were used for contaminants with multiple studies (and LOAECs) for an individual species, and therefore are considered conservative estimates of "thresholds for observed adverse effects." Consistent with approaches used by the U.S. Environmental Protection Agency (e.g., USEPA 1995), and depending on the available data, LOAECs were adjusted downward using uncertainty factors between 3 and 10, to obtain an estimate of a NOAEC. The fact that the estimated NOAECs are for use to evaluate risks to waterbirds was considered in the selection of adjustment factors. The derived NOAECs are considered conservative estimates of concentrations below which no adverse effects are expected for waterbirds and therefore may serve as screening levels for identifying contaminants of potential concern. Some values are based on data from very few studies and may change as more data becomes available.

Screening levels for PCBs

PCBs constitute a synthetic mixture of up to 209 congeners. Harris and Elliott (2011) identified ranges of total PCB concentrations in eggs that are associated with reduced hatching and/or fledging success (8 species), reduced productivity (3 species), and reduced parental care (2 species). Thresholds used to evaluate total PCB levels in seabird eggs from this study were selected using the low ends of ranges identified by Harris and Elliott (2011) for reproductive effects in high, intermediate and low sensitivity species. The estimated NOAEC for use in this study of waterbirds was based on the lowest LOAEC for terns, gulls and raptors. Basing the estimated NOAEC on LOAECs for least sensitive species raises some concern about ensuring that potentially more sensitive species in that group are protected. Consequently, to be protective, the LOAEC was adjusted downward by a factor of 3 for species differences in sensitivity, and downward by a factor of 3 again for LOAEC to NOAEC extrapolation, producing a final value that approaches an estimated NOAEC based on birds of intermediate sensitivity. Thresholds and screening levels used for total PCBs in this study on seabirds were:

NOAEC (estimated) - waterbirds (intermediate sensitivity) – 2600 ng/g fw

LOAEC - Reduced hatching/fledging or productivity - 23000 mg/kg fw

Screening levels for PBDEs

PBDEs are flame-retardant chemicals that were added to many types of consumer products to reduce potential for burning. Studies relating PBDE levels in avian eggs to adverse effects are very few in number, and the only readily available egg-based threshold is for reduced hatching success in American kestrel. Although no effect levels were reached, studies by McKernan et al. (2009) and Rattner et al. (2011) indicate that mallards and chickens are less sensitive than kestrels to PBDEs, and that the common tern, (and probably terns in general) are no more sensitive, and probably less sensitive than kestrels to PBDEs in eggs. The threshold used to evaluate total PBDE concentrations in seabird eggs for this study is a recommended threshold for reduced pipping and hatching success in American kestrel. The threshold based on American kestrel was adjusted downward by a factor of three for uncertainty about species differences in sensitivity and by a factor of three for the LOAEC to NOAEC extrapolation. The selected thresholds and screening levels for PBDEs were:

NOAEC (estimated) reduced hatching success – 200 ng/g fw

LOAEC - Reduced hatching success in a sensitive species – 1000 ng/g fw

Screening levels for DDTs

DDT is a legacy organochlorine pesticide that was manufactured and widely used between the early 1940s and 1972 for control of disease-carrying insects and insects on agricultural crops. Studies associating DDT concentrations in eggs with adverse effects have been conducted on numerous avian species (USDOI 1998, Blus 2011). Sensitivity to DDT can be highly variable depending on the species and the effect. USDOI (1998) and Blus (2011) provide ranges of thresholds for population-level effects and eggshell thinning with breakage in several avian species. The lowest threshold concentration for each species was used to rank species from most sensitive to least sensitive, and ranges were identified based on percentile rankings. The estimated NOAEC is based on the lowest threshold for sensitive species adjusted downward by a factor of three. Low-end thresholds used to evaluate DDT levels in seabird eggs, with a focus on reduced productivity and critical eggshell thinning (18%), were as follows:

NOAECs (estimated)-

eggshell thinning - 200 ng/g fw

reduced productivity - 1000 ng/g fw

LOAECS –

eggshell thinning in less-sensitive species - 10000 ng/g fw

reduced productivity in less-sensitive species - 5000 ng/g fw

Screening levels for mercury

Mercury naturally cycles in coastal and marine environments, but levels within those cycles have increased due to anthropogenic activity. Effect levels for mercury in avian eggs have been most recently reviewed by Shore et al. (2011) and include data for multiple species. Values selected for use in this study of seabird eggs are a combination of general guidelines, as well as LOAECs and NOAECs for piscivorous avian species, specifically snowy egret (*Egretta thula*), common loon (*Gavia immer*), and common tern (*Sterna hirundo*) (as cited by Shore et al. 2011). Few adjustments to the LOAEC were deemed necessary, given the available data, particularly on piscivorous birds. The lowest LOAEC for fish eating birds (egret) was adjusted downward by a factor of three for an estimated NOAEC. The thresholds and screening values used to assess mercury concentrations in seabird eggs for this study were:

NOAEC (estimated) - 500 ng/g fw

LOAECs in more-sensitive species - 800 ng/g fw (field based)

Screening levels for selenium

Selenium is an essential trace nutrient that supports beneficial metabolic functions, but it is also toxic to animals at exposure levels not much higher than those considered to be beneficial (USDOI 1998). Effect levels for selenium in avian eggs have been the subject of several reviews including those by USDOI (1998) and Ohlendorf and Heinz (2011). Values selected for use in this study of seabird eggs are ranges identified by USDOI (1998) and Ohlendorf and Heinz (2011) as representative for species of varying sensitivities. The estimated NOAEC is based on the lowest threshold for sensitive species adjusted downward by a factor of three, producing a value comparable to background. Assuming an average moisture content of 70% for fresh seabird eggs (from Zeeman et al. 2008), the screening values used in this study for seabird eggs were as follows:

NOAEC – 900 ng/g fw

LOAEC – 3000 ng/g fw

Screening levels for arsenic

Arsenic is generally known more for its toxicity to mammals (including humans) than to birds. Although based on limited data, USDOI (1998) was able to identify ranges for screening arsenic concentrations in avian eggs (as mg/kg dw). Assuming an average moisture content of 70% for fresh seabird eggs (from Zeeman et al. 2008), the screening values used in this study for seabird eggs were as follows:

NOAEC - 910 ng/g fw

LOAEC - >910 ng/g fw

Methods: Inter-laboratory Comparison

Prior to the measurement of field samples, an inter-laboratory comparison was performed to ensure the measurements between laboratories were comparable. The comparison utilized two reference materials: 1) NIST SRM 1946 Lake Superior Fish Tissue, a frozen fish tissue homogenate prepared from lake trout, and 2) frozen spiked chicken egg homogenate provided by the USFWS Analytical Control Facility. The spiked contaminant levels in the egg homogenate were set to mimic levels typically observed in other surveys (Tables S11 and S12). The exercise for metals required that laboratories obtain concentrations within 30% of the certified value (SRM 1946) or group mean (egg homogenate). The exercise for organics required measured concentrations within 40% of the certified or reference value (SRM 1946), or group mean (egg homogenate), for 70% of the target compounds with each class (organochlorine pesticides or PCBs). Both of these data quality objectives are consistent with laboratory comparability expectations for sediment (which all laboratories passed). Required reporting levels for both materials were: 20 ng arsenic/g ww, 30 ng mercury/g ww, 100 ng selenium/g ww, 0.5 ng organochlorine pesticide/g ww, and 2.5 ng PCB congener/g ww. Each material was run in triplicate by each participating laboratory.

Anonymized results are shown in Tables S7 to S12, and are summarized as follows.

Table S7 SRM 1946 Metals

All six laboratories passed the criteria for the three metals.

Table S8 SRM 1946 Chlorinated Pesticides

All six laboratories passed the criteria for organochlorine pesticides.

Table S9 SRM 1946 PCBs

Five of six laboratories passed the criteria for PCBs.

Table S10 Egg Homogenate Metals

All six laboratories passed the criteria for mercury and selenium. Arsenic was problematic and required two rounds of analysis. In the first round, the arsenic results were variable, ranging from non-detect to 190 ng/g ww, and only one of six laboratories passed the criteria. The spiked concentration was 60 ng/g ww and the required reporting level was 20 ng/g ww. Differences in digestion procedures were a suspected reason. Instrumental interference was likely not a reason because all laboratories utilized ICP-MS with a collision cell. In the second round, four of six laboratories passed the criteria.

Table S11 Egg Homogenate Chlorinated Pesticides

All six laboratories passed the criteria for pesticides. One laboratory, while passing overall, was an outlier for 4,4’-DDT and DDMU, and these results were not included in the group mean.

Table S12 Egg Homogenate PCBs

The PCB exercise as a whole was rejected because a majority of laboratory values were non-detects, or did not otherwise correspond to spiked levels. Based on these results, it was not certain the material had been properly spiked.

Only laboratories passing the metal inter-calibration exercises performed measurements on the field samples. Due to uncertainty in the organics inter-calibration exercise, particularly for PCBs, a single laboratory with prior experience analyzing eggs was selected to perform all organics measurements on the field samples.

Table S1. Analyte list, method detection level (MDL), and reporting level (RL) for egg samples. MDL and RL were converted from wet weight basis to ng/g dry weight assuming 75% moisture. % detect refers to the percentage of samples in which each analyte was detected.

PCBs	% detect	MDL	RL
PCB 018	0%	0.0125	0.025
PCB 028	33%	0.0125	0.025
PCB 037	23%	0.0125	0.025
PCB 044	11%	0.0125	0.025
PCB 049	41%	0.0125	0.025
PCB 052	84%	0.0125	0.025
PCB 066	92%	0.0125	0.025
PCB 070	71%	0.0125	0.025
PCB 074	85%	0.0125	0.025
PCB 077	56%	0.0125	0.025
PCB 081	3%	0.0125	0.025
PCB 087	72%	0.0125	0.025
PCB 099	98%	0.0125	0.025
PCB 101	84%	0.0125	0.025
PCB 105	89%	0.0125	0.025
PCB 110	96%	0.0125	0.025
PCB 114	26%	0.0125	0.025
PCB 118	99%	0.0125	0.025
PCB 119	0%	0.0125	0.025
PCB 123	12%	0.0125	0.025
PCB 126	6%	0.0125	0.025
PCB 128	87%	0.0125	0.025
PCB 138	100%	0.0125	0.025
PCB 149	87%	0.0125	0.025
PCB 151	54%	0.0125	0.025
PCB 153	100%	0.0125	0.025
PCB 156	72%	0.0125	0.025
PCB 157	31%	0.0125	0.025
PCB 158	97%	0.0125	0.025
PCB 167	65%	0.0125	0.025
PCB 168	86%	0.025	0.05
PCB 169	5%	0.0125	0.025
PCB 170	66%	0.0125	0.025
PCB 177	54%	0.0125	0.025
PCB 180	99%	0.0125	0.025
PCB 183	88%	0.0125	0.025
PCB 187	93%	0.0125	0.025
PCB 189	11%	0.0125	0.025
PCB 194	56%	0.0125	0.025
PCB 201	67%	0.0125	0.025
PCB 206	25%	0.0125	0.025

PBDEs	% detect	MDL	RL
PBDE 017	1%	0.0125	0.025
PBDE 028	74%	0.0125	0.025
PBDE 047	100%	0.0125	0.025
PBDE 049	68%	0.0125	0.025
PBDE 066	73%	0.0125	0.025
PBDE 071	6%	0.0125	0.025
PBDE 085	7%	0.0125	0.025
PBDE 099	95%	0.0125	0.025
PBDE 100	98%	0.0125	0.025
PBDE 138	9%	0.0125	0.025
PBDE 153	76%	0.0125	0.025
PBDE 154	75%	0.0125	0.025
PBDE 183	9%	0.0125	0.025
PBDE 190	2%	0.0125	0.025
PBDE 209	10%	0.0125	0.025

OCs	% detect	MDL	RL
Chlordane, cis-	51%	0.0125	0.025
Chlordane, trans-	6%	0.0125	0.025
DDD(o,p)	100%	0.0125	0.025
DDD(p,p)	24%	0.0125	0.025
DDE(o,p)	35%	0.0125	0.025
DDE(p,p)	100%	0.0125	0.025
DDMU(p,p)	88%	0.0125	0.025
DDT(o,p)	13%	0.0125	0.025
DDT(p,p)	19%	0.0125	0.025
Nonachlor, cis-	65%	0.0125	0.025
Nonachlor, trans-	80%	0.0125	0.025
Oxychlordane	1%	0.0125	0.025

Metals	% detect	MDL	RL
Mercury (CVAA)	100%	0.25	5
Mercury (EPA7473)	100%	2	2
Selenium	100%	0.25	12.5
Arsenic	100%	2.5	25

Table S2. Standard methods and instruments used to quantify each target class.

Target class	Method or instrument	Labs performed
PCBs	EPA8270Cm	PEL[a]
PBDEs	EPA8270Cm	PEL
OCs	EPA8270Cm	PEL
Mercury	EPA7473	CSD[b]
Mercury	CVAA[c]	LACSD[d]
Selenium	ICPMS[e]	LACSD
Arsenic	ICPMS	LACSD
Lipid	EPA160.3	PEL
Solids	SM2540D	PEL

[a]PEL = Physis Environmental Labs

[b]CSD = City of San Diego

[c]CVAA = Cold vapor atomic absorption

[d]LACSD = Los Angeles County Sanitation District

[e]ICPMS = Inductively coupled plasma mass spectroscopy

Table S3. Quality control data quality objectives. A batch was defined as not more than 20 samples. Metals includes mercury, arsenic, and selenium. For the organics, the accuracy was evaluated by individual contaminant (not the class sum). The reference material was either NIST SRM 1946: Lake Superior Fish Tissue, or a custom laboratory control material made from bird eggs. MDL = method detection limit and RPD = relative percent difference.

QC Material	Objective	Metal Criteria	Organics Criteria
Method Blank	Frequency	1/batch	1/batch
Method Blank	Accuracy	blank < 5 times MDL or blank < 5 times the minimum field concentration	blank < 10 times MDL
Spiked Blank	Frequency	1/batch	Not required
Spiked Blank	Accuracy	+/- 25% of spike value	NA
Reference Material	Frequency	1/batch	1/batch
Reference Material	Accuracy	+/- 20% of true value	+/- 30% of true value for 70% of compounds
Matrix Spike	Frequency	>= 10% of field samples	1/batch
Matrix Spike	Accuracy	+/- 25% of true value	+/- 50% of true value
Sample Duplicate	Frequency	>= 10% of field samples	1/batch
Sample Duplicate	Accuracy	< 25% RPD	< 25% RPD

Table S4. Data quality objective success rates for each contaminant class. Metals includes mercury, arsenic, and selenium. For the organics, the accuracy was evaluated by individual contaminant (not the class sum).

QC Material	Objective	PCB Success	OC Success	PBDE Success	Metal Success
Method Blank	Frequency	100%	100%	100%	100%
Method Blank	Accuracy	100%	100%	100%	85%
Spiked Blank	Frequency	NA	NA	NA	100%
Spiked Blank	Accuracy	NA	NA	NA	84%
Reference Material	Frequency	100%	100%	100%	100%
Reference Material	Accuracy	50%[1]	100%	92%	93%
Matrix Spike	Frequency	100%	100%	100%	100%
Matrix Spike	Accuracy	86%	91%	89%	86%
Sample Duplicate	Frequency	92%	92%	92%	100%
Sample Duplicate	Accuracy	82%	84%	75%[2]	94%

[1]Accuracy success was 100% if +/- 40% of the true value for 70% of the compounds, instead of +/- 30% of the true value for 70% of the compounds.

[2]Accuracy success was 82% if the RPD was < 30%, instead of < 25%.

Table S5. Summary statistics of analytes by species and site. Analyte units are ng/g dry weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
PCBs	California least tern	Entire SCB	55	709.3	524.8	124	3041
PCBs	California least tern	Batiquitos	5	351.6	276.4	144	704
PCBs	California least tern	Bolsa Chica	8	575.4	204.6	390	932
PCBs	California least tern	Chula Vista	4	1143.4	289.6	892	1408
PCBs	California least tern	D-Street Fill	6	661.0	287.7	411	1136
PCBs	California least tern	LA Harbor	6	764.5	717.2	245	2175
PCBs	California least tern	Lindbergh Field	5	1233.0	1028.4	574	3041
PCBs	California least tern	Pt Mugu	9	589.4	458.7	185	1625
PCBs	California least tern	Salt Works	3	1030.7	503.0	519	1524
PCBs	California least tern	TJ Estuary	7	663.4	318.9	190	1118
PCBs	California least tern	Vandenberg	2	158.5	48.5	124	193
PCBs	Caspian tern	Entire SCB	15	3413.1	2851.3	645	9967
PCBs	Caspian tern	Bolsa Chica	5	6634.3	2696.2	3191	9967
PCBs	Caspian tern	Salt Works	10	1802.5	876.7	645	3192
PCBs	Double-crested cormorant	Salt Works	8	4794.7	596.8	1436	11448
PCBs	Western gull	Entire SCB	24	1235.5	1225.9	91	3863
PCBs	Western gull	Anacapa Isl	5	311.7	203.5	91	494
PCBs	Western gull	NAS North Isl	8	2165.2	847.9	1141	3340
PCBs	Western gull	Pismo Beach	1	362.7	NA	363	363
PCBs	Western gull	Santa Barbara Isl	9	1115.3	1438.2	176	3863
PCBs	Western gull	Vandenberg	1	370.8	NA	371	371
PBDEs	California least tern	Entire SCB	55	198.0	181.4	34	824
PBDEs	California least tern	Batiquitos	5	95.7	40.9	60	166
PBDEs	California least tern	Bolsa Chica	8	78.5	26.8	55	130
PBDEs	California least tern	Chula Vista	4	287.0	95.2	173	398
PBDEs	California least tern	D-Street Fill	6	180.2	66.1	108	268
PBDEs	California least tern	LA Harbor	6	151.4	43.2	75	206
PBDEs	California least tern	Lindbergh Field	5	183.1	42.9	140	241
PBDEs	California least tern	Pt Mugu	9	79.4	18.3	46	112
PBDEs	California least tern	Salt Works	3	433.9	261.2	282	736
PBDEs	California least tern	TJ Estuary	7	517.5	220.1	268	824
PBDEs	California least tern	Vandenberg	2	46.2	17.4	34	58
PBDEs	Caspian tern	Entire SCB	15	1130.9	540.6	330	2070
PBDEs	Caspian tern	Bolsa Chica	5	1506.6	644.9	433	2070
PBDEs	Caspian tern	Salt Works	10	943.0	390.1	330	1459
PBDEs	Double-crested cormorant	Salt Works	8	573.9	596.8	90	1645
PBDEs	Western gull	Entire SCB	24	676.0	742.5	71	3421
PBDEs	Western gull	Anacapa Isl	5	812.3	733.3	71	1772
PBDEs	Western gull	NAS North Isl	8	737.0	373.3	241	1200
PBDEs	Western gull	Pismo Beach	1	583.7	NA	584	584
PBDEs	Western gull	Santa Barbara Isl	9	615.2	1067.0	110	3421
PBDEs	Western gull	Vandenberg	1	145.3	NA	145	145

Table S5. Summary statistics of analytes by species and site. Analyte units are ng/g dry weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
DDTs	California least tern	Entire SCB	55	1185.2	1224.8	238	6866
DDTs	California least tern	Batiquitos	5	396.5	137.7	259	563
DDTs	California least tern	Bolsa Chica	8	1537.8	640.0	758	2842
DDTs	California least tern	Chula Vista	4	530.5	53.7	457	572
DDTs	California least tern	D-Street Fill	6	344.7	103.5	238	528
DDTs	California least tern	LA Harbor	6	2187.0	1708.1	883	5423
DDTs	California least tern	Lindbergh Field	5	603.3	200.6	425	944
DDTs	California least tern	Pt Mugu	9	2392.1	1874.0	922	6866
DDTs	California least tern	Salt Works	3	808.1	538.5	430	1425
DDTs	California least tern	TJ Estuary	7	701.2	439.8	308	1548
DDTs	California least tern	Vandenberg	2	857.0	504.9	500	1214
DDTs	Caspian tern	Entire SCB	15	12149.7	12157.3	2282	42230
DDTs	Caspian tern	Bolsa Chica	5	25139.5	13409.4	9191	33039
DDTs	Caspian tern	Salt Works	10	5654.8	3063.0	2282	10739
DDTs	Double-crested cormorant	Salt Works	8	8683.0	7148.4	3036	24286
DDTs	Western gull	Entire SCB	24	2042.7	2122.7	447	9749
DDTs	Western gull	Anacapa Isl	5	1091.6	483.3	447	1619
DDTs	Western gull	NAS North Isl	8	1803.2	1062.3	591	4126
DDTs	Western gull	Pismo Beach	1	583.7	NA	584	584
DDTs	Western gull	Santa Barbara Isl	9	2943.2	3186.9	667	9749
DDTs	Western gull	Vandenberg	1	1673.8	NA	1674	1674
CHLs	California least tern	Entire SCB	55	30.7	26.4	0	126
CHLs	California least tern	Batiquitos	5	6.8	3.9	0	9
CHLs	California least tern	Bolsa Chica	8	31.8	20.2	19	80
CHLs	California least tern	Chula Vista	4	29.9	6.8	21	38
CHLs	California least tern	D-Street Fill	6	17.3	7.5	9	28
CHLs	California least tern	LA Harbor	6	36.5	26.9	13	82
CHLs	California least tern	Lindbergh Field	5	28.3	6.5	22	37
CHLs	California least tern	Pt Mugu	9	40.3	34.7	8	124
CHLs	California least tern	Salt Works	3	55.5	61.7	14	126
CHLs	California least tern	TJ Estuary	7	36.5	30.5	10	95
CHLs	California least tern	Vandenberg	2	16.7	10.8	9	24
CHLs	Caspian tern	Entire SCB	15	101.2	117.0	9	394
CHLs	Caspian tern	Bolsa Chica	5	230.7	122.5	58	394
CHLs	Caspian tern	Salt Works	10	36.5	25.2	9	96
CHLs	Double-crested cormorant	Salt Works	8	8.8	9.5	0	24
CHLs	Western gull	Entire SCB	24	8.0	13.0	0	58
CHLs	Western gull	Anacapa Isl	5	6.8	8.4	0	18
CHLs	Western gull	NAS North Isl	8	6.9	6.1	0	16
CHLs	Western gull	Pismo Beach	1	2.4	NA	2	2
CHLs	Western gull	Santa Barbara Isl	9	11.2	19.7	0	58
CHLs	Western gull	Vandenberg	1	0.0	NA	0	0

Table S5. Summary statistics of analytes by species and site. Analyte units are ng/g dry weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
Mercury	California least tern	Entire SCB	55	949.4	205.7	463	1667
Mercury	California least tern	Batiquitos	4	759.0	88.6	650	856
Mercury	California least tern	Bolsa Chica	8	939.7	199.0	624	1271
Mercury	California least tern	Chula Vista	4	805.5	264.0	579	1180
Mercury	California least tern	D-Street Fill	5	948.2	118.5	776	1070
Mercury	California least tern	LA Harbor	6	1091.1	104.1	905	1203
Mercury	California least tern	Lindbergh Field	5	1056.0	115.5	890	1200
Mercury	California least tern	Pt Mugu	9	878.2	157.5	683	1195
Mercury	California least tern	Salt Works	3	845.7	335.6	463	1090
Mercury	California least tern	TJ Estuary	6	1010.5	78.6	877	1090
Mercury	California least tern	Vandenberg	2	1260.8	573.9	855	1667
Mercury	Caspian tern	Entire SCB	15	2183.8	1116.1	1210	4617
Mercury	Caspian tern	Bolsa Chica	5	3115.3	1439.0	1602	4617
Mercury	Caspian tern	Salt Works	10	1718.0	542.3	1210	3170
Mercury	Double-crested cormorant	Salt Works	8	482.4	349.3	247	1280
Mercury	Western gull	Entire SCB	24	276.6	213.3	67	1105
Mercury	Western gull	Anacapa Isl	5	116.7	36.4	67	149
Mercury	Western gull	NAS North Isl	8	255.3	147.3	86	512
Mercury	Western gull	Pismo Beach	1	329.6	NA	330	330
Mercury	Western gull	Santa Barbara Isl	9	286.5	92.3	170	455
Mercury	Western gull	Vandenberg	1	1104.8	NA	1105	1105
Selenium	California least tern	Entire SCB	29	2495.4	333.4	1883	3307
Selenium	California least tern	Batiquitos	4	2551.7	279.6	2199	2867
Selenium	California least tern	Bolsa Chica	8	2380.7	456.1	1883	3307
Selenium	California least tern	Chula Vista	0				
Selenium	California least tern	D-Street Fill	0				
Selenium	California least tern	LA Harbor	6	2605.0	185.7	2391	2868
Selenium	California least tern	Lindbergh Field	0				
Selenium	California least tern	Pt Mugu	9	2523.2	338.8	1983	3047
Selenium	California least tern	Salt Works	0				
Selenium	California least tern	TJ Estuary	0				
Selenium	California least tern	Vandenberg	2	2387.3	332.2	2152	2622
Selenium	Caspian tern	Entire SCB	5	3165.1	1980.4	1566	6500
Selenium	Caspian tern	Bolsa Chica	5	3165.1	1980.4	1566	6500
Selenium	Caspian tern	Salt Works	0				
Selenium	Double-crested cormorant	Salt Works	0				
Selenium	Western gull	Entire SCB	16	1751.9	198.7	1480	2160
Selenium	Western gull	Anacapa Isl	5	1677.9	139.2	1483	1842
Selenium	Western gull	NAS North Isl	0				
Selenium	Western gull	Pismo Beach	1	1689.1	NA	1689	1689
Selenium	Western gull	Santa Barbara Isl	9	1830.2	210.5	1508	2160
Selenium	Western gull	Vandenberg	1	1479.8	NA	1480	1480

Table S6. Summary statistics of analytes by species and site. Analyte units are ng/g fresh weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
Arsenic	California least tern	Entire SCB	29	97.8	25.0	55	147
Arsenic	California least tern	Batiquitos	4	128.7	19.4	107	147
Arsenic	California least tern	Bolsa Chica	8	85.4	17.5	62	113
Arsenic	California least tern	Chula Vista	0				
Arsenic	California least tern	D-Street Fill	0				
Arsenic	California least tern	LA Harbor	6	114.2	21.7	80	142
Arsenic	California least tern	Lindbergh Field	0				
Arsenic	California least tern	Pt Mugu	9	84.5	18.9	55	107
Arsenic	California least tern	Salt Works	0				
Arsenic	California least tern	TJ Estuary	0				
Arsenic	California least tern	Vandenberg	2	95.5	25.6	77	114
Arsenic	Caspian tern	Entire SCB	5	82.7	25.1	45	105
Arsenic	Caspian tern	Bolsa Chica	5	82.7	25.1	45	105
Arsenic	Caspian tern	Salt Works	0				
Arsenic	Double-crested cormorant	Salt Works	0				
Arsenic	Western gull	Entire SCB	15	43.6	33.4	14	150
Arsenic	Western gull	Anacapa Isl	4	61.7	60.1	14	150
Arsenic	Western gull	NAS North Isl	0				
Arsenic	Western gull	Pismo Beach	1	33.7	NA	34	34
Arsenic	Western gull	Santa Barbara Isl	9	36.9	19.1	20	82
Arsenic	Western gull	Vandenberg	1	41.0	NA	41	41

Table S6. Summary statistics of analytes by species and site. Analyte units are ng/g fresh weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
PCBs	California least tern	Entire SCB	55	236.7	127.7	105	877
PCBs	California least tern	Batiquitos	5	149.6	49.1	115	223
PCBs	California least tern	Bolsa Chica	8	205.1	47.0	152	294
PCBs	California least tern	Chula Vista	4	346.1	90.1	260	431
PCBs	California least tern	D-Street Fill	6	241.0	99.2	150	408
PCBs	California least tern	LA Harbor	6	237.2	130.3	137	482
PCBs	California least tern	Lindbergh Field	5	384.4	276.7	232	877
PCBs	California least tern	Pt Mugu	9	205.4	101.8	120	458
PCBs	California least tern	Salt Works	3	296.0	80.9	205	361
PCBs	California least tern	TJ Estuary	7	214.4	77.6	123	351
PCBs	California least tern	Vandenberg	2	109.2	5.3	105	113
PCBs	Caspian tern	Entire SCB	15	954.4	597.8	231	2368
PCBs	Caspian tern	Bolsa Chica	5	1592.1	514.1	948	2368
PCBs	Caspian tern	Salt Works	10	635.5	315.4	231	1276
PCBs	Double-crested cormorant	Salt Works	8	926.8	837.1	276	2358
PCBs	Western gull	Entire SCB	23	443.9	368.2	102	1252
PCBs	Western gull	Anacapa Isl	4	155.8	63.1	102	227
PCBs	Western gull	NAS North Isl	8	680.7	273.6	370	1078
PCBs	Western gull	Pismo Beach	1	181.5	NA	181	181
PCBs	Western gull	Santa Barbara Isl	9	420.8	435.5	137	1252
PCBs	Western gull	Vandenberg	1	171.6	NA	172	172
PBDEs	California least tern	Entire SCB	55	37.5	31.4	7	166
PBDEs	California least tern	Batiquitos	5	18.8	6.9	13	30
PBDEs	California least tern	Bolsa Chica	8	15.7	7.3	9	30
PBDEs	California least tern	Chula Vista	4	56.7	18.0	38	77
PBDEs	California least tern	D-Street Fill	6	36.5	16.0	21	67
PBDEs	California least tern	LA Harbor	6	28.9	8.2	16	38
PBDEs	California least tern	Lindbergh Field	5	38.8	10.5	30	52
PBDEs	California least tern	Pt Mugu	9	15.2	2.3	11	20
PBDEs	California least tern	Salt Works	3	76.1	22.4	61	102
PBDEs	California least tern	TJ Estuary	7	92.4	39.6	45	166
PBDEs	California least tern	Vandenberg	2	8.4	1.9	7	10
PBDEs	Caspian tern	Entire SCB	15	261.2	112.9	83	414
PBDEs	Caspian tern	Bolsa Chica	5	296.0	121.9	101	407
PBDEs	Caspian tern	Salt Works	10	243.8	110.5	83	414
PBDEs	Double-crested cormorant	Salt Works	8	88.9	99.1	9	280
PBDEs	Western gull	Entire SCB	23	171.1	210.3	17	981
PBDEs	Western gull	Anacapa Isl	4	193.3	208.4	17	435
PBDEs	Western gull	NAS North Isl	8	176.2	96.2	53	303
PBDEs	Western gull	Pismo Beach	1	146.2	NA	146	146
PBDEs	Western gull	Santa Barbara Isl	9	174.9	307.4	29	981
PBDEs	Western gull	Vandenberg	1	32.3	NA	32	32

Table S6. Summary statistics of analytes by species and site. Analyte units are ng/g fresh weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
DDTs	California least tern	Entire SCB	55	225.7	228.2	47	1382
DDTs	California least tern	Batiquitos	5	76.5	15.8	61	101
DDTs	California least tern	Bolsa Chica	8	302.0	145.6	164	570
DDTs	California least tern	Chula Vista	4	106.2	18.5	88	125
DDTs	California least tern	D-Street Fill	6	70.8	30.9	47	132
DDTs	California least tern	LA Harbor	6	406.1	272.0	190	867
DDTs	California least tern	Lindbergh Field	5	129.3	50.9	78	213
DDTs	California least tern	Pt Mugu	9	453.0	363.0	190	1382
DDTs	California least tern	Salt Works	3	140.6	53.0	92	197
DDTs	California least tern	TJ Estuary	7	125.3	85.8	66	311
DDTs	California least tern	Vandenberg	2	153.6	70.2	104	203
DDTs	Caspian tern	Entire SCB	15	2594.9	2135.7	517	8303
DDTs	Caspian tern	Bolsa Chica	5	4799.5	2264.2	2335	8303
DDTs	Caspian tern	Salt Works	10	1492.6	875.5	517	2793
DDTs	Double-crested cormorant	Salt Works	8	1279.4	1098.0	294	3647
DDTs	Western gull	Entire SCB	23	527.0	566.3	118	2538
DDTs	Western gull	Anacapa Isl	4	243.7	113.9	118	369
DDTs	Western gull	NAS North Isl	8	431.1	272.6	130	1013
DDTs	Western gull	Pismo Beach	1	245.5	NA	246	246
DDTs	Western gull	Santa Barbara Isl	9	786.7	820.9	198	2538
DDTs	Western gull	Vandenberg	1	371.9	NA	372	372
CHLs	California least tern	Entire SCB	55	5.8	4.5	0	25
CHLs	California least tern	Batiquitos	5	1.4	0.8	0	2
CHLs	California least tern	Bolsa Chica	8	6.3	4.2	3	16
CHLs	California least tern	Chula Vista	4	6.0	1.8	4	8
CHLs	California least tern	D-Street Fill	6	3.5	1.9	2	7
CHLs	California least tern	LA Harbor	6	6.9	4.6	2	13
CHLs	California least tern	Lindbergh Field	5	6.0	1.6	5	8
CHLs	California least tern	Pt Mugu	9	7.6	6.8	2	25
CHLs	California least tern	Salt Works	3	8.9	7.6	3	18
CHLs	California least tern	TJ Estuary	7	6.2	4.5	2	13
CHLs	California least tern	Vandenberg	2	3.0	1.5	2	4
CHLs	Caspian tern	Entire SCB	15	21.3	21.2	2	65
CHLs	Caspian tern	Bolsa Chica	5	44.8	20.9	14	65
CHLs	Caspian tern	Salt Works	10	9.5	6.6	2	25
CHLs	Double-crested cormorant	Salt Works	8	1.4	1.5	0	3
CHLs	Western gull	Entire SCB	23	2.2	3.8	0	17
CHLs	Western gull	Anacapa Isl	4	2.0	2.4	0	5
CHLs	Western gull	NAS North Isl	8	1.7	1.5	0	4
CHLs	Western gull	Pismo Beach	1	0.6	NA	1	1
CHLs	Western gull	Santa Barbara Isl	9	3.2	5.7	0	17
CHLs	Western gull	Vandenberg	1	0.0	NA	0	0

Table S6. Summary statistics of analytes by species and site. Analyte units are ng/g fresh weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
Mercury	California least tern	Entire SCB	52	185.5	43.1	108	279
Mercury	California least tern	Batiquitos	4	165.2	33.1	117	187
Mercury	California least tern	Bolsa Chica	8	180.7	39.4	125	238
Mercury	California least tern	Chula Vista	4	158.1	40.0	112	209
Mercury	California least tern	D-Street Fill	5	182.8	32.9	130	206
Mercury	California least tern	LA Harbor	6	211.7	41.8	160	255
Mercury	California least tern	Lindbergh Field	5	224.0	39.2	176	270
Mercury	California least tern	Pt Mugu	9	172.6	40.7	115	246
Mercury	California least tern	Salt Works	3	156.5	51.9	108	211
Mercury	California least tern	TJ Estuary	6	187.2	41.5	130	229
Mercury	California least tern	Vandenberg	2	228.4	71.6	178	279
Mercury	Caspian tern	Entire SCB	15	504.3	221.2	295	1020
Mercury	Caspian tern	Bolsa Chica	5	610.8	237.3	295	877
Mercury	Caspian tern	Salt Works	10	451.0	204.1	326	1020
Mercury	Double-crested cormorant	Salt Works	8	71.1	56.0	31	192
Mercury	Western gull	Entire SCB	23	71.2	50.2	16	246
Mercury	Western gull	Anacapa Isl	4	27.7	9.3	16	35
Mercury	Western gull	NAS North Isl	8	61.2	37.1	18	126
Mercury	Western gull	Pismo Beach	1	82.5	NA	83	83
Mercury	Western gull	Santa Barbara Isl	9	78.9	27.7	50	141
Mercury	Western gull	Vandenberg	1	245.5	NA	246	246
Selenium	California least tern	Entire SCB	29	487.1	63.1	356	624
Selenium	California least tern	Batiquitos	4	549.2	67.2	474	624
Selenium	California least tern	Bolsa Chica	8	453.7	56.8	356	536
Selenium	California least tern	Chula Vista	0				
Selenium	California least tern	D-Street Fill	0				
Selenium	California least tern	LA Harbor	6	501.0	62.4	426	590
Selenium	California least tern	Lindbergh Field	0				
Selenium	California least tern	Pt Mugu	9	489.6	57.4	386	545
Selenium	California least tern	Salt Works	0				
Selenium	California least tern	TJ Estuary	0				
Selenium	California least tern	Vandenberg	2	443.3	6.1	439	448
Selenium	Caspian tern	Entire SCB	5	591.5	244.5	398	989
Selenium	Caspian tern	Bolsa Chica	5	591.5	244.5	398	989
Selenium	Caspian tern	Salt Works	0				
Selenium	Double-crested cormorant	Salt Works	0				
Selenium	Western gull	Entire SCB	15	470.5	79.5	329	636
Selenium	Western gull	Anacapa Isl	4	438.7	16.4	421	454
Selenium	Western gull	NAS North Isl	0				
Selenium	Western gull	Pismo Beach	1	423.0	NA	423	423
Selenium	Western gull	Santa Barbara Isl	9	505.7	79.2	403	636
Selenium	Western gull	Vandenberg	1	328.8	NA	329	329

Table S6. Summary statistics of analytes by species and site. Analyte units are ng/g fresh weight.

Analyte	Species	Site	N	Mean	SD	Min	Max
Arsenic	California least tern	Entire SCB	29	97.8	25.0	55	147
Arsenic	California least tern	Batiquitos	4	128.7	19.4	107	147
Arsenic	California least tern	Bolsa Chica	8	85.4	17.5	62	113
Arsenic	California least tern	Chula Vista	0				
Arsenic	California least tern	D-Street Fill	0				
Arsenic	California least tern	LA Harbor	6	114.2	21.7	80	142
Arsenic	California least tern	Lindbergh Field	0				
Arsenic	California least tern	Pt Mugu	9	84.5	18.9	55	107
Arsenic	California least tern	Salt Works	0				
Arsenic	California least tern	TJ Estuary	0				
Arsenic	California least tern	Vandenberg	2	95.5	25.6	77	114
Arsenic	Caspian tern	Entire SCB	5	82.7	25.1	45	105
Arsenic	Caspian tern	Bolsa Chica	5	82.7	25.1	45	105
Arsenic	Caspian tern	Salt Works	0				
Arsenic	Double-crested cormorant	Salt Works	0				
Arsenic	Western gull	Entire SCB	15	43.6	33.4	14	150
Arsenic	Western gull	Anacapa Isl	4	61.7	60.1	14	150
Arsenic	Western gull	NAS North Isl	0				
Arsenic	Western gull	Pismo Beach	1	33.7	NA	34	34
Arsenic	Western gull	Santa Barbara Isl	9	36.9	19.1	20	82
Arsenic	Western gull	Vandenberg	1	41.0	NA	41	41

Table S7. Metal inter-calibration results for reference material SRM 1946 Fish Tissue. All units are in ng/g ww.

Analyte	Required RL	Target Value	(+) 30% of Target	(-) 30% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
					Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Arsenic	20	277	360	194	269	280	288	512	536	513	340	344	334	337	336	364	279	303	288	290	280	280
Mercury	30	433	563	303	320	319	323	554	507	522	393	337	359	431	497	466	514	489	505	400	380	370
Selenium	100	491	638	344	408	365	355	430	430	520	601	607	576	670	658	723	521	508	515	530	490	500

Table S8. Chlorinated pesticides inter-calibration results for reference material SRM 1946 Fish Tissue. All units are in ng/g ww. ND = non-detect and empty = not reported.

Analyte	Target Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3		
				Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
4,4'-DDT	37.2	52.08	22.32	27.6	30.8	31.2	26.0	28.0	27.0	30.4	33.4	29.5
2,4'-DDT	22.3	31.22	13.38	11.3	12.3	12.5	14.0	19.0	13.0	18.6	21.5	15.7
4,4'-DDD	17.7	24.78	10.62	5.7	6.2	6.2	9.8	10.0	10.0	23.6	24.9	19.2
2,4'-DDD	2.20	3.08	1.32	0.4	0.5	0.5	1.2	1.3	1.2	ND	ND	ND
4,4'-DDE	373	522.2	223.8	354.9	342.1	350.8	310	300	300	399	492	460
2,4'-DDE	1.04	1.456	0.624	0.7	0.7	0.7	1.1	1.3	1.2	1.47	ND	1.11
4,4'-DDMU				1.5	1.6	1.7	3.7	4.3	3.9	3.05	4.11	4.21
alpha-Chlordane	32.5	45.5	19.5	31.9	35.0	37.7	27.0	27.0	22.0	32.5	31.2	31.3
gamma-Chlordane	8.36	11.704	5.016	5.2	5.7	6.4	6.8	8.3	5.9	4.38	8.34	10.3
cis-nonachlor	59.1	82.74	35.46	38.8	43.7	46.8	50.0	54.0	48.0	52.2	59.2	47.2
trans-nonachlor	99.6	139.44	59.76	92.6	90.4	97.5	75.0	70.0	75.0	120	114	112
oxychlordane	18.9	26.46	11.34	11.7	12.7	14.3	24.0	20.0	19.0	24.5	23.7	20.5
dieldrin	32.5	45.5	19.5	25.6	32.0	24.4	30.0	32.0	26.0	28.1	26.4	29
Total Passing				**9**	**9**	**9**	**10**	**10**	**9**	**9**	**9**	**11**

Table S9. PCB inter-calibration results for reference material SRM 1946 Fish Tissue. All units are in ng/g ww. ND = non-detect and empty = not reported.

Analyte	Target Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
				Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
PCB-18	0.84	1.176	0.504	ND	ND	ND				ND	ND	ND	0.415	0.391	0.353	0.35	0.35	0.29			
PCB-28	2.00	2.8	1.2	4.9	5.3	5.4	1.8	1.3	1.7	1.16	1.36	1.44	1.17	1.14	1.28	1.44	1.57	1.31	1.7	1.9	2
PCB-37										ND	ND	ND	ND	ND	0.0126	ND	ND	ND			
PCB-44	4.66	6.524	2.796	3.0	3.5	3.6	4.0	3.7	3.7	4.3	4.11	3.94	2.7	2.7	3.51	3.2	3.49	3.04	6.8	7.5	6.9
PCB-49	3.80	5.32	2.28	2.4	2.6	2.7	2.4	2.0	2.5	4	3.46	3.58	2.47	2.37	3.09	2.5	2.84	2.32	3	3.6	3.5
PCB-52	8.1	11.34	4.86	6.1	6.5	6.6	6.6	7.6	6.9	9.7	9.17	9.25	4.76	4.7	6.21	6.27	7.06	5.86	4.4	4.8	4.3
PCB-66	10.8	15.12	6.48	7.4	8.3	8.5	10.7	10.1	10.7	10.8	10.9	10.5	6.05	5.63	7.66	7.67	8.73	6.96	0	0	0
PCB-70	14.9	20.86	8.94	10.9	12.1	12.6	9.9	11.0	11.1	12.6	12.3	13.1	9.7	9.11	12.2	10.5	12.1	9.92	8.7	10	10
PCB-74	4.83	6.762	2.898	4.0	4.5	4.6	4.1	4.3	4.8	4.4	4.69	4.91	3.13	2.93	3.93	4.18	4.56	3.67	5.4	6.4	6.5
PCB-77	0.327	0.4578	0.1962	0.5	0.5	0.5	3.5	2.8	3.8	ND	ND	ND	0.201	0.202	0.257	0.35	0.41	0.45	6.6	7.8	6.5
PCB-81										ND	ND	ND	0.0669	0.0557	0.0613	ND	ND	ND			
PCB-82										ND	ND	ND	0.162	0.0798	0.145	ND	ND	ND			
PCB-87	9.4	13.16	5.64	9.0	9.6	9.7	8.9	7.6	7.6	9.6	9.86	8.45	9.15	9.02	8.3	8	8.63	7.69	8.4	11	9.8
PCB-92										ND	ND	ND	10.3	9.92	9.32	9.88	9.75	7.93			
PCB-99	25.6	35.84	15.36	21.6	23.6	23.6	28.3	24.8	24.6	26.9	24.8	27.3	27.7	26.7	25.1	25.6	25.4	20.9	25	29	30
PCB-101	34.6	48.44	20.76	35.9	36.9	37.9	32.5	33.7	34.1	35	36.2	33.6	39.5	37.7	35.6	37.7	38.1	34.1	30	36	31
PCB-105	19.9	27.86	11.94	20.1	22.1	22.2	18.7	19.3	19.4	17	16.9	18	17.3	16.8	18.2	19.6	20	16.9	14	18	17
PCB-110	22.8	31.92	13.68	20.4	22.0	22.3	23.8	22.0	23.5	22.7	19.5	20.4	26.4	25.7	24	23.2	23.9	20.3	20	22	23

Analyte	Target Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
PCB-114				ND	ND	ND				ND	ND	ND	1.16	1.12	1.23	ND	ND	ND	14	18	17
PCB-118	52.1	72.94	31.26	56.0	55.2	56.6	54.1	54.6	56.0	48.4	52.6	50.3	55.4	53.4	52.1	56.2	55.2	47.3	0	0	0
PCB-119										ND	ND	ND	1.22	1.15	1.11	ND	ND	ND			
PCB-123										ND	ND	ND	1.23	1.13	1.14	8.77	8.34	7.01			
PCB-126	0.380	0.532	0.228	0.7	0.8	0.7				ND	ND	ND	0.341	0.331	0.344	0.39	0.42	0.38	2.5	2.6	2.6
PCB-128	22.8	31.92	13.68	20.9	22.1	23.0	24.3	23.3	24.8	23.5	24.6	26.1	16.9	16.2	17.1	22.7	21	20.2	7.9	11	9
PCB-138	115	161	69	116.4	126.1	125.3	135.0	132.0	135.0	125	128	119	126	122	128	114	116	107	99	130	110
PCB-146	30.1	42.14	18.06	12.1	13.3	13.7				22.4	24.1	20.9	23.7	22.8	23.7	31.5	31.4	29.1	16	21	21
PCB-149				21.4	23.8	24.1	28.7	27.1	26.9	24.5	26.6	28.3	32	31.2	27.5	25.6	28.4	25.4	19	21	22
PCB-151				6.5	7.1	7.3	8.5	8.8	8.3	5.33	6.61	4.94	10.5	10.3	9.11	8.71	8.96	8.05			
PCB-153	170	238	102	193.5	210.2	202.2	176.5	169.6	169.2	162	179	168	163	154	163	193	184	167	90	110	97
PCB-156	9.52	13.328	5.712	8.4	9.1	9.2	9.4	9.4	9.5	8.31	7.25	9.16	7.52	7.27	7.68	8.67	8.24	7.71	4.3	6.6	5.9
PCB-157				2.2	2.4	2.5	3.7	2.8	3.6	4.1	3.5	3.4	2.13	2.1	2.19	2.62	2.35	2.44			
PCB-158	7.66	10.724	4.596	6.0	6.9	6.6				4.98	6.22	5.57	6.73	6.36	6.76	6.99	7	6.29	89	110	110
PCB-167							6.9	7.2	6.0	ND	ND	ND	4.56	4.51	4.65	5.21	4.9	4.81	7.9	11	9
PCB-168										ND	ND	ND	0.184	0.163	0.217						

Analyte	Target Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
PCB-169	0.106	0.1484	0.0636	ND	ND	ND	5.4	5.5	3.9	ND	ND	ND	0.0848	0.1	0.0701	ND	ND	ND			
PCB-170	25.2	35.28	15.12	31.7	33.7	35.2	30.7	33.5	31.5	24.3	22.6	27.3	26.4	25.5	24.6	24.1	25.2	21.8	16	17	14
PCB-177				13.5	14.6	14.9	13.2	12.5	12.1	14.2	10.6	12.2	14	13.7	13.4	12.5	13.3	12			
PCB-180	74.4	104.16	44.64	79.1	84.7	85.0	75.4	72.2	74.3	76.4	73.2	71.8	78.1	76.1	74.9	71.4	75.7	71.4	10	13	12
PCB-183	21.9	30.66	13.14				21.3	21.4	20.7	19.5	21.3	18.5	23.9	23.2	21.8	18.7	20.8	17.6	11	12	12
PCB-187	55.2	77.28	33.12	57.0	62.0	59.2	55.7	53.7	54.5	53.3	55.5	50.6	67.8	66	62.3	54.2	53.8	50.7	30	42	37
PCB-189				1.6	1.6	1.7	2.5	2.3	2.9	ND	ND	ND	1.42	1.38	1.34	ND	ND	ND			
PCB-194	13.0	18.2	7.8	13.2	14.2	14.1	13.5	12.8	12.2	13.7	12.4	14.1	13.7	13.1	13.1	13.9	15	13.5	4.6	6.4	5.5
PCB-201	2.83	3.962	1.698	2.0	2.2	2.1	3.6	3.1	2.7	2.55	2.89	3.1	3.2	3.1	2.89	16.4	19.8	16.5	4.1	4.9	5.5
PCB-206	5.40	7.56	3.24	5.1	5.6	5.7	7.1	7.5	5.9	6.4	6.33	6.74	4.79	4.67	4.77	5.96	6.13	5.72	0.13	0.6	0.55
Total Passing				**23**	**23**	**23**	**24**	**23**	**24**	**25**	**26**	**26**	**25**	**25**	**29**	**27**	**27**	**27**	**10**	**16**	**14**

Table S10. Metal inter-calibration results for the spiked egg homogenate. All units are in ng/g ww. Arsenic results are from the second round of analyses.

Analyte	Required RL	Spike Level	Mean Value	(+) 30% of Target	(-) 30% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
						Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Arsenic	20	60	76	98	53	104.6	86.7	92.7	69	68	65	61	56	58	88.819	103.23	105.455	69.3	70.1	64.3	62	75.3	59.8
Mercury	30	100	96	125	67	88	97	92	103	104	NA	92	90	91	99.6	98.8	99.1	118	121	114	75	77	80
Sele-nium	100	500	715	930	501	563	531	513	646	640	NA	836	828	877	779	833	849	772	829	830	640	620	570

Table S11. Chlorinated pesticide inter-calibration results for the spiked egg homogenate. All units are in ng/g ww. ND = non-detect and empty = not reported.

Analyte	Spike Level	Mean Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
					Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
4,4'-DDT	5	5.61	7.9	3.4	4.21	3.54		3.9	4.9	3.9	8.29	11.2	9.91	4	4.55	3.66	5.9	5.44	5.2	20	15	17
2,4'-DDT	5	7.86	11.0	4.7	7.83	6.51		9.7	10	8.9	5.83	8.15	7.14	7.64	8.11	6.92	9.75	9.22	9.27	7.7	5.3	5.6
4,4'-DDD	5	6.17	8.6	3.7	6.00	5.21		5.8	6.3	5.8	5.27	6.29	5.57	4.16	3.39	2.17	8.86	8.32	7.79	8.6	7.6	7.7
2,4'-DDD	5	6.19	8.7	3.7	3.17	2.38		5.5	6.2	6.1	5.3	6.87	5.22	5.98	6.94	5.8	8.41	8.39	8.93	7.5	6.3	6.3
4,4'-DDE	2100	1831.18	2564	1099	1984.00	1769.00		1700	1800	1800	1688	1987	1462	2400	2170	1970	2120	2350	2130	1500	1200	1100
2,4'-DDE	2	6.97	9.8	4.2	5.36	4.68		7.2	7.9	6.6	7.05	7.98	6.79	6.98	6.94	6.35	6.89	7.15	7.57	8.9	7.1	7.1
4,4'-DDMU	2	3.14	4.4	1.9	1.26	1.03		2.7	3.2	4.2	3.71	3.89	3.64	4.96	4.22	3.92	2.47	2.41	2.34	29	23	23
alpha-Chlordane	2	2.05	2.9	1.2	3.16	2.75		1.9	2.6	2.8	1.66	1.93	1.69	1.73	1.74	1.55	2.46	2.07	2.28	1.8	1.4	1.4
gamma-Chlordane	2	2.15	3.0	1.3	2.84	2.43		2.2	1.9	2.8	1.75	1.94	1.71	2.04	1.78	1.69	2.38	2.2	2.52	2.4	2	2
cis-nonachlor	20	13.22	18.5	7.9	11.59	9.38		12	13	13	8.49	13	10.4	15.2	15.4	13.3	15.2	15.1	17.7	16	13	13
trans-nonachlor	20	17.55	24.6	10.5	19.70	15.29		15	17	15	13.4	20	15	21.9	20	17.9	23	21.8	23.4	16	12	12
oxychlordane	20	21.62	30.3	13.0	20.74	18.06		15	16	19	25.2	29.8	25.4	23	20.8	19.5	24	26.1	27	22	18	18
dieldrin	20	21.03	29.4	12.6	20.96	17.76		18	19	18	14.2	20.1	15.1	19.7	19.9	18.2	31.8	31.4	34.4	21	19	19
Total Passing					**10**	**11**		**13**	**13**	**13**	**12**	**11**	**12**	**12**	**12**	**12**	**11**	**12**	**11**	**10**	**12**	**11**

Table S12. PCB inter-calibration results for the spiked egg homogenate. All units are in ng/g ww. ND = non-detect and empty = not reported.

Analyte	Spike Level	Mean Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
					Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
PCB-18	none	5.88	8.2	3.5	6.48	5.69		5.6	5.2	5.1	4.38	6.4	5.42	7.78	6.99	6.87	5.69	5.45	5.3			
PCB-28	none	6.77	9.5	4.1	1.00	ND					ND	ND	ND	ND	ND	ND	ND	ND	ND	9.4	8	8.7
PCB-37	none	0.03	0.0	0.0							ND	ND	ND	0.0279	ND	0.0317	ND	ND	ND			
PCB-44	5	13.68	19.1	8.2	16.79	14.70		12.1	12.5	11.4	11	12	8.41	14.1	12.9	17.5	16.6	16	16.5	14	13	13
PCB-49	5		0.0	0.0	ND	ND					ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-52	none	12.32	17.3	7.4	14.47	12.69		10.5	10.4	10.2	9.21	10.6	8.78	10.3	9.04	12.5	14.5	13.6	13.7	17	15	17
PCB-66	50	89.62	125.5	53.8	104.59	94.03		86.5	85.5	85.5	60.3	66.8	68.8	84.7	79	106	112	109	112	ND	ND	ND
PCB-70	5				ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-74	50				ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-77	5	42.02	58.8	25.2	ND	ND					ND	ND	ND	0.0329	0.0304	0.0381	ND	ND	ND	89	79	84
PCB-81	5										ND	ND	ND	ND	ND	ND	ND	ND	ND			

Analyte	Spike Level	Mean Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
PCB-82	100		0.0	0.0							ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB-87	none	0.06	0.1	0.0	ND	ND		ND	ND	ND	ND	ND	ND	0.067	0.0616	0.0596	ND	ND	ND	ND	ND	ND
PCB-92	5		0.0	0.0							ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB-99	100	0.02	0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	0.0255	0.0236	0.0186	ND	ND	ND	ND	ND	ND
PCB-101	100	0.29	0.4	0.2	ND	ND		ND	ND	ND	0.53	0.33	0.61	0.096	0.0874	0.0787	ND	ND	ND	ND	ND	ND
PCB-105	50				ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-110	50				ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-114	none		0.0	0.0	ND	ND					ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-118	100	0.04	0.1	0.0	ND	ND		ND	ND	ND	ND	ND	ND	0.0377	0.0383	0.0349	ND	ND	ND	ND	ND	ND
PCB-119	none		0.0	0.0							ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB-123	none	0.10	0.1	0.1							ND	ND	ND	0.107	0.102	0.0966	ND	ND	ND			
PCB-126	5		0.0	0.0	ND	ND					ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-128	100	1.03	1.4	0.6	1.02	1.04		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-138	500	0.63	0.9	0.4	ND	ND		ND	ND	ND	1.17	1.31	1.23	0.0268	0.0281	0.024	ND	ND	ND	ND	ND	ND
PCB-146	100	0.09	0.1	0.1	ND	ND					ND	ND	ND	0.0985	0.0836	0.0939	ND	ND	ND	ND	ND	ND

Ana- lyte	Spik e Level	Mean Valu e	(+) 40% of Tar- get	(-) 40% of Tar- get	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
PCB -149	50	0.06	0.1	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	0.0626	ND	ND	ND	ND	ND	ND
PCB -151	none		0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB -153	500	12.84	18.0	7.7	18.75	15.11		11.7	11.4	11	7.88	14.8	11.2	16.3	13.8	14.8	16.4	15.7	15.3	8.6	7.5	8
PCB -156	50		0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB -157	5	0.54	0.8	0.3	1.29	1.24		ND	ND	ND	ND	ND	ND	0.065	0.057	0.0582	ND	ND	ND			
PCB -158	none	0.03	0.0	0.0	ND	ND					ND	ND	ND	0.0365	0.034	0.0329	ND	ND	ND	ND	ND	ND
PCB -167	none		0.0	0.0				ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB -168	none	10.72	15.0	6.4							ND	ND	ND	ND	ND	ND	16.4	15.7	15.3	6.6	4.5	5.8
PCB -169	5		0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB -170	50	12.38	17.3	7.4	18.23	14.76		10.6	10.1	9.7	10.2	8.85	7.87	20.1	16.9	16.7	14.4	14.3	13.3	9.5	7.9	7.1
PCB -177	5		0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB -180	500	12.27	17.2	7.4	17.85	14.52		12.4	11.4	10.6	7.69	9.14	8.24	19.3	15.9	15.3	14.9	14.7	14.6	8.1	5.1	8.9
PCB -183	none	14.66	20.5	8.8				12	11.75	11.85	9.37	11.7	8.37	23.3	18.8	18.2	17.6	19.8	19.2	13	12	13
PCB -187	50	13.17	18.4	7.9	17.16	14.13		12.5	10.9	10.5	12.8	12.1	11.1	21.4	17.4	17.1	16.4	15.5	15.6	7.4	4.7	7.2
PCB -189	none		0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND			
PCB -194	50	0.17	0.2	0.1	ND	ND		ND	ND	ND	ND	ND	ND	0.192	0.156	0.162	ND	ND	ND	ND	ND	ND
PCB -201	none		0.0	0.0	ND	ND		ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

Analyte	Spike Level	Mean Value	(+) 40% of Target	(-) 40% of Target	Lab 1			Lab 2			Lab 3			Lab 4			Lab 5			Lab 6		
PCB -206	5	11.59	16.2	7.0	18.93	16.00		12.4	10.2	9.7	5.86	6	6.6	17.7	13.5	14.2	17.3	18.8	20.5	4.3	2.4	2.6

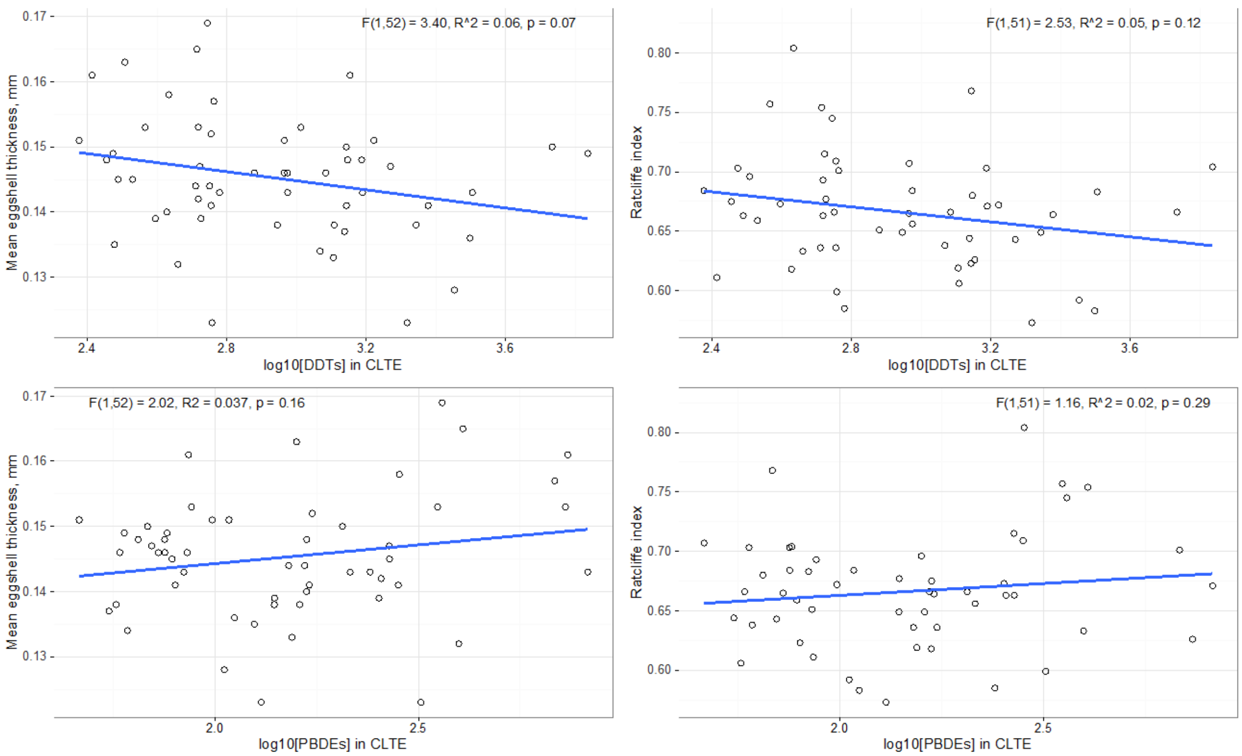

Figure S1. Relationships between eggshell thickness and toxicant concentrations in California least tern.

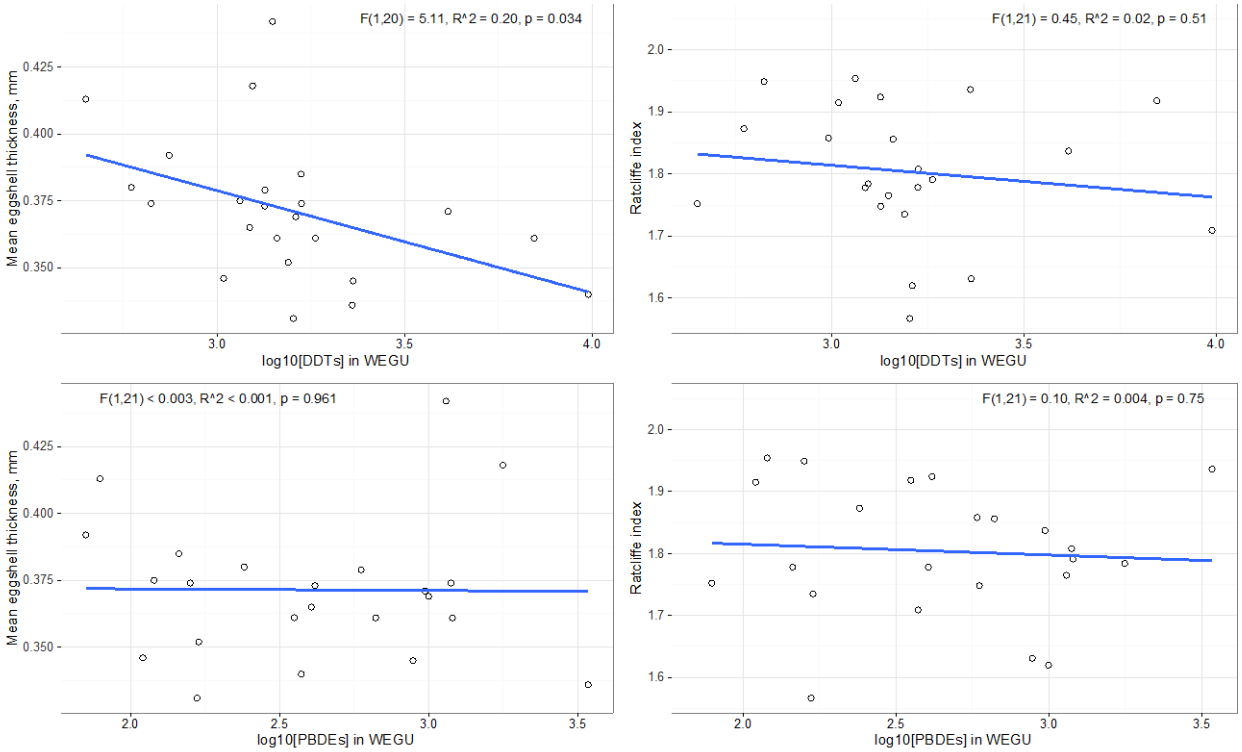

Figure S2. Relationships between eggshell thickness and toxicant concentrations in western gull.