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Terrestrial Scavenging of Marine Mammals: Cross-Ecosystem Contaminant Transfer and Potential Risks to Endangered California Condors (*Gymnogyps californianus*)

Carolyn M. Kurle,[†] Victoria J. Bakker,[‡] Holly Copeland,[§] Joe Burnett,[∥] Jennie Jones Scherbinski,[⊥] Joseph Brandt,[#] and Myra E. Finkelstein[*,@]

[†]Division of Biological Sciences, Ecology, Behavior, and Evolution Section, University of California, San Diego, La Jolla, California 92093, United States

[‡]Department of Ecology, Montana State University, Bozeman, Montana 59717-3460, United States

[§]The Nature Conservancy, 258 Main Street, Lander, Wyoming 82520, United States

[∥]Ventana Wildlife Society, 19045 Portola Drive, Suite F-1, Salinas, California 93908, United States

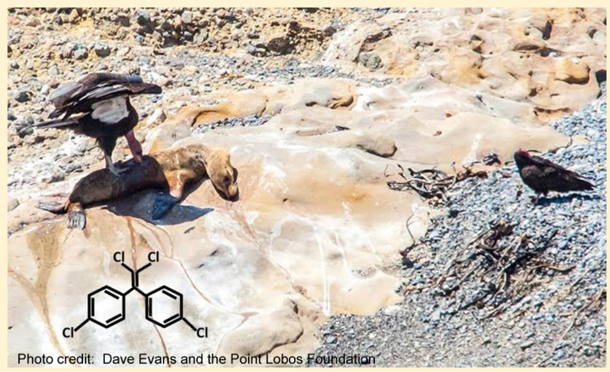
[⊥]Pinnacles National Park, National Park Service, 5000 Highway 146, Paicines, California 95043, United States

[#]United States Fish and Wildlife Service, 2493 Portola Road, Suite B, Ventura, California 93003, United States

[@]Department of Microbiology and Environmental Toxicology, University of California, Santa Cruz, California 95064, United States

Supporting Information

ABSTRACT: The critically endangered California condor (*Gymnogyps californianus*) has relied intermittently on dead-stranded marine mammals since the Pleistocene, and this food source is considered important for their current recovery. However, contemporary marine mammals contain persistent organic pollutants that could threaten condor health. We used stable carbon and nitrogen isotope, contaminant, and behavioral data in coastal versus noncoastal condors to quantify contaminant transfer from marine mammals and created simulation models to predict the risk of reproductive impairment for condors from exposure to DDE (*p,p′*-DDE), a major metabolite of the chlorinated pesticide DDT. Coastal condors had higher whole blood isotope values and mean concentrations of contaminants associated with marine mammals, including mercury (whole blood), sum chlorinated pesticides (comprised of ∼95% DDE) (plasma), sum polychlorinated biphenyls (PCBs) (plasma), and sum polybrominated diphenyl ethers (PBDEs) (plasma), 12−100-fold greater than those of noncoastal condors. The mean plasma DDE concentration for coastal condors was 500 ± 670 (standard deviation) ($n = 22$) versus 24 ± 24 (standard deviation) ($n = 8$) ng/g of wet weight for noncoastal condors, and simulations predicted ∼40% of breeding-age coastal condors have DDE levels associated with eggshell thinning in other avian species. Our analyses demonstrate potentially harmful levels of marine contaminant transfer to California condors, which could hinder the recovery of this terrestrial species.



■ INTRODUCTION

Dead-stranded marine vertebrates are a significant nutrient source for terrestrial consumers, and marine-subsidized species exhibit increased population sizes[1,2] and higher rates of survival when terrestrial foods are scarce.[3] Many avian scavengers feed on marine carcasses along the Pacific coast of North America,[4] a behavior that may have prevented the extirpation of California condors (*Gymnogyps californianus*) after the Pleistocene terrestrial megafauna disappeared.[3,5] However, marine mammal carcasses can contain high levels of persistent organic pollutants (POPs) such as DDE (*p,p′*-DDE, a major metabolite of the chlorinated pesticide DDT) and polychlorinated biphenyls (PCBs).[6,7] Thus, the nutritional benefits of marine scavenging may be offset by increased risks of contaminant-related health effects, which is of particular concern for endangered species such as the California condor.[4,8]

Nearly extinct in the 1980s,[8] the condor population has increased through intensive recovery efforts, surpassing 400 birds by 2016, approximately half of which are in the wild and associated with release sites in California, Arizona, and Baja California, Mexico. The condor population is not self-sustaining, and poisoning from feeding on carcasses contaminated with lead-based ammunition is the primary threat

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preventing its recovery.[9,10] Condors that feed along the coast are thought to be at lower risk of lead poisoning, and we have found that coastal behavior is positively associated with a condor's survival.[11] However, eggshell thinning and reduced hatching success, attributed to DDE exposure from feeding on beach-cast marine mammals, particularly California sea lions (*Zalophus californianus*; CASLs), were recently identified in coastal breeding condors and could prove to be an additional threat to condor population health.[12] Condor lead exposure is well-documented,[9,13−15] but data on exposure to DDE and other POPs are limited, despite their potential to affect condor recovery.[8]

The majority (>95%) of the U.S. population of CASLs breeds on the Channel Islands off California.[16] Nearby waters and sediments were contaminated with PCBs and DDTs discharged by several companies from the 1940s to 1970s, with Montrose Chemical Corp. expelling over 2000 t of DDTs.[17] Elevated contaminant levels persist around these islands,[18,19] and CASLs continue to exhibit high levels of DDTs, the majority of which is DDE.[7,20]

To investigate the potential risk of marine mammal feeding behavior to condor health, we (a) measured the stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope ratios in condor whole blood samples and muscle from potential diet items to estimate the degree to which condors fed on marine mammals, (b) evaluated relationships between marine-associated contaminant (e.g., PCBs and DDE) concentrations in condors and their observed feeding and spatial behavior, and (c) predicted current flock-wide DDE exposure levels and potential reproductive risks using simulation models. Our findings inform future management of condors by providing a comprehensive assessment of contaminant transfer from feeding on dead-stranded marine mammals while highlighting the broad influence of persistent pollutants in the marine environment, including cross-ecosystem risks to terrestrial species.

METHODS

Study Area and Species. All condors are assigned an ID (studbook number) and are of known sex because of their intensive management as an endangered species. We studied free-flying condors associated with two flocks: southern California ("noncoastal"), managed by the U.S. Fish and Wildlife Service Hopper Mountain National Wildlife Complex, and central California ("coastal"), managed by the Ventana Wildlife Society and Pinnacles National Park. The latter moves between the Big Sur coast and inland regions, whereas the former typically remains inland. Personnel maintain set locations where terrestrial animal carcasses are placed ("proffered feeding stations") to facilitate monitoring. Coastal condors are observed feeding on dead-stranded marine mammals along Big Sur,[12] whereas southern, noncoastal condors are not considered to feed on marine mammals. Ranges of coastal condors during the study period (2009−2013) overlap with our dead-stranded marine mammal sampling locations and are distinct from noncoastal condor ranges (Figure S1).

Sample Collection. Condor blood samples were collected opportunistically during routine health monitoring from 2009 to 2012, and condors sampled appeared to be in good body condition. All samples were used for stable isotope analysis, and a subset of these were analyzed for contaminants (Table S1). Blubber samples (∼5 g) for contaminant analysis were collected from dead-stranded marine mammals in Monterey County, California, from 2008 to 2012. Muscle samples for stable isotope analysis were collected from dead-stranded marine mammals and wild, nonproffered and domestic, proffered terrestrial carcasses in central and southern California from 2010 to 2012. Marine mammal blubber (n = 17 organochlorines; n = 7 PBDEs) and condor plasma samples were analyzed for organochlorines (n = 33) and PBDEs (n = 13); condor whole blood samples (n = 68) were analyzed for total mercury, and diet item muscle (n = 63) and condor whole blood samples (n = 83) were analyzed for stable isotope ratios (Tables S1−S3).

Sample Analysis (see also the Supporting Information). Plasma and blubber samples were analyzed for 54 congeners of PCBs, 26 chlorinated pesticides, including the seven metabolites of DDT, and 27 congeners of PBDEs (Table S4) at the California Department of Fish and Wildlife Water Pollution Control Laboratory. Condor whole blood was analyzed for total mercury by Eurofins Frontier Global Sciences, Inc. (Bothell, WA), on a Tekran 2600 Flow Injection Mercury System (FIMS). Condor whole blood and diet item muscle samples were analyzed for their $\delta^{13}C$ and $\delta^{15}N$ values using a Carlo Erba CE1108 elemental analyzer interfaced via a CONFLO III device to a Thermo-Electron Delta Plus XP mass spectrometer at the Stable Isotope Laboratory at the Department of Earth and Marine Sciences of the University of California (Santa Cruz, CA).

Condor Daily Observational Data for the Coastal Flock. Condors were monitored on a near-daily basis via visual observation at proffered feeding stations or elsewhere, aided by detection of signals from radio (VHF) and/or GPS transmitters. Daily observational data consisted of 31985 records of birds, documenting sightings in coastal Big Sur, inland in Pinnacles, and at proffered feeding stations. Observations of condors feeding on dead-stranded marine mammals in Big Sur were also made opportunistically from 1999 to 2013.

Spatial Analysis. We quantified space use with hourly GPS (PTT-100 50 g Solar Patagial Argos/GPS; Microwave Telemetry, Inc.) locations from January 1, 2003, to July 31, 2013, 05:00−20:00 PST, equaling 867498 locations from 90 unique condors. We summarized geographic ranges for coastal and noncoastal flocks using 99% fixed kernel density estimates from 2009 to 2013, concurrent with the time period of condor tissue sample collection (Figure S1), using ArcMET (Movement Ecology Tools for ArcGIS).[21] To investigate fine-scale patterns of coastal foraging, we ran a separate 99% fixed kernel density estimate for all locations within 1.5 km of the coast for the full data set (55012 locations from 37 unique condors), binning results into equal-area quantiles.

Statistical Analysis. *Coastal versus Noncoastal Condors.* Statistical tests of contaminant and stable isotope data were performed in R (version 3.0.2, R Core Team, 2013). Stable carbon and nitrogen isotope turnover from blood in birds has been shown to be complete at ∼30 days,[22−24] whereas methylmercury concentrations in avian blood decrease by ∼90% within 30 days of exposure.[25] We considered blood samples from the same individual independent as all samples were collected at intervals greater than or equal to approximately one year and no condors had more than one sample taken for organochlorine or PBDE analysis (Table S1). Only quantifiable compounds were used for determining the sum chlorinated pesticides, sum PCBs, or sum PBDEs; if all compounds within a group (e.g., PCBs) were not quantifiable,

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half the detection limit of a single compound was used as follows: p,p'-DDE was used for sum chlorinated pesticides, PCB 153 for sum PCBs, and PBDE-47 for sum PBDEs (Supporting Information). All samples had quantifiable total mercury concentrations. Variables were transformed if normality and homoscedasticity were improved, and nonparametric equivalents were utilized when data did not meet assumptions for parametric analyses.

Diet of Coastal Condors. MixSIAR, a Bayesian stable isotope mixing model incorporating discrimination factors for condors on known diets[26] and the mean [±standard deviation (SD)] $\delta^{13}C$ and $\delta^{15}N$ values from condors, marine mammals, and terrestrial proffered and nonproffered mammals, was used to estimate proportions of condor diet composed of marine versus terrestrial proffered versus terrestrial nonproffered items.

Behavior Affecting Contaminant Exposure. We assessed whether mercury, PCBs, PBDEs, and DDE levels were predicted by condor foraging behavior, focusing on DDE because it is the most abundant organochlorine in CASLs[7] and associated with reproductive failure in avian species.[27] We considered the cumulative number of observations of condors feeding on marine carcasses, overall and in one and three year windows preceding sample collection. Detecting nonproffered feeding along a wide coastal area is inherently difficult, so to minimize the influence of missing data, we also considered the cumulative number of years an individual was a marine mammal feeder prior to and inclusive of sampling year and counted an individual as a marine mammal feeder if it was observed feeding on marine mammals at least once in the year. Because gray whales have DDE levels substantially lower than those of CASLs (this study and ref 7) and condors were observed feeding on individual gray whales for several months, we also considered observations of marine mammal feeding excluding gray whales. We fit contaminant data to these marine feeding observations using linear regression models (Supporting Information), transforming response and predictor variables to improve normality and homoscedasticity and standardizing continuous predictor variables to facilitate model convergence. Model selection was guided by AICc.

We used estimated relationships between observed marine mammal feeding and measured DDE to simulate DDE levels for all condors in the coastal flock based on observed marine mammal feeding behavior from July 31, 2001, to July 31, 2013, incorporating unexplained variance due to residual SE, and summarized results from 1000 replicate runs (Matlab version 8.1) (Supporting Information). To investigate the possibility that DDE is affecting reproduction, for each simulated run we tracked the proportion of the population that, using a plasma DDE to egg DDE conversion factor reported for other raptors,[28] exceeded two DDE thresholds approximately equal to egg concentrations of 5000 and 15000 ng/g of wet weight. The lower DDE threshold is associated with 10 and 20% eggshell thinning in bald eagles[28] and condors,[29] respectively, whereas bald eagles approached 100% reproductive failure at the upper DDE threshold[28] (Supporting Information).

RESULTS

$\delta^{13}C$ and $\delta^{15}N$ and Contaminant Values. We found no differences in the $\delta^{13}C$ ($t = 1.99$; $p = 0.77$) and $\delta^{15}N$ ($t = 1.99$; $p = 0.41$) values between sexes, so we combined sexes for analyses. Coastal condors ($n = 65$) had mean (±SD) $\delta^{13}C$ (−22.1 ± 1.2‰) and $\delta^{15}N$ (9.9 ± 1.2‰) values higher than those of noncoastal condors ($n = 18$; −23.2 ± 1.2 and 8.0 ± 0.8‰, respectively), and the mean $\delta^{13}C$ and $\delta^{15}N$ values from marine mammals ($n = 17$; −17.3 ± 0.8 and 17.1 ± 1.0‰, respectively) were also higher than those from all terrestrial animals ($n = 46$; −23.7 ± 2.3 and 6.4 ± 1.5‰, respectively) (Figure 1 and Table S5), indicating the diet of coastal condors

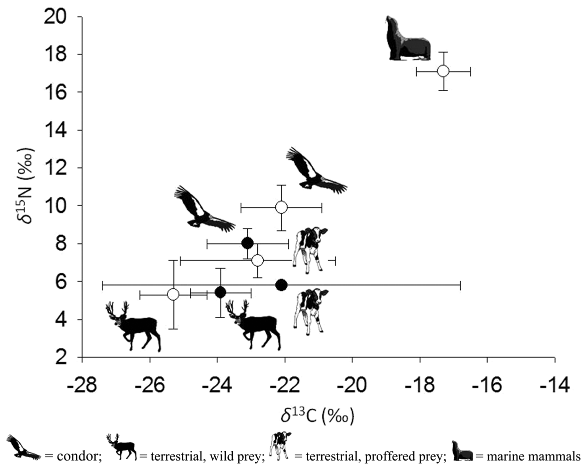

Figure 1. Mean (±SD) stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope values from whole blood from condors in the central California, coastal (○) flock ($n = 65$) and the southern California, noncoastal (●) flock ($n = 18$) and from coastal and noncoastal condor prey. See Table S5 for values.

is partially marine-derived. The estimated coastal condor diets (means) for individual birds ranged from 8 to 52% marine mammals, from 26 to 53% proffered terrestrial items, and from 16 to 63% nonproffered terrestrial items (Table S6), whereas those of noncoastal condors were 28−40% proffered and 44−68% nonproffered terrestrial items.

Coastal condors had mean concentrations of mercury, chlorinated pesticides, PCBs, and PBDEs 12−100-fold greater than those of noncoastal condors (all $p < 0.001$; Mann−Whitney U-tests) (Figure 2 and Table S7). Captive-raised condors sampled ($n = 3$) before their wild release had very low contaminant concentrations [mercury = 1.1 ± 0.3 ng/mL of whole blood; all plasma-chlorinated pesticides and PCBs were below quantification limits (Tables S7−S9)]. PBDEs were not measured in prerelease birds (Table S1). In coastal condors, methylmercury comprised the majority of total mercury measured in whole blood [99 ± 21%; $n = 5$ (Figure S2)], whereas DDE accounted for >90% of sum chlorinated pesticides, with *trans*-nonachlor being the second most abundant chlorinated pesticide detected (∼1−4% of the sum) (Table S9). PCB153 was a predominant PCB congener, accounting for 10−48% of sum PCBs, whereas PBDE-47 was the most abundant PBDE congener detected (Table S9). We found strong positive correlations among PBDEs, chlorinated pesticides, and PCBs within individual condors ($r \geq 0.951$, $p \ll 0.0001$, and $n \geq 10$) as well as marine mammals ($r \geq 0.911$, $p < 0.0006$, $n \geq 7$) (all Pearson correlations on ln-transformed data).

We found no differences between plasma lipid in coastal versus noncoastal condors (%lipid = 0.62 ± 0.18, with $n = 22$, vs %lipid = 0.56 ± 0.15, with $n = 8$; $p = 0.56$; Mann−Whitney U-test) or between years sampled ($p = 0.70$; Kruskal−Wallis), indicating the differences observed in organochlorine or PBDE concentrations were not due to the amount of lipid in a condor's plasma sample. We also found no evidence that males

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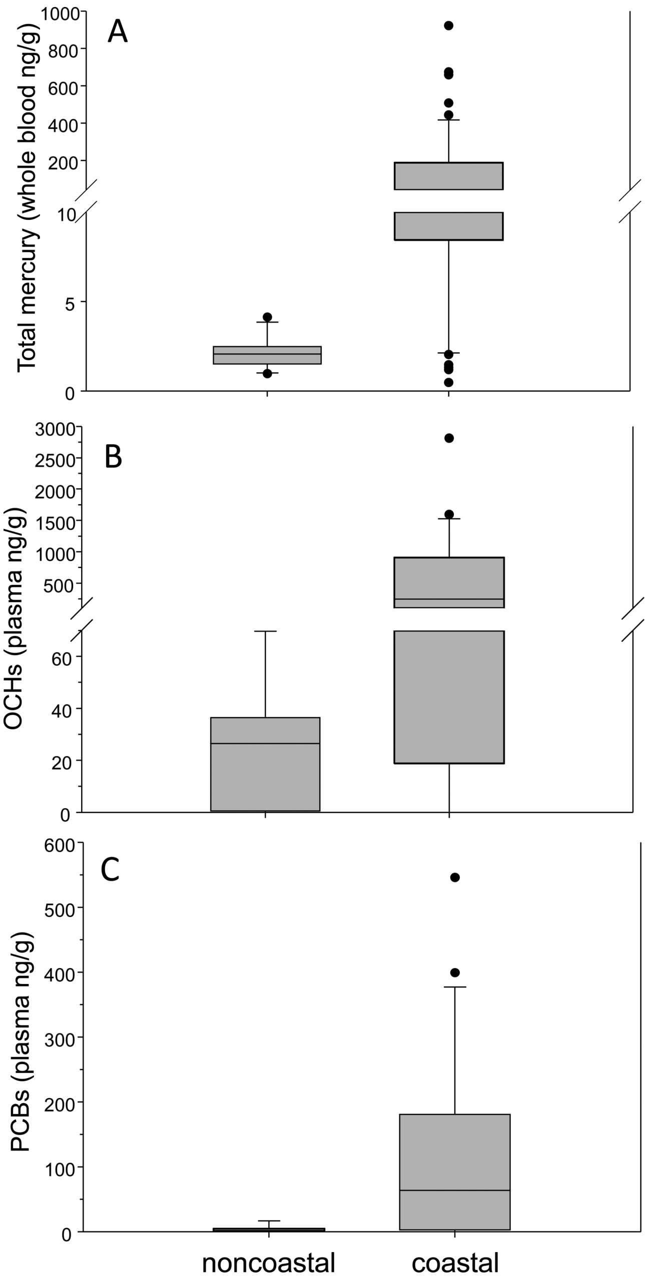

Figure 2. Contaminants measured in central (coastal) vs southern (noncoastal) California condors: (A) total mercury (n = 54 vs 11), (B) sum chlorinated pesticides (OCHs) (n = 22 vs 8), and (C) PCBs (n = 22 vs 8). Methylmercury comprised the vast majority of total mercury measured [mean ± SD = 99 ± 21%; n = 5 (Figure S2)]. Boxes indicate median, upper and lower 75th and 25th percentiles; whiskers represent 10th and 90th percentiles, and dots represent values outside the 10th and 90th percentiles (see also Tables S7–S9).

and females had different contaminant levels (all p > 0.285; Mann–Whitney U-tests).

Marine Mammal Feeding and Variation in Coastal Condor Contaminant Levels. Coastal condors exhibited large variations in contaminant levels with mercury concentrations ranging from 0.5 to 922 ng/g (whole blood; n = 54) (Table S8) and DDE concentrations from below the detection limit to 2680 ng/g (plasma; n = 22) (Table 1, Figure 2, and Table S9). We found significant positive correlations between mercury concentrations and δ^{13}C and δ^{15}N values (r = 0.329, p = 0.015, and n = 54; r = 0.652, p < 0.0001, and n = 54) and for PBDE concentrations and δ^{13}C values, but not δ^{15}N values (r = 0.816, p = 0.004, and n = 10; r = 0.374, p = 0.287, and n = 10) (all ln-transformed data). DDE and PCB concentrations were not related to δ^{13}C and δ^{15}N values (p > 0.26; n = 22).

Condors were observed feeding opportunistically on a range of marine species, including gray whales, harbor seals, and even one record of a brown pelican, but the large majority of records were CASLs. Of 1007 observations of individual condors feeding on marine carcasses gathered from 217 observation days between 1999 and 2013, most observations involved feeding on CASLs (66%), followed by gray whales (27%), unidentified pinnipeds (6%), and other marine vertebrates (e.g., harbor seal, Risso's dolphin, etc., <1%) (ref 12 and personal observations of J. Burnett).

Mercury concentrations were higher for birds observed feeding on marine mammals in the same year their blood sample was taken (CurrentYrFedMM) (n = 21) than for those not observed feeding on marine mammals (n = 33) (Mann–Whitney U-test; $p \ll 0.001$) (Figure 3), and this variable explained more variation (adjusted R^2 = 0.570) in ln mercury concentrations than lifetime cumulative marine mammal feeding behavior (ln YrsFedMM) (adjusted R^2 = 0.291; all df = 53, and all $p \ll 0.001$). In contrast, DDE, PCB, and PBDE levels were more strongly related to lifetime cumulative marine mammal feeding behavior (ln YrsFedMM) than to marine mammal feeding in the year of sample (CurrentYrFedMM) (ln DDE ~ CurrentYrFedMM, adjusted R^2 = 0.0, ln DDE ~ ln YrsFedMM, adjusted R^2 = 0.839, $p \ll 0.0001$, ln PCB ~ CurrentYrFedMM, adjusted R^2 = 0.0, ln PCB ~ ln YrsFedMM, adjusted R^2 = 0.813, $p \ll 0.0001$, all df = 20, ln PBDE ~ CurrentYrFedMM, adjusted R^2 = 0.0, p = 0.986, ln PBDE ~ ln YrsFedMM, adjusted R^2 = 0.664, p = 0.002, df = 8) (Figure 4). As such, the number of years observed feeding on marine mammals was a highly significant predictor of DDE levels in coastal condors (Table S10):

$$\ln \mathrm{DDE} = 1.894 + 2.698 \ln \mathrm{YrsFedMM} \quad (1)$$

Relationships were qualitatively similar when noncoastal and coastal flocks were combined or if gray whales were included in the ln YrsFedMM variable and currentYrFedMM variable (Figure S3).

Simulated DDE Levels of Coastal Condors. Simulated DDE levels rose steadily in coastal condors that continued to feed on marine mammals, and by 2013, 40% of breeding-age birds were predicted to exceed levels associated with eggshell thinning and almost 20% of breeding-age birds were predicted to exceed levels associated with nest failure in eagles[30] (Figure 5). Predictions indicate a leveling off of the proportion of birds exceeding these thresholds; however, this result is influenced by a growing number of new individuals, including captive-bred releases, entering the breeding population annually as the central California flock has increased from ~20 birds in 2001 to ~70 birds in 2013. The annual proportions of sampled individuals exceeding thresholds are generally consistent with predictions (Figure 5), especially given that sample sizes were small (n = 4–8 per year) and individuals were not selected randomly with respect to feeding behavior.

DISCUSSION

The successful recovery of California condors will require the reduction of multiple threats, the greatest being the incidence of lead poisoning.[8,9] Condors that spend more time on the

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Table 1. DDE (p,p'-DDE), Sum Chlorinated Pesticides (OCHs), Sum PCBs, and Sum PBDEs for Coastal Condors[a]

condor ID	gender	sample date	%lipid	DDE	sum OCHs	sum PCBs	sum PBDEs
112	female	4/12/2009	0.37	19	19	9	NA
199	male	5/17/2012	0.65	1010	1060	326	NA
204	male	5/17/2012	0.54	867	898	546	NA
219	male	5/17/2012	0.54	2680	2810	399	NA
294	female	5/24/2011	0.74	1320	1370	257	287
298	female	5/24/2011	0.66	918	952	157	128
317	female	5/12/2011	0.30	287	295	68	NA
318	male	4/12/2009	0.79	1520	1600	289	297
330	male	4/12/2009	0.73	179	199	60	50
335	male	5/24/2011	0.79	700	740	156	88
340	male	4/12/2009	0.83	169	188	61	48
345	male	5/24/2011	0.60	330	348	68	67
351	male	5/24/2011	0.69	436	455	133	NA
401	male	5/11/2010	0.80	96	102	24	15
411	male	5/11/2010	0.49	ND	ND	ND	NA
421	male	4/12/2009	0.61	ND	ND	ND	NA
431	male	5/11/2010	0.56	24	24	ND	NA
451	male	5/24/2011	0.14	ND	ND	ND	NA
470	male	5/11/2010	0.81	370	393	124	80
525	male	9/18/2012	0.60	ND	ND	8	NA
538	female	5/24/2011	0.69	19	20	3	3
566	male	9/18/2012	0.76	17	17	ND	NA

[a]Values reported in nanograms per gram of wet weight plasma. ND indicates a value below the limit of quantification (Table S9). NA indicates a value that was not measured.

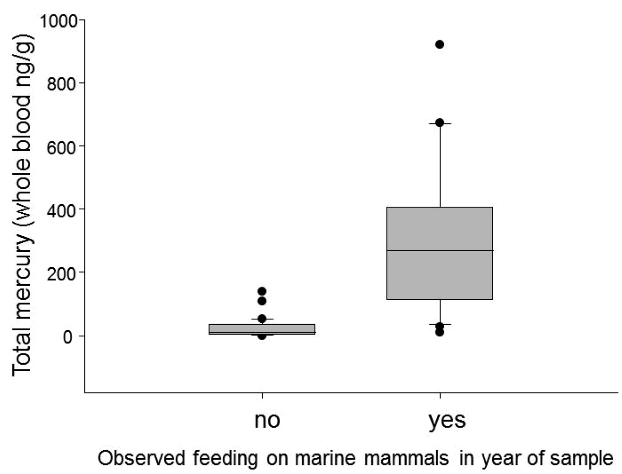

Figure 3. Total mercury whole blood concentrations from central California (coastal) condors observed (n = 21) and not observed (n = 33) feeding on dead-stranded marine mammals in the same year their blood sample was taken.

coast, where they are known to forage on marine mammals,[12] have a higher probability of survival, presumably because of a lower risk of lead poisoning.[11] However, contaminant transfer from feeding on marine mammals is an additional threat as marine-foraging coastal condors had potentially harmful concentrations of DDE (Table 1), and Burnett et al.[12] documented that condors breeding along the Big Sur coast of California had a hatching success rate 20−40% lower than that of noncoastal breeders.

Our study aimed to quantify the degree to which condors are using marine mammals as a food resource as well as to predict potential reproductive risk from this behavior. We found the diets of coastal condors included amounts of marine-derived carbon[31,32] and higher-trophic level, marine predators[32,33] greater than those of noncoastal condors based on the δ^{13}C and δ^{15}N values. Organochlorines and mercury accumulate in higher concentrations with increasing marine trophic level,[34] and accordingly, we observed higher concentrations of contaminants in coastal condors. Further, methylmercury accounted for most of the total mercury measured, consistent with the accumulation of mercury from a marine-based food web.[35,36]

The strong positive linear relationships we found between mercury concentrations and the δ^{13}C and δ^{15}N values from condors support the hypothesis that condor feeding behavior affects their contaminant exposure. We found no significant relationships between concentrations of DDE and PCBs and δ^{13}C and δ^{15}N values, potentially because of the different time scales these values represent. The whole blood δ^{13}C and δ^{15}N values reflect ingestion of diet items one to two months prior to blood collection,[22−24,37] and methylmercury blood concentrations also reflect shorter time scales, decreasing by ∼90% within a month of exposure.[25] In contrast, DDE is highly persistent, with plasma DDE concentrations establishing equilibrium with whole body lipids within days, and exhibiting a half-life of approximately one to two years in the body.[38,39]

The positive association between PBDE concentrations and the δ^{13}C, but not δ^{15}N, values, suggests marine signature influences PBDE levels more than trophic level, similar to patterns Elliot et al.[40] reported for bald eagles. However, even though chlorinated pesticides, PCBs, and PBDEs were highly correlated within individuals ($r \geq 0.95$, $n \geq 10$), as expected,[41] we did not find positive associations between the δ^{13}C values and organochlorines within the coastal flock. Coastal condors feed on a mixture of terrestrial, proffered, and marine diet items with varying frequency (Table S6). Associations between organochlorines and marine-based carbon might be partially masked by a condor's variable feeding behavior, especially in

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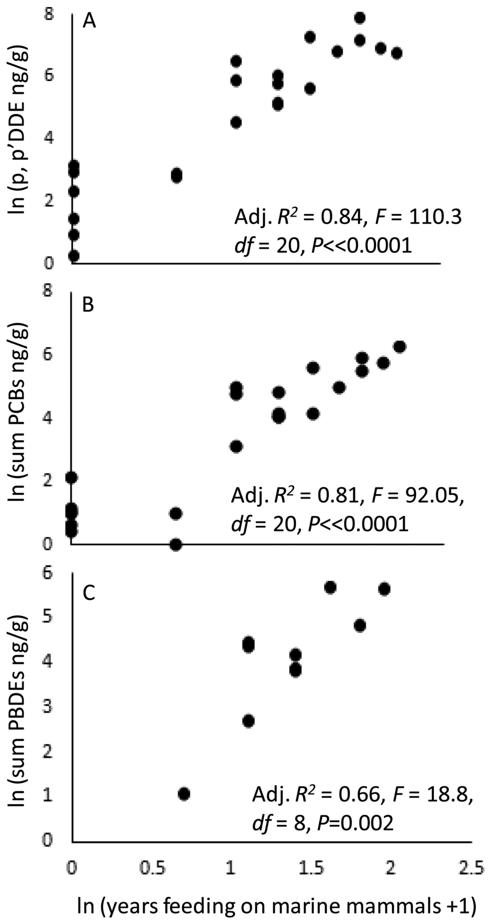

Figure 4. Cumulative number of years that central California (coastal) condors were observed feeding on marine mammals (excluding gray whales) prior to sample collection vs (A) DDE, (B) sum PCBs, and (C) PBDEs in their plasma.

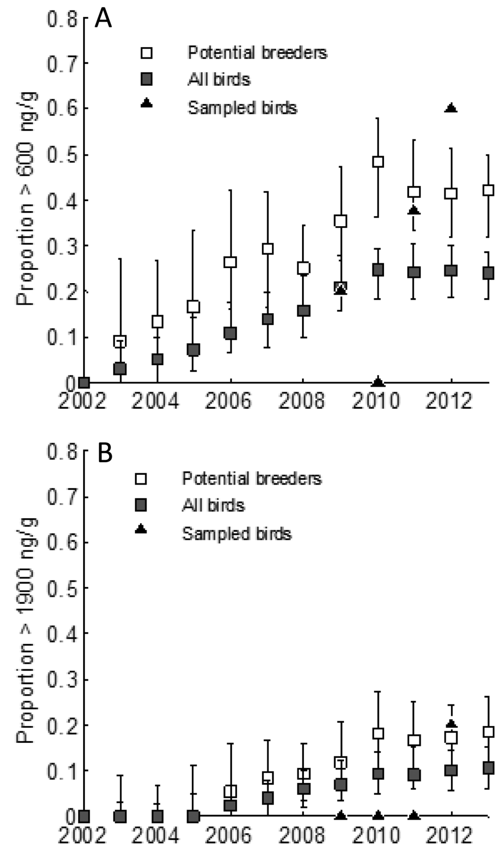

Figure 5. Proportion of the central California (coastal) flock of condors predicted to have exceeded two thresholds for DDE exposure through 2013, based on observed feeding behavior. Thresholds used were plasma DDE concentrations of (A) 600 ng/g and (B) 1900 ng/g. Filled squares show predictions for all birds, including chicks, empty squares predictions for potential breeders (≥5 years old), and triangles data for measured plasma DDE in individuals sampled for this study in 2009 (n = 5), 2010 (n = 4), 2011 (n = 8), and 2012 (n = 5) (see the Supporting Information).

light of the different sample sizes for PBDEs and organochlorines (Table S1).

The patterns of association between contaminant levels and observed condor feeding behavior support the time scales of exposure implied by the stable isotope analyses. We found that mercury levels increased with marine mammal feeding on shorter time scales (a year or less) (Figure 3), whereas DDE, PCB, and PBDE levels increased with cumulative long-term feeding behavior (Figure 4). Thus, the variation in marine-associated contaminant concentrations observed within coastal condors is related to individual variation in feeding behavior, similar to observations from Arctic foxes eating both marine and terrestrial foods.[42]

The source of DDE exposure to condors is clearly linked to foraging on marine mammals; however, the predominant sources of DDE are unconfirmed. Condors were observed to feed on CASLs at a rate higher than the rate at which they feed on other marine mammals, which was likely a function of increased access to CASLs dead-stranded in Monterey County. Marine mammal stranding data from 1997, when condors were first released in the Big Sur area, through 2008 (the end of the available data) recorded 1180 marine mammal carcasses in Monterey county, and the composition of the sample was ~57% CASLs, ~31% unidentified and other pinnipeds, ~9% oceanic dolphins, and ~3% other and unidentified cetaceans (NOAA/NMFS Protected Resources Division, unpublished data[43]). Most CASLs in California forage for at least several months of the year in southern California during their breeding season,[44] and CASLs contain concentrations of DDE higher than those of PCBs (Table 2).[7,20] Some suggested that higher DDE/PCB ratios for CASLs arise from Montrose discharges of DDTs.[45] Although a proportion of DDE in CASLs is likely from Montrose,[46] comparisons of the DDE/PCB ratios across species illustrate that high DDE/PCB ratios are not unique to Montrose-associated contamination (Figure S4). For example, CASL blubber (this study) exhibited a range of DDE/PCB ratios (1.6−5.1; n = 12) that were within the range of those observed in blubber from orcas (*Orcinus orca*) (0.89−7.7 DDE/PCB ratio; n = 5) stranded along the Oregon coast[6] and unlikely directly associated with Montrose discharges in southern California.

We found large variations (>500-fold) in DDE concentrations among blubber samples collected from dead-stranded CASLs (Table 2) that could be due to a variety of factors, including changes in body weight associated with seasonal or annual variations in the use of lipid stores, illness, or malnutrition.[47] In addition, condors feed on other marine species, such as dolphins, which contain high levels of DDE (Table 2) that may or may not have been accumulated via foraging off southern California. These factors complicate the

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Table 2. DDE (p,p'-DDE), Sum Chlorinated Pesticides (OCHs), Sum PCBs, Sum PBDEs, and %Lipid Values for Blubber Samples Collected from Marine Mammals Dead-Stranded along the Central California Coast (Figure S1)[a]

species	n	DDE[b]	sum OCHs	sum PCBs[b]	sum PBDEs	%lipid
California sea lion[c]	12					
mean ± SD		12 ± 19	13 ± 20	2.9 ± 3.6	0.46 ± 0.41[d]	34 ± 26
range		0.1−67	0.13−70	0.03−13	0.01−1.07	0.9−76
gray whale[c]	1	ND	0.02	ND	NA	24
harbor seal	1	9.9	10	2.2	NA	10
humpback whale	1	0.3	0.46	0.1	NA	24
Risso's dolphin[c]	1	33	38	11	0.84	58
unknown dolphin	1	35	39	9.6	NA	69

[a]Values reported in micrograms per gram of wet weight blubber. ND indicates a value below the limit of quantification. NA indicates a value that was not measured. [b]Limits of quantification for DDE of 0.015 and <0.007 μg/g for each PCB congener tested. [c]California condors were observed feeding on these samples. For the California sea lions, condors were observed feeding on two of the 12 animals from which samples were collected. [d]n = 6 California sea lion samples for PBDE analysis.

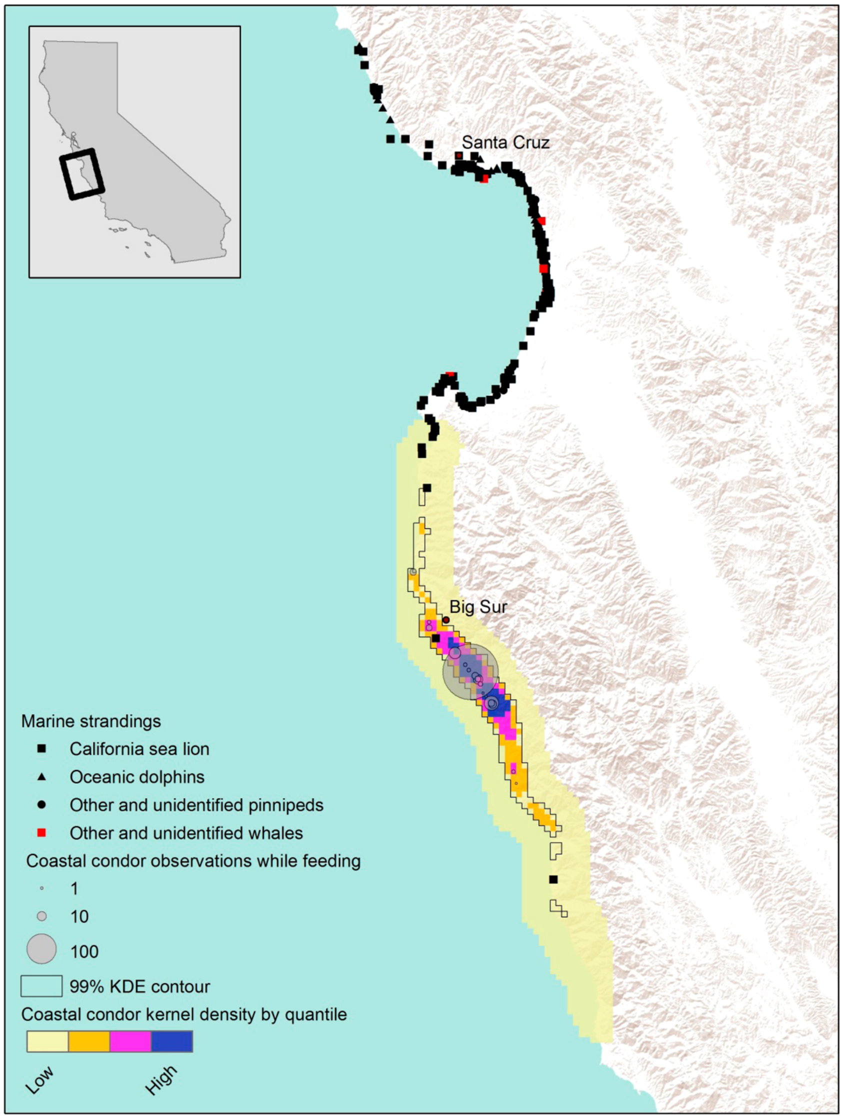

Figure 6. 99% kernel density estimate (KDE) contour and KDE equal-area quantiles of central California (coastal) condors foraging within 1.5 km of the coast from 2003 to 2013, the count of individual condor observational sightings while feeding on marine mammals, and locations of marine mammal strandings.

potential to determine the proportion of a condor's DDE derived from CASLs versus other marine mammals. Nonetheless, given that CASLs represent a predominant marine food source for condors, and CASLs frequently contain high levels of DDE (this study and ref 48), we infer that CASLs are a major source of DDE to condors.

Our simulations suggest that DDE exposure may be a concern for marine-foraging condors, especially older breeders.

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Bald eagles exhibited 10% eggshell thinning at egg DDE concentrations of 5000 ng/g, whereas eagles with DDE concentrations above 15000 ng/g approached 100% reproductive failure.[30] However, a range of sensitivity to DDE-associated eggshell thinning has been reported both between and within avian species,[27] and this sensitivity might be confounded by coexposure to other organochlorine compounds.[49] The applicability of bald eagle data to condors is also unknown, but limited data suggest condors might be more sensitive than bald eagles to DDE-associated eggshell thinning.[27] Kiff et al.[29] reported that DDE concentrations of ∼5000 ng/g (wet weight) in condor eggs were associated with 20% eggshell thinning (using a linear extrapolation) and found that the highest DDE concentration of ∼17000 ng/g (estimated wet weight) corresponded to ∼30% thinner eggshells on average compared to eggshells with nondetectable DDE. Consistent with this, coastal breeding condors were recently shown to have eggs ∼34% thinner than those of inland condors, presumably because of DDE exposure.[12] Thus, we consider DDE-induced reproductive impairment a valid concern for condors feeding on marine mammals.

Although DDE levels may be decreasing in the marine ecosystem,[45,48] DDE levels in CASLs have remained elevated over the past decade,[7,20,48] and our results indicate that ongoing marine foraging elevates DDE levels in condors, even for birds just entering the population. Analysis of condor space use confirms that condors spent the most time on the coast in areas where they were observed feeding on marine mammals and utilize only a small proportion of marine mammals reported stranded along the central California coast (Figure 6). As such, marine mammal abundance does not appear to be a limiting factor for condors. Through range expansion, condors could increase their rate of feeding on CASLs or other species such as elephant seals (*Mirounga angustirostris*), which appear to have contaminant levels lower than those of CASLs.[50]

In conclusion, marine-based subsidies are important resources for scavenging terrestrial species,[42] and we show that coastal condors in central California feed to some extent on dead-stranded marine mammals, a behavior thought to have contributed to their historic survival along the Pacific coast[3,5] as well as a proposed strategy for aiding current condor recovery.[5] However, contemporary condors eating marine mammals had elevated levels of contaminants, including DDE. Our data indicate that condors eating marine mammals may be at risk for reproductive impairment[30,34] as simulation models predict that ∼40% of breeding-age coastal condors have DDE plasma concentrations associated with eggshell thinning in other avian species.[30,51] We highlight the problem of POPs in marine systems, document transfer of these contaminants from marine to terrestrial systems via dead-stranded marine mammals, and stress the importance of considering marine mammal scavenging when managing threats to endangered California condors.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b01990.

Additional figures and tables (PDF)

AUTHOR INFORMATION

Corresponding Author

*E-mail: myraf@ucsc.edu. Phone: 831.459.1249.

Author Contributions

C.M.K. and V.J.B. are co-first authors.

Notes

The authors declare no competing financial interest.

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