9-1-2021

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Recommended Citation

Corey A. Clatterbuck, Rebecca L. Lewison, Rachael A. Orben, Joshua T. Ackerman, Leigh G. Torres, Robert M. Suryan, Pete Warzybok, Jaime Jahncke, and Scott A. Shaffer. "Foraging in marine habitats increases mercury concentrations in a generalist seabird" *Chemosphere* (2021). https://doi.org/10.1016/j.chemosphere.2021.130470

Chemosphere 279 (2021) 130470

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Foraging in marine habitats increases mercury concentrations in a generalist seabird

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HIGHLIGHTS

- Mercury (Hg) contamination in western gulls varies among habitat types.
- Foraging plasticity in generalists like gulls may reflect different exposure to Hg.
- Gulls foraging in ocean habitats had 55% higher blood Hg concentrations.
- Blood Hg concentrations were unrelated to colony, foraging fidelity and sex.
- Differential foraging habitat use may have implications for gull health.

ARTICLE INFO

Article history:
Received 9 December 2020
Received in revised form
28 February 2021
Accepted 30 March 2021
Available online 12 April 2021

Handling Editor: Michael Bank

Keywords:
Mercury
Animal movement
Coastal interface
Foraging
Larus

ABSTRACT

Methylmercury concentrations vary widely across geographic space and among habitat types, with marine and aquatic-feeding organisms typically exhibiting higher mercury concentrations than terrestrial-feeding organisms. However, there are few model organisms to directly compare mercury concentrations as a result of foraging in marine, estuarine, or terrestrial food webs. The ecological impacts of differential foraging may be especially important for generalist species that exhibit high plasticity in foraging habitats, locations, or diet. Here, we investigate whether foraging habitat, sex, or fidelity to a foraging area impact blood mercury concentrations in western gulls (*Larus occidentalis*) from three colonies on the US west coast. Cluster analyses showed that nearly 70% of western gulls foraged primarily in ocean or coastal habitats, whereas the remaining gulls foraged in terrestrial and freshwater habitats. Gulls that foraged in ocean or coastal habitats for half or more of their foraging locations had 55% higher mercury concentrations than gulls that forage in freshwater and terrestrial habitats. Ocean-foraging gulls also had lower fidelity to a specific foraging area than freshwater and terrestrial-foraging gulls, but fidelity and sex were unrelated to gull blood mercury concentrations in all models. These findings support existing research that has described elevated mercury levels in species using aquatic habitats. Our analyses also demonstrate that gulls can be used to detect differences in contaminant exposure over broad geographic scales and across coarse habitat types, a factor that may influence gull health and persistence of other populations that forage across the land-sea gradient.

1. Introduction

Contaminants are commonly used as indicators of environmental quality for wildlife species and systems (Buck 1979; Fairbrother et al., 2019). Because a primary pathway of contaminant exposure is through diet, contaminant levels are also used to

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https://doi.org/10.1016/j.chemosphere.2021.130470

describe the foraging ecology of terrestrial and marine wildlife species (Finkelstein et al., 2006; Ramos and Gonzalez-Solis 2012; Jackson et al., 2015). However, understanding the dietary, trophic, and geographic contributions to contaminant concentration can be complex, as the amount of contaminants present can vary across species ranges from local to global scales (Sunderland et al., 2009; Driscoll et al., 2013). Contaminants have been used to describe foraging habitat conditions and characterize foraging at known point sources of contamination (Anderson et al., 1975) or describe potential contaminant exposure from urban areas (Herman et al., 2005; Clatterbuck et al., 2018). Contaminant exposure has also been used to distinguish marine and terrestrial dietary sources within a population, leveraging the broad differences in potential food sources and contaminant types between marine and terrestrial foodwebs (Post 2002; McGrew et al., 2014; Kurle et al., 2016; Peterson et al., 2017). Unlike other tracers of foraging ecology, contaminant analyses can also provide information on potential downstream effects on organismal health and reproduction (Ramos and Gonzalez-Solis 2012; Kurle et al., 2016).

Mercury (Hg) is a metal that is converted to bioavailable methylmercury through biogeochemical processes largely occurring in aquatic environments, making it a potential tracer of animal foraging across the land-sea gradient (Thompson et al., 1998; Elliott and Elliott 2016; Peterson et al., 2015). As methylmercury is also bioaccumulative and biomagnified, its impacts are largely seen in high trophic organisms, like seabirds, where elevated methylmercury concentrations are associated with impaired endocrine, immune, and general physiological responses (Finkelstein et al., 2006; Goutte et al. 2014, 2015; Tartu et al., 2016). Negative impacts on breeding ecology in seabird species have been linked to elevated methylmercury concentrations, including decreased likelihood of breeding (Tartu et al., 2013; Goutte et al., 2015), lower egg hatchability (Goutte et al., 2014), and fewer fledged chicks per breeding pair (Evers et al., 2008; Goutte et al., 2014), even at methylmercury levels below those known to cause adverse effects (Tartu et al., 2013; Provencher et al., 2016). Thus differences in body burdens of methylmercury within a species or population may differentially impact animal reproduction and survival (Croxall et al., 2012; Goutte et al. 2014, 2015). However, assessment of contaminant body burdens is complex as animals integrate and offload chemical signatures over varying temporal scales due to differences in turnover rates among tissues. For example, avian blood integrates methylmercury from the diet over days and weeks, whereas avian feathers contain Hg accumulated over months and deposited during molt (Furness et al., 1986; Kahle and Becker 1999). Therefore, appropriate environmental and life history context are needed to understand variation in contaminant concentrations as a function of foraging habitat use (Bond 2010). Although previous assessments indicate that mercury concentrations are elevated in species that use marine and freshwater habitats as compared to terrestrial habitats (Evers et al., 2005; Jackson et al., 2015; Davis et al., 2016; Ackerman et al., 2016), there are still limited opportunities to document variation in mercury concentrations across populations that forage differentially along the land-sea gradient.

Developments in spatial analyses and modeling paired with improved telemetry devices have also provided an opportunity to pair movement and chemical tracing to define where animals are exposed to harmful contaminants while also providing important information on foraging locations. Combining contaminant and movement data sources may be particularly useful to characterize the foraging ecology of species that consume a variety of prey items and, therefore, may have highly integrated chemical signals (Finkelstein et al., 2006; Peterson et al., 2017). Gulls (*Larus* spp.) are opportunistic foragers known to shift foraging across a land-sea habitat gradient in response to the annual cycle or external factors including food availability and weather patterns (Isaksson et al., 2016; Spelt et al., 2019), although there is recent evidence that some gull populations may have individual foraging specialists (Bolnick et al., 2003; Masello et al., 2013; Navarro et al., 2017). In the absence of direct observations of on what and where birds are foraging, data on foraging locations is a useful alternative to understanding how food web-based contaminants vary across the landscape (Annett and Pierotti 1999; Weiser and Powell 2010). Multiple tracers, including bulk and compound-specific stable isotopes (Masello et al., 2013; Hobson et al., 2015; Corman et al., 2016; Sánchez-Fortún et al., 2020), organic contaminants (Gentes et al., 2015), telemetry (Masello et al., 2013; Camphuysen et al., 2015; Corman et al., 2016; Isaksson et al., 2016; Spelt et al., 2019), and diet samples (Annett and Pierotti 1999; Weiser and Powell 2010; Corman et al., 2016) have been used to understand changes in gull foraging activity over time and space, and in many cases, to explore potential links to observed productivity or population declines.

In this study, we examined the relationship between mercury concentrations and foraging habitat of western gulls (*Larus occidentalis*), a coastal gull native to the western United States that is known to feed on land and at sea (Annett and Pierotti 1999; Shaffer et al., 2017). We tracked movement and tested whole blood for total mercury in western gulls at three colonies, including a colony where gull population and productivity have exhibited a long-term decline (Southeast Farallon Island; Johns and Warzybok 2018). We combined movement data with blood Hg concentrations of breeding gulls to assess whether land cover features where gulls forage, foraging site fidelity, or sex may be associated with elevated mercury concentrations. Specifically, we compared data on foraging locations and fidelity based on GPS locations with Hg levels, as mercury becomes more bioavailable in aquatic systems, to explore how Hg is related to the relative use of oceanic versus terrestrial foraging areas. By pairing movement data with contaminant tracer, we identify gull foraging patterns across a large ocean area and consider how current patterns in habitat use are linked to potential exposure to contaminants and ultimately to population dynamics.

2. Methods

2.1. Field collection & lab analyses

From 2015 to 2017, we captured actively incubating western gulls (n = 59) at three colonies on the west coast of the United States: Cleft-of-the-Rock (n = 19) and Hunters Island (n = 11), off the Oregon coast and Southeast Farallon Island in California (n = 29; Fig. 1). Whereas Cleft-of-the-Rock and Hunters Island are located near rural human development on or near the Oregon mainland (respectively, human population <24,000 within ca. 60 km), Southeast Farallon Island lies ~60 km east of the urbanized San Francisco Bay Area (human population ca. ~7.6 million). Gulls were captured using a mixture of noose carpet and noose traps surrounding the nest. On the first capture, we attached one of three waterproofed GPS units: iGot-U (GT-series; www.i-gotu.com), Mr. Lee (CatLog; www.mr-lee.com), or Ornitela (Ornitrack; www.ornitela.com) to gull back or tail feathers using Tesa tape (Beiersdorf AG GmbH, Hamburg, Germany) or using a Teflon™ leg loop harness (Mallory and Gilbert 2008). All units weighed 15–25 g, which corresponded to 1.6–3.0% of body mass (mean ± SD 1060 ± 117 g) on deployment. We programmed a regular sampling rate ranging from 120s to 600 s for all GPS units. Before release, we banded unmarked gulls using steel U.S. Geological Survey (USGS) leg bands.

On re-capture, we retrieved the GPS unit and collected gull

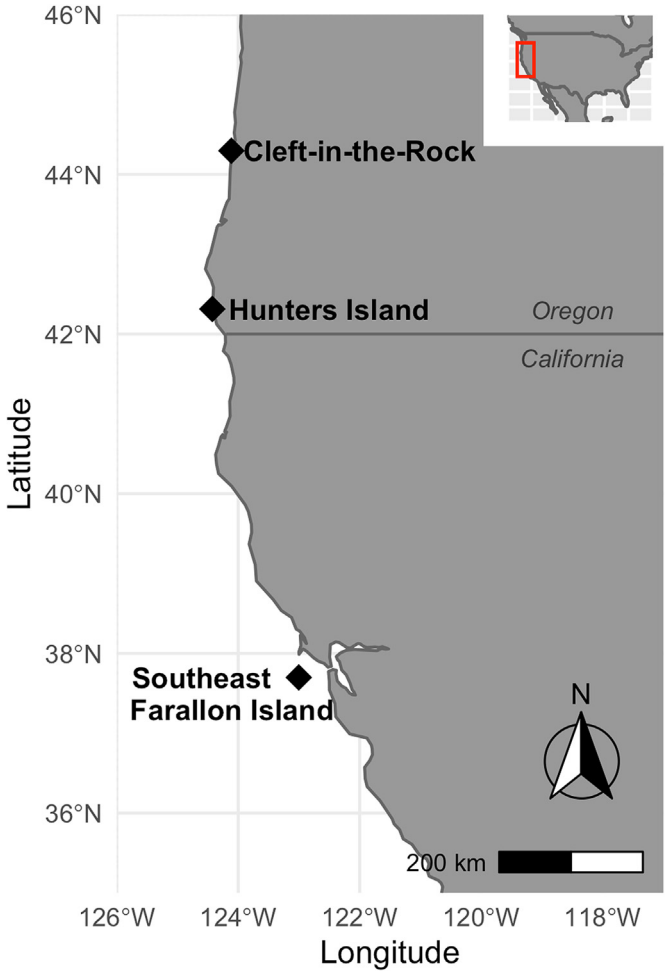

Fig. 1. Western gull colonies where gulls were trapped and sampled over the study period.

morphometric data and blood. Gull morphometrics, including culmen, skull, and tarsus length, were measured to the nearest 0.5 mm using a dial caliper. We measured gull mass using a Pesola® spring scale to the nearest 10 g during both capture and recapture when possible and collected up to 1.5 mL of gull whole blood from the tarsal or brachial vein using 24–26 gauge needles and 2 mL syringes. Birds equipped with Ornitela tags (n = 7) were not recaptured and blood samples and morphometrics were taken at first capture.

After collection, we put gull blood in vacutainers containing K_2EDTA and when possible stored the vacutainers on ice in the field and in −20 °C freezers in the lab. Due to conditions in the field, almost all gull blood samples were congealed and desiccated upon arrival for Hg analysis. Gull blood samples were analyzed wet for total mercury (THg) using a Nippon MA-3000 Direct Mercury Analyzer (Nippon Instruments North America, College Station, Texas, USA) following Environmental Protection Agency Method 7473 (U.S. Environmental Protection Agency, 2000) at the U.S. Geological Survey, Dixon Field Station Environmental Mercury Laboratory (Dixon, California; Ackerman et al., 2020). Blood samples were defrosted and allowed to warm to room temperature before weighed and analyzed for THg. THg is a suitable proxy for MeHg, as MeHg accounts for >90% of THg concentrations in avian whole blood (Rimmer et al., 2005; Renedo et al., 2021). Quality assurance measures included analysis of a certified reference material (either dogfish muscle tissue [DORM-4] or lobster hepatopancreas [TORT-3] certified by the National Research Council of Canada, Ottawa, Canada), system blank, method blank, continuing calibration verification, and duplicate with each set of about 10 samples. Quality assurance measures included machine blanks with each run (n = 28), continuing calibration verification (mean ± SD percent recovery = 99.3 ± 1.2%, n = 13), and certified reference materials (mean ± SD percent recovery = 99.5 ± 1.7%, n = 17). We were concerned that the congealed nature of the blood samples could result in high within-sample variation in Hg. However, duplicate samples indicated similarity in THg concentrations (mean ± SD relative percent difference = 2.4 ± 2.2%, n = 15) (Ackerman et al., 2016; Peterson et al., 2017). Blood Hg concentrations were non-normally distributed, so we used Tukey's ladder of powers to find the transformation that best met the assumption of normality for linear modeling: transformed THg = $-1 * [\text{THg}]^{-0.15}$. All blood Hg concentrations are reported as μg g^{-1} wet weight and are publicly available in (Ackerman et al., 2021).

All data was processed for further analysis using R software (R Core Team 2020). We determined sex based on these measurements using linear discriminant analysis, which was trained using a dataset of gulls where sex was known (Shaffer, unpublished data). Data were scaled and normalized using the "caret" package (Kuhn 2019) before predicting sex using the lda() function in the "MASS" package (Venables and Ripley 2002). Cross-validation suggested the model error rate when using the training dataset was 7.4%. We accepted the model's predictions for sex if the posteriors for either sex was 95% or greater.

2.2. Identifying putative foraging locations

We analyzed GPS data from tagged gulls to determine where gulls foraged. We retained all GPS location data points (~99.9%) that connected to three or more satellites and interpolated this data to 600 s intervals, the longest sampling rate, to ensure foraging data was comparable among individuals. For individuals equipped with Ornitela satellite tags that were not recaptured, we analyzed GPS data taken within the first 10 days after deployment. Because potential feeding areas were local to gull breeding colonies, we defined trips as any departure and return beyond a 1 km radius of the colony that lasted over 90 min using the package "trakR" (Fleishman et al., 2019). To identify locations where gulls foraged, we applied a behavioral classification system – Residence in Space and Time (RST) – with a dynamic scaling radius identified for each bird (mean ± SD radius = 1.7 ± 1.3 km) (Torres et al., 2017). RST calculates the difference of the normalized residence in time and distance to define each location as one of three potential behavioral states – rest, area-restricted search, or transit – and has been effective at defining behavior states using GPS tracks from a variety of taxa including surface foraging seabirds (Torres et al., 2017). We further split rest locations into those at the colony and included roosting movement behavior as part of the foraging classification when birds were away from the colony. Gulls are known to employ sit-and-wait foraging strategies and may remain relatively stationary when hunting in the intertidal. We then interpreted these behavioral states as the gull was either at the colony, foraging, and transit. Finally, we determined individual foraging fidelity to a geographic location by comparing maximum displacement values between foraging trips (Hazen et al., 2016; Shaffer et al., 2017), where negative values indicate low fidelity and positive values indicate high fidelity (Fleishman et al., 2019).

2.3. Classifying foraging habitat types

We conducted a cluster analysis to classify individuals based on primary foraging habitat type at putative foraging locations. To characterize the habitat type at a foraging location, we overlaid locations in the foraging (area-restricted search) state with available geographic state boundary and waterbody data, which categorized each location as either land, ocean, or freshwater (including brackish bays and estuaries; California Department of Forestry and Fire Protection 2015; Oregon Water Resources Department 2005). To normalize foraging effort among individuals, we calculated the proportion of land cover type for all foraging locations within an individual. Once all foraging locations were classified, we then considered whether there was evidence of clustering of gulls in foraging habitat categories based on the proportions of foraging locations in one of the three land cover categories using non-metric multidimensional scaling (nMDS, R package "vegan") with Bray-Curtis distances to account for zeroes in the proportional data (Oksanen et al., 2019). We used model-based clustering to confirm observed foraging habitat clusters from nMDS ordination using the R package "mclust" (Scrucca et al., 2016), where individuals were probabilistically assigned to a single cluster. We tested models of 2–4 clusters and used Bayesian Information Criterion (BIC) and similarity metrics to determine the most likely number of foraging habitat clusters. We further confirmed that these clusters separated habitat types by grouping consecutive foraging points in time as a foraging event. Like foraging locations, foraging events were also characterized by land cover, but each event consists of multiple foraging locations and potentially multiple land cover types. Therefore, foraging events were characterized as either coastal (consisting of a combination of land and ocean points), inland (land and freshwater points), mixed tidal (ocean and freshwater points), completely land, ocean, or freshwater, or all (land, ocean and freshwater). We then used pairwise t-tests and Wilcoxon rank-sum tests, depending on whether or not data were normally distributed, to examine cluster-based differences of land cover for foraging locations and foraging events, separately.

2.4. Links to THg exposure

We first asked whether differences in THg exposure were related to colony using a one-way ANOVA. We then asked whether foraging habitat type, fidelity to a foraging area, or sex were related to THg concentration. Because fidelity index was correlated with foraging habitat types (point biserial correlation, $t = -3.64$, df = 49, $p < 0.001$), we ran two separate regression models: One with foraging cluster and sex as potential independent variables and another with fidelity index and sex as potential independent variables. We were unable to test for an interaction between colony and foraging habitat type because only one individual from each Cleft-in-the-Rock and Hunters Island clustered separately from the other individuals in those colonies, which violated the assumption of independence between colony and foraging cluster (Chi-square test of independence, $\chi^2 = 18.28$, $p < 0.001$). To further explore this potential interaction, we analyzed ocean foragers alone by colony using a non-parametric ANOVA and conducted pairwise comparisons of THg for ocean foragers between each colony using t-tests. We could not perform the same analysis for inland foragers due to inadequate sample sizes from Cleft-in-the-Rock and Hunters Island. Significance was evaluated at $p < 0.05$.

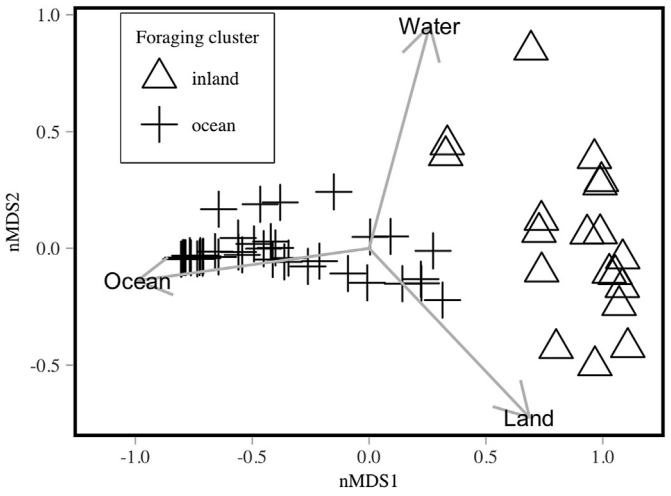

Fig. 2. Gull clusters based on nMDS ordination and model-based clustering. Loadings indicate land cover for foraging location data. Model-based clustering defined three total clusters, two of which exhibited similar mean proportions of ocean foraging. These were combined into a single ocean foraging cluster for further analysis. The final cluster identified by both nMDS ordination and model-based clustering, the inland foraging cluster, was retained for analysis.

3. Results

Using the 59 western gulls from Cleft-in-the-Rock (n = 19), Hunters Island (n = 11), and Southeast Farallon Island (n = 29), we performed linear discriminant analysis using gull body measurements to further classified these individuals as 22 males and 33 females, with four gulls (6.8%) where sex remained unknown. Gulls took a total of 584 trips away from colonies over a sum of 381 days where GPS units were deployed. Gulls generally returned to a similar geographic area (mean fidelity index = 0.49) and had a total of 14,599 GPS points classified as foraging. On average, total blood Hg concentration for all gulls was 0.637 μg g^{-1} wet weight (geometric mean; range 0.150–3.278), and we detected Hg in every individual gull.

3.1. Foraging habitat clusters

Using foraging locations, we identified two habitat type clusters using nMDS ordination – ocean and inland (Fig. 2). However, model-based clustering identified three habitat type clusters. Model-based clusters a and b were dominated by ocean foraging (mean of 97% and 71%, respectively) and cluster c that contained the same individuals as the inland habitat type cluster from nMDS ordination (Figure S1; Table S1). Because two of the three model-based clusters were dominated by ocean foraging activity and did not differ in THg concentration (Kruskal-Wallis ANOVA, $p = 0.07$; TukeyHSD, $p = 0.998$), we collapsed them into a single ocean habitat cluster that resembled the ocean habitat type cluster identified using nMDS ordination for further analysis (Figs. 2 and 3).

3.2. Foraging habitat types

Our clustering algorithms classified each gull as an ocean or inland forager based on the proportions of each habitat type at all foraging locations for that gull. Of 59 total gulls, 40 were identified

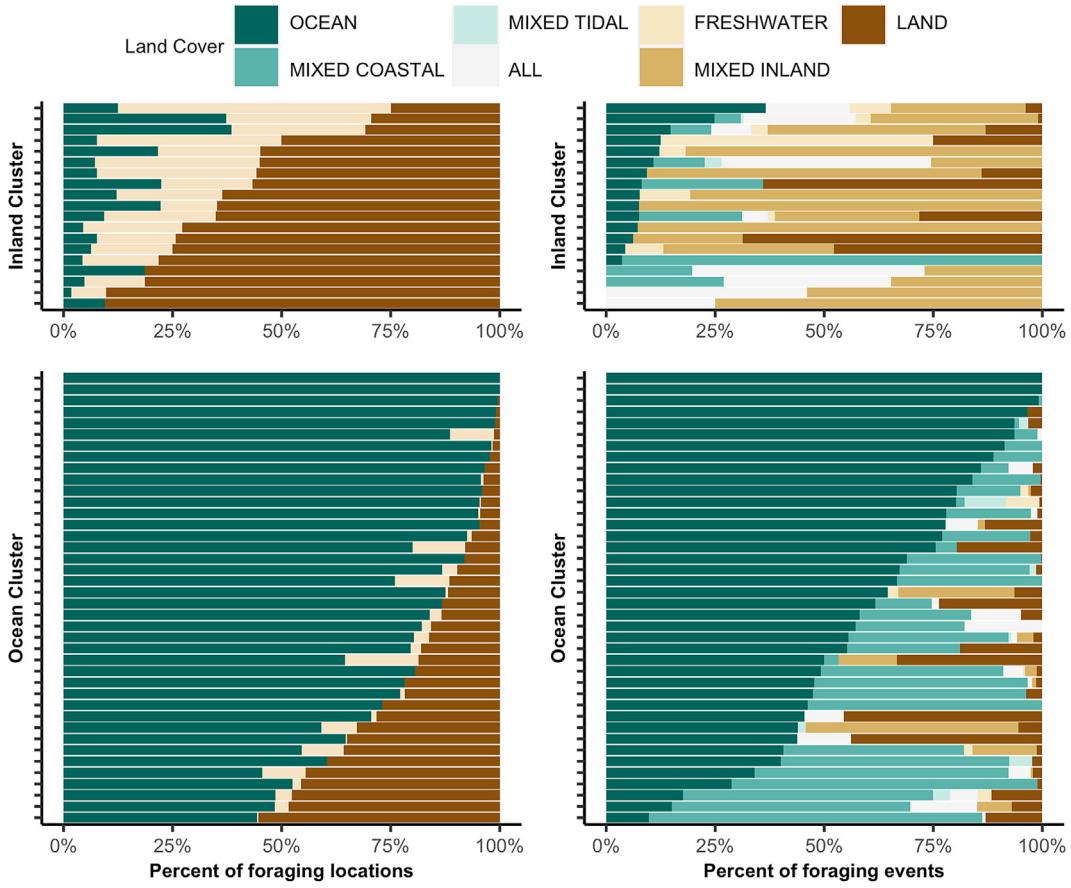

Fig. 3. Percentage land cover of gull foraging locations (left) and foraging events (right) separate by foraging cluster. Because foraging events are combinations of foraging locations, events may have mixed land cover types or all land cover types in addition to the ocean, freshwater, and land classes.

as ocean foragers while 19 were classified as inland foragers. Almost all gulls from Cleft-in-the-Rock and Hunters Island were classified as ocean-foraging gulls, while 17 of 19 inland-foraging gulls nested at Southeast Farallon Island. Gulls differed in their proportion of ocean, land, and water locations, where gulls in the ocean cluster generally had greater than 50% ocean locations and fewer land and water locations than the inland cluster (*t*-test, all $p < 0.001$, Fig. 3a). These differences were also reflected in proportions of foraging events, where gulls in the ocean cluster had greater proportions of ocean and coastal foraging events and fewer inland and water foraging events than inland gulls (Wilcoxon rank-sum tests, $p < 0.05$, Fig. 3b). Ocean-foraging gulls also exhibited significantly less fidelity (0.40 ± 0.37) to a geographic area than inland-foraging gulls (0.85 ± 0.37; Wilcoxon rank-sum test, $W = 381$, $p < 0.001$) and had higher variability in trip direction (mean azimuth = 208 ± 107°) than inland-foraging gulls (mean azimuth = 143 ± 66°), though not significantly so (Watson-Wheeler test, $W = 4.74$, df = 2, $p = 0.09$). Trip duration and normalized trip frequency also did not differ between habitat types (t-tests, $p > 0.05$, Table 1).

3.3. Links to THg exposure

We found no difference in THg among colonies ($F_{2,56} = 0.88$, $p = 0.42$). Using multiple regression analysis, we found that THg differed among gulls that foraged in ocean versus inland habitat (Fig. 4). Linear regression demonstrated that ocean foraging gulls had on average 55% higher blood THg concentrations than inland foraging gulls ($F_{2,54} = 3.91$, $p = 0.026$, adjusted $R^2 = 0.13$; $t = 2.21$, $p = 0.032$), although foraging habitat used only explained ~13% of the observed variation in THg concentrations. Sex was not significantly related to THg ($t = 1.50$, $p = 0.14$). As there was no interactive effect of sex with foraging cluster ($t = 0.27$, $p = 0.79$), we report the model without this interaction as a predictor. However, the model

Table 1
Summary statistics for gull foraging clusters. Ranges are given as standard deviations except for THg.

Cluster	N	Sex (N)			THg[a]	Mean land cover (%)			fidelity index[b]	trips per day	trip duration[c]	trip azimuth
		♀	♂	unk		ocean	water	land				
inland	19	12	6	1	0.47 ± 2.07	13 ± 11	24 ± 15	63 ± 19	0.85 ± 0.31	1.9 ± 1.3	574 ± 379	143 ± 66°
ocean	40	21	16	3	0.73 ± 1.94	80 ± 17	3 ± 4	17 ± 16	0.40 ± 0.37	1.6 ± 0.6	406 ± 239	208 ± 107°

[a] Values are the geometric mean and geometric standard deviation in μg g-1 wet weight.
[b] Values range from −1 (most dissimilar) to 1 (most similar).
[c] Values are in minutes.

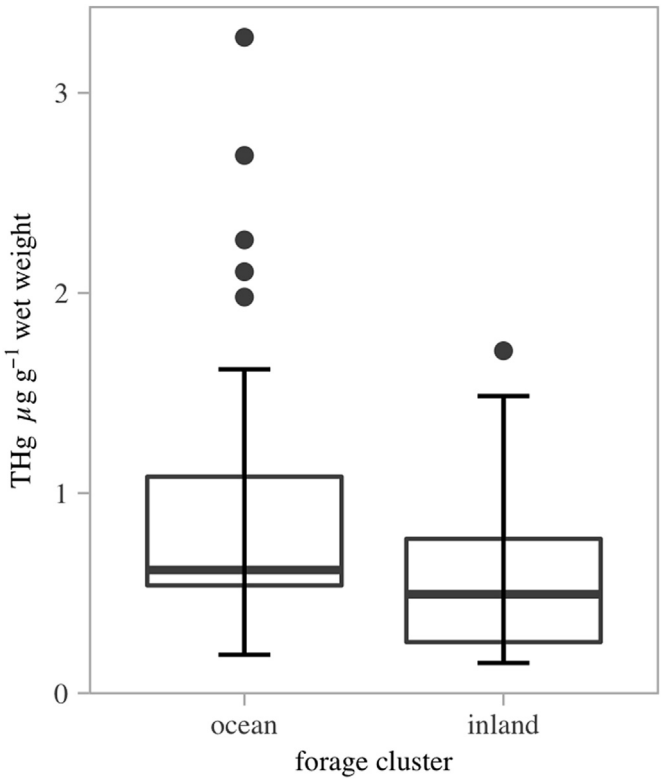

Fig. 4. Total mercury concentrations of western gulls grouped by foraging cluster. The boxplot represents the 25–75% quartile range and the median bar.

that included fidelity index and sex as predictor variables was not significantly different from the null model ($F_{2,49} = 1.51$, $p = 0.231$, $R^2 = 0.031$). Though fidelity was strongly related to foraging cluster, it was unrelated to THg ($t = -0.045$, $p = 0.964$) and there was no interactive effect of fidelity index with sex ($t = 1.13$, $p = 0.265$). Exploring the potential interaction between foraging cluster and colony, we found ocean foraging gulls had similar THg regardless of colony ($F_{2,37} = 1.02$, $p = 0.38$). Pairwise comparisons among ocean foragers from all colony combinations were not significant (Cleft-in-the-Rock and Hunters Island, $t = -1.43$, df = 16.49, $p = 0.17$; Cleft-in-the-Rock and Southeast Farallon Island ($t = -0.51$, df = 14.02, $p = 0.62$; Hunters Island and Southeast Farallon Island $t = 0.38$, df = 17.14, $p = 0.71$).

4. Discussion

Because the primary pathway for body burdens of contaminants is through diet, variables that impact diet, including geographic location, foraging habitat, and physiology are expected to influence contaminant concentrations (Finkelstein et al. 2006, 2007; Robinson et al., 2011; Ramos and Gonzalez-Solis 2012; Jackson et al., 2015). While wildlife biomonitoring efforts commonly uses these relationships to identify point sources of chemical pollution, relatively fewer studies have examined broad-scale differences in contaminant concentrations in coastal and marine waters off the urbanized west coast of the United States. Our analyses demonstrate that western gulls – generalist, avian foragers – had higher total blood Hg concentrations when foraging over ocean habitat compared to inland habitat across three geographically distinct colonies in the Northeast Pacific and that relative to colony, fidelity to foraging areas and sex, the type of foraging habitat used has the largest effect on gull Hg exposure. While there are no doubt other differences among these three colonies, our finding emphasizes the ubiquity of Hg exposure across multiple food webs (Post 2002; Kurle et al., 2016). This finding is particularly important for generalists that exploit multiple habitat types, shift foraging strategies according to food availability, or exhibit individual specialization (Annett and Pierotti 1999; Hobson et al., 2015; Bolnick et al., 2003). Our research also affirms that the use of chemical tracers can be an effective tool to identify animal foraging habitat and organismal and environmental health (Peterson et al., 2017).

Using GPS foraging locations to identify foraging habitat type, we found gulls across the three colonies fell into one of two categories: ocean-foraging gulls that foraged over ocean or coastal habitat, or inland-foraging gulls that foraged in terrestrial and freshwater habitats (Fig. 3). We detected significant differences in THg concentrations in ocean-foraging versus inland-foraging gulls (Fig. 4), a finding supported by a number of studies which also found differential Hg exposure across the land-sea foraging gradient, associated directly with the food consumed (McGrew et al., 2014; Kurle et al., 2016) and differences in foraging locations within or among colonies (Peterson et al., 2017; Sánchez-Fortún et al., 2020; Thorne et al., 2021). The differences we identified in Hg concentration based on foraging habitat have been linked to differences in methylmercury bioaccumulation between terrestrial and marine food webs (Ackerman et al., 2016). Additionally, more recent studies show Hg concentrations are predictive of foraging habitat use in other species of gulls using GPS locations or stable isotope analysis, respectively (Peterson et al., 2017; Sánchez-Fortún et al., 2020; Thorne et al., 2021). Our study supports these findings and also highlights some important areas for future research. Compared to published studies, the blood Hg values we observed in western gulls were variable, which may reflect wide variation in methylmercury exposure found in gull species (Ackerman et al., 2016). The relative importance of foraging habitat used, as compared to colony, site fidelity and sex, further support the use of Hg as a tracer of foraging ecology (Kurle et al., 2016; Peterson et al., 2017; Chételat et al., 2020).

Our findings suggest a significant link between foraging habitat and THg exposure, foraging habitat used only explained ~13% of the observed variation in THg concentrations. This is unsurprising as many other factors are known to influence methylmercury concentrations in wild birds, including biogeochemical processes that make elemental Hg bioavailable, body condition, and physiological storage and excretion mechanisms (Eagles-Smith et al., 2009; Elliott and Elliott 2016; Chételat et al., 2020). THg in avian blood represents mobilization of mercury both through the diet and from internal body tissues (Evers et al., 2005; Chételat et al., 2020). Controlled studies of seabirds that were dosed with methylmercury suggest methylmercury in whole blood has a rapid half-life of 24 h, followed by a slower half-life of 30–60 days (Monteiro and Furness 2001). Additionally, western gulls are omnivorous and their diet includes a variety of prey representing different foraging habitats and trophic levels (Annett and Pierotti 1999). In this context, it is probable that the THg concentrations we measured also assess accumulation prior to the time periods captured by the GPS tracks. Dietary shifts can occur in western gulls at the time of chick-hatching, where gulls tended to consume more marine prey; however, our study did not sample gulls over this time period (Annett and Pierotti 1989). As opportunistic foragers, gulls may exploit multiple foraging sources depending on forage availability. Longer GPS deployments or including another indicator of foraging habitat, such as sulfur stable isotopes or compound-specific stable isotopes, may strengthen the relationships we observed (Peterson et al., 2017; Sanchez-Fortun et al., 2020; Binkowski et al., 2021). Despite these complexities, a significant link between THg concentrations and oceanic versus inland foraging was detectable

(Peterson et al., 2017; Thorne et al., 2021; Chételat et al., 2020).

Previous work has suggested that western gulls that forage in terrestrial habitats have greater fidelity to a geographic foraging area than ocean foraging gulls, in part because prey distribution at sea is ephemeral whereas terrestrial food availability may be reliable and uniformly distributed (Corman et al., 2016; Shaffer et al., 2017). Our findings support these conclusions, although fidelity to a geographic foraging area was unrelated to THg concentrations. That foraging cluster, but not geographic foraging fidelity, is linked to THg concentration further suggests that methylmercury contamination in western gulls reflects broad differences in methylmercury contamination across habitat types, rather than specific geographic sources of Hg (Ackerman et al., 2016). Recent studies of gull habitat use have utilized fine-scale geographic attributes to describe gull foraging (Isaksson et al., 2016; Navarro et al., 2017; Shaffer et al., 2017; Spelt et al., 2019). While fine-scale measurements of gull habitat may be helpful to understand of local contamination for urban or point-source sites (Ricca et al., 2008; Gentes et al., 2015), our work suggests that even with coarser resolution habitat information, we can contextualize Hg concentrations with gull foraging ecology.

Despite being separated by hundreds of miles along coastlines with vastly different degrees of development, we did not detect significant differences in THg concentration among the three colonies we sampled and found that ocean-foraging gulls had similar THg concentrations regardless of colony. This aligns with other studies of seabird foraging that associate contaminant concentrations with foraging habitat used rather than site-specific signals (Soldatini et al., 2020; Thorne et al., 2021), and suggests that tracers of foraging ecology are useful because they can distinguish between multiple potential foraging strategies in and among populations (Ramos et al., 2013). Still, the possibility for an interaction between foraging habitat and colony still exists and may be easier to detect for known Hg hotspots. For example, San Francisco Bay is a well-documented area of elevated methylmercury exposure, and California gulls (*Larus californicus*) that used the estuarine waters of the bay had elevated Hg concentrations compared to terrestrial-feeding conspecifics (Peterson et al., 2017). The blood Hg values we observed in inland-foraging western gulls aligned with the range of blood Hg values found in California gulls in San Francisco Bay (Fig. 4, Peterson et al., 2017). While our cluster analysis did not distinguish between estuarine and terrestrial foraging, an interaction may be more obvious between inland foraging gulls nesting at each colony. Unfortunately, the low sample size of inland foraging gulls from the Oregon colonies did not allow us to make this comparison. Future studies may consider how variation in gull foraging strategies, likely driven by ecological pressures including intra- or interspecific competition, levels of nearby marine food resources, and fisheries activity, may influence contaminant concentrations in these populations (Hobson et al., 2015; Corman et al., 2016; Garthe et al., 2016).

5. Conclusions

Widespread production and deposition of anthropogenic compounds is expected to continue to impact seabirds and other top predators, including gulls. While the use of contaminants to identify potential point sources of pollution is needed, contaminant tracers can also inform our understanding of multi-colony or regional impacts of large-scale production and deposition of manmade compounds (Gentes et al., 2015; Peterson et al., 2017). With continued research into potential pathways for contaminant deposition and availability across the land-sea gradient, studies of contaminants and habitat use can be used to assess exposure to a wide range of contaminants (Ramos et al., 2013; Gilmour et al., 2019). Our results show that contaminant exposure is different in terrestrial, estuarine, or marine forage foods, which for a generalist species like the western gull may impact breeding, recruitment and population trajectories (Annett and Pierotti 1999; Duhem et al., 2008; Weiser and Powell 2010). With differential habitat use and thus exposure to contaminants, the adverse impacts of contaminant-associated diet may be an important consequence of foraging plasticity in gull populations. Future studies can determine how Hg and other contaminants can be used as chemical tracers to understand the ecological consequences of diet-mediated contaminant exposure for gull populations.

Author contribution

Corey Clatterbuck: Conceptualization, Methodology, Validation, Formal analysis, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization, Funding acquisition. **Rebecca Lewison**: Methodology, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration. **Rachael Orben**: Software, Formal analysis, Investigation, Resources, Writing – original draft, Writing – review & editing, Funding acquisition. **Joshua Ackerman**: Validation, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Funding acquisition. **Leigh Torres**: Software, Resources, Writing – original draft, Writing – review & editing, Funding acquisition. **Robert Suryan**: Resources, Writing – original draft, Writing – review & editing, Funding acquisition. **Pete Warzybok**: Investigation, Writing – original draft, Writing – review & editing, Funding acquisition, Project administration. **Jaime Jahncke**: Writing – original draft, Writing – review & editing, Funding acquisition, Project administration. **Scott Shaffer**: Conceptualization, Investigation, Resources, Writing – original draft, Writing – review & editing, Project administration, Funding acquisition

Declaration of competing interest

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

Acknowledgments

We thank Megan Jennings, Marisa Trego, and Tracy Grimes for feedback on study design and analysis and Matt Toney and Mark Herzog for laboratory work. The efforts of three anonymous reviewers and one internal reviewer substantially improved the final publication. JTA was supported by the U.S. Geological Survey Environmental Health Mission Area's Contaminant Biology Program. Logistical support at the Farallon Islands was facilitated by the Farallon Run Patrol and the U.S. Fish and Wildlife Service. This research was conducted under Animal Care and Use Committee permit no. 4905 at Oregon State University and no. 979 at San José State University, U.S. Federal Bird Banding and Marking Permit no. 23411, and U.S. Fish and Wildlife special use permit no. 81641. This work was funded by CSU Council on Ocean Affairs, Science, & Technology (CSU-COAST) Student Travel Award, the CSU-COAST Graduate Student Research Award, and the CSU Program for Education and Research in Biotechnology Student Travel Grant Program. The authors declare no competing interests. The scientific results and conclusions, as well as any views or opinions expressed herein, are those of the authors and do not necessarily reflect those of NOAA or the Department of Commerce. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.chemosphere.2021.130470.

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