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# Mercury contamination and stable isotopes reveal variability in foraging ecology of generalist California gulls

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## ABSTRACT

Environmental contaminants are a concern for animal health, but contaminant exposure can also be used as a tracer of foraging ecology. In particular, mercury (Hg) concentrations are highly variable among aquatic and terrestrial food webs as a result of habitat- and site-specific biogeochemical processes that produce the bioaccumulative form, methylmercury (MeHg). We used stable isotopes and total Hg (THg) concentrations of a generalist consumer, the California gull (Larus californicus), to examine foraging ecology and illustrate the utility of using Hg contamination as an ecological tracer under certain conditions. We identified four main foraging clusters of gulls during pre-breeding and breeding, using a traditional approach based on light stable isotopes. The foraging cluster with the highest  $\delta^{15}N$  and  $\delta^{34}S$  values in gulls (cluster 4) had mean blood THg concentrations 614% (pre-breeding) and 250% (breeding) higher than gulls with the lowest isotope values (cluster 1). Using a traditional approach of stable-isotope mixing models, we showed that breeding birds with a higher proportion of garbage in their diet (cluster 2: 63–82% garbage) corresponded to lower THg concentrations and lower  $\delta^{15}$ N and  $\delta^{34}$ S values. In contrast, gull clusters with higher THg concentrations, which were more enriched in <sup>15</sup>N and <sup>34</sup>S isotopes, consumed a higher proportion of more natural, estuarine prey.  $\delta^{34}$ S values, which change markedly across the terrestrial to marine habitat gradient, were positively correlated with blood THg concentrations in gulls. The linkage we observed between stable isotopes and THg concentrations suggests that Hg contamination can be used as an additional tool for understanding animal foraging across coastal habitat gradients.

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## 1. Introduction

Animals integrate unique signatures of habitat use, geography, and diet into body tissues from their prey, and therefore animal tissues can reveal elements of foraging ecology that can be unobtainable from direct observation or use of electronic tracking instruments (Ramos and González-Solís, 2012). Common ecological tracers used to study animal foraging ecology include light stable isotopes (e.g. C, N, H, and S) and fatty acids (Budge et al., 2006; Hobson, 1999; Inger and Bearhop, 2008; Peterson and Fry, 1987), and less commonly used tracers include environmental contaminants (Adams and Paperno, 2012; Calambokidis and Barlow, 1991; Catry et al., 2008). Contaminants, such as heavy metals and persistent organic pollutants, are often studied because of their potential impact on organism and ecosystem health (Tanabe, 2002; Wiener et al., 2003). However, some contaminants bioaccumulate in

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http://dx.doi.org/10.1016/j.ecolind.2016.11.025 1470-160X/Published by Elsevier Ltd. organisms and biomagnify in upper trophic level predators, which can allow these contaminants to serve as an ecological tracer and reveal elements of animal foraging ecology such as habitat type or location. Contaminants such as heavy metals and persistent organic pollutants are often non-uniformly distributed in the environment (Chasar et al., 2009; Meijer et al., 2003; Roscales et al., 2010), which may enable them to be used as tracers of habitat use at a regional or local scale.

Mercury (Hg) contamination in particular has many characteristics that may make it a useful ecological tracer. The unique processes that control methylmercury (MeHg) production from inorganic Hg are highly localized and vary substantially among habitat types (Eagles-Smith et al., 2016; Marvin-DiPasquale et al., 2003). As such, the biogeochemical processes influencing MeHg bioavailability in the environment can result in high variability in MeHg concentrations across aquatic and terrestrial habitats (Ullrich et al., 2001). Localized production of MeHg directly influences MeHg bioaccumulation in upper-trophic level predators because MeHg is the form of Hg that bioaccumulates in organisms and biomagnifies with increasing trophic level (Ullrich et al.,







2001). Consequently, different habitat types in close proximity can contain markedly different MeHg concentrations in similar organisms (Chen et al., 2005; Eagles-Smith and Ackerman, 2014). For example, fish and bird MeHg concentrations varied up to 4-fold and 11-fold, respectively, in adjacent wetlands with different biogeochemistry (Ackerman et al., 2014a; Eagles-Smith and Ackerman, 2014). Additionally, MeHg concentrations can vary among similar habitats but geographically disjunct sites based on the availability of MeHg at the base of the food web and localized bioaccumulation processes (Eagles-Eagles-Smith and Ackerman, 2014; Evers et al., 2011; Scudder et al., 2009). Furthermore, as a result of biogeochemical processes and differences between aquatic and terrestrial food webs, MeHg concentrations often are much higher in animals deriving their diet from aquatic, rather than terrestrial, habitats (Ackerman et al., 2016b; McGrew et al., 2014; Ochoa-Acuña et al., 2002; Post, 2002).

MeHg and stable isotopes in consumer tissues represent an integrated diet over varying lengths of time, depending on the tissue (Lewis and Furness, 1991; Vander Zanden et al., 2015; Wang et al., 2014), and variability in these tracers can reveal different components of animal foraging ecology (Caron-Beaudoin et al., 2013; Moreno et al., 2010; Ramos et al., 2009). Trophic position within a habitat can be established using  $\delta^{15}$ N values and MeHg concentrations (Anderson et al., 2009; Campbell et al., 2005). Habitat type and sources of primary productivity to a food web can be discriminated using carbon isotope ratios, although  $\delta^{13}$ C values also increase with trophic level but to a lesser degree than  $\delta^{15}N$  values (Inger and Bearhop, 2008; Peterson and Fry, 1987). Sulfur isotope ratios, considered to have low or undetectable levels of fractionation with trophic position, have been effectively used to reveal habitat use along a terrestrial to marine gradient for multiple taxonomic groups (Barros et al., 2010; Cotin et al., 2011; Fry and Chumchal, 2011; Lott et al., 2003; Ramos et al., 2009; Zazzo et al., 2011). MeHg concentrations can also relate to habitat use along a terrestrial to marine gradient, likely as a result of sulfate reduction and increased methylation in specific habitat types (Gabriel et al., 2014; Gilmour et al., 1992). Moreover, MeHg concentrations in organisms may reveal additional aspects of an animal's foraging ecology, including separation of foraging locations or specific habitats, than traditional ecological tracers (Adams and Paperno, 2012; Catry et al., 2008). For example, in a study where stable isotopes were inconclusive in differentiating foraging ecology of tropical seabirds, the addition of MeHg resulted in the ability to differentiate foraging locations (Catry et al., 2008). Therefore, MeHg may be a useful tracer to differentiate diet and foraging strategies for generalist species that forage across a range of diverse habitats, and complement traditional ecological tracers like light stable isotopes.

We used a combination of total Hg (THg) concentrations and light stable isotopes (nitrogen, carbon, and sulfur) of a generalist predator, the California gull (Larus californicus), to examine variability in foraging strategies and to illustrate the utility of using Hg contamination as an ecological tracer of foraging ecology. While in the San Francisco Bay Estuary (California, USA), California gulls can access marine and estuarine prey resources, as well as terrestrial anthropogenic diet sources associated with several large landfills along the bay margins. We used a traditional approach of using light stable isotopes to identify clusters of birds with similar foraging ecology during the pre-breeding and breeding time periods, and then examined if foraging clusters of gulls could be differentiated based upon their Hg contamination. We hypothesized that gulls from different foraging clusters would vary in their use of terrestrially-derived prey (from landfills), and that gulls with higher proportions of diet derived from landfills would result in lower Hg contamination than gulls with a higher proportion of diet derived from aquatic, estuarine prey. Additionally, because sulfur isotopes are strongly reflective of foraging along a terrestrial to marine gradient, we examined whether  $\delta^{34}$ S values directly related to THg concentrations to demonstrate if Hg concentrations could link to animal foraging ecology.

## 2. Methods

## 2.1. Sample collection

We captured adult California gulls in 2007 and 2008 using rocket nets (Dill and Thornsberry, 1950), remotely detonated net launchers (Coda Enterprises, Mesa, Arizona, USA), and bow nets at three breeding colonies in south San Francisco Bay, California, USA (A6, Coyote Hills, and Mowry colonies; Ackerman et al., 2014b) from 6-March to 26-April, prior to the breeding season, and from 15-May to 30-May, during the breeding season. We captured chicks by hand from 16-June to 3-July, late in the breeding season. We collected approximately 2 mL of whole blood from the brachial vein using 23-25 gauge needles with a 1 or 3 cc syringe from all birds for both total mercury (THg) and stable isotope analyses, and held blood samples on ice while in the field. Additionally, we collected a drop of blood from the majority of adult birds for sex determination using the chromo-helicase-DNA binding protein gene (Zoogen Services, Inc., Davis, California, USA). We measured culmen length, bill depth at the gonys, head-to-bill length, and flattened wing length to the nearest 0.01 mm using digital calipers. Birds were also weighed to the nearest 1.0 g using a 1-kg Pesola spring scale (Pesola AG, Baar, Switzerland), which we used as a proxy for body condition (Labocha and Hayes, 2012). We used a discriminant function, based on gull morphometric measurements (Herring et al., 2010), for sex determination of adults in the cases where we did not have genetic results. All birds were temporarily held in screen-lined and shaded poultry cages (model 5KTC, Murray McMurray Hatchery, Webster City, Iowa, USA) at the capture site until they were sampled, banded with a U.S. Geological Survey (USGS) aluminum leg band, and subsequently released. Birds were captured and marked under a California State Scientific Collection permit, Federal Bird Banding permits, Federal Fish and Wildlife permits, and research was conducted under the guidelines of the USGS Western Ecological Research Center Animal Care and Use Committee.

To examine potential dietary sources for breeding adult California gulls, we obtained reference prey samples from April to August in 2007 and 2008 from estuarine habitats adjacent to breeding colonies and from terrestrial, human-sources including local markets and the Newby Island landfill. Gull use of sampling sites was confirmed with concurrent radio telemetry tracking (Ackerman et al., 2016c). We collected potential prey samples (n = 409) from salt ponds, including brine shrimp (Artemia franciscana) and two fish species, three-spined stickleback (Gasterosteus aculeatus) and long-jawed mudsucker (Gillichthys mirabilis). We collected American avocet (Recurvirostra Americana) and Forster's tern (Sterna *forsteri*) eggs (n = 51 and n = 26, respectively) and muscle samples from pre-fledged chicks (n = 17 and n = 17, respectively), similar to methods described in Ackerman et al. (2011). Consumption of avocet and tern eggs and chicks in San Francisco Bay is well documented for California gulls (Ackerman et al., 2014b,c). To represent small mammals living in the terrestrial environment surrounding the wetland habitats frequented by gulls, we collected house mice (*Mus musculus*; n = 6). From the local landfill and food markets, we collected samples representing common foods, including chicken, turkey, pig, cow, bread, potatoes, rice, and vegetables (n = 74).

### 2.2. Mercury determination

We analyzed approximately  $100 \mu$ L of liquid whole blood (hereafter blood) for THg using a Milestone DMA-80 Direct Mercury Analyzer (Milestone, Shelton, Connecticut, USA) at the USGS Dixon Field Station Mercury Lab. Blood was analyzed for THg because >85% of Hg in blood of avian and mammalian predators is typically in the MeHg form (Alvárez et al., 2013; Rimmer et al., 2005; Soria et al., 1992; Woshner et al., 2008). During each run of samples, we included continuing calibration verifications, certified reference materials from the National Research Council of Canada, Ottawa, Canada (DORM-3 and DOLT-3), system and method blanks, and duplicate samples as quality assurance measures. The recoveries (mean  $\pm$  SE) were 101.3  $\pm$  0.9 for calibration verifications (n=23) and 104.2  $\pm$  1.3 for certified reference materials (n=16). Duplicate samples had a mean absolute relative percent difference of 3.3  $\pm$  1.1 (n=15). Blood THg concentrations were generated as wet weight (ww) values.

### 2.3. Stable isotope analysis

We dried and homogenized California gull blood and reference prey samples, and weighed two subsets of each sample into a tin capsule to 0.01 mg, to obtain stable carbon, nitrogen, and sulfur isotope ratios at the Colorado Plateau Stable Isotope Laboratory (Northern Arizona University, Flagstaff, Arizona, USA). Samples for carbon and nitrogen were run through a Thermo-Electron DELTA V Advantage IRMS configured through a Finnigan CONFLO III (Thermo Fisher Scientific Inc., Waltham, Massachusetts, USA) for automated continuous-flow analysis of  $\delta^{15}$ N and  $\delta^{13}$ C, using a Carlo Erba NC2100 elemental analyzer (Carlo Erba Instruments, Milan, Italy) for combustion and separation of C and N. Samples for sulfur were run through a DELTA plus Advantage IRMS configured through a CONFLO III (Thermo Fisher Scientific Inc., Waltham, Massachusetts, USA) for automated continuous-flow analysis of  $\delta^{34}$ S using a Costech ECS4010 elemental analyzer (Costech Analytical Technologies Inc., Valencia, California, USA). We report the results of isotope analysis in standard delta ( $\delta$ ) notation relative to international standards (atmospheric N<sub>2</sub> for nitrogen, Vienna PeeDee Belemnite for carbon, and Vienna-Canyon Diablo Troilite for sulfur), using the equation  $\delta X = [(R_{sample}/R_{standard}) - 1] \times 1000$ , with  $X = {}^{15}N$ ,  ${}^{13}C$  or  ${}^{34}S$  and  $R = {}^{15}N/{}^{14}N$ ,  ${}^{13}C/{}^{12}C$ , or  ${}^{34}S/{}^{32}S$ . The experimental precision, based on the average standard deviation of within-run internal standard replicates, was estimated to be  $\pm 0.12$ for  $\delta^{15}$ N and  $\pm 0.06$  for  $\delta^{13}$ C (NIST peach leaves) and  $\pm 0.22$  for  $\delta^{34}$ S (NIST bovine liver).

#### 2.4. Cluster analysis for foraging strategies

For each season (pre-breeding and breeding), we conducted a cluster analysis to identify unique clusters of adult California gull foraging ecology based on all three stable isotopes, and tested if clusters differed in THg concentration or body mass. We used the same approach to cluster chicks into groups representing similar parental foraging ecology. To create clusters, we used a two-step process with a principal components analysis (PCA) followed by hierarchical cluster analysis (FactoMineR package in R; Husson et al., 2015, 2010). This method generates unrotated factors from the PCA, and we retained principal components with an eigenvalue threshold of 1.0. Hierarchical clustering was then conducted on the factor scores from the PCA, using Euclidean distance and the average linking method, and intra-cluster inertia was used to identify clusters (Husson et al., 2010). Once clusters were identified, we examined if blood THg concentrations and body mass (comparing males and females separately) differed among clusters, using analysis of variance (ANOVA). We examined differences in bird mass among the clusters separately for males and females due to sexual dimorphism in the species (Herring et al., 2010; Schnell et al., 1985). Blood THg concentrations were natural-log transformed prior to analysis to

meet the assumptions of normality and homogenous variance for general linear models.

After using stable isotopes to cluster gulls into foraging strategies, we used log-transformed blood THg concentrations to cluster gulls into groups. We predetermined the number of clusters at 4 for each season, to provide consistency with the clustering based on stable isotopes. We then tested the clusters for differences in stable isotope ratios, to see if THg concentrations alone could successfully group birds that differed in their foraging ecology.

## 2.5. Stable isotope mixing models

We examined the probability of different potential sources (estuarine prey versus prey from anthropogenic sources) to the diet of identified clusters (groups) of isotopically-similar, breeding California gulls and isotopically-similar chicks, using a Bayesian stable isotope mixing model, SIAR v4 (Parnell et al., 2010), with  $\delta^{15}$ N and  $\delta^{34}$ S values. We did not include  $\delta^{13}$ C in the mixing model because reference prey samples were not lipid extracted prior to analysis. The high variability we observed for C:N ratios among potential dietary sources could have caused spurious results, as there are no standardized equations to adjust non lipid-extracted  $\delta^{13}$ C isotope values from terrestrial sources (Bearhop et al., 2002; Post et al., 2007). We identified potential sources with isotopically distinct  $\delta^{15}$ N and  $\delta^{34}$ S values, which resulted in one human-derived source (garbage:  $3.7 \pm 1.9\%$   $\delta^{15}$ N,  $2.7 \pm 4.4\%$   $\delta^{34}$ S) and five naturallyoccurring estuarine sources: aquatic prey from three different managed wetland areas (Alviso:  $17.8 \pm 2.3\%$   $\delta^{15}$ N,  $12.0 \pm 2.6\%$  $\delta^{34}$ S; Mowry: 14.1 ± 2.3‰  $\delta^{15}$ N, 15.2 ± 2.2‰  $\delta^{34}$ S; and Newark:  $9.1 \pm 1.0\% \delta^{15}$ N,  $12.0 \pm 2.4\% \delta^{34}$ S), American avocets, and Forster's terns. Mixing models for adult gulls included isotope values for avocet (14.2  $\pm$  3.3‰  $\delta^{15}$ N, 6.8  $\pm$  3.8‰  $\delta^{34}$ S) and tern eggs (18.7  $\pm$  2.1‰  $\delta^{15}$ N, 9.5  $\pm$  3.1‰  $\delta^{34}$ S) whereas mixing models for chicks included isotope values for avocet ( $12.9 \pm 2.0\% \delta^{15}$ N,  $8.7 \pm 3.2\% \delta^{34}$ S) and tern chick muscle (18.3  $\pm$  1.9%  $\delta^{15}$  N, 12.5  $\pm$  1.5%  $\delta^{34}$  S). Mouse samples (N = 6) were indistinguishable from the Alviso complex aquatic prey samples for  $\delta^{15}$ N and  $\delta^{34}$ S and therefore were not included as a potential source. For each source, we obtained a mean isotope value and standard deviation. For garbage, we obtained average isotope values for meat and non-meat products separately and then obtained an overall average isotope value. To obtain a standard deviation (SD) of the isotope values observed for each source in the mixing model, we used the following equation, where  $z_i$  is each individual isotope value,  $\hat{\mu}$  is the source mean and *n* is the sample size for the source:

$$SD = \sqrt{rac{\Sigma (z_i - \hat{\mu})^2}{n - 1}}$$

Trophic enrichment factors (TEF) can vary based on the lipid and protein characteristics of the prey (Bearhop et al., 2002; Caut et al., 2009) and selection of TEFs is critical for mixing models. In our study, for nitrogen we used a TEF of  $2.9 \pm 0.6$  for naturally-occurring estuarine prey sources (Bearhop et al., 2002; Craig et al., 2015), and we used a TEF of  $4.0 \pm 0.6$  for garbage (Bearhop et al., 2002). For sulfur we used a TEF of  $0.5 \pm 2.9$  for all sources (Craig et al., 2015). Once we established sources and TEFs, we ran the mixing model, which fit the model using Markov chain Monte Carlo (MCMC), for 500,000 iterations with a burn in at 50,000.

# 2.6. General linear models with sulfur isotope ratios and THg concentrations

After we determined that THg concentrations differed among clusters of gulls, using ANOVA, we examined if there was a predictable relationship between sulfur isotope ratios and blood THg



**Fig. 1.** Hierarchical cluster analysis on principal components of  $\delta^{34}$ S,  $\delta^{15}$ N, and  $\delta^{13}$ C values (‰) measured in the whole blood of pre-breeding California gulls (*Larus californicus*) in south San Francisco Bay, California, USA revealed four clusters. A and B show mean isotope values for each cluster ± SD, with individual gull isotope values shown for females (circles) and males (triangles). C shows the comparison of blood total mercury (THg) concentrations among clusters and significant differences are indicated by letters. Boxplots show the median and the middle 50th percentile, and whiskers encompass the full range of the data.

concentrations in adult gulls before and during the breeding season, while accounting for any potential influence of sex, year, or colony. Sulfur isotopes were chosen as the best isotopic index of individual foraging ecology for California gulls for the following reasons. In general, sulfur changes very little with trophic level, in contrast with nitrogen and carbon, and sulfur isotope ratios vary extensively along the terrestrial to marine gradient and among habitats (Fry and Chumchal, 2011; Peterson and Fry, 1987). In this study, we observed a wide range of sulfur isotope values in California gulls. Thus, we tested whether blood Hg concentrations could explain the variability in blood sulfur isotope ratios, and consequently if Hg concentrations in California gulls could explain aspects of foraging location and habitat.

## 3. Results

#### 3.1. Foraging ecology clustered by isotopes

California gull adults and chicks clustered into multiple foraging strategies, using  $\delta^{34}$ S,  $\delta^{15}$ N, and  $\delta^{13}$ C values. We observed high variability among adult California gulls (n = 146) for stable isotope values between and within seasons, with pre-breeding birds ( $\delta^{34}$ S: -2.6 to 19.2%;  $\delta^{15}$ N: 6.9 to 18.4%;  $\delta^{13}$ C: -20.5 to -15.7%) having a wider range of stable isotope values than breeding birds ( $\delta^{34}$ S: 2.6-7.6%;  $\delta^{15}$ N: 8.0-12.6%;  $\delta^{13}$ C: -19.7 to -16.2%; Table A1).

Pre-breeding California gulls grouped into four foraging clusters (Fig. 1) and breeding California gulls grouped into five foraging clusters (Fig. 2; Table 1). Cluster 1 had the lowest  $\delta^{34}$ S and  $\delta^{15}$ N values for both pre-breeding and breeding gulls. For pre-breeding gulls, mean  $\delta^{34}$ S values were enriched in <sup>34</sup>S by 30% (1.3‰), 109% (4.8%), and 236% (10.4%) between cluster 1 and clusters 2, 3, and 4, respectively. Mean  $\delta^{15}$ N values became enriched in  ${}^{15}$ N by 13% (1.1‰), 42% (3.7‰), and 78% (6.9‰), between cluster 1 and clusters 2, 3, and 4, respectively (Fig. 1). For breeding gulls, mean  $\delta^{34}$ S and  $\delta^{15}$ N values became enriched in <sup>34</sup>S and <sup>15</sup>N by 122% (3.8‰) and 32% (2.7‰), respectively, between clusters 1 and 4 (cluster 5 had one bird; Fig. 2). All four pre-breeding clusters included males and females, but two of the five breeding clusters (1 and 5) included only females. Gull chicks (n = 67) clustered into three groups (Fig. 3). Chicks in cluster 1 had the lowest  $\delta^{34}$ S and  $\delta^{15}$ N values and became enriched in  ${}^{34}$ S and  ${}^{15}$ N by 79% (4.9‰) and 37% (3.4‰), respectively, between clusters 1 and 3.

### 3.2. Isotopic clusters differed in mercury contamination

THg concentrations in gulls differed significantly among isotopic clusters of pre-breeding ( $F_{3,96} = 32.52$ , p < 0.001) and breeding birds ( $F_{4,41} = 8.40$ , p < 0.001). For pre-breeding gulls (n=100; THg concentrations  $0.02-1.02 \ \mu g/g \ ww$ ), all four clusters differed in their mean blood THg concentrations (p < 0.006; Fig. 1c). Cluster 1 had the



**Fig. 2.** Hierarchical cluster analysis on principal components of  $\delta^{34}$ S,  $\delta^{15}$ N, and  $\delta^{13}$ C values (‰) measured in the whole blood of breeding California gulls in south San Francisco Bay, California, USA revealed five clusters. A and B show mean isotope values for each cluster ± SD, with individual gull isotope values shown for females (circles) and males (triangles). C shows the comparison of blood total mercury (THg) concentrations among clusters and significant differences are indicated by letters. D shows estimated consumption of garbage by cluster from a mixing model with six possible prey sources. Violin plots show the distribution of the mixing model results with the median (dot) and the middle 50% of the output (line).

lowest blood THg concentrations and geometric mean THg concentrations increased by 100%, 229%, and 614% between cluster 1 and clusters 2, 3, and 4, respectively (Table 1). For breeding gulls  $(n = 46; \text{ THg concentrations } 0.04 - 0.62 \,\mu\text{g/g ww})$ , blood THg concentrations increased moving from cluster 1 to cluster 5, although not all clusters were significantly different (Fig. 2c). Cluster 1 had lower THg concentrations than any other cluster ( $p \le 0.006$ ), followed by cluster 2 ( $p \le 0.019$ ). THg concentrations in gulls from clusters 3 and 4 were not significantly different from each other (p=0.77), although cluster 3 had lower concentrations than the gull in cluster 5 (p = 0.044) whereas cluster 4 was not statistically different from cluster 5 (p = 0.067). Geometric mean blood THg concentrations in breeding gulls increased by 117%, 233%, 250%, and 800% between cluster 1 and clusters 2, 3, 4, and 5, respectively (Fig. 2c; Table 1). Chick blood THg concentrations (n=27) ranged from  $0.03-0.34 \,\mu g/g$  ww. Although the geometric mean blood THg concentration of cluster 3 was 67% greater than the other two clusters (Table 1), the clusters themselves were not significantly different ( $F_{2,24} = 2.56$ , p = 0.10).

## 3.3. Isotopic clusters differed in reliance on landfills

Gulls from different foraging clusters varied in their consumption of garbage, as indicated by stable isotope mixing models for breeding adults and chicks (Figs. 3; A1, A2). In general, although the first cluster did not mathematically resolve well (95% credibility interval of 7.6–80.7%), the estimated consumption of garbage in breeding adult gulls decreased and their consumption of naturallyderived estuarine prey increased moving from cluster 1 to cluster 5 (Fig. 2d). The proportion of garbage in the diet of gulls was estimated to be 63–82% for cluster 2 and 52–73% for cluster 3 (95% credibility intervals). In contrast, gull consumption of garbage was estimated to be 35–63% for cluster 4 and 15–52% for cluster 5. The differences in diet among clusters corresponded well with the resulting THg contamination of gulls. In general, gulls became more contaminated with Hg and relied less on landfills moving from cluster 1 to 5.

Adult California gulls continued to demonstrate variability in foraging ecology during late breeding. Mixing model analysis on the clusters of gull chicks, as a proxy for late-breeding adult foraging ecology, varied markedly in the estimated proportion of garbage in their diet (Fig. A2). The proportion of garbage in the diet of gull chicks was estimated to be 57–72%, 36–55%, and 14–36%, for cluster 1, 2, and 3, respectively.

Differences in diet and Hg bioaccumulation could have influenced body condition of adult California gulls, however differences in body mass (as a proxy for condition) did not align with differences in THg concentrations for either time period. Mass of

## 210 **Table 1**

Isotope values, bird mass<sup>a</sup> (g; separately for adult females and males), and total mercury (THg) concentrations<sup>b</sup> in whole blood ( $\mu$ g/g ww) by isotopically-derived cluster for pre-breeding and breeding adult California gulls (*Larus californicus*) and chicks (late in the breeding season) from south San Francisco Bay, California, USA. Isotope values and mass are shown as arithmetic mean ± SD and THg concentrations are geometric mean (geometric SD<sup>c</sup>).

	Cluster 1	Cluster 2	Cluster 3	Cluster 4	Cluster 5 <sup>d</sup>	Overall
Pre-breeding adults						
N (F, M)	16 (8,8)	39 (23,16)	27 (14,13)	20 (6,12)	-	100 (51,49)
$\delta^{34}S$	$4.4 \pm 1.9^{*}$	$5.7 \pm 1.5^{*}$	$9.2\pm3.9$	$14.8 \pm 3.1^{*}$	-	$8.1\pm4.5$
$\delta^{15}$ N	$8.8\pm1.3^{*}$	$9.9\pm0.9^*$	$12.5 \pm 1.0^{*}$	$15.7 \pm 1.1^{*}$	-	$11.5\pm2.6$
$\delta^{13}C$	$-19.2 \pm 0.8^{*}$	$-18.2 \pm 0.5$	$-17.6 \pm 0.6^{*}$	$-17.1 \pm 0.8^{*}$	-	$-18.0\pm0.9$
THg	0.07 (1.7) <sup>1</sup>	$0.14(1.7)^2$	0.23 (2.1) <sup>3</sup>	0.50 (1.6) <sup>4</sup>	-	0.18 (2.3)
Female mass	$621\pm42^1$	$616\pm53^1$	$563\pm43^2$	$576 \pm 18^{1,2}$	-	$597\pm51$
Male mass	$627\pm45^1$	$717\pm39^2$	$719\pm45^2$	$697\pm35^2$	-	$697\pm51$
Breeding adults						
N (F, M)	4 (4,0)	19 (15,4)	15 (9,6)	7 (6,1)	1 (1,0)	46 (35,11)
$\delta^{34}S$	$3.1 \pm 0.5^{*}$	$4.1 \pm 0.7^{*}$	$5.4\pm0.7$	$6.9\pm0.6^*$	6.6	$4.9 \pm 1.3$
$\delta^{15}$ N	$8.5 \pm 0.3^{*}$	$9.0 \pm 0.3^{*}$	$9.8\pm0.4$	$11.2 \pm 0.5^{*}$	12.6*	$9.6 \pm 1.0$
$\delta^{13}C$	$-18.8 \pm 0.5^{*}$	$-18.1 \pm 0.5$	$-17.7 \pm 0.4$	$-17.8\pm0.5$	-16.2*	$-17.9\pm0.6$
THg	0.06 (1.5) <sup>1</sup>	0.13 (1.5) <sup>2</sup>	0.20 (1.7) <sup>3</sup>	0.21 (1.6) <sup>3,4</sup>	$0.54^4$	0.15 (1.8)
Female mass	$609\pm86^1$	$623\pm52^1$	$594\pm57^1$	$632\pm70^1$	645 <sup>1</sup>	$616\pm59$
Male mass	-	$760\pm15$	$751\pm51$	735 <sup>b</sup>	-	$752\pm37$
Chicks						
N: isotopes	31	21	15	-	-	67
N: THg	5	15	7	-	-	27
$\delta^{34}S$	$6.2 \pm 1.5^{*}$	$8.7\pm1.2$	$11.1 \pm 1.6^{*}$	_	-	$8.1\pm2.4$
$\delta^{15}$ N	$9.1\pm0.8^{*}$	$10.6\pm1.1$	$12.5 \pm 0.8^{*}$	-	-	$10.4\pm1.6$
$\delta^{13}C$	$-19.0 \pm 0.5^{*}$	$-18.1 \pm 0.6^{*}$	$-17.6 \pm 0.4^{*}$	-	-	$-18.4\pm0.8$
THg	$0.06(1.97)^1$	$0.06(1.5)^1$	$0.10(2.1)^1$	-	-	0.07 (1.8)

<sup>a</sup> Mass was compared separately for each season and sex for adult birds using ANOVA. Significant differences are indicated by superscript numbers. Breeding males were not compared for mass, as only 11 birds were sampled and did not appear in clusters 1 or 5.

<sup>b</sup> THg concentrations for clusters were compared separately by season and age class. Significant differences are indicated by superscript numbers.

<sup>c</sup> Geometric SD is a multiplicative factor (+ to get low end, × to get high end) to describe dispersion around a geometric mean (Kirkwood, 1979). Geometric mean and SD are shown for THg concentrations because analyses were conducted on log-transformed data.

<sup>d</sup> Only one bird in this group.

\* Indicates isotope values important in identification of the cluster from mean values.



**Fig. 3.** Mean  $\pm$  SD of  $\delta^{34}$ S and  $\delta^{15}$ N values for five clusters (hierarchical cluster analysis on principal components of stable isotopes in whole blood) of breeding adult California gulls and three clusters of California gull chicks in south San Francisco Bay, California, USA, and potential dietary sources including anthropogenic-derived sources (garbage), fish and brine shrimp from three groups of salt ponds (Alviso, Mowry, and Newark), American avocet (*Recurvirostra americana*) and Forster's tern (*Sterna forsteri*) eggs and muscle samples from chicks. Potential dietary sources have been adjusted, based on individual trophic enrichment factors, to align with gull isotope values. Mixing models for adults and chicks included all three salt pond groups and garbage; however, the mixing model for adults included waterbird eggs as potential sources whereas the mixing model for chicks included waterbird chick muscle.

pre-breeding females in clusters 1 and 2 was not significantly different (p=0.77) and marginally overlapped with birds in cluster 4 (p=0.07 and p=0.06, respectively). Birds in cluster 3 were lighter than birds in clusters 1 and 2 (p ≤ 0.006) but were similar to birds in cluster 4 (p=0.57; Table 1). In contrast, pre-breeding males from cluster 1 were approximately 12% lighter than males from clusters 2–4 (p<0.001), which were not significantly different from each other (p ≥ 0.18; Table 1). Breeding gulls foraging on terrestrially-derived prey with lower Hg at landfills were in no better or worse body condition than gulls foraging on more naturally-derived marine prey with inherently higher Hg contamination, as there was no difference in female mass among clusters ( $F_{4,30}$ =0.50, p=0.74; Table 1).

#### 3.4. Mercury-derived clusters differed in stable isotopes

Blood THg concentrations in gulls alone was able to successfully cluster gulls into groups that differed in their isotope ratios. Pre-breeding and breeding gulls clustered using only their Hg contamination level resulted in significant differences in  $\delta^{34}S$  $(F_{3,96} = 64.05, p < 0.001; F_{3,42} = 9.94, p < 0.001), \delta^{15}N (F_{3,96} = 25.55, p < 0.001; F_{3,42} = 10.04, p < 0.001), and \delta^{13}C values (F_{3,96} = 2.84, p < 0.001)$ p = 0.042;  $F_{3,42} = 3.85$ , p = 0.016; Fig. 4). Of the three isotopes, sulfur demonstrated the strongest differences among clusters. Cluster 1 had the lowest  $\delta^{34}$ S values, with a mean increase during prebreeding of 58% (2.1‰), 126% (4.6‰), and 291% (10.6‰) to clusters 2, 3, and 4, respectively, and a mean increase during breeding of 29% (1.0‰), 58% (2.0‰), and 76% (2.6‰) to clusters 2, 3, and 4, respectively. When compared to the assignment of individual gulls to clusters created using all three stable isotopes, using only THg concentrations classified 46% of pre-breeding gulls and 42% of breeding gulls in the same cluster, with only 3% and 4% of gulls, respectively, moving more than into an adjacent cluster.



**Fig. 4.** Total blood mercury (THg) concentrations were used to cluster similar adult pre-breeding (A–C) and breeding (D–E) California gulls (*Larus californicus*) sampled in south San Francisco Bay. Stable isotope ratios of sulfur (A, D), nitrogen (B, E) and carbon (C, F) were compared among clusters for each season and significantly different clusters (p < 0.05) are indicated using lower-case letters. Boxplots show the median and the middle 50th percentile, and whiskers encompass the full range of the data. Note that the axes differ between pre-breeding and breeding clusters.

## 3.5. Sulfur isotopes correlated with mercury concentrations

 $δ^{34}$ S values were positively related to blood THg concentrations in adult California gulls during pre-breeding ( $F_{1,95}$  = 186.38, p < 0.001) and breeding ( $F_{1,40}$  = 31.34, p < 0.001; Fig. 5), while accounting for sex, colony, and year. For pre-breeding birds, a 10% increase in blood THg concentrations resulted in a 0.42‰ increase in  $δ^{34}$ S values. For breeding birds, a 10% increase in blood THg

concentrations resulted in a 0.15‰ increase in  $\delta^{34}$ S values. The prebreeding relationship between blood THg concentrations and  $\delta^{34}$ S values was not influenced by year ( $F_{1,95} = 3.53$ , p = 0.063) nor sex ( $F_{1,95} = 0.13$ , p = 0.72), however the Coyote Hills colony had slightly higher  $\delta^{34}$ S values for the same THg concentration in comparison to birds from the A6 colony ( $F_{1,95} = 10.77$ , p = 0.001). During breeding, the relationship between  $\delta^{34}$ S values and THg concentrations in blood was not influenced by year ( $F_{1,40} = 0.74$ , p = 0.39) nor colony



**Fig. 5.**  $\delta^{34}$ S values in whole blood of pre-breeding (black) and breeding (red) California gulls related to total mercury (THg) concentrations in whole blood. The solid lines show predicted isotope concentrations with 95% confidence limits (dotted lines) from back-transformed THg concentrations. Model predictions were generated for female birds from the A6 colony in 2007. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

( $F_{2,40}$  = 0.25), but male  $\delta^{34}$ S values were 0.8% higher than females at the same blood THg concentrations ( $F_{1,40}$  = 4.80, p = 0.034).

## 4. Discussion

Environmental contaminants are rarely used as ecological tracers, but some contaminants vary substantially among habitats, particularly those like Hg whose biological availability is dependent on the presence of certain biogeochemical processes (Marvin-DiPasquale et al., 2003). Biogeochemical processes that convert relatively biologically unavailable inorganic Hg to highly bioaccumulative MeHg and allow MeHg to biomagnify through food webs can make this contaminant particularly useful for understanding animal foraging ecology at the habitat scale. In particular, animal consumers foraging in wetland and estuarine habitats, especially in those with increased activity of sulfate- or iron-reducing bacteria, are often enriched in <sup>34</sup>S and have greater bioaccumulation of MeHg (Eagles-Smith and Ackerman, 2014; Gilmour et al., 1992). We used a generalist gull species that forages in habitats with differing MeHg contamination and showed that differences in gull Hg concentrations were strongly related to different foraging clusters. These results suggest that MeHg contamination may be an additional tool to reveal intra-species differences in foraging ecology.

Intra-annual changes in foraging ecology have been observed previously in other generalist gulls using diet studies (Bertellotti and Yorio, 1999; Lindsay and Meathrel, 2008), although the diet samples in these other studies represented a short time period of several minutes to hours prior to sampling. Light stable isotopes and MeHg can be advantageous as ecological tracers because they integrate foraging ecology over a period of weeks to months (Lewis and Furness, 1991; Vander Zanden et al., 2015). Additionally, chemical tracers can reveal animal foraging ecology without the methodological biases associated with only collecting feeding observations during daylight hours (Dierks, 1990; Jehl and Mahoney, 1983) or the highly variable digestibility of different prey types typical of many diet studies (Lindsay and Meathrel, 2008; Weiser and Powell, 2011). Thus, chemical tracers have some advantages over direct observation and the collection of diet samples, such as regurgitates or fecal matter, when quantifying foraging habitat and prey type of animals over an extended period of time.

The range of pre-breeding stable isotope values and blood THg concentrations we observed suggested a high diversity of foraging strategies among individual California gulls, similar to other generalist gulls (Auman et al., 2011; Caron-Beaudoin et al., 2013; Hebert et al., 1999; Moreno et al., 2010; Ramos et al., 2009). The variability among foraging strategies of California gulls indicated that the species foraged extensively along a coastal marine to terrestrial gradient, which encompasses a matrix of heterogeneous habitat with varying isotopic values and Hg availability. However, breeding gulls had lower blood THg concentrations and isotope values that were less enriched in <sup>15</sup>N and <sup>34</sup>S, in contrast with pre-breeding gulls, suggesting that this population of gulls shifted their foraging behavior towards increased consumption of terrestrial or anthropogenic food resources during breeding, even with marine habitats in close proximity to gull colonies. Thus, intra-annual changes in THg concentrations revealed shifts in foraging ecology and Hg contamination has been similarly used to describe foraging behavior of other waterbirds (Ackerman et al., 2014a; Lavoie et al., 2014; Winder and Emslie, 2012).

Individual gulls clustered into similar groups, regardless of whether stable isotopes or THg concentrations were used as the ecological tracer of foraging ecology. Use of a traditional ecological tracer, stable isotopes, grouped gulls into four main clusters during both the pre-breeding and breeding seasons. We used the clusters identified using this traditional technique to examine if differences in foraging ecology corresponded to differences in Hg contamination. All four clusters markedly differed in Hg contamination during pre-breeding, with a more than 600% increase in geometric mean THg concentrations between cluster 1 and cluster 4. During breeding, the range of observed isotope values and THg concentrations was narrower than that observed during prebreeding, although geometric mean THg concentrations still had a 250% increase between cluster 1 and cluster 4.

We then verified that gulls in the clusters used available habitats differently, using diet analysis through isotope mixing models. Gulls in clusters with the highest proportion of garbage in their diet had lower Hg concentrations and lower  $\delta^{34}$ S and  $\delta^{15}$ N isotope values. In contrast, gulls in clusters with a greater proportion of naturally-derived estuarine prey in their diet had higher Hg exposure and higher  $\delta^{34}$ S and  $\delta^{15}$ N values. Clustering gulls using solely blood THg concentrations successfully separated gulls into groups that differed in light stable isotope ratios. The ability of THg concentrations as a single factor to cluster individuals into groups with differences in foraging ecology provides an example of how Hg contamination can be useful for more than just quantification of toxicological risk.

The diversity of foraging strategies demonstrated by California gulls and the strong relationship we demonstrated between  $\delta^{34}$ S values and THg concentrations suggests that THg concentrations within individuals can help to identify individual gulls that are more focused on foraging in the terrestrial or marine environment. Biogeochemical processes vary extensively among habitats in the San Francisco Bay Estuary and influence MeHg incorporation into food webs (Eagles-Smith et al., 2016; Marvin-DiPasquale and Agee, 2003). The higher THg concentrations and corresponding  $\delta^{34}$ S values in some pre-breeding gulls (cluster 4) suggest that those gulls consume a higher proportion of marine prey outside of the breeding season. California gulls commonly forage on pelagic fish species in close association with fishing vessels over marine waters off of western North America in the fall (Vermeer et al., 1989; Wahl and Heinemann, 1979), in a food web that is naturally enriched in <sup>34</sup>S (Fry and Chumchal, 2011; Peterson and Fry, 1987). In contrast, the low THg concentrations and corresponding  $\delta^{34}$ S values in some gulls (cluster 1) suggest that these birds specialized on garbage outside of the breeding season as well. Obtaining fine-scale, GPS locations prior to future tissue sampling could further validate links between Hg bioaccumulation and foraging ecology (Fort et al., 2014; Peterson et al., 2015). Although, in general, breeding gulls no longer demonstrated a highly marine or estuarine diet, individual variability in foraging strategies was still evident. For example, certain gulls with higher THg concentrations likely supplemented their diet with eggs and chicks of locally-breeding waterbirds (Ackerman et al., 2006; Ackerman et al., 2014b,c), while others with lower THg concentrations likely consumed mostly garbage.

Although Hg contamination may be a useful ecological tracer of foraging ecology, not every situation is appropriate for its use and additional factors may need to be considered. In particular, life history can influence THg concentrations in animals and confound the interpretation of THg concentrations in animal tissues, in a similar manner to how life history can influence light stable isotope ratios (Hobson et al., 1993). For example, Hg contamination in rapidly growing animals can be difficult to interpret. Juvenile birds undergo rapid growth in size and production of feathers, which can dilute the body burden of Hg and make interpretation of Hg concentrations in juvenile birds difficult (Ackerman et al., 2011). We found that blood THg concentrations in gull chicks were markedly lower than adults, despite both the stable isotope values and isotope mixing models indicating that gull chicks had a higher estimated proportion of estuarine prey in their diet than adults. Additionally, the difference in THg concentrations between male and female California gulls during the breeding season is commonly observed in birds (Ackerman et al., 2016a, 2012, 2008, 2007) and underscores the importance of including sex in any interpretation. Females had lower THg concentrations than males at the same  $\delta^{34}$ S values, likely attributable to the ability of female birds to offload a portion of their Hg burden into their eggs (Ackerman et al., 2016a).

In conclusion, we found concordance among stable isotopes and Hg contamination in gull foraging ecology, demonstrating variability in diet and habitat use of coastally foraging California gulls. Some individual gulls foraged predominantly within landfills, while other gulls obtained a larger proportion of their diet from estuarine or marine habitats. For all gulls, a large proportion of their diet came from landfills, with pre-breeding adult gulls and late-season gull chicks acquiring the largest proportion of their diet from more natural, estuarine prey compared to breeding adults. The strong linkages between Hg concentrations and stable isotopes suggest that Hg contamination, often solely quantified to assess toxicological risk of organisms, may itself be a useful tool to examine animal foraging ecology.

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ecolind.2016. 11.025.

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