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Marine foraging ecology influences mercury bioaccumulation in deep-diving northern elephant seals

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Mercury contamination of oceans is prevalent worldwide and methylmercury concentrations in the mesopelagic zone (200–1000 m) are increasing more rapidly than in surface waters. Yet mercury bioaccumulation in mesopelagic predators has been understudied. Northern elephant seals (*Mirounga angustirostris*) biannually travel thousands of kilometres to forage within coastal and open-ocean regions of the northeast Pacific Ocean. We coupled satellite telemetry, diving behaviour and stable isotopes (carbon and nitrogen) from 77 adult females, and showed that variability among individuals in foraging location, diving depth and $\delta^{13}C$ values were correlated with mercury concentrations in blood and muscle. We identified three clusters of foraging strategies, and these resulted in substantially different mercury concentrations: (i) deeper-diving and offshore-foraging seals had the greatest mercury concentrations, (ii) shallower-diving and offshore-foraging seals had intermediate levels, and (iii) coastal and more northerly foraging seals had the lowest mercury concentrations. Additionally, mercury concentrations were lower at the end of the seven-month-long foraging trip ($n = 31$) than after the two-month-long post-breeding trip ($n = 46$). Our results indicate that foraging behaviour influences mercury exposure and mesopelagic predators foraging in the northeast Pacific Ocean may be at high risk for mercury bioaccumulation.

1. Introduction

Mercury, a non-essential trace element toxic to humans and wildlife [1,2], is widespread and increasing in the marine environment [3–6]. Although sources are both natural and anthropogenic, the overwhelming majority of mercury in the biota of remote marine regions, such as the Arctic, originates from human activities [5,7]. Anthropogenic atmospheric emissions occur in the form of inorganic mercury, including the gaseous elemental form (Hg^0), which can be oxidized to Hg^{II} and deposited on the ocean, where it subsequently can be converted to organic mercury (methylmercury) [8,9]. At this point, methylmercury can enter and biomagnify in oceanic food webs [5,10]. Models of global mercury cycling suggest a time lag of decades to centuries before current levels of anthropogenic emissions equilibrate between the atmosphere and ocean [11,12]. As a result, mercury concentrations in the world's oceans are expected to continue increasing [11,12].

Specific zones within the marine water column, including the epipelagic (0–200 m) and the mesopelagic (200–1000 m), differ with respect to biogeochemical cycling of mercury [9], which may have significant implications for bioaccumulation in marine predators. Increasing evidence indicates that the mesopelagic is a critical zone for entry of methylmercury into oceanic food webs, although specific mechanisms leading to oceanic mercury methylation and the subsequent integration into deep-ocean food webs are not yet fully understood [8]. The mesopelagic zone contains higher total mercury and methylmercury concentrations than either the epipelagic (0–200 m) or the zones below 1000 m [4,9,13,14], and over the past century mercury concentrations have increased

more rapidly in the mesopelagic zone than in the other oceanic zones. For example, methylmercury concentrations in water collected from the mesopelagic zone were higher in 2006 at all locations in the North Pacific than in previous studies [4]. Additionally, mercury in North Atlantic birds foraging on prey of mesopelagic origins experienced a 3.5–4.8% yr^{-1} increase in mercury concentrations over the last 100 years, a much faster rate of increase than that observed in shallower, epipelagic foraging seabirds (1.1–1.9% yr^{-1}) [3]. Mercury concentrations in large predatory fish sampled near Hawaii were highest in fish species that foraged within the mesopelagic and lowest in fish foraging in the epipelagic [14]. Despite the accumulating evidence that the mesopelagic has higher levels of mercury contamination, mercury exposure in top mesopelagic predators has been little studied. This could be problematic because mercury can adversely affect reproduction, development, behaviour and nervous system function in many organisms, and may be toxic even at low levels [15].

We followed known-age adult female northern elephant seals (*Mirounga angustirostris*) to quantify how variability in foraging behaviour, including geography, diving depth and stable isotopes (carbon and nitrogen), can explain mercury bioaccumulation in a mesopelagic predator. Specifically, we used variables to describe foraging locations and diving behaviour, in addition to stable isotopes, to determine what best explained the variability observed in mercury concentrations in blood and muscle. Additionally, we used the same variables to identify clusters of seals with similar foraging behaviours and examined if the clusters corresponded with overall differences in mercury concentrations. We studied the northern elephant seal because it is the only pinniped species (seals, sea lions and walrus) in the northeast Pacific Ocean that forages almost entirely on fish and squid in the mesopelagic [16–19], and their foraging range overlaps with more cryptic, mesopelagic marine predators that may also be vulnerable to methylmercury bioaccumulation, including cetaceans, sharks and tuna [20,21]. Importantly, adult females vary substantially both in diving behaviour (i.e. median foraging dive depth during the day can differ among individuals by nearly 300 m) and geographical location [16,17,22,23]. Annually, adult females undergo two foraging trips, upwards of 10 000 km over seven months (pre-breeding, gestational; hereafter long foraging trip) or 5000 km over two months (post-breeding; hereafter short foraging trip). Females come to shore after the long foraging trip to give birth and come to shore after the short foraging trip to moult (figure 1).

2. Material and methods

In order to relate mercury concentrations in adult female elephant seals to foraging behaviour at sea, we deployed satellite-transmitters and time-depth recorders at the start of either the short or long foraging trip [17], and non-lethally collected whole blood (hereafter referred to as blood) and muscle biopsies from all animals at the end of their foraging trip (figure 1). From 2011 to 2013, we sampled blood and muscle to represent mercury bioaccumulation over potentially different time scales, once each from 77 known-age (4–13 years) adult females. We sampled seals on average 9 days after arrival to the Año Nuevo colony (Año Nuevo State Reserve, San Mateo County, CA, USA) for breeding (5–6 days post-parturition; $n = 31$) and 2 days after arriving to the colony for the annual moult ($n = 46$). We used standard protocols to immobilize seals in order to attach or remove instruments, collect morphometrics and sample tissues [16,17,22].

We used published protocols to prepare tissue samples for mercury and stable isotope analysis [24–26]. We analysed blood and muscle samples for total mercury (Hg_T) because the mercury in these tissues from marine mammals is almost entirely methylmercury [27–29]. Tissue samples were analysed at the US Geological Survey Dixon Field Station Mercury Lab using a Milestone DMA-80 Direct Mercury Analyzer (Milestone, Shelton, CT, USA). Quality assurance measures during each batch included reference materials certified by the National Research Council of Canada, Ottawa, Canada (DORM-3, DOLT-3 or DOLT-4, and TORT-3), continuing calibration verifications, system and method blanks, and duplicate samples. Recoveries (mean ± s.e.) were 101.9 ± 0.7% ($n = 46$) for certified reference materials and 101.4 ± 0.9% ($n = 67$) for calibration verifications. Absolute relative percentage difference for duplicates averaged 4.1 ± 0.8% ($n = 53$). Mercury concentrations were generated using wet weight (ww) for blood samples and dry weight (dw) for muscle samples. We analysed red blood cells for $\delta^{13}C$ and $\delta^{15}N$, using a Carlo Erba Elemental Analyzer interfaced with a ThermoFinnigan Delta Plus XP mass spectrometer (Light Stable Isotope Lab, UC Santa Cruz, CA, USA), because they represent integrated diet over a period of weeks to months leading up to sampling [30,31]. The average experimental precision for isotope samples, calculated by averaging the standard deviation for the sets of in-house standards (Pugel) among all isotope runs, was 0.10‰ for $\delta^{13}C$ and 0.08‰ for $\delta^{15}N$.

We used time-depth recorders to determine if Hg_T concentrations varied with foraging depth or proportional use of the water column, and used satellite locations in order to determine if the geographical location of the foraging trip influenced Hg_T concentrations. Tracking and diving data were processed using standard filtering techniques and protocols [17]. All dive locations were georeferenced using the entire satellite track. We modified previously published dive type classification [17] and combined all active-bottom dives and V-shaped dives greater than 400 m as putative foraging dives, because jaw accelerometer tags deployed on elephant seals capture jaw motion events in 70–90% of all dives below 450 m [18], and an even higher percentage of V-shaped dives below 400 m (Y. Naito 2014, personal communication). Because elephant seals demonstrate a diel diving pattern [17], we assigned dives to day or night based on the solar zenith angle associated with each dive, and quantified median and 90th percentile depths for each individual separately for day and night. We quantified the percentage of the total dives that were benthic, to identify seals that spend a greater proportion of time foraging along the bottom. Additionally, we calculated an overall dimensionless dive index for each seal to quantify proportional use of the water column (0 = at the surface, 1 = on the seafloor), by dividing the maximum depth of each dive by bathymetry (using the ETOPO1 1 Arc-Minute Global Relief Model [32]) and then averaging all dives. We calculated all geographical variables using a satellite track that was linearly interpolated to one location every 8 h (three locations per day). Every location was assigned to a hydrographic ecoregion [23], including the California Current Upwelling Region, the Coastal Alaska Downwelling Region, the Subarctic Gyre and the North Pacific Polar Front (NPPF). The most common ecoregions were the California Current and the NPPF, but all seals must travel through the California Current to reach the NPPF. Because the proportions of time spent in these two ecoregions were negatively correlated, we only quantified the proportion of locations over the course of the foraging trip that occurred in the California Current to use as a geographical variable in the statistical analyses.

We set up two identical sets of candidate models to explain the variability in blood ($n = 77$ seals) and muscle ($n = 70$ seals) Hg_T concentrations, respectively, using general linear models.

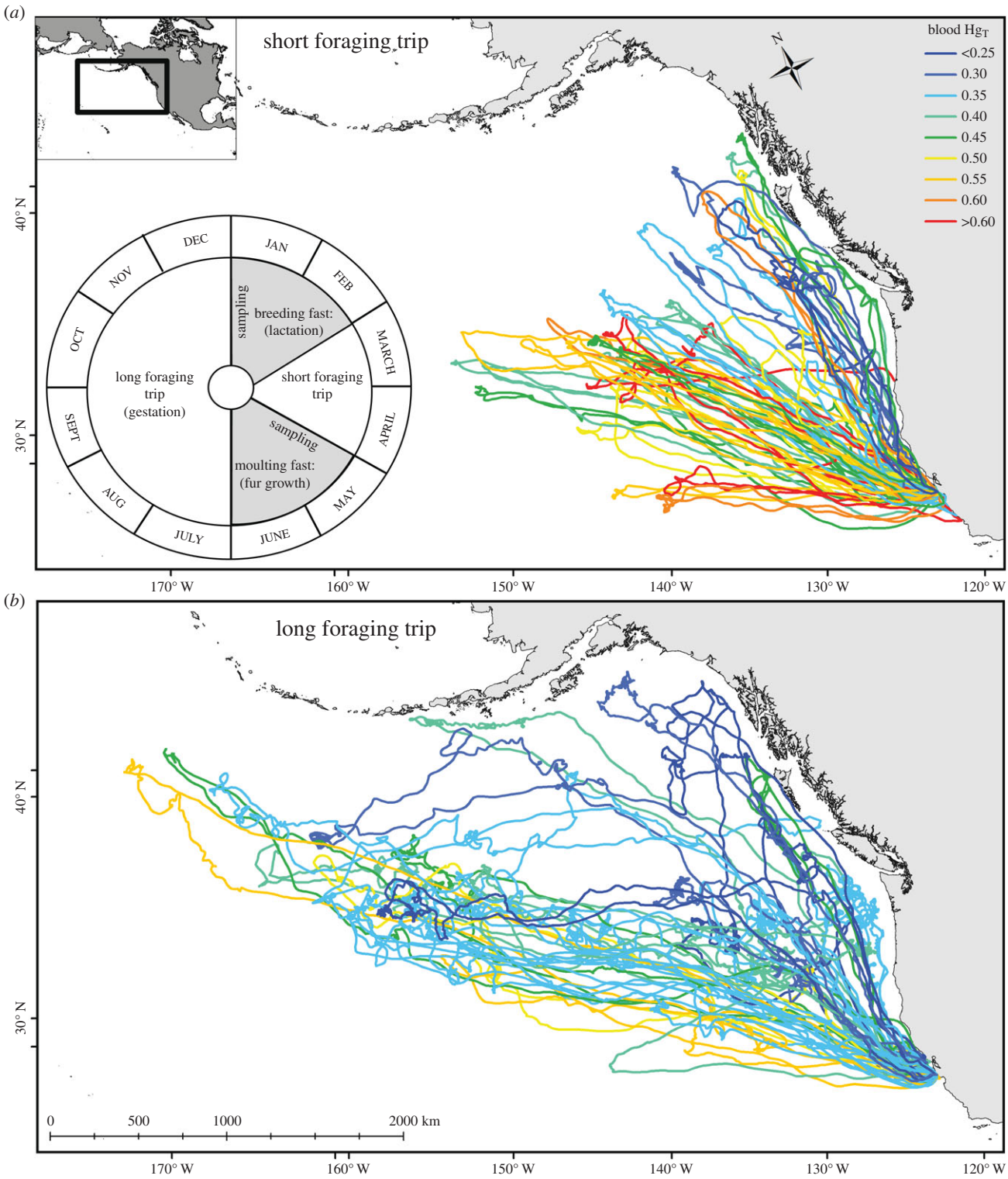

Figure 1. Blood Hg_T ($\mu g\ g^{-1}$ ww) in relation to foraging location and behaviour of adult female northern elephant seals (*M. angustirostris*) from the Año Nuevo colony, CA, USA. Seals were satellite tracked during (*a*) the short, post-breeding foraging trip ($n = 46$) and (*b*) the long, pre-breeding foraging trip ($n = 31$). The insets show 1 year in the life of adult females and the timing of sample collection. Gestation occurs during the long foraging trip. Note that we show periods of time that the majority of animals are ashore, although individual seals are ashore for less than the full period shown because seals arrive over a several-week period. These seals were at sea for a mean duration of 73 and 223 days for the short and long foraging trips, respectively.

The variables in the full model set included the foraging trip (short trip or long trip), seal age, median depth of foraging dives during the day (m), median depth of night foraging dives (m), 90th percentile depth of day foraging dives (m), 90th percentile of night foraging dives (m), maximum latitude obtained during the trip (°N), median distance of all seal locations during a trip to the continental shelf (km), median dive index (dive depth/ocean depth), proportion of time spent in the California Current ecoregion, the $\delta^{13}C$ value (‰) and the $\delta^{15}N$ value (‰). In order to determine if blood or muscle Hg_T concentrations were better explained using a more recent time scale of behaviour, we calculated all of the same diving and geographical variables using the 60 most recent days of data prior to sample collection for each foraging trip (on average, the short trips are 75 days and long trips are 220 days [17]).

We ran all possible combinations of the variables, except for a specified subset described below, in the statistical program R v. 3.0.2 [33]. Specifically, we did not allow models to contain

Table 1. Total Hg (mean ± s.d.) in whole blood (μg g^{-1} ww) and muscle (μg g^{-1} dw) of adult female northern elephant seals (*M. angustirostris*) sampled at the Año Nuevo colony (CA, USA), shown separately for the two foraging trips (short and long; figure 1). Females were clustered into three groups based on diving variables, geographical variables and stable isotope ratios (asterisks indicate variables important in distinguishing clusters). The clusters are referred to as northerly (1), shallower diving, offshore (2), and deeper diving, offshore (3).

variable	cluster 1 (northerly)	cluster 2 (shallower, offshore)	cluster 3 (deeper, offshore)	overall
blood Hg_T short foraging trip	0.36 ± 0.10 ($n = 19$)	0.45 ± 0.07 ($n = 19$)	0.56 ± 0.10 ($n = 8$)	0.43 ± 0.11 ($n = 46$)
blood Hg_T long foraging trip	0.30 ± 0.08 ($n = 11$)	0.36 ± 0.08 ($n = 18$)	0.45 ± 0.10 ($n = 2$)	0.35 ± 0.09 ($n = 31$)
muscle Hg_T short foraging trip	5.00 ± 1.52 ($n = 17$)	5.95 ± 1.41 ($n = 18$)	7.02 ± 1.94 ($n = 7$)	5.75 ± 1.67 ($n = 42$)
muscle Hg_T long foraging trip	3.86 ± 1.16 ($n = 11$)	5.12 ± 1.39 ($n = 16$)	5.77 ($n = 1$)	4.65 ± 1.41 ($n = 28$)
maximum latitude (°N)	51.5 ± 4.6*	46.0 ± 2.3*	42.9 ± 3.4*	47.8 ± 4.8
median distance to continental shelf (km)	278 ± 188*	962 ± 206*	936 ± 139*	687 ± 383
median day dive depth (m)	592 ± 38*	662 ± 28*	702 ± 30*	639 ± 52
90th percentile day dive depth (m)	710 ± 71*	818 ± 64*	829 ± 49*	777 ± 85
% benthic dives (%)	6.0 ± 4.1*	3.2 ± 1.4*	3.3 ± 2.2	4.3 ± 3.2
90th percentile night dive depth (m)	631 ± 41*	667 ± 59	722 ± 98*	660 ± 65
mean dive index (%)	22.2 ± 7.7*	13.7 ± 2.5*	15.4 ± 1.3	17.3 ± 6.4
$\delta^{13}C$ (‰)	−19.4 ± 0.4	−19.6 ± 0.2*	−19.1 ± 0.2*	−19.5 ± 0.3
$\delta^{15}N$ (‰)	14.6 ± 0.9	14.3 ± 0.5*	15.9 ± 0.9*	14.7 ± 0.9
median night dive depth (m)	516 ± 34	516 ± 42	571 ± 38*	523 ± 42

both $\delta^{13}C$ and $\delta^{15}N$ because these variables were highly correlated. We also did not allow variables calculated at different time scales (the entire trip duration versus the most recent 60 days) to appear in the same model. We used the Akaike information criterion corrected for small sample sizes (AIC_c) to rank candidate models, and considered candidate models for biological importance when $\Delta AIC_c \leq 2.0$ [34]. We calculated evidence ratios for each variable included in the top model, by dividing the Akaike weight of the top model by the Akaike weight of the same model without the variable of interest, which allows for comparison of the relative weight of support between models [34]. We also calculated the relative variable importance, which is the sum of the Akaike weights for all models containing the variable of interest, to compare the relative weight of support for different variables [35]. Foraging behaviour variables calculated over the final 60 days of a foraging trip did not fall within the top 95% of cumulative Akaike weights for blood and muscle analyses, clearly indicating that the quantification of foraging behaviour over the entire foraging trip better explained the variability in mercury concentrations than just quantifying the most recent behaviour. Thus, we report only results for models including variables calculated over the full foraging trip length.

Next, to identify unique clusters of seals based on foraging behaviour, including seals from both foraging trips, we used a combination of principal components analysis (PCA) and hierarchical cluster analysis using the FactoMineR package in R [36,37]. We used the same variables as previously described, except for the percentage of time in the California Current, and added the percentage of benthic dives, and both $\delta^{13}C$ and $\delta^{15}N$ values. The PCA produced unrotated factors and we used the eigenvalue threshold of 1.0 to retain principal components. We used PCA factor scores as input variables to the cluster analysis, which was run using hierarchical clustering on principal components using Euclidean distance and the average linking method [36]. Clusters were identified based on intra-cluster inertia [37]. We then separately tested blood and muscle Hg_T concentrations for broad-scale differences between the identified cluster groups, using AIC_c. We used AIC_c to compare five models, different only in how the seals were grouped using the three identified clusters (electronic supplementary material, table S1).

3. Results

We detected mercury in all blood and muscle samples collected from adult female elephant seals at the Año Nuevo colony from 2011 to 2013 (table 1). Upon return to land from a foraging trip, seals ranged in Hg_T from 0.18 to 0.65 μg g^{-1} ww in whole blood ($n = 77$), and from 1.90 to 10.15 μg g^{-1} dw in muscle ($n = 70$). The mean $\delta^{13}C$ value was −19.5 ± 0.3‰ (range −20.1 to −18.7‰) and the mean $\delta^{15}N$ value was 14.7 ± 0.9‰ (range 13.2 to 16.8‰).

The geographical location of the Año Nuevo colony causes all seals to spend at least a portion of the start and end of a foraging trip in the California Current as they transit to and from their at-sea foraging locations; however, the time spent in the California Current varied widely from less than or equal to 20% ($n = 19$) to more than or equal to 80% ($n = 8$; figure 1). Median depth of foraging dives ranged from 475 to 760 m during the day and 440 to 613 m at night, while the 90th percentile of foraging dives ranged from 641 to 1061 m during the day and 574 to 965 m at night (table 1).

(a) Blood mercury

Variability in blood Hg_T concentrations of seals could be explained by variability in foraging behaviour, age and whether the animal was sampled after the short or the long foraging trip. The most important variables to explain blood Hg_T included the 90th percentile of foraging dive depths

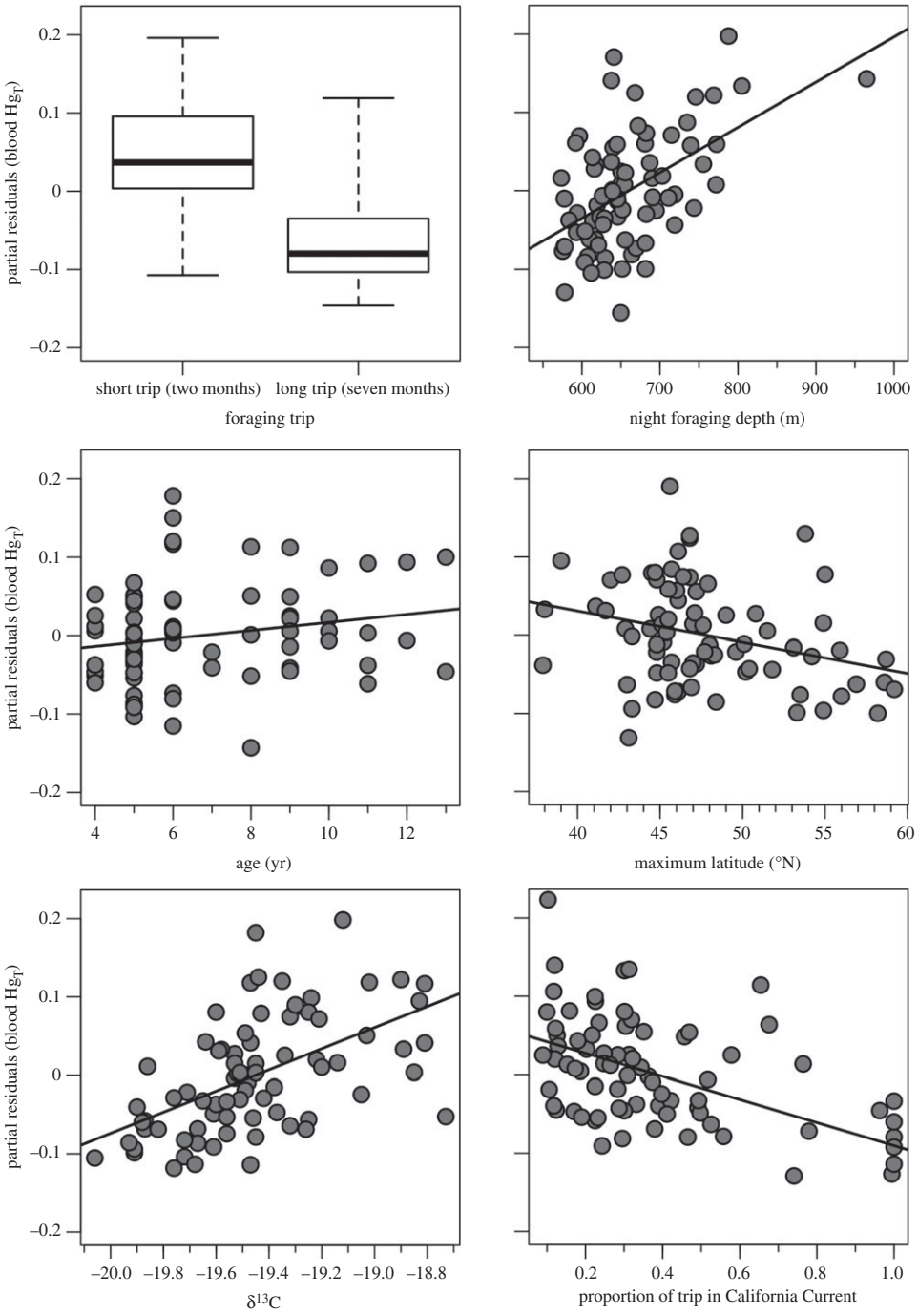

Figure 2. Panels show partial residual plots for each of the variables in the top model (each panel shows the relationship between blood Hg_T and an independent variable while accounting for all other independent variables in the top model) to explain blood Hg_T concentrations in adult female northern elephant seals (*M. angustirostris*) sampled at the Año Nuevo colony. The short foraging trip was post-breeding, whereas the long foraging trip was pre-breeding, when gestation occurs (figure 1). Night foraging depth was the 90th percentile of night foraging dive depths, maximum latitude was the furthest north location during the trip, $\delta^{13}C$ values were from red blood cells, and proportion of trip in the California Current was the proportion of time spent in that ecoregion.

during the night, the $\delta^{13}C$ value, the percentage of time in the California Current and the foraging trip (figure 2; electronic supplementary material, table S2). The top three models (within a ΔAIC_c of 2) all contained these four variables, with individual variable weights of more than 0.97, indicating their overriding importance. Removing one variable at a time from the top model and comparing this reduced model to the top model indicated that the top model was more than 6800 times more likely than similar models without each one of these four variables. By contrast, the maximum latitude was in the top three models but the top model was only 3.4 times more likely than the same model without maximum latitude. Additionally, the top model (adj-$r^2 = 0.65$; figure 2; electronic supplementary material, table S2) included age; however, age only had a variable weight of 0.51 and was only 1.1 times more likely than the same model without age. The median dive depth of foraging dives during the day was in the fifth-ranked model ($\Delta AIC_c = 2.26$) and was considered an uninformative parameter with a variable weight of 0.31. There was no support for the remaining variables. The top model was a significantly better fit than the null model, with a ΔAIC_c of 74.

Accounting for the other variables in the top model, blood Hg_T concentrations were $0.11 \pm 0.02\ \mu g\ g^{-1}$ ww higher in blood after the short compared with the long foraging trip. For every 100 m increase in the 90th percentile of foraging

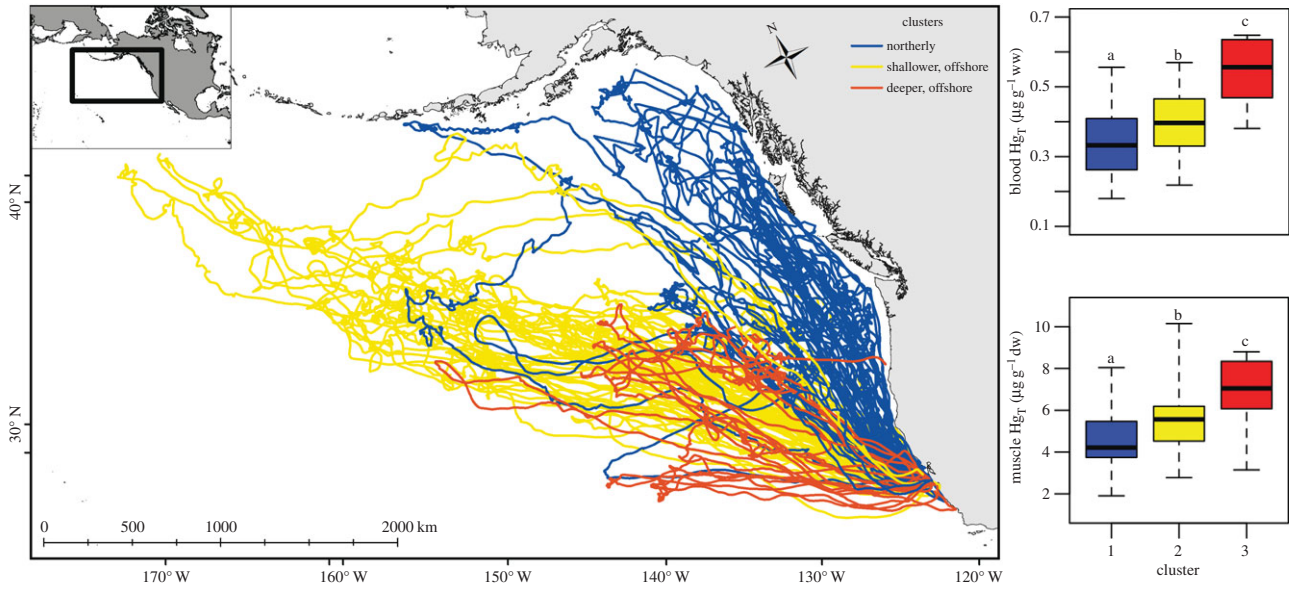

Figure 3. Hierarchical analysis on principal components, using geography, diving behaviour and stable isotopes (δ^{13}C and δ^{15}N) to describe foraging behaviour of satellite-tracked adult female northern elephant seals (*M. angustirostris*; $n = 77$) from the Año Nuevo colony, resulted in three clusters: blue (northerly; $n = 30$), yellow (shallower diving, offshore; $n = 37$) and red (deeper diving, offshore; $n = 10$). All three clusters included seals sampled after both the short and long foraging trips (figure 1), and had substantially different blood and muscle Hg_T concentrations.

dives at night, Hg_T concentrations increased 0.06 ± 0.01 μg g^{-1} ww. Concentrations of Hg_T also increased by 0.15 ± 0.03 μg g^{-1} per mil increase in δ^{13}C values (figure 2). Concentrations of Hg_T decreased by 0.04 ± 0.03 μg g^{-1} ww with each 10° increase in the maximum latitude reached during the foraging trip and decreased by 0.02 ± 0.01 μg g^{-1} ww for every 10% increase in the proportion of time spent in the California Current (figure 2). Lastly, Hg_T concentrations in blood increased with age at a rate of approximately 0.01 ± 0.01 μg g^{-1} ww per year.

Results for muscle analysis were similar to those observed in blood but had lower explanatory power than blood (lower adj-r^2 for the top model). The specific results for muscle are described in the electronic supplementary material.

(b) Cluster analysis for foraging strategies

Adult female elephant seals were grouped into three clusters based on foraging behaviour, using hierarchical clustering of factor scores from the three retained principal components (figure 3 and table 1). Seals in the first cluster ($n = 30$; hereafter northerly seals) had foraging trips closer to the continental shelf, were more northerly in maximum latitude, contained a higher proportion of benthic dives, had average dives that used a greater proportion of the water column, foraged shallower during the day and had shallower 90th percentiles of foraging dives during day and night than the overall mean values for all seals (figure 3 and table 1). The northerly cluster was not distinguished by isotope values. Seals in the second cluster ($n = 37$; hereafter shallower offshore seals) and the third cluster ($n = 10$; hereafter deeper offshore seals) foraged much further offshore than the northerly seals but differed between each other in terms of diving depth and isotope values. Mean δ^{13}C and δ^{15}N values were lower for the shallower offshore seals when compared with the deeper offshore seals (table 1). All three clusters included seals from both foraging trips (table 1).

We observed substantial differences in mercury concentrations between the three clusters of seals. The most parsimonious model for Hg_T concentrations in both blood and muscle included all three identified clusters as separate groups and had substantial weight (Akaike weight ≥ 0.88), whereas the next best model contained fewer than three separate clusters and had little weight (Akaike weight ≤ 0.08) and a $\Delta AIC_c > 6$ (electronic supplementary material, table S1). The northerly seals had the lowest median blood and muscle Hg_T concentrations of the three clusters (figure 3). Median Hg_T concentrations in blood and muscle concentrations were 19 and 35% higher in the shallower offshore seals than in the northerly seals. In turn, the deeper offshore seals had median Hg_T concentrations in blood and muscle that were 40% and 26% higher, respectively, than in the shallower offshore seals and 67% and 66% higher, respectively, than in the northerly seals (figure 3).

4. Discussion

We linked individual foraging behaviour with mercury concentrations of a top marine predator foraging in the mesopelagic. Individual foraging behaviour of adult female northern elephant seals substantially influenced mercury bioaccumulation, and seals could be broadly clustered into separate groups based on foraging behaviour. This indicates that individuals are not at equal risk to mercury exposure. We found that elephant seals that foraged offshore in the deepest parts of the mesopelagic had mercury concentrations in blood that were 40% higher than seals foraging offshore but at shallower depths, and 67% higher than seals foraging closer to the continental shelf, more northerly in latitude, and at shallower depths. Elephant seals foraged across a wide section of the northeast Pacific, overlapping with several sites that have been sampled for mercury within the water column. The lowest blood mercury concentrations from our study were from females that foraged further north, near the Subarctic Gyre, and the highest blood mercury concentrations in our study were from females that foraged further south, within the Transition Zone along the NPPF (figures 1 and 3). The

negative relationship we observed between blood mercury concentrations and maximum latitude in elephant seals (figure 2) corresponds with regional variability in mercury concentrations within water profiles [4,13]. The greatest mercury concentrations measured in water at sites to the north were between 200 and 400 m in depth, whereas the peaks of mercury measured at sites further south were between 500 and 800 m in depth, indicating that mercury concentrations at similar depths in the water column change substantially from north to south in the northeast Pacific [4,13]. Elephant seals typically forage deeper than 400 m, which may mean that seals in the Subarctic Gyre foraged deeper than the depths in the water column where the highest mercury concentrations occur, but seals further south foraged at the depths with the highest mercury concentrations. The high geographical fidelity of individual elephant seals to foraging areas [38,39] makes it highly probable that the same individuals would consistently accumulate more mercury while foraging.

In addition to geographical variability in water column mercury concentrations, individual diving behaviour strongly influenced mercury bioaccumulation. The majority of seals forage deeper during the day than at night, based on the vertical movement of prey within the water column [17]. However, some seals continued to dive to deeper depths during the night, and it was specifically these individuals that had the highest mercury concentrations. Because mercury distributions vary within the water column, seals that spent more time deeper in the water column may have foraged on a higher proportion of non-vertically migrating prey or prey migrating up in the water column at night from even deeper depths. These species could either contain greater concentrations of mercury because of their position within the water column or they could represent a higher trophic level.

Elephant seal carbon isotopes, but not nitrogen isotopes, were important to explain some of the variability in mercury concentrations. This suggests that the positive relationship between $\delta^{13}C$ values and mercury may be influenced more by a combination of oceanographic processes and latitude and less by trophic position. The region in the northeast Pacific where elephant seals foraged encompasses wide variability in $\delta^{13}C$ and $\delta^{15}N$ values at the base of the food web [40–42], which makes it difficult to directly compare isotope values to infer trophic position. Within the northeast Pacific, near shore in the California Current tends to be more enriched in ^{13}C and becomes depleted in ^{13}C moving offshore [43,44]; however, offshore ecoregions can vary significantly in $\delta^{13}C$ values as a result of both latitude and depth [40,45]. Elephant seals from all three clusters spent substantial periods of time outside of the California Current ecoregion. Elephant seals foraging outside of the California Current ecoregion that were enriched in ^{13}C could have been foraging on prey from a deeper food web because oceanographic and biological processes associated with increasing depth can cause deeper food webs to become enriched in ^{13}C [40,42,46], although it is also possible that these animals could have been foraging on prey from a higher trophic level.

The seasonality of life-history events may help explain the higher mercury concentrations we observed in blood and muscle after the short foraging trip (mean duration was 73 days) when compared with the long foraging trip (mean duration was 223 days). Mercury can be removed from circulation if it binds to tissue (like hair) that grows and subsequently becomes inert [47], and female elephant seals undergo an annual moult after the short foraging trip (figure 1). Mercury is detectable in elephant seal hair (S.H.P. 2014, unpublished data), with a mean concentration comparable with the highest mean hair concentrations documented for females of other free-ranging pinniped species [48,49], indicating that elephant seals annually offload substantial amounts of mercury into hair. Following the annual moult, female elephant seals return to the ocean for the long foraging trip, at which time gestation occurs. Maternal offloading of methylmercury in marine mammals occurs mostly via the placenta during gestation and to a much lesser extent during lactation [50,51]. For elephant seals arriving from the short foraging trip, there has been no recent maternal transfer of methylmercury and the greatest amount of time has elapsed since the prior moult (figure 1). Lastly, the increased mass and body condition (i.e. growth dilution effect due to increase in body mass) of seals at the end of the long foraging trip, in preparation for the extended lactation and fasting period associated with breeding, probably reduces mercury concentrations in internal tissues because changes in body condition can influence mercury concentrations in vertebrates [51,52]. Thus, the higher mercury concentrations we observed in seal blood and muscle after the short foraging trip (about 150 days shorter than the long foraging trip) were probably caused by decreased body mass (mass approx. 16% less) and the lack of ability to depurate mercury into developing offspring or through moult.

To our knowledge, the mean mercury concentration in the blood of female elephant seals ($n = 77$, 0.40 ± 0.11 μg g^{-1} ww) was the highest measured for any free-ranging pinniped species. Northern elephant seals bioaccumulated more mercury than their marine mammal counterparts that forage closer to the coast and within the neritic zone of the northeast Pacific. For comparison, mercury concentrations in the blood of adult females were 0.24 ± 0.21 μg g^{-1} ww (mean ± s.d.) for harbour seals (*Phoca vitulina*, $n = 27$, California, USA [49]), less than 0.30 μg g^{-1} ww for California sea lions (*Zalophus californianus*, $n = 19$, California, USA; S.H.P. 2014, unpublished data) and less than 0.36 μg g^{-1} ww, with one exception, for Steller sea lions (*Eumetopias jubatus*, $n = 30$, Alaska, USA; L. Rea 2014, personal communication). Additionally, mercury concentrations in the blood of female elephant seals were also substantially higher than in other marine mammals, such as female polar bears (*Ursus maritimus*) from the Alaskan Arctic (less than or equal to 0.21 μg g^{-1} ww, $n = 17$) [53] and sea otters (*Enhydra lutris*) from the northeast Pacific (less than or equal to 0.13 μg g^{-1} ww, $n = 20$) [54]. While fasting, lipid and muscle tissues are catabolized to fuel the energy demands of an animal, at which point mercury can move from muscle tissue into the bloodstream and increase blood mercury concentrations, as observed previously in northern elephant seal females [51]. As all of the blood samples in our study were from seals at the start of a fasting period, mercury concentrations would probably have been even greater during late fasting, which would only increase blood mercury concentrations in elephant seals, making them even higher than other free-ranging northeast Pacific pinnipeds.

We observed consistently high mercury concentrations across a range of ages, despite annual offloading of a portion of the mercury burden through reproduction and moult. This indicates that the mesopelagic is a significant and consistent source of mercury into these predators. Although blood mercury concentrations increased with age, the magnitude

of the effect was small, but suggests that female seals may not depurate or demethylate mercury at the same rate of ingestion. However, because females can reproduce every year until death [55], there is no post-reproductive period without maternal transfer of mercury during which time females would be even more vulnerable to bioaccumulation. Although mercury toxicity benchmarks for marine mammals are difficult to develop, 99% of elephant seals exceeded the prominently used clinical neurotoxicity threshold of 0.21 $\mu g\, g^{-1}$ whole blood for marine mammals [56,57], based on thresholds developed for humans [58].

Altogether, the high mercury concentrations we observed in elephant seals indicate that the mesopelagic zone in the northeast Pacific Ocean is an important source of mercury exposure to marine predators. Further, our study demonstrated that variability in individual foraging behaviours can significantly influence bioaccumulation of mercury, even within a single species. Mercury concentrations in the world's oceans are projected to increase even if anthropogenic mercury emissions are halted [11,12], thus furthering the risk of mercury exposure to predators foraging within the mesopelagic zone.

Ethics. We captured animals under NMFS permit 14636, and were approved for all procedures by the University of California, Santa Cruz Institutional Animal Care and Use Committee.

Data accessibility. The mercury concentrations in blood and muscle of individual seals, with the associated foraging behaviour data, are available through Dryad at http://dx.doi.org/10.5061/dryad.tc8j2. Raw tracking and diving data files can be available on request.

Authors' contributions. S.H.P. carried out the fieldwork and laboratory work, conducted data analysis, designed the study and drafted the manuscript. J.T.A. participated in laboratory work, assisted in the statistical analysis, participated in the design of the study and helped draft the manuscript. D.P.C. participated in fieldwork, participated in the design of the study and helped draft the manuscript. All authors gave final approval for publication.

Competing interests. We declare we have no competing interests.

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