Mercury Accumulation in Blubber and Skin from Stranded Bottlenose Dolphins (*Tursiops truncatus*) along the Florida and Louisiana Coasts (Gulf of Mexico, USA)

Meaghan A. McCormack[a*], Francesca Battaglia[b], Wayne E. McFee[c], Robert F. Young[b], Jessica Dutton[a]

[a]Department of Biology, Texas State University, Aquatic Station, San Marcos, TX 78666, USA

[b]Coastal Carolina University, PO Box 261954, Conway, SC 29528, USA

[c]National Centers for Coastal Ocean Science, National Oceanic and Atmospheric Administration, Charleston, SC 29412, USA

Meaghan McCormack: mmccormack@txstate.edu

Francesca Battaglia: fbattagli@coastal.edu

Wayne McFee: wayne.mcfee@noaa.gov

Robert Young: ryoung@coastal.edu

Jessica Dutton: jdutton@txstate.edu

Corresponding author*:

Meaghan McCormack

Department of Biology, Texas State University, Aquatic Station, San Marcos, TX 78666, USA

Email: mmccormack@txstate.edu

Abstract

Due to their long life-span and top trophic position, dolphins can accumulate high concentrations of mercury (Hg) in their tissues. This study measured the concentration of total Hg (THg) in the blubber and skin of bottlenose dolphins (*Tursiops truncatus*) that stranded along the Florida (FL) and Louisiana (LA) coasts and investigated the relationship of THg concentration in both tissues to sex, body length, age, stranding location, diet/trophic position ($\delta^{13}C$ and $\delta^{15}N$, respectively), and foraging habitat ($\delta^{34}S$). Additionally, we compared models using body length and age as explanatory variables to determine which was a better predictor of THg concentration. In both tissues, sex was not an influential predictor of THg concentration and there was a positive relationship between body length/age and THg concentration ($P < 0.001$). FL dolphins had higher mean blubber and skin THg concentrations compared to LA dolphins ($P < 0.001$). There was a modest improvement in model fit when age was used in place of body length, suggesting that while age is a better predictor of THg than body length, body length can be used as a proxy for age; however, body length is not a good proxy for dolphins that have reached asymptotic length. $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$ differed between locations and together with age were significant predictors of THg concentrations ($R^2 = 0.52$, $P < 0.001$). FL dolphins were $\delta^{13}C$ enriched compared to LA dolphins ($P < 0.001$) and THg concentrations were positively correlated with $\delta^{13}C$ ($R^2 = 0.22$, $P < 0.001$). Our results demonstrate spatial variability in THg concentrations from stranded bottlenose dolphins from the northern Gulf of Mexico; however, future research is required to understand how fine scale population structuring of dolphins from the northern Gulf of Mexico impacts THg accumulation.

Keywords: Odontoceti, Mercury, Bioaccumulation, Northern Gulf of Mexico, Stable isotope ratios

1. Introduction

Mercury (Hg) is a pollutant of great concern for both human and ecosystem health due to its toxicity and persistence in the environment (Selin, 2009). In marine systems, sulfate-reducing bacteria convert inorganic Hg (Hg^{2+}) to methylmercury (CH_3Hg^{2+}; MeHg) which enters the food web via phytoplankton and bioaccumulates in organisms and biomagnifies up marine food webs (Hammerschmidt and Fitzgerald, 2006; Fitzgerald et al., 2007). Due to their long-life span and top trophic position, dolphins can accumulate high concentrations of Hg in their tissues (Das et al., 2003; Stravos et al., 2007; Hong et al., 2013; Monterio et al., 2016). Mercury, particularly MeHg, can cause adverse neurological, behavioral, and reproductive effects in wildlife; therefore, it is important to monitor Hg levels in dolphins to determine potential threats to individual and population health, especially given that dolphins are long-lived species, which have few offspring; consequently, populations would be slow to recover from external threats (Scheuhammer et al., 2007).

In the Gulf of Mexico, certain fish species [e.g., golden tile fish (*Lopholatilus chamaeleonticeps*), king mackeral (*Scomberomours cavalla*), and Spanish mackerel (*S. maculatus*)] and bottlenose dolphins (*Tursiops truncatus*) have higher Hg concentrations compared to populations from the Atlantic Ocean (Hall et al., 1978; Stein et al., 2003; Adams & McMichael, 2007). While atmospheric wet Hg deposition rates along the Gulf of Mexico coast are amongst the highest in the U.S., the Atlantic Ocean—which delivers Hg via the Loop Current— is estimated to be predominant source of Hg to the Gulf of Mexico as a whole (National Atmospheric Deposition Program, 2007; Selin and Jacob, 2008; Harris et al., 2012). However, within the Gulf of Mexico, Hg is not evenly distributed, and some regions are more influenced by riverine (e.g., Mississippi River) and atmospheric Hg sources (Harris et al., 2012).

71 Spatial differences in Hg concentrations within the Gulf of Mexico have been reflected in
72 resident fish [e.g. red drum (*Sciaenops ocellatus*); red snapper (*Lutjanus Ceamechanus*)] and
73 oysters [e.g. American oyster (*Crassostrea virginica*)]; these differences may be carried up the
74 food web and reflected in the tissues of top predators such as bottlenose dolphins (Adams and
75 Onorato, 2005; Apeti et al., 2012; Sluis et al., 2013).
76 Bottlenose dolphins are the most abundant cetacean species in the Gulf and Mexico and are
77 broken up into inshore (e.g. bay, sound, and estuarine), coastal (up to 20-m isobath), continental
78 shelf (20 – 200 m depth) and oceanic (> 200 m depth) stocks (Vollmer and Rosel, 2013; Waring
79 et al., 2015). For management purposes, in the northern Gulf of Mexico, the Marine Mammal
80 Protection Act has designated 31 distinct bay, sound, and estuary and 3 coastal bottlenose
81 dolphin stocks (Vollmer and Rosel, 2013; Waring et al., 2015). In bays, sounds, and estuaries,
82 stocks are delineated according to observed residency patterns that in many locations have been
83 confirmed by photo-identification and/or tagging studies (Wells and Scott, 1990; Hubard et al.,
84 2004; Irwin and Würsig 2004; Balmer et al., 2008; Bassos-Hull et al., 2013; Wells et al., 2017).
85 Previous studies have identified that body length, age, sex, diet, and habitat can influence Hg
86 concentrations in dolphins; this warrants further investigation in the northern Gulf of Mexico
87 because inshore populations of bottlenose dolphins show strong habitat associations (Meador et
88 al., 1999; Stavros et al., 2007; Hong et al., 2012; Monterio et al., 2016; Damseaux et al., 2017)
89 Dolphins are primarily exposed to Hg through their diet, incorporating Hg, mostly in the
90 form of MeHg, from the muscle tissue of their prey (Hong et al., 2012). To understand
91 differences in Hg accumulation between dolphin populations, it is important to identify
92 differences in dietary sources and foraging habitats. Traditionally, dietary studies for dolphins
93 were limited to direct observation or analysis of stomach contents (Silva, 1999; Blanco et al.,

2001; Tollit et al., 2010). More recently, carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$), and sulfur ($\delta^{34}S$) stable isotope ratios in dolphin tissues have been used to obtain estimates of an individual's dietary carbon source, trophic position, and foraging habitat, respectively (Loesto et al., 2008; Newsome et al., 2010; Tollit et al., 2010). Therefore, stable isotope ratios may help explain variation in Hg concentrations between dolphin populations. Cetacean skin is a common tissue utilized in stable isotope studies, reflecting the diet, trophic position, and foraging habitat of an individual over a period of approximately 6 to 8 weeks and Hg concentrations in dolphin skin have been successful utilized to differentiate between populations (Browning et al., 2014; Dirtu et al., 2016; Damseaux et al., 2017; Hohn et al., 2017). While there have been several studies that report Hg concentrations in bottlenose dolphins from the Gulf of Mexico, particularly those focused on inshore populations along the Florida (FL) peninsula and Texas coast, there are no studies on Hg concentrations in dolphins from inshore populations along the FL panhandle and only one study which reported Hg concentrations in a single dolphin from Louisiana (LA) (Kuehl and Haebler, 1995; Meador et al., 1999, Stein et al., 2003; Bryan et al., 2007; Woshner et al., 2008; Damseaux et al., 2017). In addition, there are no bottlenose dolphin studies which analyze Hg in combination with stable isotope ratios in the Gulf of Mexico.

The objective of the study was to compare total Hg (THg) accumulation in stranded bottlenose dolphins along the FL panhandle and LA coasts. We investigated the influence of body length, age, sex, and stranding location on THg concentration in blubber and skin. In addition, we measured the $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$ stable isotope ratios in dolphin skin to determine if differences in dietary carbon source, trophic position, and foraging habitat could help explain variation in THg concentrations. Finally, general linear models (GLM) were used to determine whether body length or age is a better predictor of THg concentration.

2. Methods

2.1. Sample collection

In total, tissues samples from 185 bottlenose dolphins which stranded along the FL panhandle (n=64) and LA (n=121) coasts between 2011 and 2016 (Fig. 1) were collected by local stranding networks authorized by the National Oceanic and Atmospheric Agency (NOAA). Collecting samples from large numbers of dolphins is rare and was only possible due to the Northern Gulf of Mexico Cetacean Unusual Mortality Event (2010-2014). However, for many individuals, both tissues were not available for the present study [blubber: FL (n=48), LA (n=112); skin: FL (n=36), LA (n=93)]. At the time of sampling, total body length, sex, and decomposition code were recorded. Decomposition codes ranged from 2-6 with the majority (77%) being code 4 [Smithsonian Institution Coding System] (Geraci and Lounsbury, 2005). Samples were stored at -20° C at a NOAA facility and shipped to Texas State University where they were held at -20° C until THg analysis. Teeth were also collected at the time of necropsy.

2.2. Age Determination

Teeth were collected from the left lower mandible (generally teeth positioned at numbers 13-16 in the row), stored in 10% neutral-buffered formalin for up to 48 h, rinsed in tap water, and archived in 70% ethyl alcohol. Teeth were then prepared for sectioning using standard procedures (Myrick et al., 1983; Hohn et al., 1989). A 1-2 mm thick section (slab) was taken from each tooth of dolphins >140 cm body length. For dolphins with a body length of <140 cm, a slab was not taken, but rather decalcified whole and then thin sectioned. The slabs were cut

using a diamond wafer blade mounted on a Buehler Isomet low speed saw (Emerson Industrial Automation, Lake Bluff, IL), rinsed in tap water for approximately 6 h, and then decalcified in RDO (rapid decalcifying agent of acids; Apex Engineering Products Corporation, Aurora, IL) for 6-12 h based on the thickness of the slab. The slabs were then rinsed overnight and thin-sectioned on a Leica SM2000R sledge microtome (Leica, Inc., Nussloch, Germany) attached to a Physitemp freezing stage (Physitemp, Inc., Clifton, New Jersey). Thin sections were stained in Mayer's hematoxylin, blued for 30 s in a weak ammonia solution, dried on a slide, and mounted in 100% glycerin.

Sections were read three times using a Nikon SMZ1500 stereomicroscope (Nikon Instruments, Inc., Lewisville, Texas). At least one week elapsed between readings to eliminate bias. Teeth were aged based on Hohn et al. (1989); if two of the three readings were the same, that was used as the age estimate, whereas if differences between readings were <2 growth layer groups (GLG's), a fourth reading was made. Differences >2 GLG's required another tooth to be sectioned and the process repeated. Age estimates <1 GLG were calculated from measurements using SPOT Imaging software (Diagnostic Instruments, Inc., Sterling Heights, Michigan) while others >1 GLG were rounded to 0.50 GLG. Most teeth >5 GLG's were estimated to the last GLG.

2.3. THg Analysis

Samples were thawed, the skin (epidermis and dermis) was separated from the blubber using a ceramic knife, and the wet weight of each tissue recorded. Samples were then freeze dried (Labconco FreezeZone 2.5; Labconco, Kansas City, MO) for 48 h at -54°C and the dry weight recorded. The % water content (mean ± 1 SD) was 41 ± 15% and 47 ± 13% for blubber and skin,

respectively. Using a clean stainless-steel scalpel, both skin and blubber samples were cut into 4-5 mm pieces.

The THg concentration in blubber (10-15 mg) and skin (10-20 mg) was determined using a Direct Mercury Analyzer (DMA-80; Milestone Inc., Shelton, CT) using thermal decomposition, gold amalgamation, and atomic absorption spectrometry as described in EPA method 7473 (U.S. EPA, 2007). The DMA was calibrated using certified reference materials [CRM; MESS-4 marine sediment, 0.08 µg/g THg; TORT-3 lobster hepatopancreas, 0.292 µg/g THg; and PACS-3 marine sediment, 2.98 µg/g THg; National Research Council Canada (NRCC)] as needed. Quality control included blanks (n=70) and CRMs (n = 71) [DORM 4 (fish protein, NRCC, 0.412 µg/g THg) or ERM-CE464 (tuna fish, European Reference Material, 5.24 µg/g THg)] with every 10 samples analyzed. Blanks were below the detection limit (< 0.0000 µg/g THg) and the recovery values for the CRM/SRM was 96.8 ± 3.8% for DORM-4 (n = 41) and 98.7 ± 2.1% ERM-CE464 (n = 30). In addition, duplicates of blubber (n=26) and triplicates of skin (n = 133) samples were analyzed. The mean ± SD of the relative difference for blubber duplicates was 0.16 ± 0.14 (range = 0.001 – 0.55%); differences in oil content in blubber among individual dolphins may account for this wide range in relative percent differences. Triplicates of skin samples were analyzed because of the heterogenous condition of the samples and all triplicate samples had < 10% relative difference.

2.4. Stable Isotope Analysis

Freeze dried skin samples from FL (n=34) and LA (n=90) dolphins were lipid extracted using methanol and chloroform following the method described in Post et al. (2007) and cut into

184 approximately 1-2 mm pieces. Between 0.5-1.0 mg and 2.5-3.5 mg of each sample was weighed
185 and packaged into tin capsules for dual $\delta^{13}C/\delta^{15}N$ and $\delta^{34}S$ analysis, respectively. A duplicate
186 sample was included with every 20 samples analyzed. Stable isotope ratios were determined
187 using an elemental analyzer [$\delta^{13}C/\delta^{15}N$ (PDZ Europa ANCA-GSL); $\delta^{34}S$ (Elementar vario
188 ISOTOPE cube)] interfaced to a continuous-flow isotope ratio mass spectrometer [$\delta^{13}C/\delta^{15}N$
189 (PDZ Europa 20-20; Sercon Ltd., Cheshire, UK); $\delta^{34}S$ (SerCon 20-22 IRMS; Sercon Ltd.,
190 Cheshire, UK)] at the UC Davis Stable Isotope facility (Davis, CA). Results were expressed in δ-
191 notation using the following equation:

192 $$\delta_{Sample}(‰) = [(R_{Sample}/R_{Standard}) - 1] \times 1000$$

193 where R is the molar ratio of heavy to light isotopes (C^{13}/C^{12}, N^{15}/N^{14}, or S^{34}/S^{32}). The standards
194 used were Vienna Pee Dee Belemnite, atmospheric nitrogen, and Vienna-Canyon Diablo Troilite
195 for $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$, respectfully. To determine the analytical accuracy of $\delta^{13}C$ and $\delta^{15}N$,
196 replicate samples of bovine liver (nominal $\delta^{13}C = -21.7‰$, measured $\delta^{13}C = -21.7‰ \pm 0.08$;
197 nominal $\delta^{15}N = +7.72‰$, measured $\delta^{15}N = +7.63‰ \pm 0.07$; n=4), glutamic acid (nominal $\delta^{13}C$
198 $= -16.7‰$, measured $\delta^{13}C = -16.6‰ \pm 0.12$; nominal $\delta^{15}N = -6.8‰$, measured $\delta^{15}N = -$
199 $6.77‰ \pm 0.06$; n=11), enriched alanine (nominal $\delta^{13}C = +43.0‰$, measured $\delta^{13}C = +43.0‰ \pm$
200 0.13; nominal $\delta^{15}N = +41.41‰$, measured $\delta^{15}N = +41.1‰ \pm 0.06$; n=8), and nylon-6 (nominal
201 $\delta^{13}C = -27.7‰$, measured $\delta^{13}C = -27.8‰ \pm 0.04$; nominal $\delta^{15}N = -10.5‰$, measured $\delta^{15}N = -$
202 $10.5‰ \pm 0.06$; n=46) were analyzed. To determine the analytical accuracy of $\delta^{34}S$, replicate
203 samples of cysteine (nominal $\delta^{34}S = +32.2‰$; measured $\delta^{34}S = +34.2‰ \pm 0.28$; n = 25), hair
204 (nominal $\delta^{34}S = +2.7‰$; measured $\delta^{34}S = +2.8‰ \pm 0.32$; n = 63), Mahi-Mahi muscle (nominal
205 $\delta^{34}S = +19.5‰$; measured $\delta^{34}S = +19.5‰ \pm 0.25$; n = 71), whale baleen (nominal $\delta^{34}S = +$
206 $17.5‰$; measured $\delta^{34}S = +17.7‰ \pm 0.43$; n = 69), and taurine (nominal $\delta^{34}S = -2.5‰$;

measured $\delta^{34}S = -2.5‰ \pm 0.20$; n = 30) were analyzed. Duplicates (n=18) were run for every 20 samples and the relative % difference among duplicate samples was 5% for $\delta^{13}C$, 3% for $\delta^{15}N$, and 3% for $\delta^{34}S$.

2.5. Statistical Analysis

All data was explored for outliers and collinearity following Zurr et al. (2010). For both stranding locations, a one-way ANOVA was performed to determine if mean THg concentrations differed between blubber and skin tissues. One-way ANOVAs were also used to compare THg concentrations between stranding locations for both tissues. To compare THg accumulation between FL and LA dolphin populations, for both blubber and skin tissues, multiple linear regressions were used to determine the effect of sex (categorical, reference: female), stranding location (categorical, reference: FL), stranding year (categorical: reference: 2011), body length (continuous), and age (continuous) on THg concentration. Body length and age were not included in the same model due to the correlation between these two covariates. To assess potential differences in dietary carbon source, trophic position, and foraging habitat between FL and LA dolphin populations, multiple linear regressions were used to determine the effect of sex, stranding location, stranding year, stranding month (categorical: reference: January), and body length or age on $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$ in skin tissues. Multiple linear regressions were also used to describe the combined influence of body length or age and stable isotope ratios on skin THg concentrations. Month was included as a predictor in stable isotope models because, the isotopic values in dolphin skin reflects prey consumption from the previous 6-8 weeks (Browning et al., 2014); however, month was excluded from models in which THg

229 was the response variable because, Hg, particularly MeHg, which has an estimated biological
230 half-life of 1000 days in stripped dolphins (*Stenella coeruleoalba*), is retained in the body over
231 time (Itano and Kawai, 1981; Nigro et al., 2002).

232 Because the response variables (THg, $\delta^{13}C$, $\delta^{15}N$, or $\delta^{34}S$) could be influenced by several
233 predictors, an Akaike Information Criterion (AIC) model selection was used determine which
234 combination of explanatory variables best explained the variation in the response variable. To
235 account for small sample sizes, AIC_c was estimated. For each set of models, after all
236 combinations of explanatory variables were considered, an optimal model was chosen based on
237 the lowest AIC_c value (Akaike, 1974; Symonds and Moussalli, 2011). However, because the
238 penalty for one additional parameter is +2 AIC it is possible to obtain a competing model within
239 2 AIC units of the top model that differs in only one parameter. In some cases, the additional
240 parameter is uninformative and does not explain enough variation to justify inclusion in the
241 model (Arnold, 2010). Following Arnold (2010), all models were reported in the supplementary
242 data (Table S1-S12), but uninformative parameters ($P > 0.157$) were removed from the final
243 model reported in the text. A Gaussian distribution was applied as all response variables (THg,
244 $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$) were continuous variables. Models were validated by checking the
245 assumptions of normality and homoscedasticity through the visual inspection of residual plots; if
246 models failed to meet the assumptions data was Log_{10} transformed. Final models were compared
247 using R^2 values to determine whether body length or age best explained the variation in Hg
248 concentrations. A Gompertz growth curve was fit to age and body length data for each stranding
249 location to determine asymptotic growth at each location using the following equation:

250 $$\text{Body length} = \text{Asym}*\exp(-b2*b3^{x})$$

where asym = asymptote, b2 = the x axis displacement and b3 = growth rate (R core Team, 2018; R packages: nlme and nlshelper). Finally, to account for potential ontogenetic effects which could influence the interpretation of isotopic results a two-factor ANOVA was used to determine the effect of life stage and stranding location on stable isotope ratios. Life stage was categorized based on body length (calves ≤ 180 cm; juveniles/adults > 180 cm) and age (calves ≤ 3.5 years; juvenile/adults > 3.5 years) and analysis was performed using both categorization methods (Wells et al., 1987; Knoff et al., 2008). Data analysis was performed in R v.3.4.0 (R core Team, 2018) and the level of significance was set at $\alpha = 0.05$ for all analysis except for model parameter selection as described above.

3. Results

3.1. Blubber and skin THg concentrations

Mean blubber THg concentrations were significantly lower than mean skin THg concentrations in both FL (ANOVA; $P < 0.001$) and LA (ANOVA; $P = 0.005$) (Fig. 2). In FL dolphins, the mean ± SD blubber THg concentration was 2.36 ± 2.71 µg/g dry wt (range: 0.0378 – 13.9 µg/g dry wt) and the mean ± SD skin THg concentration was 4.36 ± 3.56 µg/g dry wt (range: 0.562 – 14.7 µg/g dry wt). In LA dolphins, the mean ± SD blubber THg concentration was 1.32 ± 2.78 µg/g dry wt (range: 0.0163 – 24.9 µg/g dry wt) and the mean ± SD skin THg concentration was 1.94 ± 3.51 µg/g dry wt (range: 0.0531 – 25.2 µg/g dry wt). There were significant differences in mean THg concentrations between FL and LA dolphins for both blubber (ANOVA; $P < 0.001$) and skin (ANOVA; $P < 0.001$).

3.2. Blubber and skin THg concentrations in relation to body length, age, sex, stranding year, and location

Overall, the bottlenose dolphins in the present study had a mean body length of 212 cm ± 52 (range: 74 – 285 cm). Dolphins that stranded in FL and LA had a mean body length of 205 cm ± 54 (range: 90 – 278 cm) and 216 cm ± 51 (range 74 – 285 cm), respectively. With regards to sex, there were 34 females, 27 males, and 3 unidentified individuals from FL and 32 females, 68 males, and 21 unidentified individuals from LA. Age was determined for 124 dolphins for which teeth were available. Dolphins ranged from < 1 month to 33 years old with the mean ± SD being 13.5 ± 9.85 years. A Gompertz growth curve provided a good fit to the data for both FL and LA dolphins (Fig. 3). In both locations, there was rapid increase in growth though age 5 followed by a decrease in growth until an asymptotic body length was reached (241 cm and 254 cm in FL and LA dolphins, respectfully).

For both blubber and skin, when using body length as a proxy for age, the optimal model based on the lowest AIC_c value included body length, stranding location, and stranding year as significant predictors of THg concentration. Sex was not an influential predictor of THg for either tissue. In both tissues, THg concentrations increased with body length, but there was greater variation in THg concentrations among larger (> 225 cm) individuals compared to smaller individuals (≤ 225 cm) (Fig. 4). When body length was used as a covariate, FL dolphins had higher THg concentrations compared to LA dolphins (Table 1). Stranding year was also an influential predictor of THg, but the influence of stranding year was likely driven by differences in the proportions of calves and juveniles/adults among sampling years. For both blubber and skin, THg concentrations were higher in 2016 compared to other samplings years; however, in

2016 only juveniles/adults were sampled whereas in all other years between 30-60% of sampled individuals were calves which have lower THg concentrations compared to juveniles/adults.

Similarly, when using age as a predictor, in both blubber and skin, THg concentrations significantly increased with age and FL dolphins had higher THg concentrations compared to LA dolphins when age was used as a covariate. For both tissues, sex was not a significant predictor of THg concentrations (Table 1). However, stranding year was not determined to be an influential predictor in age models. Since the influence of stranding year appeared to be driven by sample collection and was not significant in age models, stranding year was excluded in the multiple linear regressions shown in Fig. 5. For both blubber and skin models, there was slight improvement in model fit when age was used as predictor of THg concentration instead of body length (Table 1; Fig. 5). Overall, skin models explained more of the variation in THg concentrations than blubber models (Fig. 5).

3.3. Stable isotope ratios in relation to body length, age, sex, stranding location, month, and year

Optimal models based on the lowest AICc values revealed that sex, stranding year, and month were not influential predictors of $\delta^{13}C$, $\delta^{15}N$, or $\delta^{34}S$. Body length and age were significant predictors of $\delta^{13}C$ and $\delta^{15}N$, but not of $\delta^{34}S$ (Table 1). $\delta^{13}C$ was positively influenced by body length/age while $\delta^{15}N$ was negatively influenced by body length/age. Stranding location was a significant predictor of $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$ (Table 1). Overall, FL dolphins were $\delta^{13}C$ enriched (−15.5‰ ± 1.60), $\delta^{15}N$ deplete (+14.6‰ ± 1.21), and $\delta^{34}S$ deplete (+11.5‰ ± 3.04) relative to LA dolphins ($\delta^{13}C$ = − 17.2‰ ± 1.58; $\delta^{15}N$ = +16.5‰ ± 1.05; $\delta^{34}S$ = +13.3‰ ± 1.65).

316 To explore potential ontogenetic effects a two-way ANOVA was performed to determine
317 the effect of life stage and location on stable isotope ratios. Boxplots for isotopic data using body
318 length and age to categorize life stage are shown in Figure 6. For $\delta^{13}C$, there was no difference
319 between using body length and age. In both analyses, life stage and location were significant
320 predictors of $\delta^{13}C$ (ANOVA; $P < 0.01$). Florida dolphins were $\delta^{13}C$ enriched compared to LA
321 dolphins and calves were $\delta^{13}C$ deplete relative to juveniles/adults in both stranding locations. For
322 $\delta^{15}N$, when using body length to categorize life stage, there was a significant interaction term
323 which prohibited the statistical interpretation of the main effects (ANOVA; $P = 0.004$). It
324 appeared, based on the boxplots, that life stage influenced $\delta^{15}N$ in FL, but not in LA. However,
325 when age was used to categorize dolphins, the interaction term was not significant and only
326 location significantly influenced $\delta^{15}N$, with LA dolphins being $\delta^{15}N$ enriched compared to FL
327 dolphins. Similarly, for $\delta^{34}S$ analysis, when body length was used to categorized life stage there
328 was a significant interaction term and life stage appeared to be influential in FL, but not in LA.
329 However, when age was used to categorize life stage only stranding location was a significant
330 predictor of $\delta^{34}S$ with FL dolphins being $\delta^{34}S$ deplete relative to LA dolphins

331

332 *3.4. Skin THg concentrations in relation to stable isotope ratio and body length/age*

333 Together, $\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$, and body length/age were found to be significant predictors of
334 skin THg concentrations. $\delta^{13}C$, $\delta^{34}S$, and body length/age positively influenced THg
335 concentrations while $\delta^{15}N$ negatively influenced THg concentrations (Table 1). Individually,
336 there was no relationship between $\delta^{15}N$ and THg concentrations ($P = 0.01$; $R^2 = 0.04$) and $\delta^{34}S$

and THg concentrations ($P = 0.61$); however, there was a significant positive relationship between $\delta^{13}C$ and THg ($P < 0.001$; $R^2 = 0.22$) (data not shown).

4. Discussion

Within a species, THg concentrations can vary widely and several biological (e.g., tissue type, body length/age, sex) and ecological (e.g., foraging behavior, environmental THg concentrations) factors have been used to explain variation in THg concentrations (Meador et al. 1999; Hong et al., 2012; Monterio et al., 2016; Dirtu et al., 2016; Damseaux et al., 2017). Independently, THg concentrations and stable isotope ratios have been be used to distinguish between populations of bottlenose dolphins in the Gulf of Mexico (Barros et al., 2010; Wilson et al., 2012, 2013; Rossman et al., 2015, 2016; Hohn et al., 2017; Damseaux et al., 2017); however, the present study is unique in that it utilized both Hg and stable isotope analysis to better understand Hg accumulation in bottlenose dolphins from the northern Gulf of Mexico.

4.1. Mercury tissue distribution and comparison to other studies

Consistent with previously reported distribution patters, in both FL and LA dolphins, mean THg concentrations were higher in the skin compared to the blubber (Carvalho et al., 2002; Aubail et al., 2013; Borrell et al., 2015; Dirtu et al., 2016). Methylmercury, the predominant form of Hg found in marine organisms, demonstrates preferential binding to tissues rich in sulfhydryl groups, particularly muscle tissue (Bloom, 1992). Although MeHg is lipid soluble, it has a moderate octanal-water distribution coefficient (log Kow = 1.7-2.5) compared to other

lipophilic contaminants such as PCBs (log Kow = 6-7.5) (Halbach, 1985; Major et al., 1991; Metcalf and Metcalf, 1997). Therefore, because MeHg has a low octanal-water distribution coefficient and is preferentially distributed in muscle tissues, blubber generally has a lower THg concentration compared to other tissues (e.g., kidney, liver, muscle, and skin) (Cardellicchio et al., 2002; Carvalho et al, 2002; Aubail et al., 2013; Borrell et al., 2015). In contrast, cetacean skin which is composed of two layers: epidermis and dermis, has been shown to accumulate THg overtime and between 70-100% of the THg found in bottlenose dolphin skin is in the form of MeHg (Yang et al., 2002; Stravos et al., 2007, 2011; Woshner et al.,2008; Aubail et al., 2013; Borrell et al., 2015).

Blubber and skin THg concentrations were compared to THg concentrations in bottlenose dolphins reported in the literature (Table 2). There were no studies which reported blubber THg concentrations in bottlenose dolphins from the Gulf of Mexico or nearby Atlantic Ocean. In the present study, mean skin THg concentrations for FL bottlenose dolphins (4.4 ± 3.7 µg/g dry wt) were comparable to the mean skin THg concentration found in bottlenose dolphins from Sarasota Bay, FL (4.0 ± 2.9 µg/g dry wt), but higher than the mean skin concentrations reported in bottlenose dolphins off the South Carolina (SC) coast (1.7 ± 0.92 µg/g dry wt) (Bryan et al., 2007, Stavros et al., 2007, 2011). In contrast, the mean skin THg concentration found in LA dolphins in the present study (1.9 ± 3.5 µg/g dry wt), was lower than the mean concentration reported in Sarasota Bay, FL, but comparable to concentrations reported in dolphins off the SC coast (Bryan et al., 2007; Stavros et al., 2007; 2011). Annual wet Hg deposition across the northern Gulf of Mexico is greater than the wet deposition in SC which may explain the higher THg concentration found in FL dolphins (Selin and Jacob, 2008); however, wet deposition patterns do not explain why THg concentrations in dolphins from LA and SC are similar.

Both FL and LA bottlenose dolphins in the present study had lower mean skin THg concentrations compared to those reported in bottlenose dolphins from the Florida Coastal Everglades (11.1± 7.7 µg/g dry wt) and Indian River Lagoon (IRL), FL (7.0 ± 5.9 µg/g dry wt) (Stavros et al., 2007, 2011; Damseaux et al., 2017). Intermediate mean skin THg concentrations were reported for dolphins from the lower FL Keys (2.9 ± 2.1 µg/g dry wt) (Damseaux et al., 2017). Skin Hg concentrations reported by Damseaux et al. (2017) from bottlenose dolphins from the Florida Coastal Everglades are the highest in the literature; mangrove forests in this region are rich in organic content, supporting anaerobic bacteria which in turn facilitates the conversion of Hg^{2+} to MeHg that can incorporated into the food web (Bergamaschi et al., 2012). The IRL— a shallow estuary on the east coast of FL—has low flushing rates which allows for the accumulation of Hg (Smith, 1993).

Compared to blubber THg concentrations reported worldwide, both FL (2.36 ± 2.71 µg/g dry wt) and LA (1.32 ± 2.78 µg/g dry wt) bottlenose dolphins in the present study had mean blubber THg concentrations that were higher compared to those reported in the Northeast Atlantic Ocean (0.8 ± 0.7 µg/g dry wt), but lower than those reported in the Mediterranean Sea (2.54 ± 5.42 µg/g dry wt) and Canary Islands (83.4 ± 35.5 µg/g dry wt) (Roditi-Elasar et al., 2003; Aubail et al., 2013; García-Alvarez et al., 2015). It has been suggested that dolphins in the Mediterranean have elevated concentrations of THg compared to those in the Atlantic due to natural cinnabar deposits (Bacci, 1989, Andre et al., 1991; Cardellicchio et al., 2002; Pompe-Gotal et al., 2009). The authors did not provide an explanation for the exceptionally high THg values reported in blubber tissues from Canary Islands. In contrast, both FL and LA dolphins in the present study had mean skin THg concentrations which were lower than the Northeast Atlantic Ocean (5.7 ± 2.9 µg/g dry wt); however, similar to blubber, both FL and LA dolphins in

404 the present study had mean skin THg concentrations that were lower than those reported in the
405 Mediterranean Sea (7.9 ± 5.7 μg/g dry wt) (Roditi-Elasar et al., 2003; Aubail et al., 2013).

406

407 *4.3. Mercury concentrations in relation to body length, age and sex*

408 The present study found no difference in THg concentration between sexes, consistent
409 with the findings of previous studies (Woshner et al, 2008; Aubail et al., 2013; García-Alvarez et
410 al., 2015; Monterio et al., 2016). This is most likely because although Hg can be maternally
411 transferred via gestation and lactation, the amount of Hg is small compared to Hg derived from
412 dietary sources (Storelli and Marcotrigiano, 2000; Frodello et al., 2002; Hong et al., 2012).

413 The present study also found significant positive relationships between THg
414 concentration and body length/age in both the blubber and skin, supporting the findings of
415 previous studies (Yang et al., 2002; Stavros et al., 2007, 2011; Wosher et al., 2008; Aubail et al.,
416 2013; Borrell et al., 2015). The increase in THg concentration with increasing body length/age
417 likely reflects bioaccumulation as a result of continuous dietary exposure and the low excretion
418 rate of Hg, particularly MeHg, from the body (Itano and Kawai, 1981; Nigro et al., 2002);
419 however, it may also be a result of larger dolphins eating larger and/or higher trophic level prey
420 which inherently have higher THg concentrations [e.g. pinfish (*Lagodon rhombodies*) vs. spotted
421 seatrout (*Cynoscion nebulsosus*)] (Berens McCabe et al., 2010; Miller et al., 2011). Lower rates
422 of THg accumulation in smaller/younger dolphins may be explained by growth dilution (Andre
423 et al., 1991). The wide variation in THg concentrations among dolphins > 225 cm is likely
424 because after reaching asymptotic body length, individuals continue to age and accumulate THg
425 (Andre et al., 1991). Rapid growth followed by a period of slowed growth until an asymptotic

426 body length is reached is consistent with the literature (Read et al., 1993; Stolen et al., 2002;
427 McFee et al., 2012). This suggests that age may be a more accurate predictor of THg compared
428 to body length which was supported by the increase in model fit when using age as a predictor in
429 place of body length. However, the increase in model fit was not substantial and body length can
430 be a good proxy for age when age is not available, especially for smaller individuals before
431 asymptotic body length is reached (approximately 250 cm).

432

433 *4.4. Variation in THg accumulation between FL and LA dolphins*

434 On average, when body length or age was used as a covariate, dolphins from FL had
435 higher THg concentrations compared to dolphins from LA. Similar spatial patterns of THg
436 accumulation were found in American oysters tissues from the northern Gulf of Mexico; Apeti et
437 al. (2012) measured the THg concentrations in oyster tissues from the northern Gulf of Mexico
438 and found that oysters from certain regions of FL (Apalachee Bay, Florida Bay, Tampa Bay, the
439 Florida Everglades, and Pensacola Bay) had the highest the highest THg concentrations, whereas
440 oysters in Louisiana, Alabama and Mississippi had the lowest THg concentrations. The highest
441 median THg concentrations were reported in Apalachee Bay, FL which is included in the spatial
442 extent of the present study.

443 Lower THg concentrations in dolphins from LA may be a result of indirect influences
444 from the Mississippi River. The Mississippi River which drains 41% of the contiguous United
445 States delivers large amounts of sediments and nutrients to the central northern Gulf of Mexico
446 (Presley et al., 1998; Apeti et al., 2012). Large amounts of sediment dilutes atmospheric
447 deposited Hg with material that is lower in Hg concentration and large influxes of nutrients

448 support higher phytoplankton productivity which can reduce THg concentrations in fish through
449 growth dilution and lower concentrations of THg in fish can be reflected in dolphins (Presley et
450 al., 1998; Chen and Folt, 2005; Apeti et al., 2012). In addition, seasonally occurring hypoxic
451 zones which influence areas from the Mississippi delta west through upper coastal Texas can
452 release hydrogen sulfide in the sediment which inhibits Hg methylation (Benoit et al., 1999;
453 Rabalais et al., 2001; Fitzgerald et al., 2007, Sluis et a., 2013). In contrast, in the FL panhandle
454 there are no major rivers delivering sediments and nutrients to dilute atmospheric Hg inputs;
455 therefore, dolphins inhabiting FL would be expected to have higher THg concentrations
456 compared to dolphins from LA.

457

458 *4.5. Stable isotope ratios and relationship with THg concentration*

459 The isotopic values reported in the present study were within the ranges reported by other
460 studies in the northern Gulf of Mexico for bottlenose dolphin skin (Wilson et al., 2012; Wilson et
461 al., 2013; Hohn et al., 2017). However, categorizing dolphins by stranding location as proxy for
462 source stock is inherently flawed. This categorization failed to account for differences within
463 stranding locations. For example, stranded dolphins may include dolphins from different estuary,
464 barrier island, and coastal populations within FL and LA, which cannot easily be distinguished
465 from one another based on their morphology. It would be beneficial in future studies to
466 isotopically identify dolphins based on source stock and determine if Hg concentrations change
467 when moving from inshore to offshore habitats.

468 In marine systems, differences in $\delta^{13}C$ between benthic (i.e., high $\delta^{13}C$) and pelagic (i.e.,
469 low $\delta^{13}C$) producers can be carried up the food web, providing an indirect way to assess the

foraging habitat of a predator (Barros et al., 2010). Florida bottlenose dolphins were $\delta^{13}C$ enriched compared to LA bottlenose dolphins which may indicate that FL dolphins utilize a mixture of benthic/seagrass habitats as well as pelagic based food webs (Barros et al., 2010; Rossman et al., 2015). These findings are consistent with seagrasses distribution patterns in the northern Gulf of Mexico. Seagrasses are moderately present in western FL panhandle, but are less common in LA (Love et al., 2013). Additionally, LA dolphins may be receiving deplete dissolved organic carbon (DIC) from the Mississippi River (Chantam and Lewis, 1991). There was a decrease in $\delta^{13}C$ with increasing body length/age suggesting that there may be changes in foraging behavior between age classes (Rossman et al., 2015). The was a positive relationship between $\delta^{13}C$ and THg which may be driven by differences in $\delta^{13}C$ between stranding locations suggesting that FL dolphins are feeding on prey with higher THg concentrations compared to LA dolphins. However, sampling of dolphin prey in both locations would be necessary to support this hypothesis.

$\delta^{15}N$ is used as a measure of trophic position with more enriched $\delta^{15}N$ being associated with higher trophic positions, but differences in the organic matter sources within an ecosystem can confound the interpretation of trophic position (Wilson et al., 2009; Newsome et al., 2010; Wilson et al., 2012). If organic matter sources differ in $\delta^{15}N$ by more than 4‰, a consumers' nitrogen values are reflective of both isotopic fractionation and a mixture of organic matter sources (Wilson et al., 2009). Bottlenose dolphins from FL were $\delta^{15}N$ deplete (+ 14.61‰ ± 1.22) compared to LA dolphins (+ 16.45‰ ± 1.05); however, they differed less than the 3‰ which is approximately the difference between trophic levels (Peterson and Fry, 1987). Enriched $\delta^{15}N$ in LA may also be a result high levels of nutrient runoff (e.g. wastewater, fertilizers) from the Mississippi River (Valiela et al., 1997). Multiple linear regression analysis revealed there was a

493 decrease $\delta^{15}N$ with increasing body length/age which is consistent with calves transitioning from
494 nursing to independently foraging (Knoff et al., 2008). However, we did not see similar trends in
495 the ANOVA analysis when determining the influence of life stage and location on $\delta^{15}N$. We
496 categorized calves as being either ≤ 180cm or ≤ 3.5 years old, but calves may nurse for longer
497 periods of time which could have altered our results (Mann et al., 2000). Contrary to what was
498 expected, we did not find a positive relationship between $\delta^{15}N$ and THg concentration which
499 may be due to differences in $\delta^{15}N$ signatures between stranding locations and the wide variety of
500 prey consumed by bottlenose dolphins. Further research would be needed to sample the food
501 webs of both populations to better understand the relationship between $\delta^{15}N$ and THg. Finally,
502 our samples were from stranded animals, potentially adding a confounding factor to our results
503 as $\delta^{15}N$ values may be higher than expected in free ranging populations due to nutritional stress
504 and resultant catabolism of tissues (Payo-Payo et al., 2013).

505 In nearshore marine sediments, sulfate reduction results in^{34}S-deplete sulfides, between
506 the range of 0 to -20‰; these $\delta^{34}S$ products are then taken up by benthic producers and the $\delta^{34}S$
507 deplete signature is carried up the food web (Chanton et al, 1987; Chasar et al., 2005). In
508 contrast, seawater sulfate has a $\delta^{34}S$ value of +21‰ (Peterson et al., 1985). As a result, a
509 consumers' $\delta^{34}S$ is reflective of the relative importance of benthic and water column production.
510 Both FL and LA dolphins in the present study had $\delta^{34}S$ values consistent with nearshore habitats
511 dominated by benthic production. Florida bottlenose dolphins were $\delta^{34}S$ deplete compared to LA
512 dolphins which may reflect differences between estuary and barrier island stocks. Hohn et al.
513 (2017) utilized stable carbon, nitrogen and sulfur stable isotope ratios to assign stranded dolphins
514 from the northern Gulf of Mexico to source stocks. According to their results, most FL dolphins
515 from the present study would be assigned to estuary stocks whereas most of the LA dolphins

would be assigned to barrier island stocks. There was no relationship between $\delta^{34}S$ and body length suggesting there was no differences among age classes in foraging habitat. There was also no relationship between $\delta^{34}S$ and THg concentration suggesting that there were no differences in Hg accumulation between inshore and offshore foraging habitat; however, this interpretation is limited because most individuals from the present study $\delta^{34}S$ most appear to be from estuarine and barrier island stocks. Differences in stable isotope ratios and THg concentrations between FL and LA dolphins suggests that differences in dietary sources and foraging habitat influence THg concentrations in northern Gulf of Mexico bottlenose dolphins. However, further research is required to understand how fine scale population structuring of dolphins from the northern Gulf of Mexico impacts THg accumulation, particularly if THg accumulation differs between inshore and offshore populations.

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872 **Table 1**. Best fit generalized linear model (GLM) and parameter estimates selected based on
873 the lowest Akaike Information Criteria (AICc). *Indicates variables were Log10 transformed.
874

Final Model	R^2	p value
(1) Hg in relation to sex, body length and stranding location		
Blubber Hg* = − 5.8 + 2.5(Body Length*) + − 0.4(Stranding Location: LA) + 0.3(Year: 2016)	0.43	<0.001
Skin Hg* = – 4.8 + 2.3(Body Length*) + − 0.62(Stranding Location: LA) + 0.39(Year: 2016) + 0.25(Year: 2015) + 0.26(Year: 2014) + 0.17*Year: 2013	0.56	<0.001
(2) Hg in relation to sex, age, and stranding location		
Blubber Hg* = − 0.39 + 0.58(Age*) + − 0.34(Stranding Location: LA)	0.47	<0.001
Skin Hg* = 0.02 + 0.58 (Age*) + − 0.56(Stranding Location: LA)	0.63	<0.001
(3) Stable isotope ratios in relation to sex, body length, and stranding location		
δ^{13}C Skin = − 15.5 + 3.8(Body Length*) + − 1.7(Stranding Location: LA)	0.23	<0.001
δ^{15}N Skin = 20.0 + − 2.4(Body Length*) + 2.0(Stranding Location: LA)	0.40	<0.001
δ^{34}S Skin = 11.5 + 1.7(Stranding Location: LA)	0.11	<0.001
(4) Stable isotope rations in relation to sex, age, and stranding location		
δ^{13}C Skin = − 16.4 + 0.88(Age*) + − 1.5(Stranding Location: LA)	0.22	<0.001
δ^{15}N Skin = 14.6 + − 0.3(Age*) + 2.0(Stranding Location: LA)	0.45	<0.001

$\delta^{34}S$ Skin = 11.1+ 2.2(Stranding Location: LA)	0.11	<0.001
(5) Hg in relation to stable isotope ratios		
Skin Hg* = −1.5 + 1.9(Body Length*) + 0.11($\delta^{13}C$) + − 0.09($\delta^{15}N$) + 0.04($\delta^{34}S$)	0.47	<0.001
Skin Hg* = 2.3 + 0.48(Age*) + 0.10($\delta^{13}C$) + − 0.10($\delta^{15}N$) + 0.05($\delta^{34}S$)	0.52	<0.001

Table 1 continued

887 **Table 2**. Comparison of THg concentrations (mean ± SD) in bottlenose dolphin blubber and
888 skin between the present study and previously published studies in dry wt. *values were
889 converted from wet wt using a moisture content of 41% and 47% for blubber and skin,
890 respectively as determined in the present study.

Tissue	THg (µg/g dry wt)	n	Location	Reference
Blubber	83.4± 35.5	29	Canary Islands	García-Alvarez et al., 2015
	2.36 ± 2.71	48	Florida – panhandle	This study
	1.32 ± 2.78	112	Louisiana	This study
	2.54 ± 5.42*	14	Mediterranean Sea– Israel	Roditi-Elasar et al., 2003
	0.8 ± 0.7	16	Northeast Atlantic Ocean – Portugal and France	Aubail et al., 2013
Skin	11.1 ± 7.7	22	Florida – Coastal Everglades	Damseaux et al., 2017
	4.4 ± 3.7	36	Florida – panhandle	This study
	7.0 ± 5.9	75	Florida – Indian River Lagoon	Stavros et al., 2007
	8.6 ± 7.0	15	Florida – Indian River Lagoon	Stavros et al., 2011
	2.9 ± 2.1	9	Florida – Lower FL Keys	Damseaux et al., 2017
	4.0 ± 2.9*	40	Florida –Sarasota Bay	Bryan et al., 2007
	1.9 ± 3.5	93	Louisiana	This study
	7.9 ± 5.7*	13	Mediterranean Sea– Israel	Roditi-Elasar et al., 2003
	5.7 ± 2.9	16	Northeast Atlantic Ocean – Portugal and France	Aubail et al., 2013

	1.7 ± 0.92	74	South Carolina – Charleston	Stavros et al., 2007
	1.8 ± 1.8	12	South Carolina	Stavros et al., 2011

Table 2 continued

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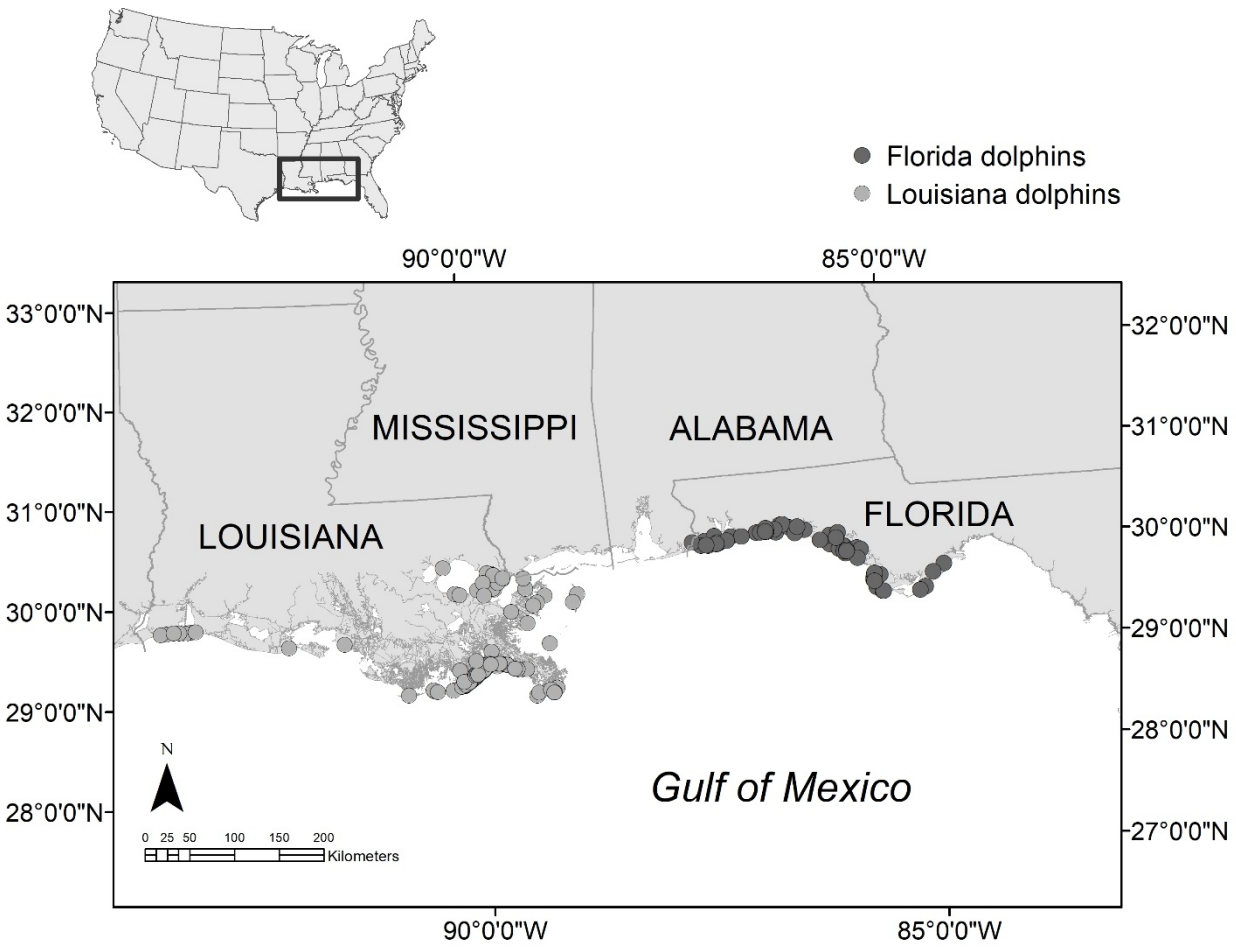

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904

905 **Figure 1**. Bottlenose dolphin (*Turisops truncatus*) stranding locations in Florida (FL; n = 64) and

906 Louisiana (LA; n = 121).

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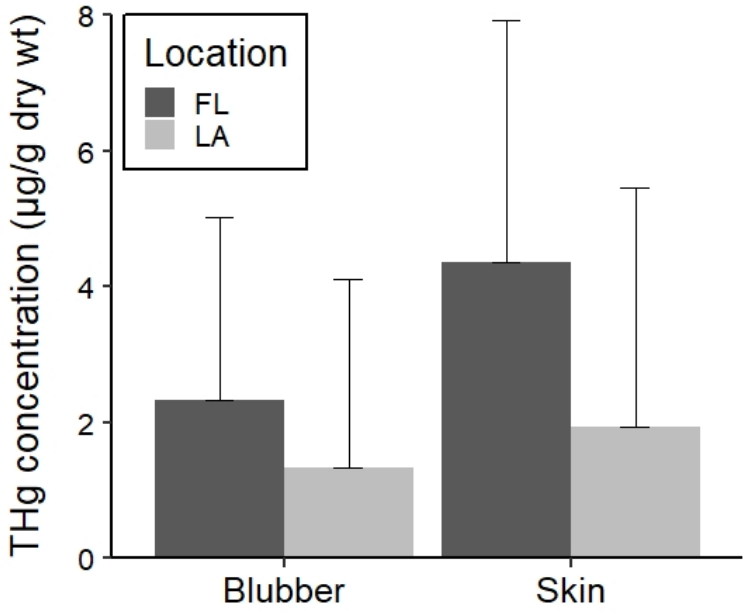

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911

912 **Figure 2**. THg concentration (mean + SD) in blubber and skin of Florida (FL) and Louisiana

913 (LA) bottlenose dolphins.

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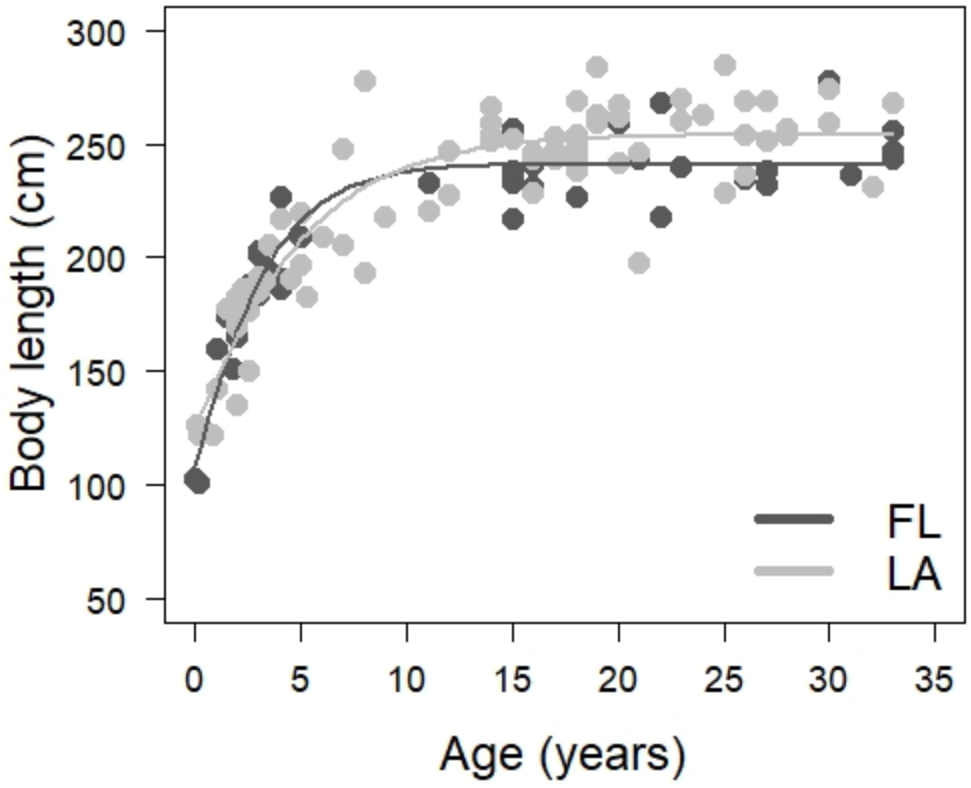

917

918 **Figure 3**. Relationship between body length and age in Florida (FL) and Louisiana (LA)

919 bottlenose dolphins with growth curves fitted using the Gompertz model.

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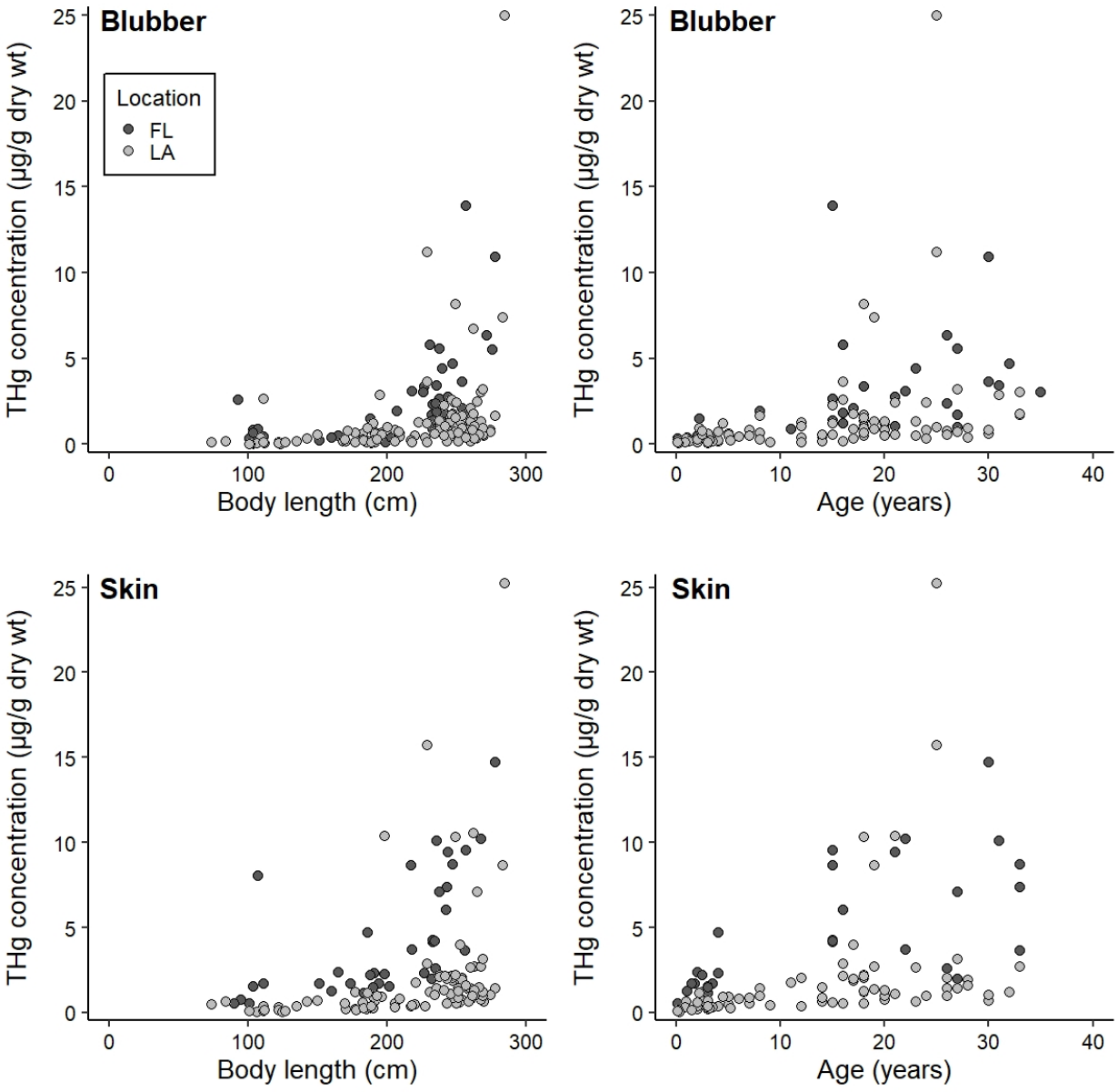

Figure 4. Relationship between THg concentrations in blubber and skin of bottlenose dolphins from Florida (FL) and Louisiana (LA) in relation to body length (left column) and age (right column).

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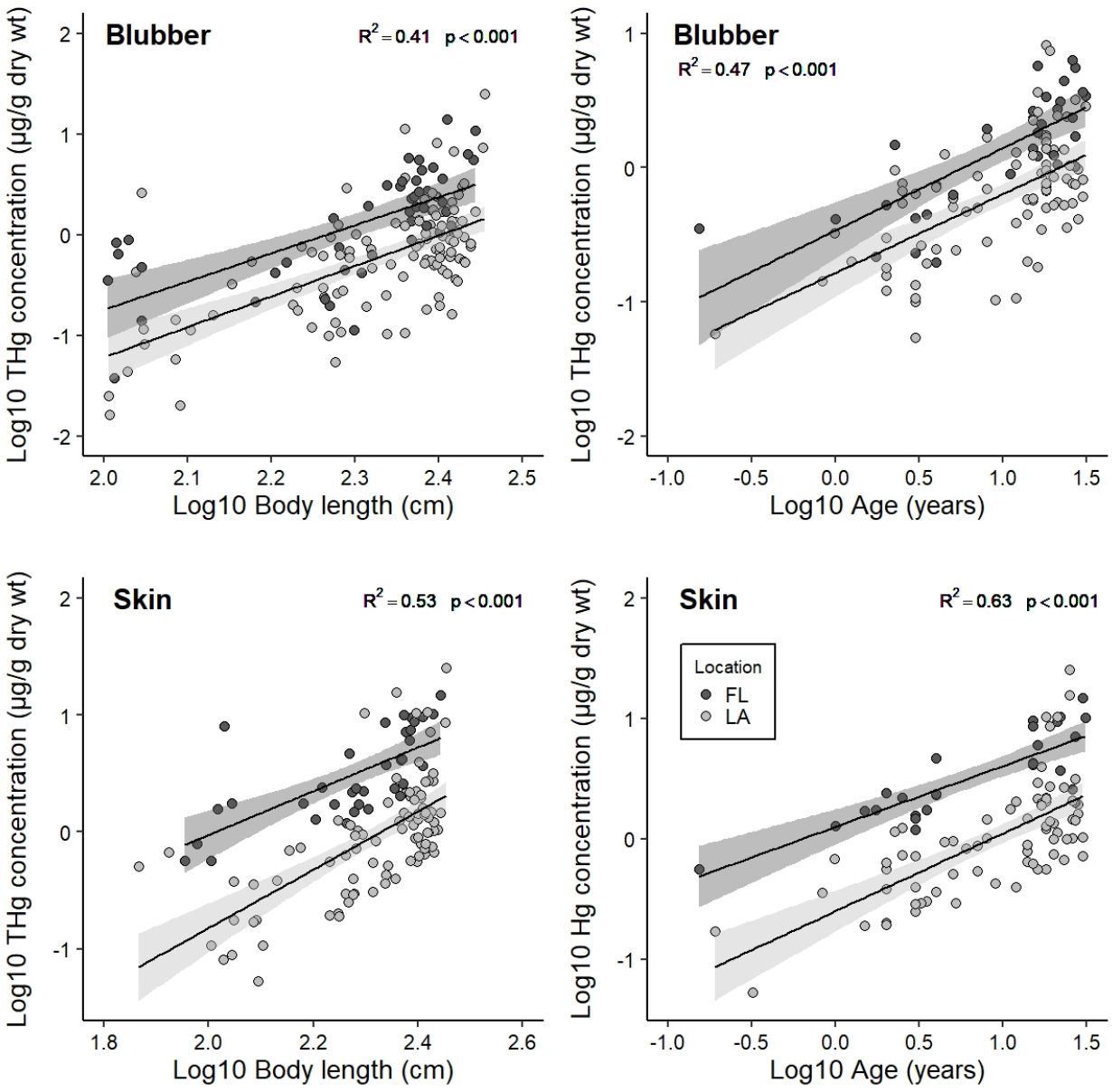

928

929 **Figure 5**. Relationship between Log10 THg concentrations in blubber and skin and Log10 body
930 length (left column) and Log10 age (right column). Regression lines (Log_{10} Hg = β_0 + Stranding
931 Location + Body length/Age) and 95% CI are shown.

932

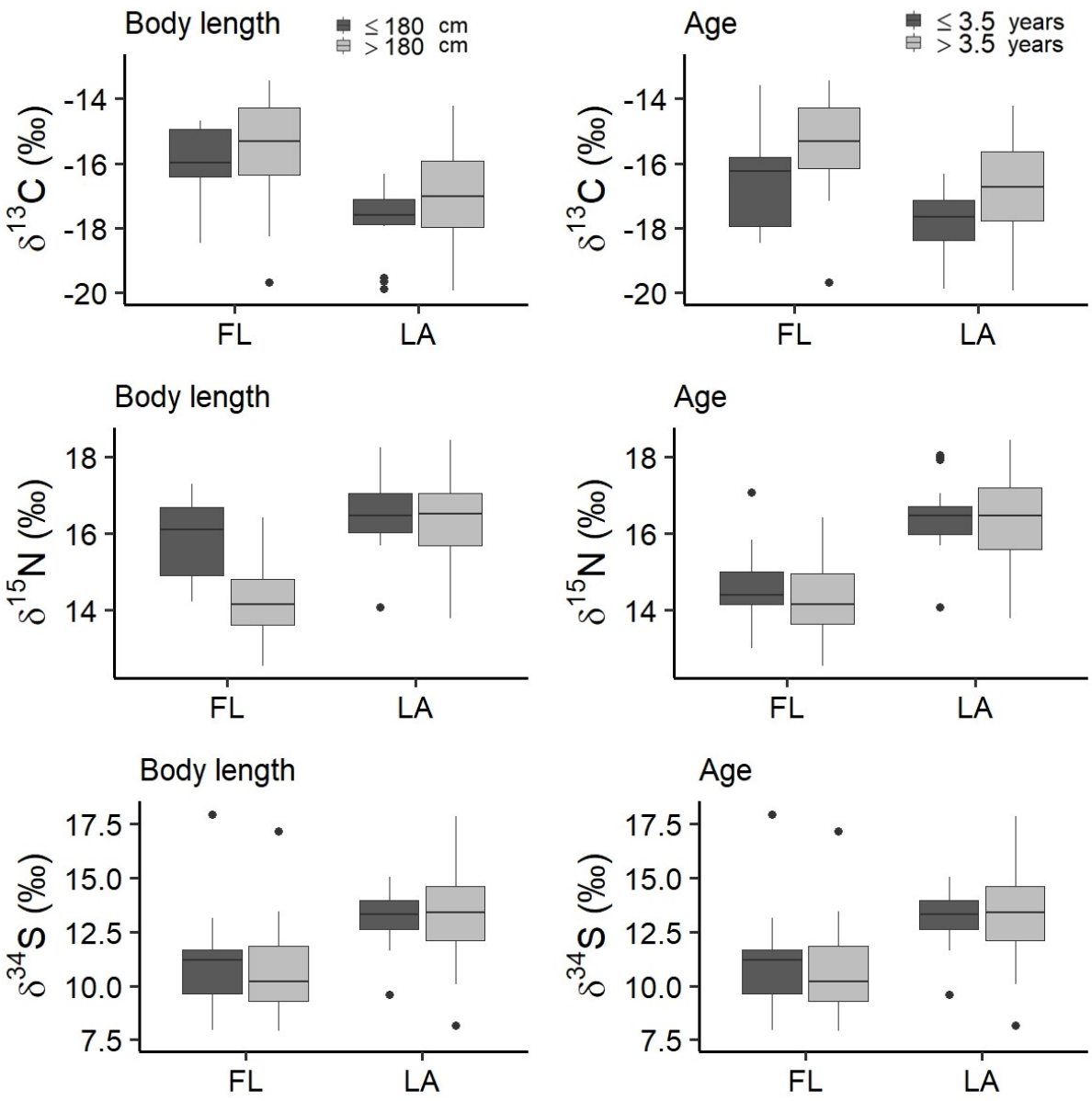

Figure 6. Florida (FL) and Louisiana (LA) bottlenose dolphin $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$ values in relation to body length (left column) and age (right column). Dark gray boxes represent calves (≤ 180 cm or ≤ 3.5 years) and light gray boxes represent juveniles/adults (>180 cm or >3.5 years). Whiskers show the minimum and maximum values, excluding outliers which are shown as black dots.