Mercury and selenium concentrations, and selenium:mercury molar ratios in small cetaceans taken off St. Vincent, West Indies

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ABSTRACT

This study measured the concentration of total mercury (THg) and selenium (Se), and calculated the Se:Hg molar ratios in the muscle, blubber, liver, and kidney of small cetaceans (false killer whale, *Pseudorca crassidens*; killer whale, *Orcinus orca*; Risso's dolphin, *Grampus griseus*; short-finned pilot whale, *Globicephala macrorhynchus*; and dolphins of the genus *Stenella*) taken for human consumption off St. Vincent, West Indies. Overall, 122 samples were analyzed; mean THg concentrations (μg/g dry weight) were highest in the liver (730), followed by the kidney (274), muscle (76.4), and blubber (4.57). To explain variability in muscle THg concentrations, carbon (δ^{13}C) and nitrogen (δ^{15}N) stable isotope ratios were analyzed to explore differences in dietary carbon source and relative trophic position, respectively, among species. There was no relationship between δ^{15}N and THg concentration, but there was a positive relationship between δ^{13}C and THg concentration. On average for each species, the Se:Hg molar ratios were > 1 in blubber and < 1 in muscle. All liver samples and the majority of kidney, muscle, and blubber samples exceeded the FAO/WHO human consumption advisory level of 1 μg/g wet weight. Based on our estimations, consuming only 6.6 g of muscle a week would exceed the MeHg provisional tolerable weekly intake of 1.6 μg MeHg/kg body weight/week for a 60 kg person. Given the high THg concentration in these cetaceans and the frequency at which these tissues are consumed, this is a potential human health issue that warrants further investigation.

1. Introduction

Mercury (Hg) is a toxic nonessential trace element that is released into the environment from both natural (volcanic activity, geothermal sources, and topsoil) and anthropogenic (coal-fired power plants and small artisanal gold mining operations) sources (Pirrone et al., 2010). In the marine environment, inorganic Hg (Hg^{2+}) can be converted to methylmercury (CH_3Hg^+; MeHg), primarily by sulfate-reducing bacteria in the sediment, which is readily assimilated into marine phytoplankton and transferred to higher trophic levels (Hammerschmidt and Fitzgerald, 2006; Fitzgerald et al., 2007). Mercury, particularly MeHg, bioaccumulates in marine organisms and biomagnifies up marine food webs; as a result, long-lived, high trophic level predators such as odontocetes (toothed cetaceans) can accumulate high concentrations of

Hg in their tissues (Endo et al., 2002, 2005; 2006; Kershaw and Hall, 2019).

Human exposure to Hg largely results from the consumption of contaminated seafood, which can have deleterious neurological, cardiovascular, immunological, and developmental effects (Rice et al., 2014). In response to these health concerns, the Food and Agricultural Organization of the United Nations (FAO) and the World Health Organization (WHO) have recommended that people avoid the frequent consumption of predatory species (e.g., sharks, swordfish, and tuna) with Hg concentrations > 1 μg/g wet weight (wet wt) (FAO/WHO, 2011). Odontocetes are consumed by several coastal communities and island nations around the world (Robards and Reeves, 2011), and while the FAO/WHO advisory does not specifically address the consumption of odontocetes, Hg concentrations in odontocete tissues have been

reported as high as 81 μg/g wet wt, 81-times higher than the 1 μg/g wet wt FAO/WHO recommendation (Endo et al., 2005).

Currently, Hg consumption advisories are based on the concentration of Hg alone, but several studies have proposed that the selenium:mercury (Se:Hg) molar ratio can also be used in risk assessments. Selenium (Se), an essential element, if present in molar excess, may have a protective effect against Hg toxicity through the formation of a mercury selenide (HgSe) complex (Kaneko and Ralston, 2007; Ralston, 2008; Berry and Ralston, 2008; Peterson et al., 2009; Ralston and Raymond, 2010). However, due to the high intra- and interspecies

variability in the Se:Hg molar ratios, other studies have questioned the reliability of using Se:Hg molar ratios in risk assessments and have argued that before ratios can be used, more research is needed to understand the interaction between Hg and Se, and the relationship between the molar ratio and human health (Burger et al., 2012, 2013; Burger and Gochfeld, 2011, 2013). While the majority of studies have focused on the Se:Hg molar ratios in commonly consumed fish (Kaneko and Ralston, 2007; Burger and Gochfeld, 2013; Polak-Juszczak, 2015; Azad et al., 2019), other studies have investigated the Se:Hg molar ratio in odontocetes (Endo et al., 2002, 2006; Seixas et al., 2008; Cáceres-
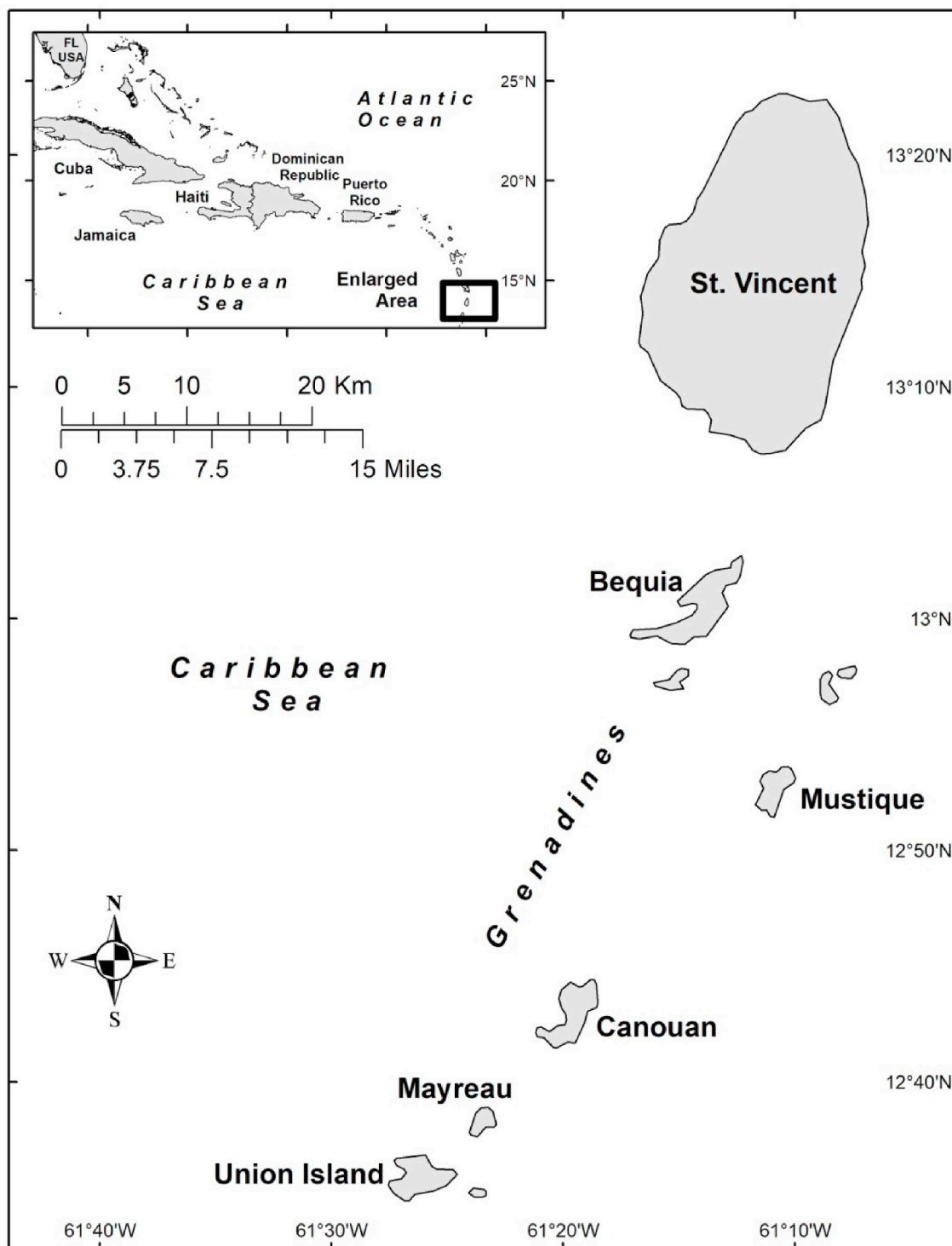

Fig. 1. Map showing the inhabited islands of St. Vincent & the Grenadines, along with the country's location in the Eastern Caribbean.

Saez et al., 2018).

Odontocetes are primarily exposed to Hg through their diet and differences in foraging habitats and dietary sources can lead to variability in Hg concentrations both within and among species (Das et al., 2003b; Capelli et al., 2008; Hong et al., 2012). An individuals' dietary carbon source and relative trophic position can be estimated by the determining the carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope ratios, respectively, in odontocete tissues (e.g., muscle, skin) (Newsome et al., 2010; Kiszka et al., 2011; Mèndez-Fernandez et al., 2012). Stable isotope ratios, particularly $\delta^{15}N$, have been increasingly utilized to trace contaminants, including Hg, through the food web, as $\delta^{15}N$ increases with trophic level (Deniro and Epstein, 1981; Dehn et al., 2006; Dirtu et al., 2016).

In St. Vincent & the Grenadines, an archipelagic nation in the Eastern Caribbean, Vincentian whalers take a variety of small cetaceans not protected by the International Whaling Commission (IWC) for human consumption, with the short-finned pilot whale (*Globicephala macrorhynchus*) being the main target of the whaling operation (Fielding, 2018). Muscle, blubber, kidney, and liver from small cetaceans are all consumed regularly in St. Vincent & the Grenadines. The muscle and blubber are sold in established markets and by mobile vendors throughout the island (Fielding, 2013); whereas liver and kidney are distributed through less formal networks. While some data exists on the frequency of muscle and blubber consumption (Fielding, 2013), no data is available on the frequency of liver and kidney consumption.

A pilot study by Fielding and Evans (2014) determined the concentration of total Hg (THg) in muscle and blubber tissue from spinner dolphins (*Stenella longirostris*; 1.57 and 1.42 µg/g wet wt, respectively) and Atlantic spotted dolphins (*Stenella frontalis*; 1.14 and 0.92 µg/g wet wt, respectively) taken by the St. Vincent whaling operation in 2009. Only one other study has investigated the concentration of contaminants in cetaceans taken for human consumption in the Eastern Caribbean. Five short-finned pilot whales and two *Stenella* spp. taken from the neighboring island of St. Lucia had a combined species mean THg concentration of 3.16 µg/g wet wt in the muscle, 7.49 µg/g wet wt in the kidney, and 66.1 µg/g wet wt in the liver (Gaskin et al., 1974). As a result of the limited data, expected high Hg concentrations in cetaceans, and the importance of the artisanal whaling operation in St. Vincent, this detailed study is warranted.

The objective of this study was to determine the THg and Se concentrations, and the Se:Hg molar ratios in muscle, blubber, kidney, and liver from several species of small cetaceans [false killer whale (*Pseudorca crassidens*), killer whale (*Orcinus orca*), Risso's dolphin (*Grampus griseus*), short-finned pilot whale, and unidentified dolphins (*Stenella* spp.)] taken for human consumption from waters near St. Vincent. In addition, the intra- and interspecies variability in THg concentration in muscle tissue was evaluated using carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope ratios to determine the influence of foraging habitat and relative trophic position, respectively, on observed THg concentrations.

2. Materials and methods

2.1. Sample collection

Muscle, blubber, liver, and kidney samples were collected between August 2015 and July 2016 from small cetaceans caught for human consumption by artisanal whalers operating off St. Vincent (Fig. 1). Samples were collected throughout the year, depending on the availability of carcasses that were landed on the beach, by a local assistant. Permission was obtained from the vendors who processed and sold the cetacean products prior to sampling. Due to the collection taking place at various stages of the processing, it is difficult to estimate the number of individual cetaceans from which we took samples. We are certain, however, that there are no duplications in our sample set; that is to say that no two samples of the same tissue type came from the same individual cetacean. Muscle tissue is more heavily processed in comparison to other tissues before being consumed; therefore, samples were collected at three processing stages: fresh (raw or unprocessed), dried (cured outdoors for 2–5 days), and cooked (heated and rehydrated by steaming; locally referred to as "scalled"). Blubber was collected with the skin attached since this is how blubber is normally consumed in St. Vincent; however, following previous toxicological studies, including those which report THg concentrations in cetacean tissues as well as those focused on the human health implications of consuming cetacean food products, the skin was removed before THg and Se analysis (Julshamn et al., 1987; Dam and Bloch, 2000; Carvalho et al., 2002; Stavros et al., 2007; Damseaux et al., 2017). Furthermore, because it is well known that THg concentrations are higher in the skin (epidermis) compared to the blubber (dermis and subcutis) and that Hg accumulation patterns in skin closely follow that of muscle, we felt it appropriate to isolate the blubber from the skin (Carvalho et al., 2002; Cáceres-Saez et al., 2015; Dirtu et al., 2016; Cozzi et al., 2017). A detailed breakdown of sample sizes for each tissue type and species is shown in Table 1.

Approximately 20 g of each tissue was sampled and stored individually in a plastic bag at −20 °C. All samples were shipped to the University of the South (Sewanee, TN) in December 2016 and stored at −80 °C until further processing and analysis. Samples were exported to the United States under CITES Permit 16US774223/9 and imported under NMFS Permit 19091.

2.2. THg and Se analysis

All processing for THg and Se analysis occurred at Texas State University (San Marcos, TX). Tissue samples were thawed and weighed to obtain the wet wt, after which they were freeze-dried (Labconco

Table 1
Sample size for each tissue type and species with the corresponding tissue moisture content (%; mean ± SD) in parentheses. FKW = false killer whale, KW = killer whale, RD = Risso's dolphin, SPW = short-finned pilot whale, SS = *Stenella* spp. ND = not determined due to sample size ≤ 2.

Species	Muscle fresh	Muscle dried	Muscle cooked	Blubber	Liver	Kidney	Total
FKW	4 (75 ± 5)	17 (27 ± 13)	0	5 (46 ± 14)	2 (71 ± ND)	2 (75 ± ND)	30
KW	9 (74 ± 6)	2 (25 ± ND)	1 (48 ± ND)	6 (53 ± 12)	5 (68 ± 4)	2 (64 ± ND)	25
RD	0	2 (32 ± ND)	1 (55 ± ND)	1 (49 ± ND)	0	0	4
SPW	17 (72 ± 9)	0	4 (59 ± 5)	23 (42 ± 9)	9 (75 ± 5)	5 (80 ± 5)	58
SS	2 (73 ± ND)	0	0	1 (41 ± ND)	1 (65 ± ND)	1 (68 ± ND)	5
Total	32	21	6	36	17	10	122

FreeZone[2.5]; Labconco, Kansas City, MO) for 48 h and the dry weight (dry wt) recorded. To allow for the conversion between dry and wet weight THg and Se concentrations, the mean percentage moisture content for each tissue type and species is shown in Table 1.

The concentration of THg and Se in each sample was determined using microwave acid digestion and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Approximately 0.25 g of sample was digested in 5 ml of acid (4.5 ml of nitric acid and 0.5 ml of hydrochloric acid) in an ETHOS UP microwave digestion system (Milestone Inc., Shelton, CT) for 75 min (25 min ramp time to 200 °C, 20 min hold time at 200 °C, and 30 min cool down time). Once cool, the muscle, kidney, and liver samples were diluted with 45 ml of Milli-Q water (MilliporeSigma, Burlington, MA), whereas the blubber samples were diluted with 25 ml of Milli-Q water, to obtain a final sample volume of 50 ml (dilution factor ~ 200) and 30 ml (dilution factor ~ 120), respectively. All samples were then sent to the Trace Element Analysis Core Laboratory at Dartmouth College (Hanover, NH) for ICP-MS analysis (Agilent 7900 and 8900; Agilent Technologies, Santa Clara, CA) following EPA method 6020A (U.S. EPA, 1998). Both instruments were calibrated using NIST-traceable primary standards with six calibration standards covering the expected calibration range; if the THg or Se concentration exceeded the calibration range individual samples were re-analyzed at a greater dilution. The Se:Hg molar ratio for each sample was calculated by dividing the concentration of Se and THg (μg/g dry wt) by the molecular weight (Hg = 200.59, Se = 78.96), and the ratio determined (μmol Se/μmol Hg).

Quality control included blanks (digested acid, no sample; n = 8), certified reference materials [CRM; DORM-4 fish protein (n = 4) and DOLT-5 dogfish liver (n = 4); National Research Council Canada], spiked samples (n = 8), and duplicate samples (n = 8). All blanks were below the detection limit for both elements (Hg = < 0.05 μg/g, Se = < 0.04 μg/g). The CRM mean THg recovery was 84.0% for DORM-4 and 88.2% for DOLT-5 and the mean Se recovery was 96.4% for DORM-4 and 97.0% for DOLT-5. The mean spike recovery was 94.5% for THg and 96.9% for Se. The mean relative % difference between duplicate samples was 7.9% for Hg and 4.7% for Se for muscle, kidney, and liver (n = 6), and 19.7% for Hg and 24.7% for Se for blubber (n = 2).

2.3. Stable isotope analysis

All stable isotope analysis (SIA) was completed at the Southeast Environmental Research Center Stable Isotope Laboratory at Florida International University (North Miami, FL). Muscle tissue (false killer whale: n = 3; killer whale: n = 8; short-finned pilot whale: n = 16; *Stenella* spp.: n = 2) was analyzed because it incorporates the isotopic signature of dietary sources from the previous few months prior to capture (Newsome et al., 2010). Samples were dried, homogenized into a fine powder, and lipid extracted prior to SIA because lipids are depleted in $\delta^{13}C$ (DeNiro and Epstein, 1978). Lipids were extracted by agitating muscle tissue in a 2:1 chloroform:methanol mixture for 1 min with a solvent volume 5-times greater than the sample, after which the samples were left undisturbed at room temperature for 1 h, and then centrifuged and the supernatant removed. After repeating this process two more times, each sample was rinsed in deionized water, dried, and 0.4–0.5 mg of sample added to a 4 × 6 mm tin capsule for SIA using a ThermoFinnigan Delta V isotope ratio mass spectrometer (IRMS) coupled with a NA 1500 NE elemental analyzer. Analytical reproducibility was based on replicates of internal standards including bovine liver (NBS standard reference material) and glycine (Alfa Aesar); variation among standards was 0.07‰ and 0.08‰ for $\delta^{13}C$ and $\delta^{15}N$, respectively. The mean muscle C:N values were less than 3, indicating an adequate lipid extraction (Lesage et al., 2010). Isotopic ratios (R) are reported in the standard delta (δ) notation relative to the international standards of Vienna Pee Dee belemnite ($\delta^{13}C$) and atmospheric nitrogen ($\delta^{15}N$) using the following equation:

$$\delta(\%_0) = [(R_{Sample}/R_{Standard}) - 1] \text{ X } 1000$$

2.4. Statistical analysis

All statistical analyses were performed using R version 3.4 and the significance level for all tests was set at α = 0.05. An analysis of variance (ANOVA) was used to determine whether there was a significant difference in the THg and Se concentrations, and Se:Hg molar ratios among tissue types within a species, and for each tissue type among species. All the data failed to meet the assumptions of parametric tests after being examined for normality using the Shapiro-Wilk test and homogeneity using the Levene's test, and therefore the data was natural log transformed prior to statistical analysis. If the data then passed the assumptions of parametric tests, a one-way ANOVA followed by a Tukey post-hoc test was used to determine if there were mean differences within and among species, whereas, if assumptions were not met, a Kruskal-Wallis ANOVA by Ranks followed by Dunn's pairwise comparison was used. Within each tissue type, a Spearman rank correlation was used to determine the relationship between the Se:Hg molar ratio and THg concentration. A Kruskal-Wallis test followed by Dunn's pairwise comparison was also used to determine if isotopic signatures ($\delta^{13}C$ and $\delta^{15}N$) differed among species. Multiple linear regressions were performed to determine the influence of species and Se concentration on THg concentration in each tissue, the influence of $\delta^{13}C$ and species on $\delta^{15}N$, and the influence of $\delta^{13}C$ and species and $\delta^{15}N$ and species on THg concentration in muscle. Risso's dolphin and *Stenella* spp. were excluded from analyses due to their small sample size. Except for comparisons to the FAO/WHO Hg advisory, all data was analyzed and discussed on a dry wt basis.

3. Results

3.1. THg and Se concentrations and Se:Hg molar ratios

This study measured the concentration of THg and Se, and calculated the Se:Hg molar ratio in 122 tissue samples from several species of small cetaceans. The highest percentage of samples were collected from short-finned pilot whales (47.5%), followed by false killer whales (24.6%), killer whales (20.5%), *Stenella* spp. (4.1%) and Risso's dolphins (3.3%). The majority of samples analyzed were muscle (48.4%), followed by blubber (29.5%), liver (13.9%), and kidney (8.2%) (Table 1).

The THg and Se concentrations, and Se:Hg molar ratios in each species and tissue type are shown in Table 2. In false killer whales, killer whales, short-finned pilot whales, and *Stenella* spp., the mean THg concentration was greatest in the liver or kidney and lowest in the blubber. For all species, the concentration of THg in the muscle exceeded that in the blubber. There was a difference (p < 0.001) in mean THg concentration between liver and blubber in false killer whale, killer whale, and short-finned pilot whale, with the liver on average being 167-times more enriched in THg than the blubber. The greatest mean THg concentration was measured in false killer whale liver (2032 μg/g dry wt) and the lowest in Risso's dolphin blubber (0.524 μg/g dry wt). All liver samples and the majority of kidney (false killer whale, killer whale, and *Stenella* spp.: 100%; short-finned pilot whale: 40%) and muscle (killer whale, Risso's dolphin, and *Stenella* spp.: 100%; short-finned pilot whale: 95.2%; false killer whale: 85.7%) samples exceeded the FAO/WHO advisory level of 1 μg/g wet wt, whereas the percentage of blubber samples that exceeded this limit ranged from 0% in Risso's dolphin to 100% in false killer whale, killer whale, and *Stenella* spp (short-finned pilot whale: 69.6%). When all species were combined, THg concentrations [mean ± standard deviation (SD); μg/g dry wt] were greatest in the liver (730 ± 1058), followed by the kidney (274 ± 535), muscle (76.4 ± 173), and blubber (4.57 ± 5.00).

Table 2
Total Hg (THg) and Se concentrations (µg/g dry wt and µg/g wet wt), and the Se:Hg molar ratios, for each tissue type and species. Sample sizes are shown in Table 1; for muscle, all three processing stages were combined. FKW = false killer whale, KW = killer whale, RD = Risso's dolphin, SPW = short-finned pilot whale, SS = *Stenella* spp., SD = standard deviation, ND = not determined due to sample size ≤2.

Species	Tissue	Element	(µg/g dry wt)				(µg/g wet wt)			
			Mean	SD	Min	Max	Mean	SD	Min	Max
FKW	Muscle	THg	18.4	28.0	2.74	132	4.35	5.72	0.684	26.0
		Se	6.11	10.1	1.51	48.2	1.43	2.04	0.378	9.49
		Se:Hg molar ratio	0.92	0.36	0.59	2.24				
	Blubber	THg	6.11	6.05	2.42	16.8	2.72	1.79	1.57	5.89
		Se	7.30	4.54	3.35	14.6	3.71	2.29	2.30	7.76
		Se:Hg molar ratio	4.22	2.83	1.32	8.72				
	Liver	THg	2032	ND	690	3373	632	ND	175	1088
		Se	959	ND	355	1562	297	ND	90.1	504
		Se:Hg molar ratio	1.24	ND	1.18	1.31				
	Kidney	THg	232	ND	24.0	440	60.0	ND	5.58	114
		Se	131	ND	5.03	256	33.9	ND	1.17	66.6
		Se:Hg molar ratio	1.01	ND	0.53	1.48				
KW	Muscle	THg	196	285	24.5	805	48.4	71.0	6.50	202
		Se	86.9	155	4.33	451	21.3	38.5	1.15	110
		Se:Hg molar ratio	0.76	0.34	0.32	1.42				
	Blubber	THg	11.9	6.78	1.66	19.5	5.04	2.55	1.13	7.63
		Se	5.53	2.86	1.19	8.50	2.41	1.24	0.807	4.19
		Se:Hg molar ratio	1.30	0.38	0.92	1.82				
	Liver	THg	1396	1084	14.8	2690	472	381	4.01	964
		Se	572	417	5.14	1076	193	147	1.39	386
		Se:Hg molar ratio	1.06	0.21	0.88	1.43				
	Kidney	THg	945	ND	154	1737	357	ND	52.4	661
		Se	372	ND	42.3	701	141	ND	14.4	267
		Se:Hg molar ratio	0.86	ND	0.70	1.03				
RD	Muscle	THg	40.8	56.0	4.16	105	11.0	15.1	1.12	28.4
		Se	10.2	13.2	2.25	25.4	2.74	3.55	0.609	6.85
		Se:Hg molar ratio	0.94	0.72	0.44	1.76				
	Blubber	THg	0.524	ND	ND	ND	0.270	ND	ND	ND
		Se	4.17	ND	ND	ND	2.15	ND	ND	ND
		Se:Hg molar ratio	20.2	ND	ND	ND				
SPW	Muscle	THg	77.3	167	3.24	664	20.5	45.8	0.906	186
		Se	33.7	73.2	1.64	245	8.75	19.4	0.468	68.7
		Se:Hg molar ratio	0.92	0.45	0.45	2.31				
	Blubber	THg	2.42	1.32	0.194	5.98	1.35	0.671	0.125	3.24
		Se	6.80	5.93	1.28	24.2	3.82	3.44	0.586	15.1
		Se:Hg molar ratio	8.62	10.2	1.22	49.6				
	Liver	THg	149	243	20.9	655	35.7	61.9	4.75	185
		Se	82.6	137	4.19	364	18.5	30.2	0.985	77.4
		Se:Hg molar ratio	1.22	0.96	0.46	3.37				
	Kidney	THg	18.7	26.6	2.48	64.6	4.15	5.88	0.293	14.0
		Se	13.4	17.8	2.67	44.1	2.93	3.94	0.316	9.59
		Se:Hg molar ratio	2.21	0.45	1.73	2.74				
SS	Muscle	THg	11.7	ND	8.71	14.7	3.22	ND	2.09	4.34
		Se	4.24	ND	3.89	4.59	1.15	ND	0.934	1.36
		Se:Hg molar ratio	0.96	ND	0.79	1.13				
	Blubber	THg	6.47	ND	ND	ND	3.79	ND	ND	ND
		Se	11.0	ND	ND	ND	6.47	ND	ND	ND
		Se:Hg molar ratio	4.33	ND	ND	ND				
	Liver	THg	29.1	ND	ND	ND	10.2	ND	ND	ND
		Se	8.70	ND	ND	ND	3.07	ND	ND	ND
		Se:Hg molar ratio	0.76	ND	ND	ND				
	Kidney	THg	296	ND	ND	ND	94.8	ND	ND	ND
		Se	119	ND	ND	ND	38.2	ND	ND	ND
		Se:Hg molar ratio	1.02	ND	ND	ND				

Dry wt concentrations were converted to wet wt concentrations for each sample based on the individual % moisture content, except for dried and cooked muscle samples which were converted to wet wt using the mean % moisture content in fresh muscle for each respective species. Because no fresh Risso's dolphin muscle samples were available, dried and cooked Risso's muscle samples were converted to wet wt using the mean % moisture content in fresh *Stenella* spp. muscle.

In killer whales and short-finned pilot whales, mean Se concentrations were greatest in liver and lowest in blubber; in false killer whales, mean concentrations were greatest in liver and lowest in muscle; in *Stenella* spp., mean concentrations were greatest in kidney and lowest in muscle; and in Risso's dolphin, mean concentrations were greater in muscle than blubber (Table 2). In false killer whales and killer whales, the concentration of Se in blubber was significantly different to the concentration in liver and kidney ($p < 0.05$), whereas there were no differences ($p > 0.05$) found in Se concentrations between tissue types

in short-finned pilot whales. Like THg, the mean Se concentration was greatest in false killer whale liver (959 µg/g dry wt) and lowest in Risso's dolphin blubber (4.17 µg/g dry wt). When all species were combined, Se concentrations (mean ± SD; µg/g dry wt) were greatest in the liver (325 ± 455), followed by the kidney (119 ± 219), muscle (32.5 ± 85.8), and blubber (6.70 ± 5.16).

The relationship between THg and Se concentrations in each tissue is shown in Fig. 2. In muscle, liver, and kidney regression models, species was not an influential predictor of THg concentration and was
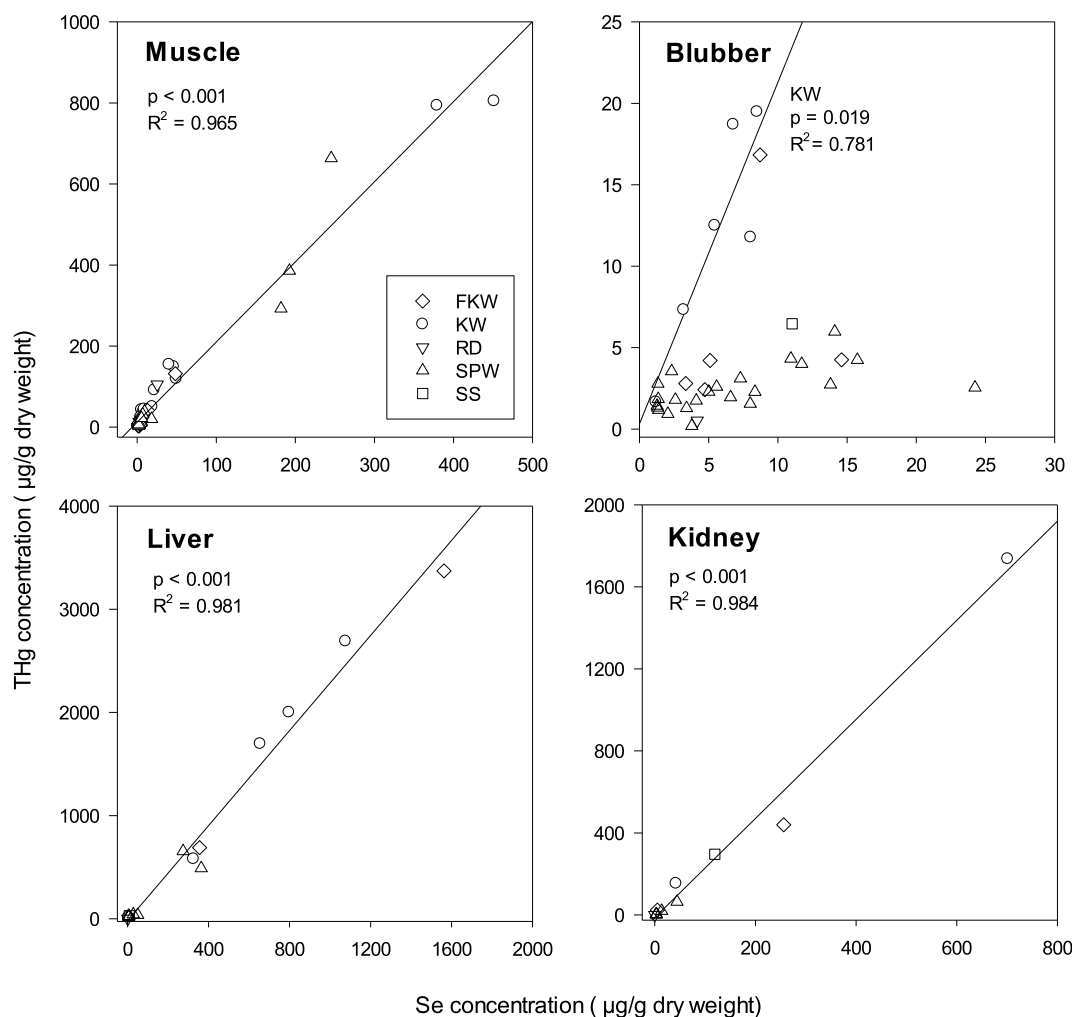
Fig. 2. Relationship between THg and Se concentrations in muscle, blubber, liver, and kidney. Regression lines in muscle, liver, and kidney are for combined species. The regression line for blubber corresponds to killer whales, the only species that had a significant relationship between THg and Se. FKW = false killer whale, KW = killer whale, RD = Risso's dolphin, SPW = short-finned pilot whale, SS = *Stenella* spp.

therefore removed as a predictor. Selenium positively influenced THg concentrations ($p < 0.001$) in muscle, liver, and kidney, whereas a positive relationship between THg and Se concentrations was only observed in blubber in killer whales ($p = 0.019$).

On average, the Se:Hg molar ratios were greatest in the blubber and lowest in the muscle tissue of all species except *Stenella* spp., which was lowest in the liver (Fig. 3; Table 2). The mean Se:Hg molar ratios ranged from 0.76 in killer whale muscle and *Stenella* spp. liver to 20.2 in Risso's dolphin blubber. The blubber Se:Hg molar ratio in false killer whale, killer whale, and short-finned pilot whale differed ($p < 0.001$) from muscle in each species. Additionally, blubber was also different from liver ($p = 0.028$) and kidney ($p = 0.003$) in false killer whales. None of the species examined had a mean Se:Hg molar > 1 for muscle, whereas all of the species had a mean Se:Hg molar ratio > 1 in the blubber, and two species (short-finned pilot whale and Risso's dolphin) had a mean blubber Se:Hg molar ratio > 5. In the kidney and liver, the majority of mean Se:Hg molar ratios for each species were close to 1 (Fig. 3; Table 2). When all species were combined, the Se:Hg molar ratio (mean ± SD) was greatest in the blubber ($6.99 ± 8.96$), followed by the kidney ($1.58 ± 0.77$), liver ($1.15 ± 0.70$), and muscle ($0.89 ± 0.40$).

For liver, there were no differences ($p > 0.05$) in THg and Se concentrations and Se:Hg molar ratios among species. For kidney, THg concentrations in killer whales differed from short-finned pilot whale ($p = 0.047$) and Se:Hg molar ratios in short-finned pilot whales differed

from both killer whales and false killer whales ($p < 0.05$), but there were no differences in Se concentrations among species. While killer whales had a higher ($p < 0.05$) mean THg and Se concentration in muscle tissue than false killer whales and short-finned pilot whales, no differences ($p = 0.344$) in Se:Hg molar ratios were found among species. Killer whales had a greater mean blubber THg concentration and lower Se:Hg molar ratios than short-finned pilot whales ($p < 0.01$), however, no differences ($p = 0.758$) in Se concentrations were found between these two species. There was a positive correlation between the Se:Hg molar ratio and THg concentration in the liver ($p = 0.029$), a negative relationship in the kidney and blubber ($p = 0.019$ and $p = 0.008$, respectively), and no relationship ($p = 0.072$) in the muscle (Fig. 4).

3.2. Interspecies variability in muscle stable isotope ratios and relationship with THg

Mean ± SD stable isotope ratios indicated that killer whales were $\delta^{13}C$ ($-14.6 ± 0.67‰$) and $\delta^{15}N$ ($+12.1 ± 1.07‰$) enriched compared to false killer whales ($\delta^{13}C = -15.9 ± 1.07‰$; $\delta^{15}N = 11.4 ± 0.80‰$), short-finned pilot whales ($\delta^{13}C = -15.6 ± 0.68‰$; $\delta^{15}N = 10.9 ± 0.49‰$), and *Stenella* spp. ($\delta^{13}C = -16.0‰$; $\delta^{15}N = 10.5‰$) (Fig. 5A and B); however, the only significant difference among $\delta^{13}C$ and $\delta^{15}N$ values were between killer whales and short-finned pilot whales ($p = 0.022$ and $p = 0.024$,
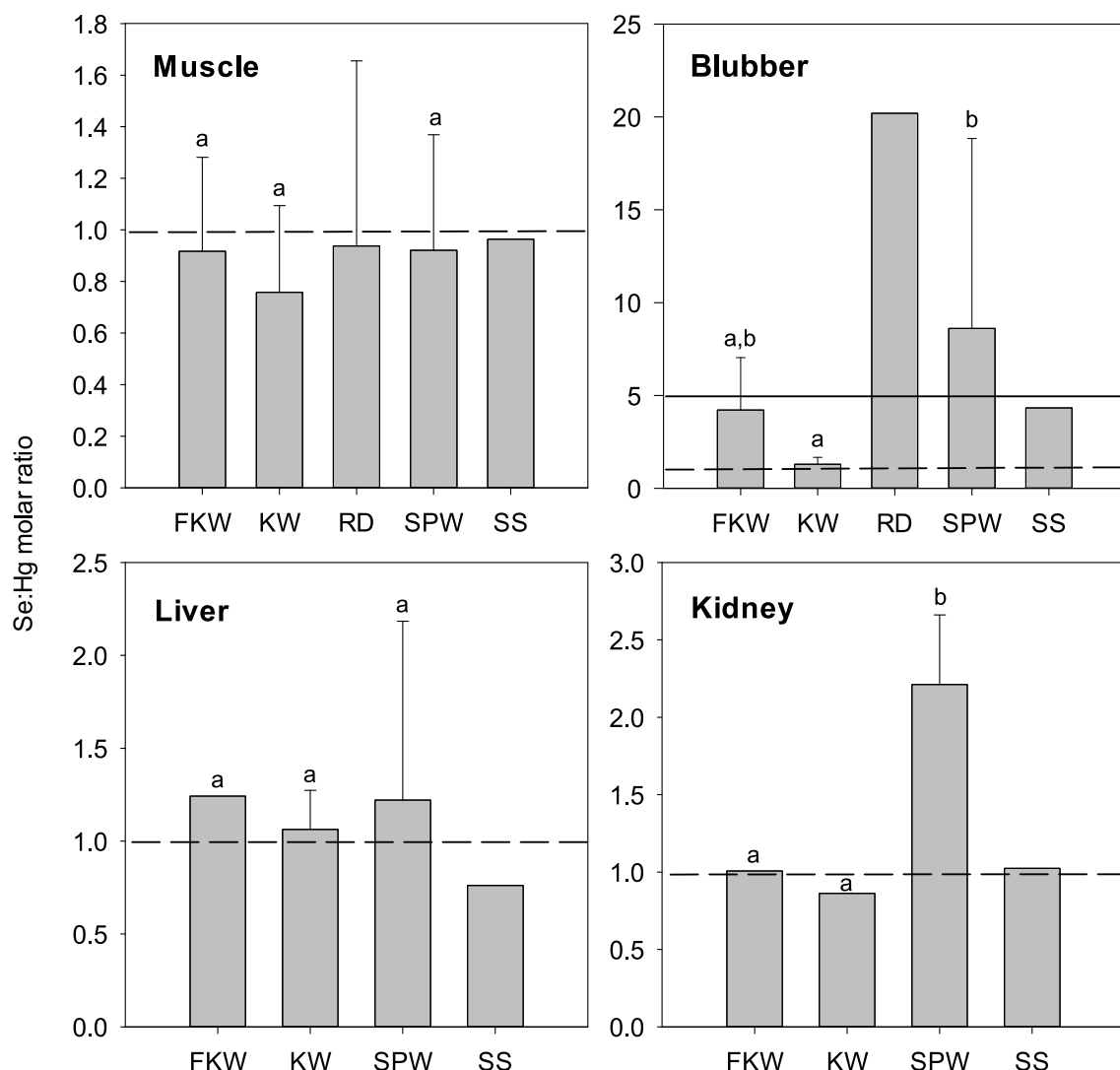
Fig. 3. Se:Hg molar ratios (mean + SD) in muscle, blubber, liver, and kidney of small cetaceans. The solid horizontal line represents the 5:1 Se:Hg molar ratio and broken horizontal line represents the 1:1 Se:Hg molar ratio. FKW = false killer whale, KW = killer whale, RD = Risso's dolphin, SPW = short-finned pilot whale, SS = *Stenella* spp. Lowercase letters indicate species grouped by similar Se:Hg molar ratios.

respectively). In all multiple regression analyses (Fig. 5C–E), species was not a significant predictor of THg concentration and was removed as a predictor. There was a positive relationship between $\delta^{13}C$ and $\delta^{15}N$ (p < 0.001; Fig. 5C), and between $\delta^{13}C$ and THg concentration (p = 0.006; Fig. 5D), but no relationship between $\delta^{15}N$ and THg concentration (p = 0.129; Fig. 5E).

4. Discussion

This is the first study to investigate THg and Se concentrations, and the corresponding Se:Hg molar ratios, in at least five small cetacean species taken for human consumption in the Eastern Caribbean. This study does not recommend any specific Hg dietary guidelines, but expands on the work of Fielding and Evans (2014) by examining Hg concentrations in four additional small cetacean species targeted in the St. Vincent whaling operations. In addition, this study utilizes stable isotope analysis to determine whether ecological factors such as foraging habitat and relative trophic position could help explain the observed THg concentrations in muscle tissue. Total Hg and Se concentrations and Se:Hg molar ratios were provided for individual species and all species combined. In St. Vincent, people do not necessarily know which species they are consuming since food products from

multiple odontocete species are marketed under the same local name: "blackfish" (Fielding, 2014); therefore it is important to report the mean THg concentration people can be exposed to.

4.1. Mercury concentrations in St. Vincent odontocetes

Odontocetes accumulate high concentrations of Hg; however, Hg concentrations can vary widely both within and among species (Das et al., 2003a; Endo et al., 2005; Kershaw and Hall, 2019). Factors including age, diet, sex, geographic location, and tissue type have been used to explain variation in Hg concentrations (Andre et al., 1991; Meador et al., 1999; Zhou et al., 2001; Endo et al., 2010; Dirtu et al., 2016). In this study, the interpretation of THg concentrations and stable isotope ratios was limited to species identification because no biological data (e.g., length, age, sex) or take location other than near St. Vincent was reported.

The tissue distribution found in the present study, with the greatest THg concentrations in the liver or kidney and lowest THg concentration in the blubber, is typical of the THg tissue distribution found in odontocetes (Itano et al., 1984; Cardellicchio et al., 2002). The liver and kidney are important filtration and detoxification organs and therefore can accumulate THg to relatively high concentrations. Muscle also
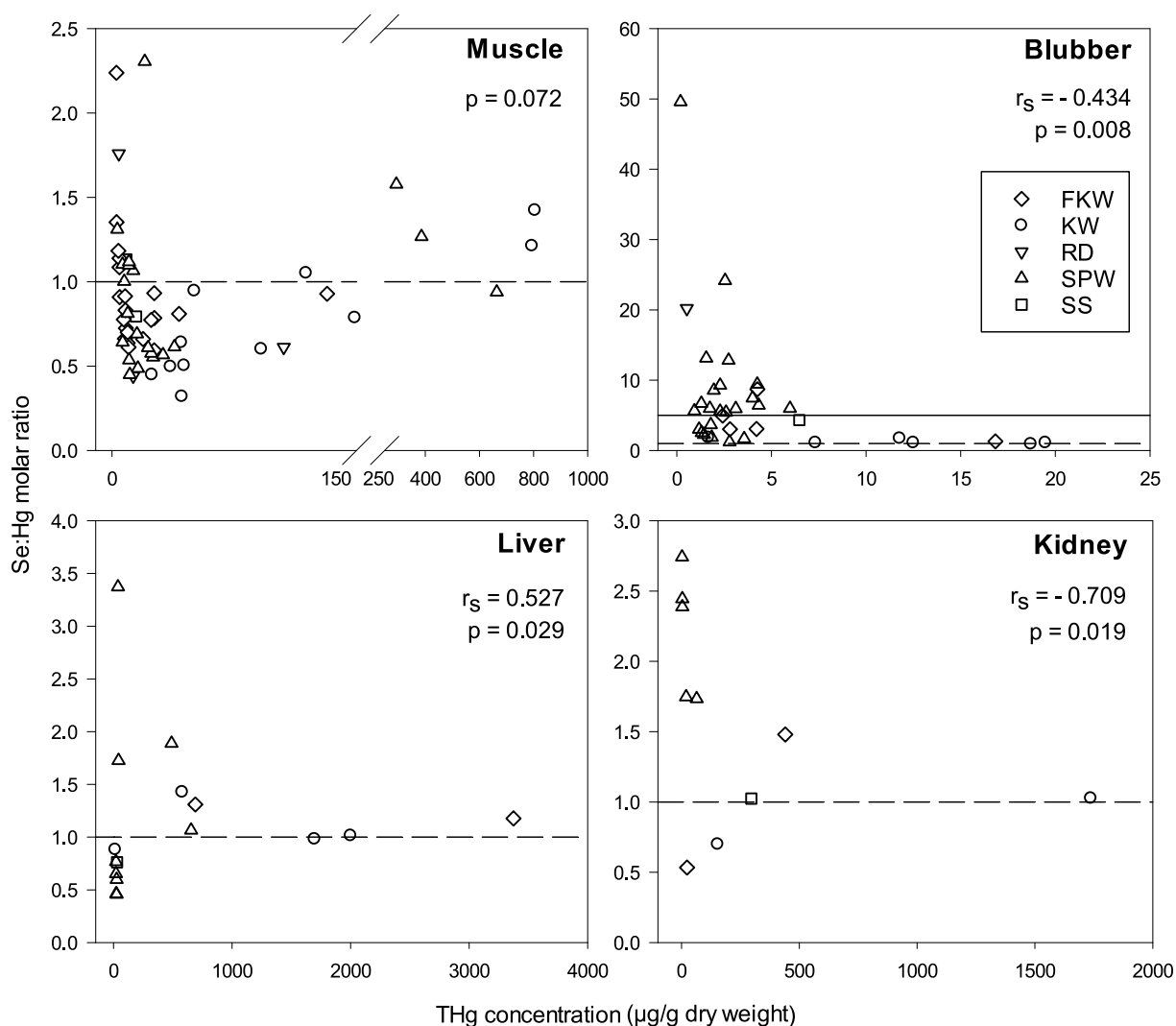
Fig. 4. Relationship between the Se:Hg molar ratio and THg concentration in the muscle, blubber, liver, and kidney tissue of small cetaceans. The solid horizontal line represents the 5:1 Se:Hg molar ratio and broken horizontal line represents the 1:1 Se:Hg molar ratio. FKW = false killer whale, KW = killer whale, RD = Risso's dolphin, SPW = short-finned pilot whale, SS = *Stenella* spp.

accumulates THg to high concentration because MeHg, the predominant form of Hg in muscle tissue, has a high binding affinity for sulfhydryl (-SH) groups found in proteins within muscle tissue (Bloom, 1992). In contrast, even though MeHg is lipophilic, due to its preferential binding to –SH and low octanol-water partition coefficient (Bienvenue et al., 1984), MeHg does not accumulate to a high concentration in blubber.

In this study, killer whales had the greatest mean muscle, blubber, and kidney THg concentrations of all the species examined, which is most likely due to this species having the highest relative trophic position as shown by the higher $\delta^{15}N$ values. For comparison to other studies, when necessary, wet wt THg concentrations were converted to dry wt concentrations using the mean % moisture content provided for each species in Table 1. The THg concentrations reported in killer whales in this study are the highest in the literature. In comparison to other studies, the mean liver THg concentration reported in killer whales in this study was 5–times higher than reported in the United Kingdom (275 μg/g dry wt; Law et al., 1997) and 7–times higher than reported in Japan (194 μg/g dry wt; Endo et al., 2006). Similarly, the mean kidney and muscle concentrations in this study were approximately 41–times greater than those reported in Japan (22.5 and 4.73 μg/g dry wt, respectively; Endo et al., 2006).

Mean short-finned pilot whale muscle THg concentrations reported

in this study were between 1.8–14.4-times greater than those reported for the same species in New Caledonia (30 μg/g dry wt; Bustamante et al., 2003) and Japan (5.36–41.4 μg/g dry wt; Endo et al., 2005; Sakamoto et al., 2015). In contrast, THg concentrations in the liver and blubber in this study were 9.6- and 2.9-times lower, respectively, than mean liver (1432 μg/g dry wt) and blubber (7.10 μg/g dry wt) THg concentrations reported for short-finned pilot whales from New Caledonia (Bustamante et al., 2003). In the Faroe Islands, long-finned pilot whale, a closely related species to the short-finned pilot whale, had a mean muscle THg concentration for adult whales of 8.23 μg/g dry wt (Dam and Bloch, 2000), 9.4-times lower than reported in this study.

The mean muscle THg concentration in false killer whales in this study was 4.5- to 8.6-times lower than reported in Japan (82.8–158 μg/g dry wt; Endo et al., 2005, 2010). In Chile, the mean false killer whale muscle and kidney THg concentration was 6.4- and 1.2-times higher than this study (118 and 272 μg/g dry wt, respectively), whereas the liver concentration (1068 μg/g dry wt) was 1.9-times lower (Cáceres-Saez et al., 2018). Although the number of samples was small, our findings indicated slightly higher THg concentrations than previously reported for muscle and blubber in *Stenella* spp. in St. Vincent (Fielding and Evans, 2014). Risso's dolphins had a muscle THg concentration that was higher than previously reported in Japan and Taiwan (11.7–16.5 μg/g dry wt; Chen et al., 2002; Endo et al., 2005; Sakamoto
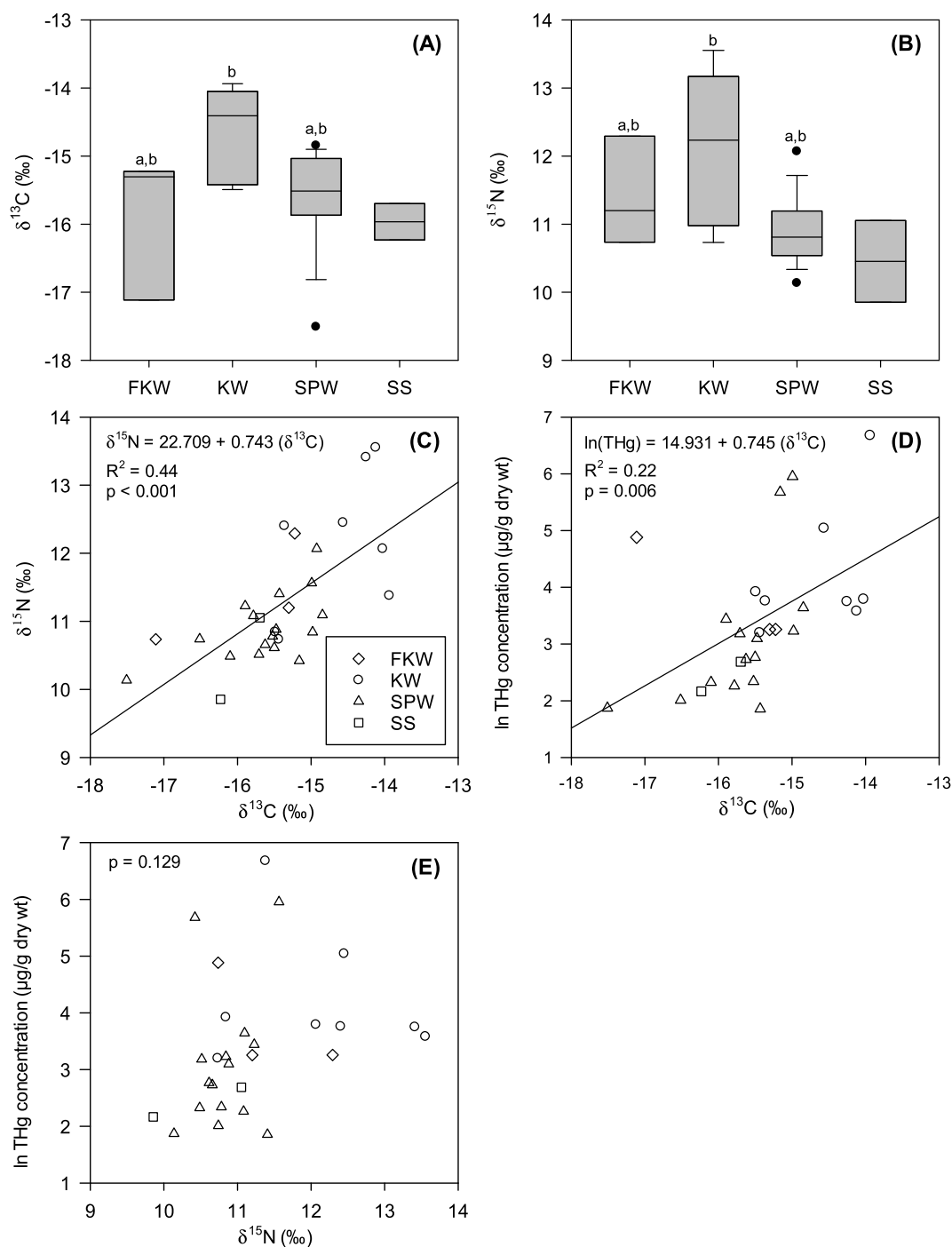
Fig. 5. Muscle $\delta^{13}C$ (A) and $\delta^{15}N$ (B) values for each species, and the relationship between $\delta^{15}N$ and $\delta^{13}C$ (C), and muscle THg concentration and $\delta^{13}C$ (D) and $\delta^{15}N$ (E). Lowercase letters indicate species grouped by similar $\delta^{13}C$ or $\delta^{15}N$. FKW = false killer whale, KW = killer whale, SPW = short-finned pilot whale, SS = *Stenella* spp.

et al., 2015). The highest muscle THg concentrations in Risso's dolphins (1463 µg/g dry wt) have been reported in the Mediterranean Sea which is naturally high in Hg (Nriagu, 1979; Shoham-Frider et al., 2002).

The reasons for the differences between the THg concentrations in this study and previous studies are not apparent, but could be a result of differences in environmental Hg concentrations, body length, age, dietary preferences, and whether pods are residential or migratory. Whalers in St. Vincent preferentially target larger cetaceans, owing to the increased value of the catch. Because all our tissue samples were collected from odontocetes taken in the Vincentian whaling operation, this could result in our data being skewed toward the larger (and

therefore, older and more likely male) individuals. While this data might not represent mean THg concentrations in the overall population of each species, they do bear directly upon concerns related to food safety, as our samples were collected only from odontocetes in the Eastern Caribbean taken for human consumption.

4.2. Interspecies variability in stable isotope ratios and relationship with THg

Killer whales had significantly higher $\delta^{15}N$ values in comparison to the other sampled species, reflecting their higher trophic position. In

the Caribbean, killer whales have been observed preying on cetaceans and other high trophic level prey (Bolaños-Jiménez et al., 2014). In comparison, the other species are known to feed on lower trophic level prey; short-finned pilot whales forage primarily on mesopelagic squids, while false killer whales and *Stenella* spp. forage on fish and squid of various trophic levels (Dolar et al., 2003; Mintzer et al., 2008; Baird, 2018; Olson, 2018). The enriched $\delta^{13}C$ values in killer whales in comparison to false killer whales, short-finned pilot whales, and *Stenella* spp. may also reflect differences in diet and foraging habitats; $\delta^{13}C$ values are generally higher in species from coastal or benthic food webs (Lesage et al., 2001). Our values were $\delta^{13}C$ enriched and $\delta^{15}N$ depleted compared to muscle from the same species off Japan (Endo et al., 2010). This could be due to geographic differences in the dietary carbon (e.g., seagrass versus phytoplankton; Newsome et al., 2010), nitrogen source, and the rate of nitrogen fixation between the two regions (McMahon et al., 2013). No other studies have reported stable isotope ratios in small cetaceans from the Caribbean, but $\delta^{15}N$ values in this study are comparable to those reported in Caribbean sharks which feed at similar trophic levels (Tilley et al., 2013). The linear covariation between $\delta^{15}N$ and $\delta^{13}C$ found in the present study is characteristic of marine systems (Kelly, 2011).

A positive relationship between $\delta^{15}N$ and THg concentration in muscle tissue was expected as THg biomagnifies with increasing trophic position; however, no significant relationship was found. Prior studies have found similar results and suggested that while THg is found in higher concentration in higher trophic level organisms differentiating between cetacean trophic positions can be challenging because it is unclear how much of the variation in THg can be explained by biomagnification, bioaccumulation, and differences in environmental Hg concentrations between geographic locations (Das et al., 2003a; Endo et al., 2010).

4.3. Se:Hg molar ratios

Prior studies have found positive correlations between Hg and Se concentrations in odontocete tissues, suggesting there is an interaction between these elements which may play a role in Hg detoxification (Endo et al., 2005; Cáceres-Saez et al., 2018). Selenium can reduce Hg toxicity through the formation of inert Se–Hg complexes, by competing with Hg for binding sites, and through its antioxidant properties (Cuvin-Aralar and Furness, 1991). It has been suggested that Se:Hg molar ratios in excess of 1:1 indicate conditions in which Se may have a protective effect against Hg toxicity (Berry and Ralston, 2008).

The Se:Hg molar ratios differed between the tissue types examined, reflecting the accumulation patterns of Hg and Se. Like other non-essential trace elements, Hg is not under homeostatic control and therefore accumulates in odontocete tissues (Das et al., 2003a). In contrast, Se, an essential element used in selenoproteins, is under homeostatic control and has tissue specific regulation. However, in some cases, especially when Hg concentrations are high, Se concentrations can exceed what is physiologically required, further suggesting that Se plays a key role in Hg detoxification (Cáceres-Saez et al., 2018).

On average for each species, Se:Hg molar ratios were > 1 in blubber, < 1 in muscle, and more variable in kidney and liver with some species having a mean ratio above 1 and others below 1. The Se:Hg molar ratios are > 1 in blubber because Se is in molar excess due to Hg not accumulating to high concentration in blubber as previously discussed. Mercury was in molar excess in the muscle due to it accumulating to higher concentration than Se. In the liver of marine mammals, previous studies have shown that Se:Hg molar ratios approach 1 only after Hg concentrations are exceedingly high, suggesting that a physiological threshold exists for which Se and Hg may exist in equal molar concentration (Wagemann et al., 1998; Ikemoto et al., 2004; Cáceres-Saez et al., 2018). Similar to Ikemoto et al. (2004), Se:Hg molar ratios in the liver, kidney, and muscle approached or exceeded 1

when THg concentrations exceeded approximately 200 μg/g dry wt.

For muscle and liver, the Se:Hg molar ratios were not statistically different among killer whales, short-finned pilot whales, and false killer whales because even though the THg and Se concentrations varied among species, they were proportionally the same. However, short-finned pilot whales had a higher Se:Hg molar ratio in the blubber and kidney compared to false killer whales and killer whales; this is most likely a result of short-finned pilot whales having a lower THg concentration than the other two species, but a comparable Se concentration in the blubber and equimolar concentrations of Se and Hg in the kidney.

Compared to other studies that calculated mean Se:Hg molar ratios for the same species in different geographic areas, the findings of this study are within the range previously reported for liver (this study = 0.76 to 1.24; previous studies = 0.68 to 1.03) and kidney (this study = 0.86 to 2.21; previous studies = 0.54 to 2.17) (Stoneburner, 1978; Cardellicchio et al., 2002; Endo et al., 2006; Cáceres-Saez et al., 2018). Endo et al. (2005) reported Se:Hg molar ratios in muscle tissue that were above 1 (1–2.6) suggesting that Se may have a protective effect against Hg toxicity; however, the mean Se:Hg molar ratios calculated in the present study (0.76–0.96) do not support that finding and are more consistent with those reported by Cáceres-Saez et al. (2018) (0.85–1.10). With the exception of Risso's dolphin in Israel (0.33; Shoham-Frider et al., 2002), the mean blubber Se:Hg molar ratios reported in this study (1.3–20.2) are within the range or higher than previously reported (6.68–8.51; Stoneburner, 1978; Cardellicchio et al., 2002).

High intra- and interspecies variability in Se:Hg molar ratios are commonly reported and have raised concerns over the use of these ratios in risk assessment (Burger et al., 2012, 2013; Burger and Gochfeld, 2011, 2013). Se:Hg molar ratios can be negatively correlated with fish length, suggesting that Se provides less protective effects against Hg toxicity in older fish (Burger et al., 2013). Body length/age was unknown in the present study, but could account for some of the variability in Se:Hg molar ratios observed within and among species. Furthermore, it is unclear if a Se:Hg molar ratio of 1:1 is protective, as this assumes that all available Se is bound to Hg. In addition, it is also uncertain whether a different ratio is needed for those who are more susceptible to Hg toxicity including pregnant women and young children (Burger et al., 2013). Therefore, a Se:Hg molar ratio greater than 1:1 may be necessary to see the protective effect of Se against Hg toxicity (e.g., 5:1). If we consider this more conservative 5:1 ratio, only Se concentrations in blubber could potentially provide protection against Hg toxicity. More research which incorporates the length/age of the investigated species is necessary to be able to fully assess the applicability of using Se:Hg molar ratios in risk assessment, especially when THg concentrations are exceedingly high.

4.4. Human health implications in St. Vincent

Cetaceans are an important food source for select human populations around the globe. While commercial whaling for large cetaceans has decreased dramatically since the International Whaling Commission (IWC) moratorium in 1986, the taking of small cetaceans, which are not protected under the IWC moratorium, has increased (Robards and Reeves, 2011). Due to their high trophic position and long life span, small cetaceans can accumulate high concentrations of Hg which may pose a threat to human health if frequently consumed (Grandjean et al., 1997; Weihe and Joensen, 2012). The risk to human health from consuming marine mammals, including cetaceans, high in Hg has been studied in the Faroe Islands (Grandjean et al., 1997; Weihe and Joensen, 2012), Greenland (Johansen et al., 2004), the Canadian Arctic (Boucher et al., 2012), and Japan, which has the world's largest direct take of small cetaceans (Kasuya, 2007; Endo and Haraguchi, 2010; Robards and Reeves, 2011), but little is known about the Hg concentrations and associated health risks in human consumers of small

cetaceans in St. Vincent. One of the few published studies did report elevated blood Hg concentrations in pregnant women from St. Vincent & the Grenadines compared to subjects in the U.S., Canada, and other Caribbean countries (Forde et al., 2014).

In contrast to the Faroe Islands where the government has provided Hg guidelines in relation to the consumption of long-finned pilot whales, there are currently no Hg guidelines regarding the consumption of small cetaceans in St. Vincent. Dietary recommendations to cease the consumption of long-finned pilot whale in the Faroe Islands were made when muscle THg concentrations exceeded 2 μg/g wet wt (Weihe and Joensen, 2012), 10-times lower than the mean THg concentration in all species combined in this study.

According to the findings of this study, consumption of liver, kidney, and muscle presents the greatest risk of Hg exposure to humans; however, it should be noted that kidney and liver generally are not sold through commercial vendors and therefore only a select group of St. Vincentians will consume these organs. While blubber contained the lowest concentration of THg, 80% of the investigated species still had between 69.5 and 100% of samples that exceeded the FAO/WHO Hg advisory level. In addition, blubber can accumulate lipophilic contaminants such as PCBs and DDT (Dam and Bloch, 2000) which were not investigated in this study, but can also pose a threat to human health as reported in the Faroe Islands (Weihe and Joensen, 2012). Furthermore, this study did not measure the concentration of THg in skin which is consumed with blubber and likely has a higher THg concentration. Thus, the findings reported here should not be viewed as an indicator of the relative safety of blubber consumption except when strictly applied to THg in blubber alone.

To estimate the annual cetacean catch in St. Vincent during the 2015–2016 collection period for this study, values have been extrapolated from records from a single whaler because there is no centralized record keeping. Whaling occurs off of St. Vincent approximately 221 days per year, and approximately 410 small cetaceans were taken during the sampling period. We were unable to estimate the species composition of the total catch except to distinguish among short-finned pilot whales, killer whales, and other delphinids. While the primary target of the St. Vincent whaling operation is the short-finned pilot whale, only 25% of the catch was comprised of this species, the remainder were killer whales (1%) and other delphinid species (74%) (Fielding, 2018).

A 2013 interview-based study, which surveyed 211 post-secondary school students throughout St. Vincent & the Grenadines found that muscle and blubber were consumed on average 25 times per year by 55–64% of respondents and 68% believed cetacean meat (muscle tissue) to be a healthy food (Fielding, 2013); a broader dietary survey is currently ongoing in St. Vincent. The FAO/WHO provisional tolerable weekly intake (PTWI) for MeHg is currently 1.6 μg MeHg/kg body weight/week. While THg was not speciated to calculate the percentage MeHg in this study, based on previous studies for muscle (75%; Endo et al., 2006; Fielding and Evans, 2014), blubber (6.5%; Fielding and Evans, 2014), liver (1.9%; Endo et al., 2006) and kidney (3.2%; Endo et al., 2006), the mean MeHg wet wt concentration for all species combined was 14.5 μg/g in the muscle, 0.1426 μg/g in the blubber, 4.42 μg/g in the liver, and 3.04 μg/g in the kidney. Based on these estimations, consuming more than 6.6 g of muscle, 673 g of blubber, 21.7 g of liver, or 31.6 g of kidney a week would exceed the MeHg PTWI for a 60 kg person.

5. Conclusions

The high THg concentrations in cetacean tissues in this study, along with the high number of small cetaceans that are taken for human consumption each year, and the frequency at which cetacean products are consumed, suggests that consumption of small cetaceans in St. Vincent is a human health issue that warrants further investigation. Future policy changes or advisories may be needed to inform the public,

especially regarding the consumption of killer whales and short-finned pilot whales.

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