



ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

JES

JOURNAL OF
ENVIRONMENTAL
SCIENCESwww.jesc.ac.cn

Analysis of PFAAs in American alligators part 2: Potential dietary exposure of South Carolina hunters from recreationally harvested alligator meat

Jessica J. Tipton¹, Louis J. Guillette Jr², Susan Lovelace¹, Benjamin B. Parrott³, Thomas R. Rainwater⁴, Jessica L. Reiner^{5,*}

1. College of Charleston, Charleston, SC 29724, USA

2. Medical University of South Carolina, Department of Obstetrics and Gynecology, Charleston, SC 29425, USA

3. University of Georgia, Odum School of Ecology, Savannah River Ecology Laboratory, Jackson, SC 29831, USA

4. Tom Yawkey Wildlife Center & Belle W. Baruch Institute of Coastal Ecology and Forest Science, Clemson University, P.O. Box 596, Georgetown, SC 29442, USA

5. National Institute of Standards and Technology, Chemical Sciences Division, Hollings Marine Laboratory, Charleston, SC 29412, USA

ARTICLE INFO

Article history:

Received 6 March 2017

Revised 4 May 2017

Accepted 9 May 2017

Available online 29 June 2017

Keywords:

Public hunt

American alligator

Dietary exposure

PFOS

Consumption advisory

Contaminant consumption

ABSTRACT

Exposure to perfluorinated alkyl acids (PFAAs) has been linked to many harmful health effects including reproductive disorders, developmental delays, and altered liver and kidney function. Most human exposure to environmental contaminants, including PFAAs, occurs through consumption of contaminated food or drinking water. This study uses PFAA data from meat samples collected from recreationally harvested American alligators (*Alligator mississippiensis*) in South Carolina to assess potential dietary exposure of hunters and their families to PFAAs. Consumption patterns were investigated using intercept surveys of 23 hunters at a wild game meat processor. An exposure scenario using the average consumption frequency, portion size, and median perfluorooctane sulfonic acid (PFOS) concentration in alligator meat from all hunt units found the daily dietary exposure to be 2.11 ng/kg body weight per day for an adult human. Dietary PFOS exposure scenarios based on location of harvest suggested the highest daily exposure occurs with alligator meat from the Middle Coastal hunt unit in South Carolina. Although no samples were found to exceed the recommended threshold for no consumption of PFOS found in Minnesota state guidelines, exposure to a mixture of PFAAs found in alligator meat and site-specific exposures based on harvest location should be considered in determining an appropriate guideline for vulnerable populations potentially exposed to PFAAs through consumption of wild alligator meat.

© 2017 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences.

Published by Elsevier B.V.

* Corresponding author. E-mail: jessica.reiner@nist.gov (Jessica L. Reiner).

Introduction

Perfluorinated alkyl acids (PFAAs) are a group of man-made chemicals with fully fluorinated carbon bonds. The chain length of PFAAs can vary, along with the potential for linear or branched isomers. The two PFAAs most ubiquitous in the environment and wildlife are perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) (Reiner and Place, 2015). In addition to wildlife studies, human surveys like the National Health and Nutrition Examination Survey, NHANES, found PFOS in greater than 98% of human blood samples in the general U.S. population (Calafat et al., 2007). The consequences of exposure to PFAAs have been studied in wildlife and humans. Previous studies on laboratory animals have found relationships between PFAA exposure and endocrine disruption, developmental disorders, and hepatic tumors (Butenhoff et al., 2012; Kim et al., 2013; Kjeldsen and Bonefeld-Jørgensen, 2013; Zhang et al., 2016). Wildlife studies have found PFAA exposure to result in altered renal and hepatic function, immune suppression, and reproductive disorders (Fair et al., 2013; Persson and Magnusson, 2015). Regarding human health, in a study of PFAA exposed children from the Faroe Islands (high seafood diet), antibody production in response to vaccinations was used to determine decreased immune health in highly exposed individuals (Grandjean et al., 2012). Ingestion of contaminated food or drinking water is cited as the most common PFAA exposure pathway for humans in the U.S. (Haug et al., 2011). Exposure to PFAAs in high-risk populations such as pregnant women and children is especially concerning due to the potential impacts of these chemicals on development and immune function.

Apex predators in ecosystems can be used as sentinels of habitat quality in areas impacted by anthropogenic activities (Mazzotti et al., 2009). In South Carolina (SC), the American alligator (*Alligator mississippiensis*) is not only a sentinel species important for understanding the influence of human impacts on aquatic ecosystems throughout the coastal plain, it is also a valuable harvested resource. Of the many anthropogenic impacts facing wildlife conservation and management agencies today, few are as widely spread and directly applicable to human health as environmental pollution (Fox, 2001).

Concern about the impacts of contaminants on human health has led many regulatory agencies to issue recommended consumption guidelines in an attempt to decrease potential exposure. Recreational harvesting of wild game species through hunting and fishing activities is an important source of protein for many hunters and anglers in SC (Burger, 2002). The SC Department of Natural Resources (SC DNR) is responsible for managing the state's alligators and overseeing hunts on public and private lands through the SC Alligator Management Program. Many of the alligators harvested in these yearly hunts are transported by the hunter to wildlife processing centers, where the whole alligator is processed and the meat is prepared for human consumption (for the respective hunters). Alligators are top-level predators, and contaminant concentrations in their muscle tissue (meat) should be reflective of their exposure to pollutants through dietary intake and contact with other environmental matrices (e.g., water, sediment) (Smith et al., 2007; Chumchal et al., 2011). Alligators are long lived,

aquatic, non-migratory species and have been shown to bioaccumulate environmental contaminants in previous studies (Campbell, 2003). Since the SC public hunt season opened in 2007 thousands of pounds of alligator meat have been harvested annually. According to the SC DNR, 440 alligators were harvested in the 2013 hunting season with a meat yield of 11,591 lb (Butfiloski, 2014). A total of 319 alligators were harvested in the 2015 hunting season with a meat yield of over 9000 lbs. (Butfiloski, 2015). Alligators in coastal SC can live to 50 and possibly >70 years of age (Wilkinson et al., 2016) and therefore have a great potential to bioaccumulate persistent environmental contaminants over time. Because of this pollutant accumulation potential and the potential human health risk associated with consuming contaminated meat, information on PFAA concentrations in meat of wild alligators in SC is critical so that management agencies may provide appropriate consumption guidelines that are protective of human health.

To date, there have been no studies on human consumption of alligator meat harvested in SC. Previous studies examining wild game consumption by SC hunters and anglers were conducted prior to the opening of a public alligator hunt season in 2007, and indicated that the diet of some recreationalists relied heavily on large game species (Burger, 2002). Dietary exposure assessments can be used by consumers and public health managers to estimate chemical exposure to specific populations based on game consumption rates and contaminant concentration data. When the source of potential exposure to a chemical is an identifiable food source (e.g., a particular game species) and contaminant data on that food source are available, it is appropriate to survey the population at risk for consumption of that food source. Surveys providing consumption data can be combined with existing contaminant data to determine risks under different consumption scenarios, as is often done for fish harvested from contaminated waterbodies (Harris and Jones, 2008). Upper bound and lower bound hypotheses have been used to determine major contributors to PFAA exposure in food exposure assessments (Yamada et al., 2014). This study employed consumption surveys of SC hunters that harvested alligators in the 2015 public hunt, combined with analysis of PFAA concentrations in meat from animals harvested during that time, to examine the potential dietary exposure of hunters and their families to PFAAs from consumption of alligator meat. Meat consumption and PFAA concentration scenarios were also explored to describe the range of potential PFAA exposures to the population and the main contributing factors. Understanding the range of potential exposures based on frequency of consumption, consumption amount and site-specific contaminant concentrations is essential for protecting human health through informed consumption recommendations. This study provides a first look at the consumption patterns of alligator hunters in SC and potential dietary exposure to PFOS through consumption of harvested meat.

1. Materials and methods

1.1. Study population

A consumption survey was conducted with 23 hunters that harvested an alligator in the 2015 public hunt season

(Sept. 12–Oct. 10) and who were visiting a wild game processor to have the meat prepared for consumption (Appendix A). The survey was administered in two ways: (1) an intercept survey administered at the location where alligator carcasses were dropped off for processing, and (2) a telephone survey of hunters unable to take the survey at the time of alligator carcass drop-off. The survey included questions on prior and future consumption rates, meal sizes, family composition, meat sharing, other dietary intake of fish and game species, as well as the location of alligator harvest. Self-reported harvest locations were checked against state hunt permit locations for validation. Surveys took approximately 15 min to complete. The survey was reviewed by the College of Charleston Institutional Review Board and found to be exempt. The survey was administered by researchers trained on survey administration and data collection. Surveys were either recorded on paper for later data entry or entered directly into Qualtrics Survey Software (Provo, UT). Qualtrics was also used to analyze the data individually and then collectively for percent responses. Similar response categories for consumption frequency were combined for data analysis.

1.2. Assessment of potential dietary exposure

PFAAs were analyzed using liquid chromatography tandem mass spectrometry (LC-MS/MS) as previously described in Reiner et al. (2012) and values are provided in Tipton et al. (accompanying paper). Consumption data obtained from surveys were combined with PFOS contaminant concentrations for corresponding alligator meat samples to determine actual dietary exposure based on the predicted consumption responses ($n = 19$). Four surveys with no corresponding meat samples were removed from the analysis. Predicted alligator meat consumption rates were converted to amount per day and multiplied by the predicted meal size (in ounces) and the concentration for the corresponding meat sample to determine daily dietary exposure. Daily dietary exposure was multiplied by 30.4 to determine predicted monthly dietary exposure. These data represent the actual dietary PFOS exposure hunters would experience if they consumed the alligator meat they harvested in 2015 in the amounts that they identified over the ensuing year.

Dietary exposure scenarios were used to analyze the range of consumption rates and contaminant concentrations to produce the average predicted dietary exposure, upper bound dietary exposure and lower bound dietary exposure. The SC coastal plain is divided into four hunt units: Unit 1 – Southern Coastal Counties, Unit 2 – Middle Coast Counties, Unit 3 – Midlands Counties and Unit 4 – Pee Dee Counties. Five scenarios were described for all hunt units combined and two scenarios were described for the Southern Coastal and Middle Coastal hunt units separately, to examine site-specific dietary exposure based on alligator harvest location. Average consumption rate and average meal size for all hunters was multiplied by the median PFOS concentration to determine the average dietary consumption rate per day and month. Each scenario was divided by 80 kg for an adult exposure and 15 kg for child exposure. Similar consumption rate categories were combined for analysis of average consumption data, and the average rate was used for the range of choices selected.

For example, for the “four to six times a year” category, five times a year was used to then determine frequency per day. For the “several times a month” category, three times a month was used as the frequency of consumption. The lower bound scenario for all hunt units used the lowest consumption rate, lowest meal size and lowest PFOS concentration in all meat samples to determine the lowest potential dietary exposure per day and per month. For the upper bound scenario for all hunt units, the highest consumption rate, largest meal size and highest PFOS concentration from all hunters and all meat samples in the study were used to determine the highest potential dietary exposure per day and per month. Lower bound and upper bound scenarios were calculated using the same method for specific hunt units, but included only the low and high PFOS concentrations from samples harvested within that unit.

2. Results and discussion

2.1. Potential dietary exposure

All hunters were over the age of 18 years old ($n = 23$) and the majority of hunters harvested their alligator from the Southern Coastal and Middle Coastal hunt units ($n = 13$, $n = 7$, respectively). More male hunters (82.6%) participated in the consumption survey than female hunters (17.4%). We found a wide range (1–8 years) reported for the number of years hunters have participated in harvesting an alligator in SC, with some being first-time hunters and others participating each year from the opening of the public hunt season in 2007. Most alligator hunters also harvested other wild game species in SC including deer (*Odocoileus virginianus*), turkey (*Meleagris gallopavo*), wild hogs (*Sus scrofa*), coyotes (*Canis latrans*), rabbit (*Sylvilagus spp.*), squirrel (*Sciurus carolinensis*), raccoon (*Procyon lotor*), waterfowl, doves (*Zenaidura macroura*) and quail (*Colinus virginianus*) (Fig. S1). Deer was the most common wild game harvested followed by turkey and wild hogs (94.7%, 73.7%, 68.4%, respectively; Fig. S1). These findings are similar to a survey of SC hunters from 1998 that found hunters most consumed wild caught fish, followed by deer, squirrel, quail and raccoon (Burger, 2002). No alligator consumption was reported in the previously described survey because the hunt season had not yet been established in the state.

When hunters were asked about their consumption of alligator meat over the previous year, answers ranged from none (21.7%) to at least once a month (21.7%), with the highest number of respondents eating meat 2–6 times throughout the past year (43.5%). When hunters consumed alligator meat over the previous year the most common portion size at one meal was 3 oz (44.4%) followed by 20.8 oz (27.8%). All but one respondent planned on eating alligator meat in the upcoming year and only one respondent planned on consuming meat from an alligator other than the one they harvested. When asked about planned alligator meat consumption for the upcoming year, hunters planned on consuming meat more frequently than in prior years, but the amount consumed per meal remained consistent (Table 1). Planned consumption of meat increased from two to six times in the previous year (18.2%) to at least once a month in the upcoming year (63.6%).

Table 1 – Survey responses for prior and expected alligator meat consumption.

Prior consumption (% response)		
Frequency (n = 23)	≥once a month	21.7
	7–11 times/year	8.7
	2–6 times/year	43.5
	None	21.7
Amount/meal (n = 18)	3 oz	44.4
	8 oz	16.7
	10.4 oz	11.1
	20.8 oz	27.8
Expected consumption (% response)		
Frequency (n = 22)	≥once a month	63.6
	7–11 times/year	18.2
	2–6 times/year	18.2
	N/A	
Amount/meal (n = 22)	3 oz	45.5
	8 oz	13.6
	10.4 oz	13.6
	20.8 oz	27.3

N/A: not applicable.

The planned portion size for the upcoming year remained similar to the previous year's sizes, with most hunters planning on consuming 3 oz per meal (45.5%), followed by 20.8 oz (27.3%). Considering the increased access to meat from the recent harvest, it is logical that frequency of consumption would increase. The large portion sizes reported (20.8 oz) may be the result of confusion about the survey question. Hunters may have misinterpreted the survey question regarding predicted portions size per meal to include family consumption rather than consumption by one individual, as most individuals do not in a meat portion of 20.8 oz. The increase in frequency of consumption of large portion sizes, 20.8 oz, could be a potential concern for human health and exceeds the SC alligator meat consumption guideline for sensitive populations of one, 8 oz meal per month (Butfiloski, 2016). This consumption guideline was established in 2016, a year after the sampling event; however, the exposure scenarios can be used to address the range of both dietary intake and contaminant concentrations in alligator meat, providing resource managers and risk assessors with valuable data for high and low consumers, based on locations of harvest.

When examining exposure scenarios there is a wide range of predicted exposures derived from dietary intake (consumption rates) for each survey participant and PFOS concentrations in their corresponding alligator meat samples. Dietary exposure based on the predicted consumption rates, meal sizes and corresponding meat sample concentrations ranged from 1.19 ng PFOS/day to 190 ng PFOS/day with a mean exposure of 81.3 ng PFOS/day (Table 2). The dietary exposure for PFOS showed quite a wide range based on the predicted consumption rates from the survey while other compounds showed a narrower range and lower amount dietary exposure (Table 3). Important to note, compounds such as PFUnA and

Table 2 – Potential dietary exposure for the minimum, maximum, mean, and median PFOS values based on predicted consumption rates and corresponding alligator meat PFOS concentrations.

	Daily exposure (ng/kg body weight per day)			Monthly exposure (ng/kg body weight per month)		
	Total (ng/day)	Adult	Child	Total (ng/month)	Adult	Child
Min	1.19	0.015	0.079	36.2	0.453	2.42
Max	190	2.37	12.7	5770	72.1	385
Mean	81.3	1.02	5.42	2470	30.9	165
Median	45.2	0.565	3.01	1370	17.2	91.6

PFDoA have a relatively high maximum daily exposure (83.1 and 97.4 ng/day, respectively) compared to the other PFAAs measured. For these compounds, which potentially have a high daily exposure, there is little research done on the toxicity. For all hunt units combined the lower bound dietary exposure scenario for an adult was found to be 0.006 ng PFOS/kg body weight per day and the upper bound 38.3 ng PFOS/kg body weight per day (Table 4). Under a scenario including the average consumption frequency, amount and median PFOS concentration, dietary exposure for an adult was 2.10 ng PFOS/kg body weight per day. Using the average consumption scenario we also determined dietary exposure based on the range of PFOS concentrations found in all samples and found that for adults the low concentration with an average consumption scenario resulted in a dietary exposure of 0.282 ng PFOS/kg body weight per day and the high concentration with an average consumption scenario resulted in a dietary exposure of 10.1 ng PFOS/kg body weight per day. The average dietary exposure scenario (168 ng PFOS/day; Table 4) overestimated the actual average predicted dietary exposure (81.3 ng PFOS/day), and may be more indicative of an exposure for high-end consumers. However, the average predicted dietary exposure was performed on a small sample size ($n = 19$) and included less than half of the contaminant concentrations reported from all harvested meat included in the study ($n = 43$). The average dietary exposure scenario uses the median of all PFOS concentrations and is a better indicator of potential exposures based on the inclusion of more PFOS tail meat data (Table 4). Also, considering that over one quarter of the predicted dietary exposure amounts per day exceeded 100 ng PFOS per day, it may be appropriate to provide consumption guidelines that address the subset of hunters that are on the high end of consumption rates.

The large range in exposure scenarios can be attributed to PFOS concentrations, which vary based on location of harvest, with the highest concentrations coming from the Middle Coastal hunt unit. This area has been previously recorded as a hotspot for PFOS contamination in wildlife and sediment studies (Fair et al., 2012; White et al., 2015; Bangma et al., 2017). The extremes in PFOS concentrations within alligator meat harvested for consumption results in a subset of hunters that experience increased dietary exposure from meat harvested in the Middle Coastal unit compared to other units in the state. When management agencies are producing wild game consumption guidelines, site-specific contamination

Table 3 – Potential dietary exposure for the minimum, maximum, mean, and median PFDA, PFUnA, PFDoA, PFTriA, and PFHxS values based on predicted consumption rates and corresponding alligator meat concentrations.

	PFDA dietary exposure (n = 19)		PFUnA dietary exposure (n = 19)		PFDoA dietary exposure (n = 19)	
	Daily exposure (ng/day)	Monthly exposure (ng/month)	Daily exposure (ng/day)	Monthly exposure (ng/month)	Daily exposure (ng/day)	Monthly exposure (ng/month)
Min	0.362	11.0	0.343	10.4	0.147	4.48
Max	25.2	767	83.1	2526	97.4	2961
Mean	8.41	256	15.0	456	14.6	443
Median	4.32	131	4.75	145	4.98	151
	PFTriA dietary exposure (n = 18)		PFHxS dietary exposure (n = 19)			
	Daily exposure (ng/day)	Monthly exposure (ng/month)	Daily exposure (ng/day)	Monthly exposure (ng/month)		
Min	0.085	2.59	0.0896	2.72		
Max	34.5	1048	5.78	176		
Mean	5.73	174	1.33	40.5		
Median	2.99	91	0.836	25.4		

and consumption rates should be considered (Conder and Arblaster, 2016). In the case of alligator meat in SC, the wide range of consumption rates within the population of hunters surveyed provides an impetus for a maximum consumption guideline/recommendation based on location of harvest. The vast difference between low and high PFOS concentrations based on the location of harvest and their strong influence over dietary exposure demonstrates why consumption guidelines for wild game should be site specific. Fish consumption guidelines in SC are based on both harvested species and waterbody of harvest; this study has shown that consumption guidelines for other aquatic game species, like alligators, should be managed in the same manner.

2.2. Potential exposure in children

In this study, we found that more than 45% of hunters were sharing alligator meat with children under 15 years old, with a

range of 1–4 children under 15 years old per household (Fig. S2). Half of the hunters responded that their children, under the age of 15, consumed alligator meat at the same frequency as they did. Children under 15 are of particular concern due to their vulnerability to health effects from contaminant exposure (Suk et al., 2015). A low body weight, but similar frequency of consumption can make dietary exposure for a child much higher than that of an adult (Tables 4 and 5). One consumption scenario for children exceeded the European Food Safety Authority (EFSA) PFOS consumption recommendation for adults of no more than 150 ng/kg body weight per day (European Food Safety, 2008). However this scenario is unlikely based on the large portion size used in our analysis. The wide range of consumption frequencies and portion size amounts can lead to higher levels of dietary exposure to PFOS in children. The European Food Safety also reported PFOA consumption recommendations of no more than 1500 ng/kg body weight per day (2008).

Table 4 – Potential dietary exposure from harvested alligator meat in 2015.*

		Total (ng/day)	Adult (ng/kg body weight per day)	Child (ng/kg body weight per day)
<i>All hunt units</i>				
LB	Low frequency × Small serving × Low conc.	0.475	0.00594	0.032
UB	High frequency × Large serving × High conc.	3070	38.3	204
Avg. low	Avg. frequency × Avg. serving × Low conc.	25.5	0.282	1.51
Avg.	Avg. frequency × Avg. serving × Median conc.	168	2.10	11.2
Avg. upper	Avg. frequency × Avg. serving × High conc.	911	10.1	53.7
<i>Southern coastal</i>				
LB	Low frequency × Small serving × Low conc.	0.475	0.006	0.032
Avg.	Avg. frequency × Avg. serving × Median conc.	75.3	0.942	5.02
UB	High frequency × Large serving × High conc.	1450	18.2	96.9
<i>Middle coastal</i>				
LB	Low frequency × Small serving × Low conc.	1.57	0.019	0.101
Avg.	Avg. frequency × Avg. serving × Median conc.	399	4.99	26.6
UB	High frequency × Large serving × High conc.	3070	38.3	204

* Lower bound (LB), upper bound (UB), and average (Avg.) consumption scenarios for PFOS in all alligator hunt units and for the Southern Coastal and Middle Coastal hunt units.

Table 5 – Lower bound, middle bound, and upper bound consumption scenarios for all alligator hunt units for PFDA, PFUnA, PFDoA, PFTriA, and PFHxS.

		Lower bound	Middle bound	Upper bound
		Low frequency × Small serving × Low conc.	Avg. frequency × Avg. serving × Median conc.	High frequency × Large serving × High conc.
PFDA	Adult	0.001	0.801	4.28
	Child	0.005	4.27	22.8
PFUnA	Adult	0.001	0.764	9.76
	Child	0.006	4.07	52.1
PFDoA	Adult	0.001	0.565	9.57
	Child	0.003	3.01	51.0
PFTriA	Adult	0.0004	0.412	2.71
	Child	0.002	2.20	14.5
PFHxS	Adult	0.0003	0.0935	0.286
	Child	0.002	0.499	1.53

PFOA was below the detection limit in all samples measured, so scenarios for PFOA were not determined. The other PFAAs detected in alligator meat samples were run through the potential exposure scenarios (Table 5). Since consumption recommendations do not exist for these compounds (PFDA, PFUnA, PFDoA, PFTriA, and PFHxS), little can be said if the potential consumption scenarios are below the recommended daily exposure. Consumption guidelines for fish species have specific warnings for children and women who are nursing, pregnant or may become pregnant to protect them from negative health effects during vulnerable developmental stages. Considering the extreme difference in potential dietary exposure to children based on the waterbody of harvest, future alligator consumption guidelines may consider restricting dietary intake by vulnerable populations for meat harvested specifically from the Middle Coastal hunt unit. These factors could have significant implications for the developmental, reproductive and immune health of the children exposed.

2.3. Potential exposure in adults

There are no adult dietary consumption scenarios (Table 4) from any hunt unit that exceed the current PFOS and PFOA consumption recommendations. The EFSA has described the tolerable daily intake (TDI) for PFOS to be 150 ng/kg body weight per day and 1500 ng/kg body weight per day for PFOA (European Food Safety, 2008). In this study we found no exposure, actual or in scenarios, to exceed these guidelines. We also found no PFOS concentrations that exceeded the “no restriction limit” of greater than 40 ppb, established by the Minnesota Department of Health (2008). However, the large ranges described in exposure scenarios are reflective of the wide range of consumption rates and contaminant concentrations. Additionally, no TDI for the other compound scenarios calculated this study exist (Table 5), so there is an uncertainty if these may lead to harmful health effects. All these consumption patterns show that there is a substantial portion of consumers on the low and high ends of consumption distribution (Tables 4 and 5). The large range in low and high concentrations demonstrates the potential for site-specific

exposure based on the location of harvest. Based on the results of this study, the consumption patterns of alligator meat by hunters and PFOS concentrations do not exceed levels that may be harmful to the health of adult humans from dietary exposure to PFOS. Although there was one predicted consumption amount that exceeded the SC DNR alligator meat consumption guideline for adults and one exposure scenario for children that exceeded the TDI level set by the EFSA, these were likely the result of a misinterpreted survey question (because the hunter may have responded with family consumption, instead of individual consumption) and a consumption scenario not likely to occur. However, we did find evidence that the location of alligator harvest and body weight of consumer were important factors in dietary exposure which should be considered when issuing future consumption recommendations.

2.4. Mixtures and other contaminants

Current consumption and drinking water guidelines only address PFOS and PFOA concentrations and do not consider the health impacts from other PFAAs or combined effects from mixtures. PFAA dosing studies have found that exposure to specific PFAAs result in different toxic effects (Kim et al., 2013; Jantzen et al., 2016). Fish exposed to PFNA alone were shown to have reproductive-related health effects (Zhang et al., 2016). There were five PFAAs (PFDA, PFUnA, PFDoA, PFHxS and PFOS; Tipton et al., *accompanying paper*) routinely detected in alligator meat from SC. Previous studies have reported similar results with a variety of PFAAs detected in wildlife (Ye et al., 2008; Reiner and Place, 2015; Bangma et al., 2017; Christie et al., 2016). Exposure to a mixture of PFAAs has been described to have a stronger effect on hormone receptors than doses of a single PFAA (Kjeldsen and Bonefeld-Jørgensen, 2013). Further, mixtures of PFAAs have been observed to result in health effects at concentrations lower than those resulting in toxicity from a single PFAA (Gorochategui et al., 2014). Future health advisories should consider the implications from exposure to PFAAs other than only PFOS and PFOA and include guidelines that reflect the mixture of PFAAs found in wildlife.

A prior study of SC hunters and anglers found that there is a subset of the hunting and fishing population that relies heavily on harvested meat in its diet (Burger, 2002). Risk assessments require site-specific data to account for regional and cultural differences in consumption combined with contaminant data that address the appropriate species and locations of harvest. This study used a multidisciplinary approach to evaluate the concentrations of PFAAs within the tail meat of alligators harvested and processed for consumption during the 2015 hunt season and the consumption patterns of hunters. Contaminant data revealed site-specific differences in PFAA concentrations that potentially lead to site-specific dietary exposure in hunters and their families. Although no PFAA concentrations exceed available consumption guidelines for adults, respondents from the high end of the consumption scenario distribution and vulnerable populations consuming harvested alligator meat may be at risk from other environmental pollutants (e.g., mercury, organochlorine pesticides, polychlorinated biphenyls, dioxins). This study focused only on PFAA concentrations in alligator meat

and did not examine potential dietary exposure to other contaminants or mixtures of contaminants (mercury, PCBs, pesticides, etc.) that may be harmful to human health at the described consumption patterns. Future studies of other chemical contaminants in these samples can utilize the consumption data collected from alligator hunters in the 2015 season to evaluate potential dietary exposure to these other environmental contaminants and mixtures.

Disclaimer

Certain commercial equipment or instruments are identified in the paper to specify adequately the experimental procedures. Such identification does not imply recommendations or endorsement by the NIST nor does it imply that the equipment or instruments are the best available for the purpose. This paper represents Technical Contribution No. 6540 of the Clemson University Experiment Station.

Acknowledgments

Funding and support for this research was provided by the Graduate School at the College of Charleston. We would like to give special thanks to Cordray's Processing and Taxidermy for allowing us to utilize their facility and to collect samples.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.jes.2017.05.046>.

REFERENCES

- Bangma, J.T., Bowden, J.A., Brunell, A.M., Christie, I., Finnell, B., Guillette, M.P., et al., 2017. Perfluorinated alkyl acids in plasma of American alligators (*Alligator mississippiensis*) from Florida and South Carolina. *Environ. Toxicol. Chem.* 36, 917–925.
- Burger, J., 2002. Daily consumption of wild fish and game: exposures of high end recreationists. *Int. J. Environ. Health Res.* 12, 343–354.
- Butenhoff, J.L., Chang, S.C., Olsen, G.W., Thomford, P.J., 2012. Chronic dietary toxicity and carcinogenicity study with potassium perfluorooctanesulfonate in Sprague Dawley rats. *Toxicology* 293, 1–15.
- Butfiloski, J., 2014. Alligator hunting season report 2013. F&AP Report. South Carolina Department of Natural Resources, Wildlife and Freshwater Fisheries Division.
- Butfiloski, J., 2015. Alligator hunting season report 2015. F&AP Report. South Carolina Department of Natural Resources, Wildlife and Freshwater Fisheries Division.
- Butfiloski, J., 2016. South Carolina alligator hunting guide for 2016. F&AP Report. South Carolina Department of Natural Resources, Wildlife and Freshwater Fisheries Division.
- Calafat, A.M., Wong, L.Y., Kuklennyik, Z., Reidy, J.A., Needham, L.L., 2007. Polyfluoroalkyl chemicals in the U.S. population: data from the national health and nutrition examination survey (NHANES) 2003–2004 and comparison with NHANES 1999–2000. *Environ. Health Perspect.* 115, 1596–1602.
- Campbell, K.R., 2003. Ecotoxicology of crocodylians. *Appl. Herpetol.* 1, 45–163.
- Christie, I., Reiner, J.L., Bowden, J.A., Botha, H., Cantu, T.M., Govender, D., et al., 2016. Perfluorinated alkyl acids in the plasma of South African crocodiles (*Crocodylus niloticus*). *Chemosphere* 154, 72–78.
- Chumchal, M.M., Rainwater, T.R., Osborn, S.C., Roberts, A.P., Abel, M.T., Cobb, G.P., et al., 2011. Mercury speciation and biomagnification in the food web of Caddo Lake, Texas and Louisiana, USA, a subtropical freshwater ecosystem. *Environ. Toxicol. Chem.* 30, 1153–1162.
- Conder, J.M., Arblaster, J.A., 2016. Development and use of wild game consumption rates in human health risk assessments. *Hum. Ecol. Risk Assess.* 22, 251–264.
- European Food Safety, 2008. Perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA) and their salts scientific opinion of the panel on contaminants in the food chain. *EFSA J.* 653, 1–131.
- Fair, P.A., Houde, M., Hulse, T.C., Bossart, G.D., Adams, J., Balthis, L., et al., 2012. Assessment of perfluorinated compounds (PFCs) in plasma of bottlenose dolphins from two southeast US estuarine areas: relationship with age, sex and geographic locations. *Mar. Pollut. Bull.* 64, 66–74.
- Fair, P.A., Romano, T., Schaefer, A.M., Reif, J.S., Bossart, G.D., Houde, M., et al., 2013. Associations between perfluoroalkyl compounds and immune and clinical chemistry parameters in highly exposed bottlenose dolphins (*Tursiops truncatus*). *Environ. Toxicol. Chem.* 32, 736–746.
- Fox, G.A., 2001. Wildlife as sentinels of human health effects in the Great Lakes – St. Lawrence basin. *Environ. Health Perspect.* 109 (Suppl. 6), 853–861.
- Gorochategui, E., Pérez-Albaladejo, E., Casas, J., Lacorte, S., Porte, C., 2014. Perfluorinated chemicals: differential toxicity, inhibition of aromatase activity and alteration of cellular lipids in human placental cells. *Toxicol. Appl. Pharmacol.* 277, 124–130.
- Grandjean, P., Andersen, E.W., Budtz-Jørgensen, E., Nielsen, F., Mølbak, K., Weihe, P., et al., 2012. Serum vaccine antibody concentrations in children exposed to perfluorinated compounds. *J. Am. Med. Assoc.* 307, 391–397.
- Harris, S.A., Jones, J.L., 2008. Fish consumption and PCB-associated health risks in recreational fishermen on the James River, Virginia. *Environ. Res.* 107, 254–263.
- Haug, L.S., Huber, S., Becher, G., Thomsen, C., 2011. Characterisation of human exposure pathways to perfluorinated compounds - comparing exposure estimates with biomarkers of exposure. *Environ. Int.* 37, 687–693.
- Jantzen, C.E., Annunziato, K.A., Bugel, S.M., Cooper, K.R., 2016. PFOS, PFNA, and PFOA sub-lethal exposure to embryonic zebrafish have different toxicity profiles in terms of morphometrics, behavior and gene expression. *Aquat. Toxicol.* 175, 160–170.
- Kim, M., Son, J., Park, M.S., Ji, Y., Chae, S., Jun, C., et al., 2013. *In vivo* evaluation and comparison of developmental toxicity and teratogenicity of perfluoroalkyl compounds using *Xenopus* embryos. *Chemosphere* 93, 1153–1160.
- Kjeldsen, L.S., Bonefeld-Jørgensen, E.C., 2013. Perfluorinated compounds affect the function of sex hormone receptors. *Environ. Sci. Pollut. Res.* 20, 8031–8044.
- Mazzotti, F.J., Best, G.R., Brandt, L.A., Cherkiss, M.S., Jeffery, B.M., Rice, K.G., 2009. Alligators and crocodiles as indicators for restoration of everglades ecosystems. *Ecol. Indic.* 9, S137–S149.
- Minnesota Department of Health, 2008. Fish consumption advisory program: meal advice categories based on PFOS in fish. <http://www.health.state.mn.us/divs/eh/fish/eating/mealadvicetables.pdf> (Accessed January 11, 2017).
- Persson, S., Magnusson, U., 2015. Environmental pollutants and alterations in the reproductive system in wild male mink (*Neovison vison*) from Sweden. *Chemosphere* 120, 237–245.

- Reiner, J.L., Place, B.J., 2015. Perfluorinated alkyl acids in wildlife. In: DeWitt, J.C. (Ed.), *Toxicological Effects of Perfluoroalkyl and Polyfluoroalkyl Substances*. Springer, Switzerland, pp. 127–150.
- Reiner, J.L., O'Connell, S.G., Butt, C.M., Mabury, S.A., Small, J.M., De Silva, A.O., et al., 2012. Determination of perfluorinated alkyl acid concentrations in biological standard reference materials. *Anal. Bioanal. Chem.* 404, 2683–2692.
- Smith, P.N., Cobb, G.P., Godard-Codding, C., Hoff, D., McMurry, S.T., Rainwater, T.R., et al., 2007. Contaminant exposure in terrestrial vertebrates. *Environ. Pollut.* 150, 41–64.
- Suk, W., Ahanchian, H., Ansong Asante, K., Carpenter, D., Diaz-Barriga, F., Ha, E., et al., 2015. Environmental pollution: an under-recognized threat to children's health, especially in low- and middle-income countries. *Environ. Health Perspect.* A42–A45.
- Tipton, J.J., Guillette Jr., L.J., Lovelace, S., Parrott, B.B., Rainwater, T.R., Reiner, J.L., 2017. Analysis of PFAAs in American alligators part 1: concentrations in alligators harvested for consumption during South Carolina public hunts. *J. Environ. Sci.* 61, 24–30.
- White, N.D., Balthis, L., Kannan, K., De Silva, A.O., Wu, Q., French, K.M., et al., 2015. Elevated levels of perfluoroalkyl substances in estuarine sediments of Charleston, SC. *Sci. Total Environ.* 521–522, 79–89.
- Wilkinson, P.M., Rainwater, T.R., Woodward, A.R., Leone, E.H., Carter, C., 2016. Determinate growth and reproductive lifespan in the American alligator (*Alligator mississippiensis*): evidence from long-term recaptures. *Copeia* 104, 843–852.
- Yamada, A., Bemrah, N., Veyrand, B., Pollono, C., Merlo, M., Desvignes, V., et al., 2014. Dietary exposure to perfluoroalkyl acids of specific French adult sub-populations: high seafood consumers, high freshwater fish consumers and pregnant women. *Sci. Total Environ.* 491–492, 170–175.
- Ye, X.B., Schoenfuss, H.L., Jahns, N.D., Delinsky, A.D., Strynar, M.J., Varns, J., et al., 2008. Perfluorinated compounds in common carp (*Cyprinus carpio*) filets from the Upper Mississippi River. *Environ. Int.* 34, 932–938.
- Zhang, W., Sheng, N., Wang, M., Zhang, H., Dai, J., 2016. Zebrafish reproductive toxicity induced by chronic perfluorononanoate exposure. *Aquat. Toxicol.* 175, 269–276.