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Analysis of PFAAs in American alligators part 1: Concentrations in alligators harvested for consumption during South Carolina public hunts

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ABSTRACT

Environmental contamination resulting from the production or release of harmful chemicals can lead to negative consequences for wildlife and human health. Perfluorinated alkyl acids (PFAAs) were historically produced as protective coatings for many household items and currently persist in the environment, wildlife, and humans. PFAAs have been linked to immune suppression, endocrine disruption, and developmental toxicity in wildlife and laboratory studies. This study examines the American alligator, *Alligator mississippiensis*, as an important indicator of ecosystem contamination and a potential pathway for PFAA exposure in humans. Alligator meat harvested in the 2015 South Carolina (SC) public hunt season and prepared for human consumption was collected and analyzed for PFAAs to determine meat concentrations and relationships with animal body size (total length), sex, and location of harvest. Of the 15 PFAAs analyzed, perfluorooctane sulfonate (PFOS) was found in all alligator meat samples and at the highest concentrations (median 6.73 ng/g). No relationship was found between PFAA concentrations and total length or sex. Concentrations (of one or all compounds varied significantly across sampling locations, with alligators harvested in the Middle Coastal hunt unit having the highest PFOS concentrations (median 16.0 ng/g; $p = 0.0001$). Alligators harvested specifically from Berkley County, SC (located in the Middle Coastal hunt unit) had the highest PFOS concentrations and the greatest number of PFAAs detected ($p < 0.0001$). The site-specific nature of PFAA concentrations in alligator meat observed in this study suggests a source of PFAA contamination in Berkley County, SC.

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Introduction

Perfluorinated alkyl acids (PFAAs) are synthetic chemicals characterized as having C–F bonds of varying chain length and have been historically used in the production of many common household items (Prevedouros et al., 2006). The water and stain resistant properties of PFAAs have led to their use as surfactants in many commercial products. Additionally, PFAAs and their precursors can be found in aqueous film forming foams, carpets, non-stick cookware, and paper used for food packaging (Prevedouros et al., 2006). PFAAs are stable because of the strong carbon–fluorine bond and persistent in the environment. Chain lengths equal to or greater than 8 carbons are known to accumulate in ecosystems and wildlife (Butt et al., 2008). PFAAs have been found in rainwater, dust, fresh and saltwater bodies, and are believed to be capable of long-range transport by ocean currents and atmospheric transport (Armitage et al., 2006, 2009; Yamashita et al., 2008). Concern about environmental accumulation and potential human health effects has led to strict regulations on the production of PFAAs in the United States and a strategic plan for a phase-out of production was planned for completion in 2015 (EPA, 2000). PFAAs have an extremely long half-life in humans and the environment and are resistant to breakdown from many thermal and chemical processes (Olsen et al., 2012).

PFAAs have been measured in wildlife and humans around the world. The two most common PFAAs found in the environment and wildlife are perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA). The stability and long-range transport potential of PFAAs and their precursors has contributed to elevated levels of these contaminants in Arctic wildlife, far removed from point sources (Butt et al., 2010). Biomagnification of PFAAs in wildlife has been well recorded, with PFOS concentrations positively related to trophic level and risk of negative health effects (Keller et al., 2012; Muller et al., 2011; Loi et al., 2011; Yordy et al., 2013). Marine mammal studies have linked PFAA exposure to altered renal and hepatic function, as well as immune suppression (Fair et al., 2013). Studies conducted on laboratory animals have also shown immune suppression and increased mortality in PFOS exposed mice (Guruge et al., 2009), decreases in reproductive success in zebrafish from estrogenic effects after exposure to perfluorononanoic acid (PFNA) (Jantzen et al., 2016; Zhang et al., 2016), and developmental abnormalities and liver damage in clawed frogs exposed to PFAAs (Kim et al., 2013).

The impacts on human health from exposure to PFAAs need further investigation; however, current research suggests that exposure to PFAAs could result in endocrine disruption (Kjeldsen and Bonefeld-Jørgensen, 2013), developmental toxicity (Kjeldsen and Bonefeld-Jørgensen, 2013), and immune suppression (Grandjean et al., 2012). Most human exposure to PFAAs in the United States (U.S.) is through ingestion of contaminated food or drinking water (Lindstrom et al., 2011). Exposure of high risk populations, such as pregnant women and children, is especially concerning due to the nature of health implications from exposure to PFAAs, including impacts to development and immune function.

Charleston Harbor in South Carolina (SC), U.S., is a potential hotspot for PFAA contamination. A study examining PFAA levels in bottlenose dolphins (*Tursiops truncatus*) found some of the highest serum concentrations reported in marine mammals, at similar levels to humans occupationally exposed (Fair et al., 2012). Estuarine sediments tested from the Charleston Harbor watershed were found to have the highest PFAA levels of any urban area recorded in the U.S., with levels increasing from 2004 to 2014 (White et al., 2015). Despite the alarming levels of PFOS and PFOA concentrations being reported in the sediment and wildlife in waterbodies of SC, few studies have examined PFAA contaminant exposure of other aquatic and terrestrial species (Bangma et al., 2017a; Yordy et al., 2013). PFAA accumulation in apex predators such as bottlenose dolphins and contaminant input from unknown sources provide an impetus for a closer investigation of SC wildlife potentially at risk.

Few studies have been conducted on PFAA contamination levels in reptiles (Keller et al., 2012; Wang et al., 2013; Christie et al., 2016; Bangma et al., 2017a, 2017b), and all of these have used blood as a focal tissue for analysis; none have examined PFAA concentrations in other tissues, including those potentially consumed by other wildlife or humans. The American alligator (*Alligator mississippiensis*) is a highly sought after wild game species for which a hunting season has recently (2007) been established in SC. Alligators are apex predators in many freshwater and brackish water ecosystems and play a key role in maintaining a balanced aquatic food web (Nifong and Silliman, 2013). The American alligator can be used as a sentinel species and indicator of contaminants within an ecosystem due to its long lifespan, non-migratory range, and high trophic status (Milnes and Guillette, 2008). There is no current research available on PFAA concentrations in alligator meat harvested in SC's recreational hunts and collected for consumption despite high meat yields from hunts in 2013 (>11,000 lb) and 2015 (>9000 lb) (Butfiloski, 2014, 2015). Obtaining exposure information for harvested alligators is critical in determining areas of environmental concern, negative impacts on wildlife health, and potential exposure to humans through consumption of harvested meat. In this study we sampled 43 American alligators recreationally harvested throughout the SC coastal plain during the 2015 public hunt season to determine the concentration of PFAAs in tail meat collected for consumption. Additionally, we examined the relationships between PFAA concentration in tail meat and alligator body size, sex, and location of harvest.

1. Materials and methods

1.1. Sample collection

Tail meat samples (approximately 500 g) were collected opportunistically during the SC recreational hunt season from September 12, 2015 to October 10, 2015 at a local wild game meat processor. Each sample was consistently collected by a licensed processor from the base (anterior end) of the tail using a clean filet knife. A total of 43 samples were collected, individually wrapped in methanol rinsed tin foil, placed on

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