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HIGHLIGHTS

• Seafood items had unique Hg-nutrient signatures reflecting ecology and physiology.

• Seafood consumers had unique blood Hg-nutrient signatures reflecting diet habits.

• Top predator consumers had high blood Hg, similar nutrients than other consumers.

• Consumers with salmon diet had distinct, high % omega-3 fatty acids in blood.

• Seafood type is necessary to understand risks and benefits of seafood consumption.

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ABSTRACT

Dietary recommendations for seafood are confusing due to the desire to balance both benefits from nutrients and risks from contaminants. The overall health value of different fish and shellfish items depends on concentrations of multiple nutrients (e.g., selenium (Se), omega-3 fatty acids) and contaminants (e.g., mercury (Hg)). However, few studies have examined the connections between human exposure to multiple nutrients and contaminants and the consumption of specific types of seafood. Our goals were to compare 1) Hg, Se and omega-3 fatty acid concentrations (Hg-nutrient signatures) among common fish and shellfish items and 2) Hg-nutrient signatures in the blood of avid seafood consumers, based on seafood consumption habits. We compiled nutrient and Hg concentration data for common fish and shellfish items from the literature. We also measured blood concentrations of Hg and seafood nutrients collected from adult, avid seafood consumers on Long Island, NY. Canonical discriminant analyses revealed distinct Hg-nutrient signatures among seafood items, and these signatures were reflected in the blood of consumers based on different consumption habits. For example, consumers with a salmon-dominated seafood diet had relatively high percentage of omega-3 fatty acids in blood, and consumers who tend to eat top predator seafood have higher Hg, but similar blood nutrient concentrations compared to consumers who tend to eat low trophic level seafood. These results provide direct evidence of links between the ecological characteristics of the type of seafood consumed and Hg-nutrient exposure. This approach helps assess the overall human health value of specific seafood types, leads to specific diet recommendations, and can be used to characterize risk:benefit status among seafood consumers.

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1. Introduction

The overall human health value of seafood is complex, but important to understand as seafood consumption is increasing worldwide (Food and Agriculture Organization of the United Nations, 2010). Seafood is a omega-3 fatty acids that have well known health benefits (Oken et al., 2004; Olsen et al., 1993; Simopoulos, 2002; Siscovick et al., 1995). However, some seafood items are also primary sources of contaminants, including mercury (Hg), PCBs, and other persistent organic pollutants that have known adverse health effects (Grandjean et al., 1997; Nyland et al., 2011; Oken et al., 2005; Salonen et al., 1995; Stern, 2005; Stewart et al., 2008; Turyk et al., 2006). Thus, the overall risk and benefits of seafood consumption depend on the extent of exposure and resultant health effects of multiple nutrients and contaminants, many of which vary among seafood items (Karimi et al., 2012; Mahaffey et al., 2008). Studies examining patterns of individual seafood nutrients

lean source of protein and a primary source of other nutrients including

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Abbreviations: Hg, mercury; Se, selenium; EPA, eicosapentaenoic acid; DHA, docosahexaenoic acid; USDA, U.S. Department of Agriculture.

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or contaminants are valuable in their focus, but make it difficult to compare the overall nutritional and toxicological quality among seafood items. Studies that simultaneously compare patterns of multiple seafood nutrients and contaminants will allow us to compare the relative, overall health value of different seafood items, and can help inform seafood consumption advice.

One consequence of the inherent complexity of the health value of seafood is that seafood consumption advice, namely mercury advisories, can dissuade consumers from eating seafood (Engelberth et al., 2013; Lando et al., 2012; Oken et al., 2003; Shimshack et al., 2007) in some cases below current recommended intake levels (American Heart Association, 2013; U.S. Department of Agriculture U.S. Department of Health Human Services, 2010). These studies suggest that consumers are more sensitive to potential health risks of eating seafood even when the potential benefits outweigh the risks. Moreover, many studies examining the health value of eating fish focus on risk from exposure to Hg and other contaminants (Turyk et al., 2012), with more studies needed to examine risks and benefits together. A fundamental challenge to developing consumption advice is that contaminant and nutrient content varies among and within seafood items (Karimi et al., 2012; Mahaffey et al., 2008), leading to discrepancies in advice specific for individual seafood items (Gerber et al., 2012). To help characterize the overall risk-benefits of individual seafood items and develop consumption advice, there is a need to compare the relative, overall quality of seafood items based on multiple nutrient and contaminant factors, and to examine exposure to these nutrients and contaminants in seafood consumers.

Our goal was to take an initial step toward fulfilling this need by examining Hg-nutrient patterns in seafood items and in the blood of avid, or regular seafood consumers. First, we compare Hg-nutrient signatures (relative concentrations of Hg, Se, and omega-3 fatty acids eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA)) among individual seafood items by summarizing data from the literature. We focus on Hg, Se, EPA and DHA because seafood is the primary source of these substances to humans (Mahaffey, 2004; Mozaffarian and Rimm, 2006; Svensson et al., 1992; USDA, 2012), and they have known health implications. Second, we examine whether Hg-nutrient signatures in the blood of avid seafood consumers reflect different seafood consumption habits, such as the dominant seafood type consumed. In general, we expect seafood items to have unique Hg-nutrient signatures that reflect their ecological characteristics, such as position in the food web, and taxonomic identity. Similar unique signatures have been observed in other aquatic organisms (Karimi and Folt, 2006). In addition, we expect seafood consumers to have Hg-nutrient signatures that match their consumption habits, such as the preferred seafood item consumed. These analyses will help identify the individual seafood items that are important sources of Hg and individual nutrients to consumers. This information is useful to better compare the relative risk-benefits of individual seafood items (Gerber et al., 2012; Ginsberg and Toal, 2009), and provides a framework for research that goes further by including other contaminants and nutrients.

2. Methods

2.1. Mercury–nutrient signatures in seafood

We collected a mean and standard deviation concentration for Hg, Se and omega-3 fatty acids in raw, edible tissue for each common seafood item from published studies and public databases. Direct measurements of Hg and nutrient content of the seafood consumed by study participants are difficult to obtain and were beyond the scope of this study. Therefore, we used estimates from the literature that would best represent U.S. commercial seafood items. Means and standard deviations of seafood concentrations are more commonly reported than raw data in the literature (Karimi et al., 2012). Mercury values were obtained from the Seafood Hg Database of commercial seafood (Karimi et al., 2012), and for alewife and sturgeon using the same

method as that used for seafood items in the database. Similarly, most omega-3 estimates were obtained from the USDA Nutrient Database of commercial foods, and others from the peer-reviewed literature (Ackman, 2000). We assumed that alewife had the same EPA and DHA content as herring (Crawford et al., 1986). For omega-3 fatty acids, we focused on EPA and DHA because fish and shellfish are a primary source of these particular fatty acids (Ackman, 2000; U.S. Department of Agriculture U.S. Department of Health Human Services, 2010). Selenium values in seafood are not as well studied as Hg or fatty acids. Therefore, we used Se estimates from the literature that were likely to be included in the U.S. commercial market (sensu Karimi et al., 2012). Selenium values were obtained from multiple data sources (Bourre and Paquotte, 2008; Burger and Gochfeld, 2005; Burger and Gochfeld, 2011; Hall et al., 1978; Kaneko and Ralston, 2007; Karimi et al., 2013; Sweden National Food Agency, 2013; USDA, 2012; Wander and Patton, 1991). For cases in which a data source reported multiple mean and standard deviations of Se for a given seafood item (Kaneko and Ralston, 2007; Karimi et al., 2013), we calculated a mean of means, and mean of standard deviations for the data source. For studies that did not report a standard deviation or standard error for Se, standard deviation was calculated from the reported mean and a coefficient of variation of 0.29, typical for Se concentrations in seafood taxa (Karimi et al., 2013). For studies that did not report standard deviation or standard error for EPA or DHA, or if sample size was 1, we calculated standard deviation from the reported mean, and a coefficient of variation of 0.58 and 0.48 for EPA and DHA, respectively, based on typical CVs for seafood items (USDA, 2012). All Hg, Se, EPA and DHA values were reported as, or converted to $\mu g g^{-1}$ (wet weight). For composite seafood items of multiple taxa (e.g., "Swordfish, Shark, Marlin", Table S1), we included one estimate for Hg or nutrients for each taxa from the literature and calculated grand mean (mean of means across taxa) and grand standard deviation (mean of standard deviations across taxa) for Hg, Se, and omega-3 fatty acid concentrations. For noncomposite seafood items, such as "canned tuna, white", we used only one estimate for Hg or nutrient from the literature.

Second, we generated raw values for Hg and individual nutrients based on means and standard deviations from the literature for each seafood item assuming lognormal distributions. Lognormal distributions are common for seafood contaminant and nutrient concentrations (Giesy and Wiener, 1977; Karimi and Folt, 2006). For each substance (Hg or nutrient) and each seafood item, we calculated lognormal standard deviations (s) and lognormal means (μ) as

$$s = \sqrt{\log\left(1 + \left(\frac{\text{grand SD}}{\text{grand mean}}\right)^2\right)} \tag{1}$$

and

$$\mu = \log_{10}(\text{grand mean}) - \frac{s^2}{2}.$$
 (2)

We used lognormal means and lognormal standard deviations to generate 50 replicate, random values for each substance (Hg or nutrient), and each seafood item using the rnorm function in R (R Core Team, 2013). Studies of aquatic organisms use approximately 10–100 replicates (individual organisms) to statistically compare Hg or nutrients in concentrations among taxa (Budge et al., 2002; Karimi et al., 2013). Therefore, we chose to generate 50 replicate values in order to be within this range, and to be likely to distinguish Hg–nutrient signatures among seafood items.

2.2. Mercury-nutrient signatures in the blood of avid seafood consumers

We conducted a study on Hg and nutrient exposure from seafood consumption in which we measured mercury and nutrient concentrations in blood samples from adult, avid seafood consumers. The study was

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