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Association of methylmercury intake from seafood consumption and blood mercury level among the Asian and Non-Asian populations in the United States

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ABSTRACT

Background: MeHg is a well-established neurotoxicant for fetal brain growth and development and has been shown to increase the risk of cardiovascular disease in aging populations. In the U.S., Asian populations are of particular concern because of their seafood consumption behaviors.

Objectives: Our objective was to calculate the average daily MeHg intake (ADMI) from seafood and to assess the relationship between ADMI with blood methylmercury (BMeHg) concentrations, specifically among women of reproductive age (WORA) and adults ≥ 50 years of age.

Methods: We estimated ADMI from seafood using the 30-day fish consumption data from the NHANES 2011–2014 datasets. Using multivariable linear regression, we estimated the proportional change in mean BMeHg associated with a doubling of the ADMI. Further, correlations between ADMI and BMeHg were compared between Asians and other racial/ethnic groups.

Results: Our analysis found both Asian WORA and Asian adults age ≥ 50 years old had significantly higher BMeHg levels and ADMI than their Non-Asian counterparts. Correlations between ADMI from seafood and blood Hg levels were stronger among Asian WORA than among Non-Asian WORA. Key fish species that influenced the dietary MeHg intake for Asians were mackerel, tuna, and "other known/unknown fish species".

Conclusion: We confirmed that Asian populations have higher MeHg intake than the Non-Asian population in the U.S. and seafood intake is a key predictor of blood Hg concentration, especially among Asian women of reproductive age. Future studies should incorporate information on other known and unknown fish species that are frequently consumed by Asian populations and different parts and fish organs eaten to better understand determinants of MeHg exposure.

1. Background

Mercury (Hg) is a naturally occurring metal found throughout the environment. Release of Hg into the environment is mainly from human activities, particularly coal-fired power stations, residential coal burning, industrial processes, waste incinerators and as a result of mining for mercury, gold and other metals (WHO, 2016). Mercury combines with carbon and hydrogen to make organic mercury compounds. The most common organic mercury compound, methylmercury (MeHg), is produced mainly by microscopic organisms in sediments and soil and can accumulate up the aquatic food chain, leading to elevated concentrations in predatory fish (ATSDR 2016; Patrick, 2002). For the general human population, the predominant route for MeHg exposure is dietary exposure via consumption of seafood, especially fish (Diez, 2009).

MeHg is of particular public health concern because it is a well-

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Abbreviations: ADMI, average daily MeHg intake; A/P/N/M, Asian, Pacific Islander, Native American, or multiracial; BMeHg, blood methylmercury; CI, confidence interval; DHA, docosahexaenoic acid; FAO/WHO, Food and Agriculture Organization of the United Nations and World Health Organization; Hg, mercury; MEC, Mobile Examination Center; MeHg, methylmercury; N/A, not available; NHANES, The National Health and Nutrition Examination Survey; R^2, coefficient of multiple correlation; Se, selenium; TBHg, total blood mercury; WHO, World Health Organization; WORA, women of reproductive age

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established neurotoxicant (ATSDR 2016; Myers et al., 2000). MeHg can cross the placenta, penetrate the blood-brain barrier and concentrate in the fetus (Diez, 2009; Morrissette et al., 2004; Stern and Smith, 2003), which can increase the risk of behavioral and cognitive impairment in offspring (Grandjean et al., 1997; Myers et al., 2000; Oken et al., 2005; Prpić et al., 2017; Stokes-riner et al., 2012). However, seafood also contain nutrients, such as docosahexaenoic acid (DHA), an omega-3 long chain polyunsaturated fatty acid that is important for fetal brain growth and development and has cognitive benefits for children (Daniels et al., 2004; Oken et al., 2008). Therefore, balancing the risks and benefit of seafood consumption is critical for healthy neurological development of the fetus. In 2010, the joint Food and Agriculture Organization of the United Nations and World Health Organization (FAO/WHO) report found that maternal fish consumption is associated with optimal neurodevelopment in her offspring (WHO and FAO, 2010). Fish is also highly recommended to prevent cardiovascular diseases (Djoussé et al., 2012; WHO and FAO, 2010; Zheng et al., 2012). At the same time, a growing body of evidence has been shown that the risk of cardiovascular disease increases with elevated levels of blood MeHg (Genchi et al., 2017; Roman et al., 2011; Stern, 2005)

Investigations in Asian countries [e.g., Cambodia (Agusa et al., 2007), Taiwan (Hsu et al., 2007), Japan (Sakamoto et al., 2007)] have reported fish/shellfish consumption levels greater than average worldwide consumption. Studies have also demonstrated that in the U.S., people of Asian descent, whose food choices are influenced by Asian dietary patterns (Sechena et al., 2003; Xu and Newman, 2015), consume seafood more frequently, in greater variety, and in greater quantity than non-Asian Americans. Asian populations in the United States have higher Hg levels compared to other racial/ethnic groups due to their higher seafood consumption (Buchanan et al., 2015a; Lin et al., 2013; Mahaffey et al., 2009; Mckelvey et al., 2007; Sechena et al., 2003; Tsuchiya et al., 2008a, 2008b; Xu and Newman, 2015). McKelvey et al. found that Asians in New York City have higher blood mercury levels than other racial/ethnic groups and that foreign-born Chinese New Yorkers, in particular, had higher mercury levels than non-Asian frequent fish consumers (Mckelvey et al., 2007). Buchanan, Anglen and Turyk (Buchanan et al., 2015a) found that both Asian women of reproductive age (WORA) and Asian adults older than 50 years had higher BMeHg levels than other racial/ethnic groups. About a quarter of Asian WORA had BMeHg greater than 3.5 μg/L, a proposed level of concern that accounts for fetal concentration of maternal MeHg (Mahaffey et al., 2004; Stern and Smith, 2003).

The National Health and Nutrition Examination Survey (NHANES) is designed to assess the health and nutritional status of adults and children in a nationally representative sample (US NHANES). Since 2011–2012, NHANES has measured total blood mercury (TBHg) and, in addition, speciated blood methylmercury (BMeHg) to include a direct measurement of MeHg (NCHS, 2011). Because blood mercury is predominantly in the methylated form in the seafood consuming population (Mahaffey et al., 2009), past epidemiological studies have used TBHg or TBHg minus urinary inorganic Hg rather than BMeHg to estimate exposure to MeHg (Ayotte et al., 2011; Miller et al., 2016; Valera et al., 2008).

The 2011–2012 NHANES survey also initiated a separate race/ethnicity category for Asians, who were previously included in a category with "other race" (NCHS, 2011). In prior investigations using the new Asian race category, 2011–2012 NHANES data have been used to assess the distribution of BMeHg concentrations for Asian and non-Asian women of reproductive age (WORA) and the older adult population (Buchanan et al., 2015a), and in the overall US population (Mortensen et al., 2014) and exposure to TBHg among Asian subgroups (Awata et al., 2017)

MeHg concentrations within and among fish species are known to vary by more than 10-fold, therefore, understanding patterns of MeHg exposure is challenging (Mahaffey et al., 2004). Numerous studies have investigated the correlations of dietary Hg intake and blood Hg concentrations among NHANES participants (e.g. Awata et al., 2017; Davis et al., 2014; Mahaffey et al., 2004; Xue et al., 2012). Awata et al. identified associations of TBHg and dietary intake of MeHg from fish in Asian subpopulations using the NHANES 24-h dietary recall data and fish MeHg concentrations from the U.S. Federal Drug Administration Total Dietary Study data (Awata et al., 2017). NHANES also provides detailed data on consumption frequencies of 31 seafood species in the previous 30 days, which may reflect habitual fish consumption more accurately than the 24-h dietary recall data (US NHANES). However, recent NHANES datasets have not previously been used to characterize the association between MeHg daily intake from seafood based on past 30 day fish consumption survey and blood MeHg levels among the Asian and non-Asian US population.

In this paper we extended Buchanan, Anglen and Turyk's (Buchanan et al., 2015a) analyses of mercury exposures in vulnerable population subgroups, WORA and adults age ≥ 50 years old, by 1) adding NHANES 2013–2014 dataset to the original NHANES 2011–2012 data; 2) calculating the average daily MeHg intake (ADMI) from different seafood (fish and shellfish) species using the 30 day seafood consumption data; and 3) assessing the relationship between seafood meals and ADMI with blood MeHg and TBHg biomarker concentrations, stratified by subgroup and Asian and non-Asian race/ethnicity.

2. Materials and methods

2.1. Study population

NHANES is a cross-sectional survey designed to provide a representative sample of the US non-institutionalized civilian population (US NHANES). For this study, the two recent NHANES cycles that assessed BMeHg, TBHg, and seafood consumption information were combined: 2011/2012 and 2013/2014. BMeHg and TBHg were measured in eligible participants aged 1 year and older at the physical examination. Seafood consumption frequency was collected from 30-day dietary recalls for participants 18 years of age and older (NCHS, 2011) (NCHS, 2013). For this paper we included participants ≥ 18 years older, which excluded 7954 participants aged younger than 18 years old. Our analysis was restricted to participants in whom both BMeHg and TBHg were measured, which resulted in the additional exclusion of 3839 participants. Participants outside of the age/gender categories selected for subgroup analysis and with missing covariates were further excluded. The final study population in this analysis consisted of 1284 WORA (females 18–44 years of age), among whom 183 participants self-reported their race/ethnicity as "Non-Hispanic Asian", and 2642 adults ≥ 50 years of age, among whom 249 participants self identified their race/ethnicity as "Non-Hispanic Asian". These two vulnerable groups (WORA and adults ≥ 50 years of age) were chosen are because of their potential risk for behavioral and cognitive impairment in offspring and cardiovascular disease, respectively. NHANES was approved by the NCHS Institutional Review Board, and all participants provided written informed consent.

Seafood consumption was collected using self-reported dietary recall interview over the past 30 days in-person in the Mobile Examination Center (MEC). The survey included the number of fish and shellfish meals consumed, broken out by specific type of shellfish (clams, crabs, crayfish, lobsters, mussels, oysters, scallops, shrimp, other known and unknown shellfish) and fish (breaded fish, tuna, bass, catfish, cod, flatfish, haddock, mackerel, perch, pike, pollock, porgy, salmon, sardines, sea bass, shark, swordfish, trout, walleye, and other known and unknown fish).

2.2. Hg intake, Hg exposure and covariates

Average daily MeHg intake (ADMI) for each fish species was estimated using the following equation developed by EPA risk assessment module (EPA, 2011b)

$$ADMI = (CF*MS*THgC*CAF)/(BW*30\ \text{days})$$

where ADMI = average daily MeHg intake (μg/kg-day); CF = consumption frequency (number of seafood meals), which was derived from self-reported survey over the past 30 days; MS = meal size (g/meal) was estimated from EPA Exposure handbook for different gender and age groups (EPA, 2011a), (Table 10–121,10–122. WORA: 109 g/meal for seafood except tuna, 60 g/meal for tuna; Women age ≥ 50 years: 108 g/meal for seafood except tuna, 64 g/meal for tuna; Men age ≥ 50 years: 120 g/meal for seafood except tuna, 68 g/meal for tuna); THgC = total mercury concentration from seafood (μg/g), was estimated from the database published in Birch et al. (2014); CAF = Cooking Adjustment Factor of 1.5, adjusting for the MeHg increase due to water and fat loss during cooking and fish skin removal (Morgan et al., 1997); BW = body weight (kg), which was obtained at the MEC by trained health technicians.

The limit of detection (LOD) for TBHg concentration and MeHg concentration was 0.16 μg/L and 0.12 μg/L respectively. Measurements below the LOD were imputed as the LOD divided by the square root of two (NCHS). Overall, TBHg and MeHg measurements were below the LOD in 8.8% and 15.2% of WORA (2.8% and 9.2% among Asian WORA), and 5.2% and 9.8% of adults ≥ 50 years of age (2.8% and 4.6% among Asian adults ≥ 50 years of age).

Demographic characteristics included in this analysis were sex, age (continuous variable), annual family income (< $20,000, $20,000-$34,999, $35,000 -$54,999, $55,000–99,000, ≥ $100,000; missing for 133 people), country of birth and years living in the U.S. (born in the U.S., born in other country and living in the U.S. for less than 10 years, born in other country and living in the U.S. for 10–20 years, born in other country and living in the U.S. for 20–30 years, born in other country and living in the U.S. for more than 30 years; missing for 2 people); season when the survey was conducted (November 1 through April 30 and May 1 through October 31), language spoken at home for Asians (Only English or more English than non-English; both English and non-English equally; more English than non-English; missing for 2 people).

2.3. Statistical analysis

Due to non-normality of the data, ADMI, BMeHg and TBHg concentrations were natural logarithm transformed to improve the fit of the model. Correlation between ADMI and BMeHg and TBHg were presented as R-squared. Comparisons of geometric means of BMeHg levels and ADMI between subgroups were evaluated using *t*-tests. We applied multivariable linear regression of the natural log transformed BMeHg and TBHg concentrations on the log of the mean ADMI or the number of seafood meals, adjusting for covariates, and present the adjusted R squared, which can be interpreted as the percentage of the variation of BMeHg and TBHg explained by the ADMI together with other covariates in the model. We estimated the proportional change in BMeHg for a doubling of the ADMI as $(e^{(\ln 2*\beta)}-1)*100\%$, with 95% confidence intervals (CI) estimated as $(e^{[\ln 2\ *(\beta \pm 1.96*SE)]}-1)*100\%$, where β and SE are the estimated regression coefficient and standard error. For seafood meals, we estimated the percent change in BMeHg for an increase in one meal, as $(e^{\beta}-1)*100\%$, and 95% confidence intervals (CI) estimated as $(e^{(\beta \pm 1.96*SE)}-1)*100\%$. We imputed ADMI for those who did not consume seafood in the past 30 days by dividing the lowest ADMI by 2, and did sensitivity analysis of the total population. Due to the uncertainty in the role of Se on Hg toxicokinetics, we also did a sensitivity analysis with additional adjustment for measured serum selenium for all subgroups.

NHANES uses a complex survey design that represents the U.S. non-institutionalized civilian population. Each sampled person is assigned a numerical sample weight that quantifies the number of people in the population represented by that specific person, so that estimates computed from that sample are unbiased and nationally representative (US NHANES). We applied survey design variables and sampling weights for MEC exam to adjust for differential selection probabilities in NHANES two cycles of data for descriptive statistics and estimates from regression analyses. All statistical analyses were conducted with SAS (version 9.3; SAS Institute Inc., Cary, NC).

3. Results

The final analysis included a total of 3926 participants, among whom 1284 were WORA, and 2642 were participants ≥ 50 years old. The geometric means of BMeHg and TBHg were greater among the Asian than the Non-Asian population for both subgroups. The geometric means of BMeHg were 1.69 μg/L for Asian WORA and 0.58 μg/L for WORA of other race/ethnicity (P for *t*-test < 0.0001). Asian adults age ≥ 50 years old had a geometric mean of BMeHg of 3.29 μg/L and non-Asian adults age ≥ 50 years old had geometric mean of BMeHg of 0.80 μg/L (P for *t*-test < 0.0001).

A significantly greater percentage of the Asian WORA consumed seafood than participants of other race/ethnicities. Among WORA, 83.1% of Asian and 74.0% of Non-Asians reported consuming seafood in the past 30 days (P for Chi-square test = 0.0043). Among adults ≥ 50 years of age, 87.3% of Asian and 85.3% of Non-Asian reported consuming seafood in the past 30 days (P Chi-square test = 0.39) (See Tables S1a, S1b).

In participants who consumed seafood in the past 30 days, the geometric mean of ADMI was estimated to be 0.03 μg/kg-day for the Asian WORA population and 0.02 μg/kg-day for Non-Asian WORA population (P for *t*-test < 0.0001), and 0.04 μg/kg-day for the Asian adults ≥ 50 years of age and 0.02 μg/kg-day for Non-Asian adults ≥ 50 years of age (P for *t*-test < 0.0001). Average ADMI estimations were below the EPA reference dose of 0.1 μg/kg-day for all subgroups (IRIS, 1997). The estimated ADMI for commonly consumed seafood species were tabulated in Tables 1a, 1b. The top three most frequently consumed seafood were shrimp (26.9%), salmon (13.9%) and tuna (12.1%) among Asian WORA and shrimp (23.7%), tuna (20.3%) and salmon (12.8%) among Non-Asian WORA (Table 1a). For the Asian WORA population, fish species with the highest contribution to the ADMI were “other known fish species” (17.3%), tuna (15.1%) and mackerel (13%), whereas for Non-Asian WORA, tuna (38.3%), “other known fish species” (10.6%) and salmon (7.6%) contributed the most to the ADMI (Table 1a). For adults ≥ 50 years of age, the most frequently consumed seafood are shrimp (22.3%), salmon (14.9%) and “other known fish species” (12.2%) by Asians, and tuna (19.7%), shrimp (18.7%) and salmon (14.8%) by Non-Asians (Table 1b); however the percent contribution to the ADMI was greatest for mackerel (35.4%), “other known fish species” (11.3%), and “other unknown fish species” (8.1%) for Asians, and tuna (19.4%), swordfish (15.8%) and “other known fish species” (11.0%) for Non-Asians (Table 1b). For the Asian population, even though the total meals of mackerel only comprised a small percentage of total seafood meals (1.4% for WORA and 5.6% for adults age ≥ 50 years old), the estimated MeHg intake from mackerel had a large impact on total MeHg intake (13% for WORA and 35.4% for adults age ≥ 50 years old) (Tables 1a, 1b). Tuna, which also had a sizable impact on the ADMI, was not delineated into type. Concentrations can range from the highest category of MeHg concentration (e.g. big eye, ahi) to the low moderate range (e.g. chunk light).

Table 2a and Fig. 1(a)–(d) present the results of linear regression analyses predicting blood MeHg levels as a function of log transformed ADMI from seafood in participants consuming seafood in the past 30 days. For WORA, ADMI from seafood explained 38% and 22% of the variation in BMeHg concentration among Asians and among Non-Asians, respectively. A doubling in ADMI resulted in an increase of 45.3% (95%CI: 31.4%, 60.6%) and 31.7% (95%CI: 25.4%, 38.3%) in BMeHg among Asians and among Non-Asians, respectively. Adjustment for age, family income, season of examination, country of birth and years in the U.S moderately increased the correlation to 47% and 27%,

Table 1a
ADMI from seafood and the percentage of total Hg intake and total seafood meals among WORA who consumed seafood in the past 30 days.

	Hg concentration in fish	Asian (n = 183)		Non-Asian (N = 1101)	
Seafood species	**μg Hg/g wet weight**	**% of total seafood ADMI**	**% of total seafood meals**	**% of total seafood ADMI**	**% of total seafood meals**
Clams	0.03	1.4	3.8	0.8	2.3
Crabs	0.06	3.7	4.8	4.7	6
Lobster	0.19	6.6	2.4	4.1	1.6
Shrimp	0.01	5.2	26.9	4.6	23.7
Other shellfish type known	0.03	1.3	2.9	0.9	2
Shellfish with % ADMI ≤ 0.9[a]	0.03	2.2	6.7	2	6
Shellfish Total		**20.4**	**47.6**	**17.1**	**41.6**
Bass	0.26	2.1	0.5	1.2	0.3
Catfish	0.11	4	2.9	4.9	3.5
Cod	0.09	1.8	1.4	3.4	2.7
Flatfish	0.05	0.9	1.2	1.2	1.5
Mackerel	0.64	13	1.4	1.2	0.1
Porgy	0.32	2.5	0.5	0.7	0.2
Salmon	0.04	8.1	13.9	7.6	12.8
Seabass	0.19	2.6	0.9	1	0.4
Swordfish	1.27	1.9	0.1	5.7	0.3
Tuna	0.24	15.1	8.5	38.3	20.3
Other fish type known	0.10	17.3	12.1	10.6	7.9
Other fish type unknown	0.14	7.2	3.8	3.9	1.9
Fish with % ADMI ≤ 0.9[b]	0.18	3.1	5.1	3.3	6.6
Fish total		**79.6**	**52.4**	**82.9**	**58.4**

Note: ADMI, average daily MeHg intake (μg/kg-day); WORA, women of reproductive age; Non-Asians include Non-Hispanic White, Non-Hispanic Black, Mexican American, other Hispanic and other race (including Multi- Racial); Hg concentrations in fish is adapted from Birch et al. (2014); N/A: not available.

[a] "Shellfish with % ADMI ≤ 0.9″ includes crayfish, mussels, oysters, scallop and "other shellfish type unknown".

[b] "Fish with % ADMI ≤ 0.9″ includes breaded fish, haddock, perch, pike, sardines, shark, trout and walleye.

but did not substantially change the percent change in BMeHg per doubling of the ADMI. In adjusted models, ADMI from fish with Hg level ≥ 0.2 μg/g (including bass, mackerel, pike, porgy, shark, swordfish, tuna and walleye), which is the critical value for human consumption used by the US EPA (Rothschild and Duffy, 2002) explained 43% of the variation in BMeHg for Asian WORA and 28% for Non-Asian WORA, slightly less than the proportions explained by total seafood intake (47% and 27%, respectively).

For Asian adults ≥ 50 years of age, the correlations of ADMI from seafood and BMeHg were not as strong as the correlations for Asian WORA (Table 2a). ADMI from seafood explained 28% and 35% of the variation of blood MeHg concentration among Asian and among Non-Asian adults age ≥ 50 years old respectively, after adjusting for age, sex, family income, season of examination, country of birth and years in

Table 1b
ADMI from seafood and the percentage of total Hg intake and total seafood meals among adults ≥ 50 years old who consumed seafood in the past 30 days.

	μg Hg concentration in fish	Asian (N = 249)		Non-Asian (N = 2393)	
Seafood species	**Hg/g wet weight**	**% of total seafood ADMI**	**% of total seafood meals**	**% of total seafood ADMI**	**% of total seafood meals**
Crabs	0.06	1.8	3.3	2.6	3.3
Lobster	0.19	2.2	1.2	3	1.1
Shrimp	0.01	3.1	22.3	3.7	18.7
Shellfish with % ADMI ≤ 0.9[a]	0.03	2.5	9.7	3.2	9.1
Shellfish Total	N/A	**9.7**	**36.5**	**12.5**	**32.2**
Bass	0.26	3.9	1.4	2	0.5
Catfish	0.11	3	2.8	5.4	3.8
Cod	0.09	2.2	2.4	4.8	3.9
Flatfish	0.05	1.6	2.8	1.8	2.2
Haddock	0.07	0.2	0.4	2.6	2.6
Mackerel	0.64	35.4	5.6	3.5	0.2
Perch	0.14	1.8	1.2	1.4	0.7
Pollock	0.01	0.4	2.9	0.1	0.6
Porgy	0.32	1.5	0.5	0.8	0.2
Salmon	0.04	6	14.9	8.8	14.8
Seabass	0.19	2.1	1.1	1.5	0.5
Swordfish	1.27	3.9	0.3	15.8	0.9
Tuna	0.24	6.2	4.7	19.4	19.7
Walleye	0.24	1.4	0.7	1.7	0.5
Other fish type known	0.10	11.3	12.2	11	7.8
Other fish type unknown	0.14	8.1	5.2	4.5	2.1
Fish with % ADMI ≤ 0.9[b]	0.20	0.3	4.5	2.6	6.8
Fish total	**N/A**	**90.3**	**63.5**	**87.5**	**67.8**

Note: ADMI, average daily MeHg intake (μg/kg-day); Non-Asians include Non-Hispanic White, Non-Hispanic Black, Mexican American, other Hispanic and other race (including Multi-Racial); Hg concentrations in fish is adapted from Birch et al. (2014); N/A: not available.

[a] "Shellfish with % ADMI < 0.9″ includes clams, crayfish, mussels, oysters, scallop, "other shellfish type known".

[b] "Fish with % ADMI < 0.9″ includes breaded fish, pike, sardines, shark and trout.

Table 2a
Correlation (R^2) of ADMI from seafood and BMeHg level, and percent change (95% CI) in BMeHg per doubling of ADMI among participants who consumed seafood in the past 30 days.

Seafood Type	Women of Reproductive Age (N = 1284)				Adults Age ≥ 50 years old (N = 2642)			
	Asian (N = 183)		Non-Asian (N = 1101)		Asian (N = 249)		Non-Asian (N = 2393)	
Crude Model	R^2 or adjusted R^2	Percent change (95% CI) in BMeHg by ADMI	R^2 or adjusted R^2	Percent change (95% CI) in BMeHg by ADMI	R^2 or adjusted R^2	Percent change (95% CI) in BMeHg by ADMI	R^2 or adjusted R^2	Percent change (95% CI) in BMeHg by ADMI
Per doubling of shellfish ADMI	**0.24**	37.1 (24.4, 51.0)	**0.09**	22.5 (14.0, 31.6)	**0.07**	17.2 (9.3, 25.7)	**0.10**	27.3 (20.3, 34.7)
Per doubling of fish ADMI	**0.26**	36.2 (23.8, 49.9)	**0.19**	34.5 (25.9, 43.8)	**0.17**	23.3 (13.4, 34.1)	**0.23**	41.9 (36.3, 47.8)
Per doubling of seafood ADMI[a]	**0.38**	45.3 (31.4, 60.6)	**0.22**	31.7 (25.4, 38.3)	**0.19**	24.8 (16.7, 33.4)	**0.28**	42.7 (37.2, 48.5)
Per doubling of fish Hg level ≥ 0.2 μg/g[b] **ADMI**	**0.08**	19.6 (5.3, 35.9)	**0.18**	47.6 (33.1, 63.6)	**0.10**	18.0 (8.6, 28.3)	**0.14**	42.7 (32.8, 53.2)
Adjusted model[c]								
Per doubling of shellfish ADMI	**0.31**	50.9 (45.5, 57.0)	**0.16**	44.1 (41.1, 47.4)	**0.11**	16.8 (8.9, 11.7)	**0.19**	20.9 (14.1, 28.1)
Per doubling of fish ADMI	**0.33**	49.6 (44.7, 55.2)	**0.24**	48.7 (45.4, 52.1)	**0.19**	22.3 (12.5, 33.1)	**0.31**	39.1 (33.9, 44.5)
Per doubling of seafood ADMI[a]	**0.44**	44.9 (30.0, 61.6)	**0.26**	29.1 (22.8, 35.8)	**0.22**	23.8 (15.5, 32.8)	**0.34**	39.5 (34.7, 44.4)
Per doubling of fish Hg level ≥ 0.2 μg/g[b] **ADMI**	**0.42**	41.2 (37.8, 45.0)	**0.27**	52.4 (47.7, 57.6)	**0.24**	18.1 (6.3, 31.2)	**0.27**	38.8 (30.2, 47.9)
Adjusted model[d]								
Per doubling of shellfish ADMI	**0.34**	53.6 (48.0, 59.8)	**0.16**	44.1 (41.2, 47.2)	**0.17**	14.7 (7.1, 22.9)	**0.21**	21.1 (14.1, 28.6)
Per doubling of fish ADMI	**0.35**	50.2 (45.7, 55.0)	**0.25**	48.5 (45.1, 52.1)	**0.27**	23.2 (13.8, 33.5)	**0.32**	38.7 (33.7, 44.1)
Per doubling of seafood ADMI[a]	**0.47**	54.6 (49.9, 59.8)	**0.27**	47.4 (45.0, 49.9)	**0.28**	23.9 (15.7, 32.7)	**0.35**	39.4 (34.6, 44.4)
Per doubling of fish Hg level ≥ 0.2 μg/g[b] **ADMI**	**0.43**	40.8 (37.7, 44.2)	**0.28**	51.8 (47.2, 56.9)	**0.33**	20.4 (6.3, 36.2)	**0.28**	38.3 (29.7, 47.6)

Note: ADMI, average daily MeHg intake (μg/kg-day); BMeHg, blood methylmercury (μg/L); Non-Asians include Non-Hispanic White, Non-Hispanic Black, Mexican American, other Hispanic and other race. (including Multi- Racial); The percent change in BMHg level and 95% confidence intervals (95% CI) represent percent change in BMeHg for a doubling in ADMI.

[a] Seafood includes shellfish and fish.

[b] Fish Hg level ≥ 0.2 μg/g include bass, mackerel, pike, porgy, shark, swordfish, tuna and walleye.

[c] Model adjusted for age, family income, and season in which the medical examination was conducted; additionally adjusted for sex for adults ≥ 50 years old.

[d] Model additionally adjusted for country of birth and years living in the U.S.

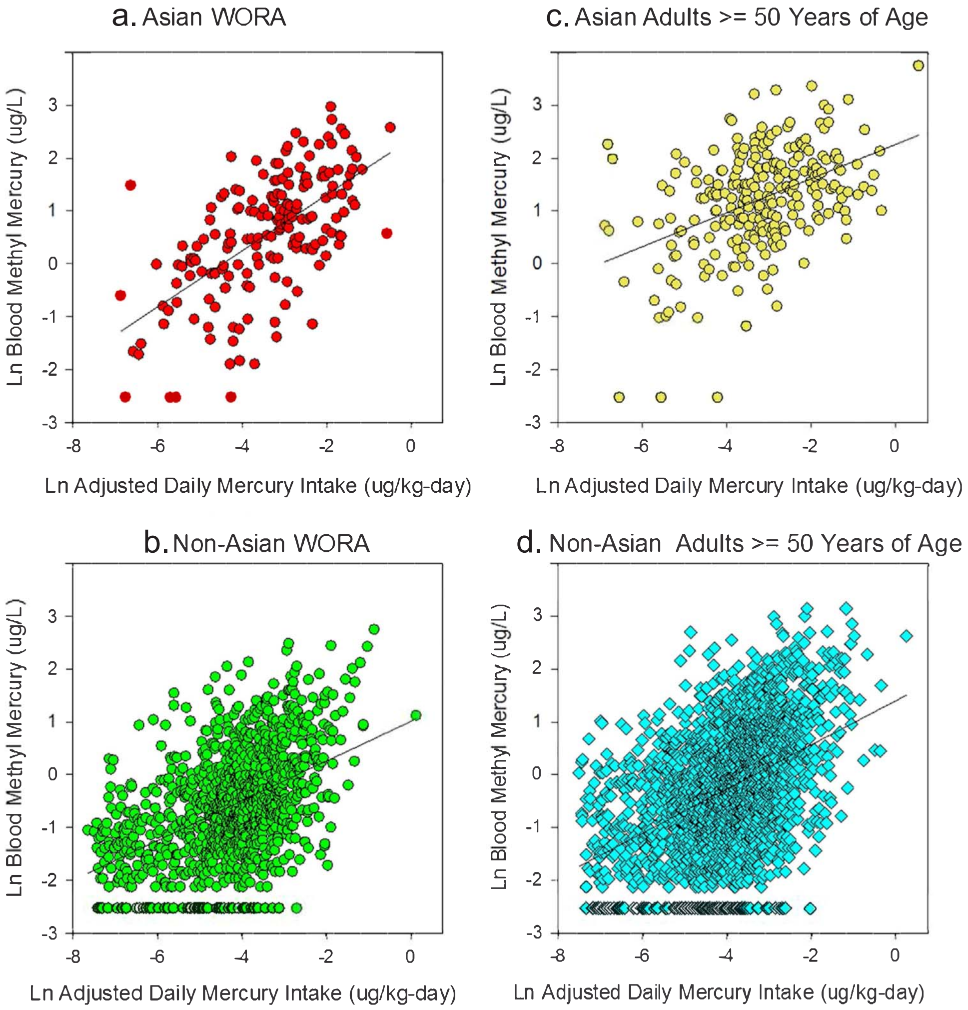

Fig. 1. (a)–(d) Log BMeHg level and log ADMI from seafood among four subgroups. Note: BMeHg: blood methylmercury (μg/L); TBHg: total blood mercury μg/L; ADMI, average daily MeHg intake (μg/kg-day); WORA: women of reproductive age.

Table 2b

Correlation (R^2) of number of seafood meals and BMeHg levels and percent change (95% CI) in BMeHg by seafood meals among participants who consumed seafood in the past-30 days.

Seafood species	Women of Reproductive Age (N = 1284)				Adults Age ≥ 50 years old (N = 2642)			
	Asian (N = 183)		Non-Asian (N = 1101)		Asian (N = 249)		Non-Asian (N = 2393)	
Crude Model	R^2 or adjusted R^2	Percent change in BMHg	R^2 or adjusted R^2	Percent change in BMHg	R^2 or adjusted R^2	Percent change in BMHg	R^2 or adjusted R^2	Percent change in BMHg
Sum of shellfish meals	**0.17**	11.2 (6.1, 16.3)	**0.08**	9.4 (4.1, 15.7)	**0.11**	11.4 (6.1, 16.6)	**0.12**	16.3 (12.3, 19.1)
Sum of fish meals	**0.26**	15.6 (9.3, 21.4)	**0.27**	18.3 (15.7, 21.4)	**0.14**	7.6 (3.7, 11.2)	**0.23**	14.6 (12.5, 16.3)
Sum of seafood[a] meals	**0.30**	9.2 (6.4, 12.7)	**0.23**	10.2 (7.7, 13.4)	**0.18**	6.6 (3.4, 8.1)	**0.25**	11.5 (9.5, 12.1)
Fish Hg level ≥ 0.2 μg/g[b] meals	**0.13**	33.2 (23.7, 44.1)	**0.16**	25.1 (17.3, 34.2)	**0.08**	14.5 (10.3, 18.7)	**0.13**	22.1 (18.2, 26.3)
Adjusted model[c]								
Sum of shellfish meals	**0.19**	11.4(6.3, 16.2)	**0.12**	9.4 (3.5, 14.7)	**0.13**	10.8 (5.4, 15.2)	**0.19**	13.6 (10.3, 16.6)
Sum of fish meals	**0.28**	15.1 (9.4, 22.3)	**0.30**	17.2 (14.7, 20.4)	**0.17**	6.8 (3.1, 10.7)	**0.29**	13.3 (11.2, 15.3)
Sum of seafood[a] meals	**0.32**	9.6 (6.4, 12.9)	**0.26**	9.6 (6.2, 12.7)	**0.20**	5.8 (3.6, 8.1)	**0.30**	9.2 (8.6, 11.4)
Fish Hg level ≥ 0.2 μg/g[b] meals	**0.16**	35.6 (24.4, 47.2)	**0.20**	23.6 (16.4, 32.1)	**0.10**	12.7 (8.1, 16.2)	**0.22**	19.4 (16.6, 23.9)
Adjusted model[d]								
Sum of shellfish meals	**0.22**	11.1 (7.5, 16.3)	**0.13**	9.5 (3.7, 13.1)	**0.17**	9.4 (5.6, 14.2)	**0.20**	13.6 (10.4, 16.2)
Sum of fish meals	**0.31**	16.4 (10.2, 22.7)	**0.31**	17.2 (14.8, 20.3)	**0.21**	6.5 (3.3, 10.5)	**0.30**	13.2 (11.5, 15.2)
Sum of seafood[a] meals	**0.36**	9.1 (6.2, 12.4)	**0.25**	9.5 (6.6, 12.5)	**0.24**	5.9 (3.2, 8.1)	**0.31**	9.3 (8.7, 11.3)
Fish Hg level ≥ 0.2 μg/g[b] meals	**0.17**	33.5 (22.6, 46.8)	**0.21**	23.4 (16.6, 32.1)	**0.26**	11.8 (7.6, 16.2)	**0.28**	20.6 (17.4, 23.5)

Note: BMeHg, blood methylmercury (μg/L); Non-Asians include Non-Hispanic White, Non-Hispanic Black, Mexican American, other Hispanic and other race (including Multi- Racial); The percent change in BMHg level and 95% confidence intervals (95%CI) represent the percentage change of the BMeHg with a one unit increase in seafood meals.

[a] Seafood includes shellfish and fish.

[b] Fish Hg level ≥ 0.2 μg/g include bass, mackerel, pike, porgy, shark, swordfish, tuna and walleye.

[c] Model adjusted for age, family income, and season in which the medical examination was conducted; additionally adjusted for sex for adults ≥ 50 years old.

[d] Model additionally adjusted for country of birth and years living in the U.S.

the U.S. ADMI from fish with Hg level $\geq$ 0.2 µg/g, together with other covariates, explained 33% and 28% variation in BMeHg for Asians and Non-Asians, respectively, compared with 28% and 35%, respectively for the proportions explained by total seafood intake (Table 2a). The associations of ADMI and TBHg concentrations were only slightly weaker than the associations of ADMI and BMeHg (See Table S2).

Table 2b presents the association of number of seafood meals and log transformed BMeHg among WORA and adults ≥ 50 years old. With a few exceptions, correlations between BMeHg and seafood meals were slightly weaker than the correlation between BMeHg and ADMI in adjusted models.

Since there are different species of mackerel, and the ADMI from mackerel contribute a substantial percentage of the total Hg intake from seafood for both Asian subgroups (13% for Asian WORA and 35% for Asian age ≥ 50 years old), we conducted sensitivity analyses applying Hg levels from different major types of mackerels to our ADMI calculation. We used Hg levels from mackerel based on three different scenarios: consumption of Atka Mackerel and no other mackerel species (Hg level 0.04 µg/g (Tsuchiya et al., 2008a, 2008b)), consumption of Spanish mackerel and no other mackerel species (Hg level 0.182 µg/g, (FDA, 2014)), and the consumption of three mackerel species - jack, chub and Atlantic- (mean Hg level 0.081 µg/g, (US EPA, 1997)). Overall, changes in the correlation of ADMI from the three mackerel consumption scenarios and BMeHg are minimal (See Table S4).

Supplemental Table 6 presents a sensitivity analysis for correlation of ADMI and BMeHg level among the total population, using imputed ADMI for those who did not consume seafood in the past 30 days. The results showed stronger correlations of ADMI and BMeHg levels than the correlations among subgroup that included only those who consumed seafood in the past 30 days. For example, ADMI explained 56% of the variance in blood MeHg level for seafood consumption among total Asian WORA, including those who did not consume seafood in the past 30 days (See Table S6).

4. Discussion

As in our previous analysis of 2011–2012 NHANES MeHg levels and fish consumption data (Buchanan et al., 2015a), our analysis of the expanded NHANES dataset including 2013–2014 found both Asian WORA and Asian adults age ≥ 50 years of age had significantly higher BMeHg levels and ADMI than their Non-Asian counterparts. Correlations between ADMI from 30-day recall of seafood consumption and blood Hg level were stronger among Asian WORA than among Non-Asian WORA, but weaker among Asian adults age ≥ 50 years old than among Non-Asian adults age ≥ 50 years old. Key fish species that contributed to dietary MeHg intake for Asians in the U.S. were mackerel, tuna, “other known fish species” and “other unknown fish species”. In contrast, for Non-Asians, key fish species that contributed to dietary MeHg intake were tuna, “other known fish species”, swordfish and salmon.

The geometric mean of ADMI for the Asian WORA population and for Asian adults age ≥ 50 years old in the US was lower in our analysis compared with other studies of Asian populations in the US, especially studies of Japanese, Korean, Vietnamese and Chinese populations, who also had higher geometric mean TBHg than did our study participants (Tsuchiya et al., 2008a, 2008b; Xu and Newman, 2015; Table 3). Given that the Asian population defined in our analysis is “Non-Hispanic Asian” which includes individuals with self-reported origins in the Far East Asia, Southeast Asia, or South Asia (the Indian subcontinent) that have various seafood consumption behaviors (NCHS, 2013), the lower ADMI levels are expected when compared with Asians who self-identify as Japanese, Korean and Chinese, who have higher seafood consumption rates than South Asians (Tsuchiya et al., 2008a, 2008b; Zhang et al., 2009). Furthermore, there may be some selection bias in these volunteer-based studies since participants who consume more fish may be more concerned about mercury exposure. Nevertheless, our results are comparable to Xue et al.'s study using the 1999–2006 NHANES and 1990–2002 FDA's Total Diet Study data (Xue et al., 2012). Awata et al. used the NHANES 2011–2012 dataset and estimated a higher ADMI for MeHg than our study, based on 24-h recall dietary data and Hg concentrations in fish from the FDA Total Diet Study. However, 24-h recall dietary data may represent only a snapshot of study participants’ dietary consumption rather than long term food consumption patterns (Awata et al., 2017). In addition, estimates of Hg concentrations in fish used in Awata et al.’s analysis were generally lower than the Hg concentrations used in our study, which was developed by US EPA by modeling data from the EPA, FDA, and peer-reviewed literature (Birch et al., 2014). These different approaches used to calculate ADMI (24-hr recall vs. 30-day consumption; different fish Hg level databases) may partially explain the different results between the two studies.

Our analysis found that for the Asian WORA population, the top three fish species contributing to MeHg intake are “other known fish species”, tuna and mackerel. For Asian adults age ≥ 50 years old, mackerel, “other known fish species”, and “other unknown fish species” made the largest contribution to the MeHg intake. Even though shark and swordfish had the highest MeHg concentrations, their contributions to MeHg exposure were small because intake of those fish types was low among Asians. Our findings are consistent with Xue et al.’s findings that tuna, other saltwater fish, and other freshwater fish are the main drivers of seafood MeHg exposure in the Asian/Pacific Islander/Native American/Multiracial NHANES 1999–2006 population (Xue et al., 2012). A study of Chinese and Vietnamese church communities in coastal Virginia found that the Chinese church community received most of their mercury exposure from commercial market fish (e.g., salmon), and the Vietnamese community was exposed from both market and local fish (e.g., striped bass and croaker) (Xu and Newman, 2015). In Seattle, Tsuchiya et al. found that the top six species representing over 50% of the total daily Hg intake for Japanese women were salmon, canned white tuna, halibut, ahi, canned light tuna, and black cod. For Korean women, they were canned white tuna, flounder/sole, squid, canned light tuna, salmon, and yellow croaker (Tsuchiya et al., 2008a, 2008b). Of note, with exception of salmon and canned tuna, all of these seafood species are categorized as “other known fish species” or “other unknown fish species” by NHANES. These findings suggest that in order to capture key sources of Hg among Asian fish consumers, NHANES should expand the species choices to include species preferred by Asian populations at risk.

We found weak to moderate correlations of ADMI with blood Hg, similar to other investigations (Table 3). The MeHg levels in fish species differ by body of water and other characteristics (Bonito et al., 2016; Burger et al., 2011). Nevertheless, numerous public databases assign one concentration to each fish species, assuming that intraspecies variability is minimal. MeHg accumulates in the aquatic food chain and is, therefore, more likely to be found in carnivorous fishes, and the amount increases with age and size of the fish (Bonito et al., 2016). However, our analysis found that some of the key fish species that influence the dietary MeHg intake, such as “other known fish species”, which accounted for 17.3% and 11.3% of total ADMI among Asian WORA and Asian adults age ≥ 50 years old, were not necessarily the carnivorous fish, suggesting that frequency of consumption and perhaps the meal portion size also play an important role in MeHg intake. Moreover, since the range of MeHg concentration can vary widely by type of tuna, fish consumption surveys should include questions about consumption of specific types of tuna (e.g. chunk light, albacore, sushi, steak, etc) so that consumption advice can target the type that is contributing the most to ADMI.

The larger contribution of ADMI to variation in mercury levels in younger than older Asian adults is intriguing. We found that ADMI explained 38% and 19% of the variance in blood MeHg level for Asian WORA and Asian adults age ≥ 50 years old, respectively. The variation in blood MeHg level is influenced by other potential sources of Hg, variability in individual metabolism and Hg toxicokinetics (Andreoli

Table 3
Comparison of ADMI and blood Hg levels across studies of U.S. populations.

Study	Population	Geometric mean of estimated mercury intake from seafood (μg/kg-day)	Geometric mean of measured BMeHg or TBHg level (μg/L)	Crude R^2 or Adjusted R^2
Current study[a]	Asian in NHANES 2011–2014, WORA	0.03	1.69	0.38
	Asian in NHANES 2011–2014, males and females aged ≥50	0.04	3.29	0.19
	Non-Asian in NHANES 2011–2014, WORA	0.02	0.58	0.22
	Non-Asian in NHANES 2011–2014, males and females aged ≥50	0.02	0.80	0.28
Current study[b]	Asian in NHANES 2011–2014, WORA	0.03	1.16	0.30
	Asian in NHANES 2011–2014, males and females aged ≥ 50	0.04	2.42	0.17
	Non-Asian in NHANES 2011–2014, WORA	0.02	0.41	0.10
	Non-Asian in NHANES 2011–2014, males and females aged ≥ 50	0.02	0.66	0.21
Mahaffey et al., 2004[c]	All race/ethnicities, NHANES 1999–2000, women 16–49 years of age	0.02	0.80	0.23
Mahaffey et al., 2004	Non-Hispanic white, NHANES 1999–2000, women 16–49 years of age	0.02	0.75	N/A
	Non-Hispanic black, NHANES 1999–2000, women 16–49 years of age	0.05	1.01	N/A
	Mexican American, NHANES 1999–2000, women 16–49 years of age	0.02	0.57	N/A
	Other Hispanic, NHANES 1999–2000, women 16–49 years of age	0.03	0.97	N/A
	A/P/N/M, NHANES 1999–2000, women 16–49 years of age	0.05	1.06	N/A
Xue et al., 2012	A/P/N/M NHANES 1999–2006, males and females aged 21–50	0.04	1.69	N/A
Xue et al., 2012	A/P/N/M NHANES 1999–2006, males and females aged ≥50	0.04	1.70	N/A
	Rest NHANES 1999–2006, males and females aged 21–50	0.02	1.04	N/A
	Rest NHANES 1999–2006, males and females aged ≥50	0.02	1.18	N/A
Awata et al., 2017[d]	Non-Hispanic Asian, NHANES 2011–2012	0.09	1.93	0.31
Awata et al., 2017	Non-Hispanic White, NHANES 2011–2012	0.06	0.71	0.33
	Non-Hispanic Black, NHANES 2011–2012	0.05	0.71	0.26
	Chinese, NHANES 2011–2012	N/A	2.58	0.45
	Asian Indian, NHANES 2011–2012	N/A	0.79	0.32
	Other Asian, NHANES 2011–2012	N/A	2.48	0.28
Tsuchiya et al. (2008a, 2008b)	Japanese WORA, WA, US	0.09	4.92[e]	0.28
Tsuchiya et al., 2008a, 2008b	Korean WORA, WA, US	0.05	2.44[e]	0.07
	Japanese and Korean WORA combined	0.06	3.44[e]	N/A
Xu and Newman, 2015	Chinese church community, VA, US	0.09	2.08[e]	N/A
Xu and Newman, 2015	US Vietnamese church community, VA, US	0.12	5.84[e]	N/A

Note: ADMI, average daily MeHg intake (μg/kg-day); BMeHg, blood methylmercury (μg/L); TBHg, total blood mercury (μg/L); A/P/N/M, participants self-identified as Asian, Pacific Islander, Native American, or multiracial; Rest NHANES 1999–2006, participants self-identified as black, non-Hispanic white, Mexican American, and Hispanic; N/A: not available.

[a] Among participants who consumed seafood in the past 30 days.

[b] Among all participants with imputation of ADMI for those who did not consume seafood in the past 30 days.

[c] Studies use both 24-h dietary recall survey and 30-day survey for fish consumption information.

[d] Studies use 24-h dietary recall survey for fish consumption information. Model was adjusted for sex, age, education, income, birth place, urbanization, and census region.

[e] Conversion of hair Hg level (ppm) to total blood Hg level (μg/L) are based on the World Health Organization (WHO) recommendation of a mercury hair-to-blood ratio of 250.

and Sprovieri, 2017; Okpala et al., 2017), types of fish consumed, variability of Hg concentration in fish, meal size, cooking methods and other factors. Sources of variation of BMeHg among Asian adults age ≥ 50 years old other than fish intake might explain the apparent moderate association of MeHg intake and blood MeHg level. There is increasing evidence that oral and gut bacteria may methylate dental amalgam, with a recent study using NHANES data showing significant associations of number of amalgams with BMeHg (Yin et al., 2016). Older individuals may be more susceptible because of increasing numbers of amalgams (Richardson et al., 2011). There is also evidence that liver (Lin et al., 2014) and kidney function (Akerstrom et al., 2016) might be associated with Hg metabolism and the blood Hg level. However, a sensitivity analysis with additional adjustment for serum creatinine levels as a marker of kidney function showed a slightly stronger correlation of ADMI from seafood and BMeHg level only among Asian WORA (See Table S5). Further investigation is needed to evaluate why older Asian Americans have higher Hg levels that are less accounted for by frequency of fish consumption and species consumed compared to younger Asians.

We found the correlation between ADMI and blood Hg level was stronger among Asian WORA than among Non-Asian WORA, which agrees with Awata et al.'s finding that the Chinese subgroup had the highest standardized regression coefficient value for the regression model between dietary Hg intake and blood Hg levels in NHANES 2011–2012 (Awata et al., 2017). Our analysis did not examine other dietary sources of Hg intake other than fish and shellfish which could influence Hg levels. Diet can vary by ethnicity and acculturation status ethnicities ((Lee et al., 1999; Sechena et al., 2003;Salant and Lauderdale, 2007). Dietary components, such as fruits or fruit juice may have a protective effect on association between fish consumption and BMeHg (He and Wang, 2011; Passos et al., 2007), while tea may accelerate the enterohepatic MeHg cycle and contribute to a temporary bioamplification of MeHg in the bloodstream (Canuel et al., 2006). But our sensitivity analysis adjusting for carotenoids (includes alpha-carotene, beta-carotene, beta-cryptoxanthin, lycopene and lutein + zeaxanthin) as potential surrogates for fruits and vegetable intake did not modify the correlation between ADMI and BMeHg levels (data not shown). Cooking methods might also affect the bioacessibility of mercury. Steaming, grilling, and frying can reduce MeHg bioaccessibility by 29.4–95.8% for certain fish species (He and Wang, 2011). This mechanism is not well understood and is not accounted for in NHANES nor other fish consumption and BMeHg investigations. Studies also have shown possible Hg contamination of herbal remedies used by Asians, and some of the herbal remedies (eg. Ginseng) may detoxify mercury (Canuel et al., 2006; Kim et al., 1992)

Some fish also contain high levels of selenium (Se), which in some studies has been demonstrated to play an antioxidant role and may have protective effects against Hg toxicity (Burger and Gochfeld, 2013; Ralston et al., 2007; Ralston and Raymond, 2010). However, there is uncertainty in the role of Se on Hg toxicokinetics. Se to Hg molar ratios in excess of 1:1 have been reported to counteract the adverse effects of Hg, protecting against Hg toxicity and/or leading to the removal of both Se and Hg from biological turnover (Kehrig et al., 2013; Ralston and Raymond, 2010; Tan et al., 2009). Hg levels could be lowered either from sequestering Hg-Se complexes in tissues or through inhibition of absorption by the formation of Hg-Se complexes in the stomach or gastrointestinal tract. Therefore, we performed a sensitivity analysis with additional adjustment for measured serum selenium in all subgroups and the results showed a slightly stronger correlation between ADMI and BMeHg together with other covariates only among Asian WORA (adjusted R^2 = 0.52 after vs. adjusted R^2 = 0.47 without adjustment for selenium), suggesting that selenium partially explained the variation of BMeHg in this subgroup and may play a role in absorption or bioavailability of Hg (See Table S5).

Our study results show moderate correlations between ADMI and BMeHg level among Asian populations in the US and slightly weaker associations in general with seafood meals and BMeHg. On one hand, the findings indicate that a seafood frequency survey is a potentially useful tool to provide estimated Hg exposure information, which would reduce the need to collect costly and invasive biometric data. On the other hand, our results suggest that the accuracy of estimates of Hg intake from seafood consumption could be improved by collection of more specific fish species and portion size of each fish meal. Our finding that TBHg explains a similar proportion of ADMI to BMeHg is not surprising given the strong correlation between these measurements ($R^2 = 0.89$), and supports TBHg as a valid screening test for elevated MeHg and for investigations of determinants of MeHg exposure (Buchanan et al., 2015a).

The major strength of our study is that it uses nationally representative data of Asian population from 2011 to 2014 NHANES datasets. Studies of dietary MeHg intake among Asian populations in the United States are limited mainly to certain single ethnicity communities (such as Chinese, Japanese, and Koreans) or certain geographic areas with high Asian populations (e.g., New York City, Washington state, Coastal Virginia, and California). Analyses focusing on the general Asian population in the United States are rare but warranted to understand the MeHg exposure among this population. Another advantage is the relatively large sample size of the Asian population, which allowed adjustment for various socio-demographic and acculturation factors.

Our ADMI calculations may be subject to errors from a number of factors. First, the 30-day dietary survey may reflect habitual fish consumption more accurately than the 24-h dietary recall data, but could be more subject to recall limitations. Second, since NHANES does not collect portion size information for 30-day fish consumption, our portion size estimates were based on established EPA estimates by gender and age categories. Alternative strategies that have been used to estimated portion sizes for the 30-day fish consumption data in NHANES include estimating average fish portion size from the 24-h recall data (Mahaffey et al., 2004). Third, fish consumption behavior in terms of quantity, species and parts of fish consumed (i.e. fillets, fish head, roe, skins, tails, bones, eggs, and/or other organs) can vary across different Asian ethnicities (Buchanan et al., 2015b; Sechena et al., 2003; Tsuchiya et al., 2008a, 2008b; Xu and Newman, 2015) and additional details about subgroups of certain fish species (eg. Mackerel, "other known fish" and "other unknown fish") which accounted for up to 54% of seafood intake could have increased the accuracy of ADMI estimates. Fourth, Hg concentrations in fish were obtained from U.S. EPA estimates that incorporated extensive data representing sampling of over 26,000 fish, gathered during 1999–2010 (Birch et al., 2014). These estimates represent geometric mean fish mercury concentrations during that time period. Estimates for "other known fish" and "other known shellfish" were generated for species not directly assessed in the 30 day recall, which could be influenced by the lack of Hg data for fish species that were consumed by participants but not available in the EPA dataset. Estimates for "other unknown fish" and "other unknown shellfish" were based on average mercury concentration in all fish weighted by 30-day consumption frequency for women aged 16–49 years in the NHANES 1999–2010 data. We conducted a sensitivity analysis substituting Hg estimates for the "other known fish" and "other unknown fish" categories from a previous investigation based on data from 1997 to assess the influence of these estimates on our findings (Mahaffey et al., 2008). Weaker correlations between ADMI and BMeHg together with other covariates were observed in all subgroups, supporting our use of the more contemporary and smaller Hg estimates from Birch et al. (eg. adjusted $R^2 = 0.38$ vs. adjusted R^2 = 0.47 for Asian WORA; adjusted $R^2 = 0.21$ vs. adjusted R^2 = 0.28 for Asian adults age ≥ 50 years old) (See Table S7).

The lack of publicly available information on Asian subgroups from NHANES prevented us from further investigating the patterns of seafood consumption among Asian subgroups, which have various seafood consumption behaviors (NCHS, 2013). Further, regional differences in fish consumption and blood Hg levels have been noted, but geographic location is also not publically available in NHANES (Cusack et al., 2017). Finally, other known and unknown factors that affect the toxicokinetics of mercury, such as foods that may increase or decrease the metabolism and absorption (Liberda et al., 2014; Passos et al., 2007), breastfeeding (Rebelo and Caldas, 2016), and specific genetic characteristics that may affect Hg metabolism (Canuel et al., 2016) were not controlled in our analysis.

In conclusion, our estimates of average daily intake of MeHg ingestion from seafood confirms that the Asian population has higher MeHg intake than the Non-Asian population in the US, and we have identified key fish species that contribute to MeHg intake in this subgroup. Correlations of average daily intake of MeHg with blood MeHg suggest that fish consumption is a key predictor of blood MeHg concentration, especially among Asian women of reproductive age. Future studies of Asian populations in the U.S. should incorporate information on other known and unknown fish species that are frequently consumed by Asian populations, as well as different parts and fish organs eaten to better understand determinants of MeHg exposure. Postconception education to avoid fish consumption has been successful at decreasing TBHg in Korean women (Kim et al., 2006), and efficacious interventions to lower Hg exposure and promote healthy fish consumption among pregnant women of other ethnicities have also been reported (Kirk et al., 2017; Oken et al., 2013). Mercury guidance and intervention among Asians in the US, particularly subgroups at higher risk of mercury exposure, is warranted due to their fish consumption behavior.

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Competing financial interests declaration

The authors declare they have no actual or potential competing financial interests.

NHANES was approved by the NCHS Institutional Review Board, and all participants provided written informed consent.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2017.09.031.

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