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Total and methyl mercury in whole blood measured for the first time in the U.S. population: NHANES 2011–2012

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Abstract

Background—Despite the public health and toxicologic interest in methyl mercury (MeHg) and ethyl mercury (EHg), these mercury species have been technically difficult to measure in large population studies.

Methods—Using NHANES 2011–2012 data, we calculated reference ranges and examined demographic factors associated with specific mercury species concentrations and the ratio of MeHg to THg. We conducted several multiple regression analyses to examine factors associated with MeHg concentrations and also with the ratio of MeHg to THg.

Results—Asians had the highest geometric mean concentrations for MeHg, 1.58 μg/L (95% CI 1.29, 1.93) and THg, 1.86 μg/L (1.58, 2.19), followed by non-Hispanic blacks with MeHg, 0.52 μg/L (0.39, 0.68) and THg, 0.68 μg/L (0.54, 0.85). Greater education attainment in adults and male sex were associated with higher MeHg and THg concentrations. Race/ethnicity, age, and sex were significant predictors of MeHg concentrations, which increased with age and were highest in Asians in all age categories, followed by non-Hispanic blacks. Mexican Americans had the lowest adjusted MeHg concentrations. The ratio of MeHg to THg was highest in Asians, varied by racial/ethnic group, and increased with age in a non-linear fashion. The amount of increase in the MeHg to THg ratio with age depended on the initial ratio, with a greater increase as age increased.

Of the overall population, 3.05% (95% CI 1.77, 4.87) had MeHg concentrations >5.8 μg/L (a value that corresponds to the U.S. EPA reference dose). The prevalence was highest in Asians at 15.85% (95% CI 11.85, 20.56), increased with age, reaching a maximum of 9.26% (3.03, 20.42) at ages 60–69 years. Females 16–44 years old had a 1.76% (0.82–3.28) prevalence of MeHg concentrations >5.8 μg/L.

Conclusions—Asians, males, older individuals, and adults with greater educational attainment had higher MeHg concentrations. The ratio of MeHg to THg varied with racial/ethnic group, increased with age, and was nonlinear. U.S. population reference values for MeHg and the ratio of

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MeHg to THg can assist in more precise assessment of public health risk from MeHg consumed in seafood.

Keywords

Blood mercury; mercury species; methyl mercury; NHANES; biomonitoring

1. Introduction

Human mercury (Hg) toxicity depends on the form or species. Blood methyl mercury (MeHg), ethyl mercury (EHg), and inorganic mercury (IHg) are distinct species of public health concern, with distinct exposure sources and toxicologic characteristics.

Methyl and ethyl mercury have been technically difficult to measure at the same time in human blood specimens. Methyl mercury (MeHg) has been responsible for poisoning outbreaks of devastating neurotoxicity (Bakir et at, 1973; Harada, 1995). Fetal and infant brain development appears to be particularly susceptible to MeHg toxicity (Amin-Zaki et al., 1974; NRC, 2000). Because MeHg accumulates in fish, shellfish, and crustaceans, U.S. dietary recommendations are to limit consumption of certain fish, especially during pregnancy (U.S. EPA, 2014). Because of its antiseptic properties, sodium ethyl mercury thiosalicylate (thimerosal) has been used as a preservative in vaccines and such blood-derived products as immunoglobulins and plasma (FDA, 2014). Although there are concerns that EHg may be a neurotoxin similar to MeHg, EHg was more rapidly cleared (within days) from the body than MeHg; and at identical doses in animals, EHg appeared less able to enter the brain tissue than MeHg (Burbacher et al, 2005; Magos, 2003). Limited EHg measurements have been reported in human specimens or populations.

Inorganic (IHg) and total mercury (THg) blood measurements have been widely available and used in epidemiologic and research studies of mercury developmental neurotoxicity (Amin-Zaki, et al., 1974; Crump, et al., 1998; Steuerwald et al, 2000). THg or organic Hg (total minus inorganic Hg) have been used to estimate MeHg in whole blood based on studies showing that 70–95% of THg in blood is in the form of MeHg and bound to hemoglobin, unless there has been a recent large exposure to inorganic or elemental Hg (Bakir et al., 1973; Hansen et al., 1990; Oskarsson et al., 1996). In the absence of information to the contrary, the relationship of THg to MeHg has been assumed to be linear and constant across population demographics.

Blood THg and IHg have been measured as part of the National Health and Nutrition Examination Survey (NHANES) for more than a decade. NHANES provides data from a representative sample of the U.S. population and various analyses have reported predictors of higher THg, including male sex, older age, non-Hispanic black race/ethnicity, greater fish consumption, and higher income (Birch et al., 2014; Caldwell, et al, 2009; Mehaffey et al., 2004; Razzaghi et al., 2014; Tyrrell et al., 2013; Xue et al., 2012). THg concentrations over time have also been useful to examine whether there may be increased risk for excessive MeHg fetal exposure, using THg or organic Hg (calculated as THg minus IHg) in women of child-bearing age as a surrogate (Schober et al., 2003).

We report results of whole blood mercury species measured for the first time in NHANES 2011–2012. To our knowledge, this is the first time MeHg and EHg have been measured in a large population–based sample, and the first opportunity to assess demographic factors affecting the ratio of MeHg to THg. This survey period is also the first time that NHANES oversampled Asians, and with the ongoing oversampling of all Hispanic persons, we could obtain separate estimates for two additional racial/ethnic groups: Asians and All Hispanics (Mexican Americans and Other Hispanics). At the time of this analysis, the NHANES 2011–2012 dietary data were not available so we could not examine fish intake relative to the blood mercury concentrations.

2. Methods

2.1 NHANES

The National Health and Nutrition Examination Survey (NHANES) is conducted by the National Center for Health Statistics of the Centers for Disease Control and Prevention (CDC). The design is a complex, multistage, probability cluster sample designed to represent the U.S. population based on age, sex, and race/ethnicity (NCHS, 2008). The survey is intended to assess the health and nutritional status of the civilian, non-institutionalized U.S. population, and as an ongoing survey, collects information from about 5000 participants annually. NHANES collects information about a wide range of health-related behaviors and includes physical examinations and specimen collection for laboratory tests. Blood specimens are collected by venipuncture from participants ages 1 year and older. Data are publicly released in 2-year cycles. The survey incorporates sample population weights to account for the unequal selection probabilities caused by the cluster design, non-response, and planned over-sampling of certain subgroups (NCHS, 2008).

In NHANES 2011–12, racial/ethnic categories were self-reported as non-Hispanic white, non-Hispanic black, Hispanic (Mexican American and Other Hispanic), non-Hispanic Asian (referred to in this analysis as Asian), and other racial—multi-racial (Other). The Asian category includes all persons having origins in any of the original peoples of the Far East, Southeast Asia, or the Indian subcontinent (NHANES, 2013). The category “Other” was excluded from racial/ethnic descriptive calculations and multiple regression analyses because the proportion of the population they represented was heterogeneous and too small to produce valid reportable results. Similarly, based on the NHANES recommendations (Mirel et al, 2013), results for the group Other Hispanic were not reported separately but subjects in this group were included in a new group (All Hispanic) composed of subjects in either the Mexican American or Other Hispanic groups. Education level in adults (ages 20+ years) was determined by asking the highest grade level of school completed, or the highest degree received. Although dietary intake, including fish consumption, is part of the NHANES, the dietary intake data files for 2011–2012 were not available at the time of this analysis so we were unable to examine reported fish consumption.

For descriptive analyses, age was categorized as follows: 1–5, 6–11, 12–19, 20–29, 30–39, 40–49, 50–59, and 60+ years. For females, we also categorized ages 1–5, 16–29, and 30–44 years. The NCHS Research Ethics Review Board approved all content for NHANES 2011–

2012, and all participants provided signed, informed consent prior to data and specimen collection.

2.2 Blood Mercury Measurements

Whole blood specimens were collected in mercury-free containers, aliquotted, and stored at = −20 °C, and then shipped on dry ice to the CDC's National Center for Environmental Health laboratory, where they were stored frozen (= −20 °C) and then typically analyzed within three weeks of collection. THg was measured using the ELAN® DRC II inductively coupled-plasma dynamic reaction cell mass spectrometer (ICP-DRC-MS) from PerkinElmer Life Sciences (Shelton, CT, USA).

Quantification of inorganic (IHg), methyl (MeHg), and ethyl (EHg) mercury in whole blood used a triple spike isotope dilution (TSID) method employing a PerkinElmer® Clarus 500™ gas chromatograph (GC) and the ELAN® DRC II ICP-DRC-MS (PerkinElmer Life Sciences, Shelton, CT, USA). We used a robotic CombiPAL® (CTC Analytics, Zwinger, Switzerland) sample handling station featuring twin fiber-based solid phase microextraction (SPME) injector heads. MeHg, EHg, and IHg were analyzed through the use of Solid Phase Micro Extraction (SPME) fiber for delivering sample to a GC coupled to an ICP-DRC-MS. Method accuracy was verified by analyzing whole blood standards (SRM 955c level 3) from the National Institute of Standards and Technology (NIST) and making an average of 60 independent replicate measurements over 10 months. The following are CDC vs. certified mean mercury concentrations in μg/L; with standard deviation (SD) and relative standard deviation (RSD) as percentages: for THg, the CDC mean was 19.1 (SD 0.9; RSD 4.6) vs. NIST-certified mean 17.8;. For MeHg, the CDC mean was 4.5 (SD 0.3; RSD 6.3) vs. NIST-certified mean 4.5. For EHg, the CDC mean was 5.0 (SD 0.3; RSD 6.2) vs. NIST-certified mean 5.1. For IHg, the CDC mean was 9.1 (SD 0.8; RSD 8.3) vs. NIST-certified mean 9.0. The limit of detection (LOD) was 0.16 μg/L for THg; 0.12 μg/L for MeHg; 0.16 μg/L for EHg, and 0.27 μg/L for IHg. Additional details including proficiency testing results have been published (Sommer et al., 2014).

2.3 Statistical Analysis

Category-specific geometric means and percentiles were calculated using SUDAAN version 11.0.0 (Research Triangle Institute, Research Triangle Park, NC, USA). SUDAAN uses sample weights and calculates variance estimates that account for the complex survey design. Ninety-five percent confidence interval (CIs) for geometric means were estimated based on the Taylor series linearization method (SUDAAN user's manual, 2001), and CIs for percentiles were adapted from the methods of Korn and Graubard (1998) and Woodruff (1952). Geometric means were computed when >60% of the samples had detectable values. The LOD divided by the square root of 2 was used for imputation of values lower than the LOD. The Spearman rank coefficient was calculated to determine the correlation between THg and MeHg.

Mercury values were log10 transformed prior to modeling. Because the Mexican American group represents a subset of the All Hispanic group, two separate multiple regression models were used to obtain model adjusted estimates. The primary model included the non-Hispanic

White, non-Hispanic Black, Asian, and All Hispanic racial/ethnic groups. Then to obtain separate estimates for the Mexican American group, a second model was used that included the non-Hispanic White, non-Hispanic Black, Asian, and Mexican American racial/ethnic groups. Adjusted geometric means of demographic groups were calculated using multiple regression models. The term “adjusted” is used throughout the manuscript to refer to model-based results and indicates that all statistically significant main effects and interactions were included in the model.

When modeling each Hg species (total and methyl), sex, race/ethnicity, and age were considered as main effects, and all two-way interactions and age^2 were included in the original model. The final models for both Hg species included sex, race/ethnicity, age, age^2, and the interaction between race/ethnicity and age. Results are presented as adjusted geometric means and shown for selected ages and by race/ethnicity or sex.

To model the ratio of MeHg to THg (MeHg:THg) we used the log10 (MeHg:THg) as the dependent variable. Sex, race/ethnicity, and age were considered as main effects, and all two-way interactions and age^2 were included in the original model. The final model included race/ethnicity and age. Because the ratio of MeHg:THg varied with age, we chose to calculate estimates for the different racial/ethnic groups at a common age. The common age we chose was the weighted mean age of the population, 39.7 years, and we used this age to display the results for each racial/ethnic category.

We use the term “mean” to refer to geometric mean and “adjusted mean” to refer to adjusted geometric mean in the following sections.

3. Results

3.1 NHANES 2011–2012

The overall response rate was 69.5% (unweighted) for participants examined in the 2011–2012 survey. There were 7920 results for THg and 7841 results for Hg species. Detection frequencies were as follows: THg 92.9%; MeHg 83.7%; EHg 3.5%; and IHg 22.7%. The overall population mean for MeHg was 0.50 μg/L (0.42, 0.59) and for THg was 0.70 μg/L (0.62, 0.80, 95% CI) (Tables 1 and 2, respectively). THg and MeHg concentrations were highly correlated (r=0.9405). Because our focus was the newly available MeHg results and these were highly correlated with THg, THg results are not further described beyond the descriptive results shown in Table 2.

MeHg concentrations increased with age and were higher in males than females (Table 1). Asians had the highest mean MeHg concentration of the racial/ethnic groups, 1.58 μg/L (1.29, 1.93) and Mexican Americans had the lowest, 0.32 μg/L (0.27, 0.39). The age-related increase in mean MeHg concentrations was also apparent in child-bearing aged females, with mean concentrations of 0.42 μg/L (0.33 0.55) in females 16–29 years old, compared to 0.50 μg/L (0.42, 0.59) in females 30–44 years old. MeHg mean concentrations also appeared to increase with educational attainment in the adults, ranging from 0.48 μg/L (0.39, 0.60) in those with <9th grade completion to 0.96 μg/L (0.76, 1.21) in adults who were college graduates and higher.

Because of low detection frequencies, means were not calculated for EHg and IHg, and only higher percentiles could be calculated (Supplemental Tables 1 and 2, respectively). EHg was detected at the 95th percentile only in Asians, and IHg was detected at the 75th or higher percentiles.

3.2 Multiple Regression Analysis

Multiple regression analysis results with the adjusted mean concentrations for both MeHg and THg at selected ages are shown in Table 3. The significant predicators of whole blood MeHg and THg were similar: race/ethnicity, age, age^2, and the interaction of race/ethnicity and age. Computed at the weighted mean age of 39.7 years, males had significantly higher adjusted mean MeHg and THg concentrations than females (MeHg, 0.66 vs. 0.61 μg/L, p=0.0119; THg, 0.90 vs. 0.84 μg/L, p=0.0037). These differences correspond to males having 8.9% and 7.4% higher adjusted mean MeHg and THg concentrations, respectively, compared with females. Adjusted means increased with age in each racial/ethnic category, and there were notable racial/ethnic differences. Asians had higher adjusted MeHg concentrations compared to any other racial/ethnic categories at similar ages. Non-Hispanic blacks had the next highest adjusted MeHg concentrations, followed by All Hispanics and non-Hispanic whites; the lowest adjusted concentrations were in Mexican Americans. Because Mexican Americans were a subset of the All Hispanic category, the adjusted MeHg and THg estimates were calculated using a separate regression model that included non-Hispanic white, non-Hispanic black, Asian and Mexican American categories only.

3.3 Ratio of MeHg to THg (MeHg:THg)

Table 4 shows the estimated ratio of MeHg:THg at the weighted mean age of 39.7 years for each racial/ethnic category. Asians had a significantly higher ratio compared to all other categories: non-Hispanic whites (p< 0.0001); non-Hispanic blacks (p=0.0050); All Hispanics (p< 0.0001); and Mexican Americans (p=0.0003). Non-Hispanic blacks had higher ratios than Mexican Americans (p=0.00983) and non-Hispanic whites (p=0.0244). In each racial/ethnic category, the estimated ratios increased slightly with age (Figure 1). The amount of increase with age depended on the initial ratio, with a greater increase as age increased. Thus, in each racial/ethnic category, the ratio of MeHg:THg was not quite linear with age, despite the appearance of the curves in Figure 1. Actually, log10(ratio) was linear with age, but when results were back transformed to ratios, the relationship was slightly non-linear.

3.4 Prevalence of MeHg Concentrations Higher than 5.8 μg/L

The U.S. Environmental Protection Agency (U.S. EPA) has used mathematical modeling and THg measurements to estimate a maximum daily intake of MeHg that is not likely to cause harmful effects during a lifetime, known as a reference dose (RdF) (U.S. EPA, 2014). The current value is 0.1 μg/kg/day and is equivalent to a blood THg concentration of 5.8 μg/L. We calculated the weighted prevalence of MeHg concentrations above this value in the U.S. population, shown in Table 5. Older adults, particularly 60–69 year olds, males, and Asians had the highest prevalence of MeHg above 5.8 μg/L. Of particular interest are young children ages 1–5 years of whom 0.05% (CI 0.00, 0.65) had MeHg concentrations higher than 5.8 μg/L. Also of interest are women in the child-bearing ages 16–44, of whom 1.76%

(0.82, 3.28) had MeHg concentrations higher than 5.8 μg/L. Among women 16–44 years old, Asians had the highest prevalence at 9.71% (6.02, 14.62), followed by non-Hispanic whites at 1.6% (0.41, 4.48), and the other racial/ethnic groups at <1% (data not shown); but the sample sizes were generally small, resulting in unstable estimates.

4. Discussion

We analyzed whole blood speciated and total Hg (THg) in a representative sample of the U.S. population. To our knowledge, speciated Hg results have not been reported previously in such a large or nationally representative sample. The NHANES 2011–2012 survey oversampled Asian and All Hispanic racial/ethnic groups, allowing us to distinguish five racial/ethnic categories, although Mexican American were a subset of the All Hispanic category. THg and MeHg concentrations were highly correlated, as expected. The overall mean MeHg and THg concentrations in Asians was approximately two to three times higher compared to other racial/ethnic groups; and regardless of age, Asians had the highest concentrations relative to other racial/ethnic groups. Non-Hispanic blacks had the next highest overall mean concentrations MeHg and THg, followed by non-Hispanic whites, All Hispanics, and then Mexican Americans. We speculate that higher MeHg and THg concentrations in All Hispanics compared to Mexican Americans may reflect the diversity of Hispanic diets, with greater fish consumption in Other Hispanics (e.g., non-Mexican Americans). In fact, most of the racial/ethnic differences likely are the result of differences in fish consumption, but we did not have dietary information to examine this exposure source. Previous NHANES analyses reported higher blood concentrations in Asians relative to other racial/ethnic groups as a result of greater fish consumption (Mehaffey et al., 2004; Mehaffey and Mergler, 1998). Higher whole blood THg concentrations in non-Hispanic blacks compared to non-Hispanic whites and Mexican Americans also have been noted in previous NHANES analyses (Birch et al., 2014; Caldwell, et al 2009; Mehaffey et al., 2004) but fish consumption does not appear to be a satisfactory explanation (Schober, et al., 2003). Several notable findings in the present analysis are the influence of higher education attainment on THg and MeHg concentrations, the addition of new racial/ethnic categories and their effects on THg and MeHg, and also the effects of age, sex, and race/ethnicity on the ratio of THg to MeHg. We speculate that higher THg and MeHg concentrations in adults with the highest education attainment (college graduate or above) may be related to higher income and to health-related dietary choices that include greater fish consumption. These possibilities can be examined when the NHANES 2011–2012 dietary survey details become available. However, other NHANES analyses have demonstrated an association between higher income, fish consumption, and higher whole blood THg (Tyrrell et al., 2013) or organic Hg (Birch et al., 2014). Increasing concentrations of THg with age have been previously noted (Caldwell et al, 2009) and presumably result from bioaccumulation of organic Hg, a consequence of the very slow elimination of MeHg (terminal half-life of 50–52 days) (Kershaw et al., 1980; Miettinen et al., 1971) that may be more prolonged with advance age (Mayersohn 1994). It is also likely that recurring fish consumption contributes to age-related THg concentrations.

The ratio of MeHg to THg differed by racial/ethnic group and increased slightly with age, with the age-related increase dependent on the magnitude of the initial value. At the

weighted mean age of 39.7 years, the ratio was higher for Asians (0.85) and lowest for Mexican Americans (0.67), and the ratio trended upward slightly more steeply with age in Asians (Figure 1). In a sample of Korean adults, You et al (2012) observed a ratio and trend similar to U.S. Asians in our analysis. The ratios calculated from measured values of THg and MeHg in the U.S. general population contrast with previous estimates by Bakir et al (1973) made in poisoned individuals, in whom as much as 90–95% of blood THg was organic Hg (e.g., MeHg) and by Phelps et al. (1980), who reported a linear correlation between total and organic Hg in heavy consumers of mercury-contaminated fish. Other studies in adults who regularly consumed fish reported organic Hg to THg ratios ranging from 0.70 to 0.80, or 70–80% (Hansen et al., 1990; Mehaffey and Mergler, 1998; Oskarsson et al., 1996). Differences in the ratio of MeHg to THg may be explained by the amount, frequency, and how recently MeHg-containing fish have been consumed. That is, frequent fish consumers may have higher MeHg intake, and recent fish consumption may transiently elevate blood MeHg concentrations for a few days while the MeHg is undergoing distribution throughout the body. This pharmacologic explanation is supported by serial measurements in humans after they consumed a fish meal containing MeHg. Blood concentrations of THg and organic Hg increased shortly after the meal, peaked 4–14 hours later at concentrations about 10 times higher than baseline, and did not return to baseline by 2.5 days post-meal (Kershaw et al., 1980). These authors cautioned that measurements of Hg may be misleading and elevated if blood is obtained within a few days after a fish meal containing MeHg. This caution is relevant to smaller studies. However, the present analysis involved a large sample size and generated population estimates, so bias attributable to time since last fish meal is likely to be minimized. Use of single blood specimens from each individual is another potential limitation, but NHANES collects blood is collected at various times during the day, which also can mitigate the effects of extreme values. Nonetheless, it may be desirable to distinguish subpopulations at risk for sustained elevated MeHg concentrations and increased body burdens of MeHg, particularly young females with the highest likelihood of becoming pregnant. This might be possible in NHANES if fish consumption in the previous week were included in the dietary questionnaire, along with the food frequency and 24-hour dietary recall currently obtained. Another limitation is that Hg data were available for only one NHANES survey period, whereas data from a full NHANES sampling period of four years provides more robust U.S. population and subgroup estimates (NHANES, 2013).

The U.S. EPA has promulgated a MeHg reference dose (RfD, which is an estimate for the maximum acceptable daily exposure that is not likely to cause harmful effects during a lifetime (U.S. EPA, 2001). The RfD of 0.1 μg/kg/day corresponds to a blood MeHg concentration of 5.8 μg/L, which has been calculated as THg minus IHg in the absence of MeHg measurements. In a report that analyzed NHANES data from 1999–2010, U.S. EPA estimated that 3.1% of women 16–49 years had calculated blood MeHg concentrations higher than 5.8 μg/L. Similar to the current analysis, older age, higher income and race/ethnicity were predictors of higher concentrations (U.S. EPA, 2013). In the same report, U.S. EPA noted that in NHANES 2009–2010, 2.14% (SE 0.36) of women 16–49 years old had calculated blood MeHg concentrations higher than 5.8 μg/L, similar to our finding of 1.76% (95% CI 0.82, 3.28) in women 16–44 years old. Further, in NHANES 2011–2012, we were

able to show that Asians overall had the highest percentage, 15.85% (11.85, 20.56) with MeHg higher than 5.8 μg/L (Table 5) and that MeHg concentrations increased with age regardless of race/ethnicity (Table 1). The increase in MeHg with age has implications for estimating the number of infants at risk for *in utero* exposure to MeHg. Because older U.S. women (e.g., 35 years and above) have lower birth rates than younger women (Martin et al., 2012), using the estimated prevalence in women 16–49 years probably overestimates the numbers of births to women with higher blood MeHg. Sample size limitations precluded our ability to reliably estimate the prevalence of MeHg > 5.8 μg/L in females by smaller age strata; with additional data in the future, this limitation may be overcome. Young children are also a population of concern for MeHg exposure because of potential developmental neurotoxicity. Notably, MeHg concentrations at the 95th percentile in children 1–5 years old were at least 5 times lower than the RfD-equivalent blood concentration of 5.8 μg/L (Table 1).

The National Research Council employed the bench mark dose (BMD) method of risk assessment and used data from a study of children born to women with MeHg exposure from seafood (NRC, 2000). They estimated the cord blood Hg concentration at which the most sensitive neurocognitive effect could be detected. The resulting BMD of 85 μg/L had a lower 95% confidence bound or benchmark dose limit (BMDL) of 58 μg/L (NRC, 2000). This is a health outcome-derived estimate of risk, and the BMDL is 4–5 times higher than the results observed in Asians, the group with the highest MeHg concentrations in NHANES 2011–2012 (95th percentile:10.49 μg/L, 95% CI 8.48, 12.51). Thus, even this group with the highest MeHg concentrations was found to have values well below those associated with the most sensitive neurocognitive toxic effect.

EHg concentrations were detectable in only 3.5% (weighted) of the overall U.S. population, and no demographic group had >6% detection frequency. These findings are not unexpected because the most likely EHg exposure sources, vaccine or medication administration, are sporadic, and EHg disappears rapidly from the blood. The reported elimination half-life is several days, and possibly even shorter in infants and young children (Burbacher et al., 2005; Magos, 2003).

IHg concentrations were frequently nondetectable, a finding noted in previous NHANES analyses (Caldwell, et al., 2009). However, Asians had a greater prevalence of detectable concentrations (34.2%, weighted), possibly related to higher THg and MeHg concentrations. In the body, THg and MeHg both can be demethylated, resulting in elimination of IHg in urine and increased blood IHg concentrations, which have been observed in populations who consume large amounts of fish (Abe, et al., 1995; Ohno et al., 2007; Sherman, et al., 2013).

4.1 Conclusions

The NHANES 2011–2012 provided unique data to examine THg, MeHg, EHg and IHg in a representative sample of U.S. population. We determined that the relationship between MeHg and THg was not fixed among racial/ethnic categories and increased with advancing age. As a group, Asians had higher THg and MeHg concentrations, regardless of age, and the highest ratio of THg to MeHg compared to other racial/ethnic groups. Non-Hispanic blacks had the next highest overall MeHg concentrations and ratio of THg to MeHg, and

Mexican Americans had the lowest. EHg was detected infrequently, only in 3.5% (weighted) of the overall sample. IHg was detected only in 24.9% (weighted) of the overall sample, and the highest detection frequency was in the Asian racial/ethnic group, possibly related to the higher blood THg and MeHg concentrations in this group. MeHg concentrations increased with age across all demographic categories and did not exceed the BMDL of 58 μg/L, even at the 95th percentile. However, at the 95th percentile, MeHg concentrations in adults older than 60 years or highly educated, and for Asians overall, were more likely to exceed 5.8 μg/L, equivalent to the U.S. EPA's RfD. Children ages 1–5 years had the lowest MeHg concentrations.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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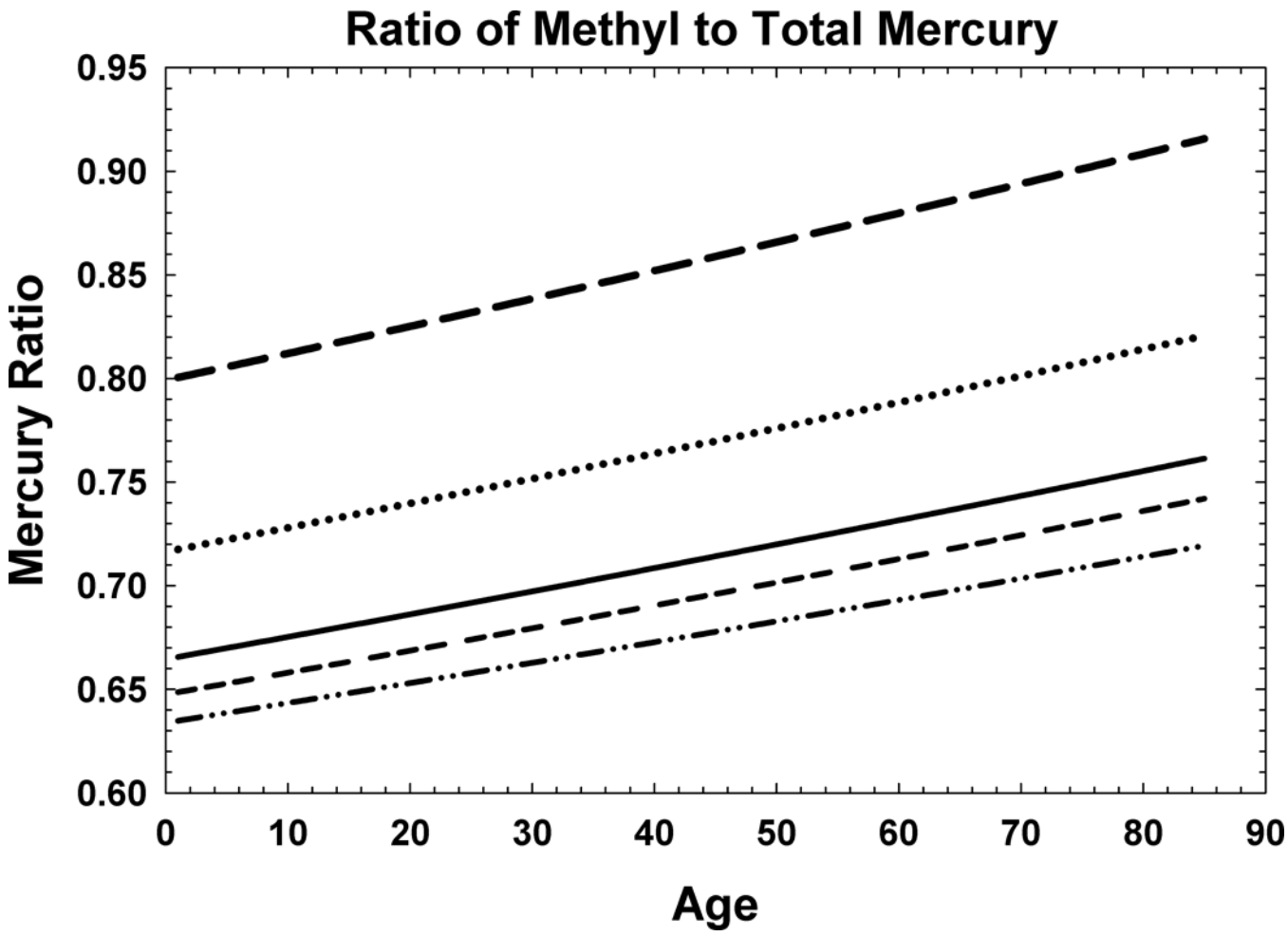

Figure 1. The Ratio of Methyl Mercury to Total Mercury in the U.S. Population for Ages 1 to 85 Years, NHANES 2011–2012

Each curve displays the estimated ratio of MeHg:THg for one of the racial/ethnic groups. The ratio is plotted against age using NHANES data for ages 1 to 85 years. To improve readability, the 95% confidence intervals are not shown. Each curve increases slightly with age, and the magnitude of the increase depends on the initial value. As a result, the curves are not parallel.

Race/ethnicity Key: AH=All Hispanic; MA=Mexican American; NHA= Non-Hispanic Asian (Asian); NHB=Non-Hispanic Black; NHW=Non-Hispanic White

Table 1

Sample-weighted Methyl Mercury Whole Blood Concentrations (μg/L) in the U.S. Population, NHANES 2011–2012

Category	Geometric mean (95% CI)	50th %tile (95% CI)	75th %tile (95% CI)	90th %tile (95% CI)	95th %tile (95% CI)	Unweighted Sample Size
All ages (in years)	0.50 (0.42, 0.59)	0.48 (0.40, 0.57)	1.25 (0.95, 1.61)	2.81 (2.29, 3.55)	4.43 (3.46, 5.49)	7841
1–5	0.17 (0.15, 0.19)	0.14 (0.12, 0.17)	0.27 (0.22, 0.35)	0.54 (0.42, 0.78)	0.97 (0.59, 1.14)	657
6–11	0.21 (0.18, 0.24)	0.18 (0.15, 0.22)	0.40 (0.33, 0.49)	0.82 (0.64, 1.06)	1.34 (0.94, 1.84)	1044
12–19	0.28 (0.24, 0.32)	0.27 (0.21, 0.31)	0.57 (0.46, 0.67)	1.27 (0.87, 1.67)	2.15 (1.41, 2.81)	1121
20–29	0.49 (0.37, 0.64)	0.49 (0.36, 0.65)	1.13 (0.73, 1.80)	2.48 (1.61, 3.64)	3.70 (2.89, 4.99)	892
30–39	0.53 (0.45, 0.63)	0.50 (0.42, 0.66)	1.25 (0.95, 1.58)	2.83 (2.09, 3.66)	4.41 (3.61, 5.96)	880
40–49	0.64 (0.51, 0.81)	0.61 (0.46, 0.83)	1.39 (0.99, 1.97)	2.91 (1.97, 4.51)	4.56 (3.30, 7.20)	834
50–59	0.73 (0.57, 0.92)	0.79 (0.58, 0.97)	2.06 (1.51, 2.52)	3.33 (2.76, 4.43)	4.74 (3.69, 6.86)	831
60–69	0.83 (0.58, 1.19)	0.77 (0.52, 1.21)	1.94 (1.13, 3.96)	4.97 (2.24, 9.97)	8.90 (3.60, 12.93)	822
70+	0.62 (0.50, 0.78)	0.65 (0.46, 0.83)	1.62 (1.24, 2.23)	3.53 (2.68, 4.43)	5.09 (4.10, 6.82)	760
Females						
All females	0.49 (0.41, 0.58)	0.47 (0.38, 0.56)	1.19 (0.90, 1.61)	2.72 (2.18, 3.46)	3.99 (3.28, 4.99)	3916
1–5 years	0.17 (0.15, 0.19)	0.13 (<LOD, 0.18)	0.29 (0.22, 0.38)	0.68 (0.43, 0.95)	1.03 (0.54, 1.68)	321
16–29 years	0.42 (0.33, 0.55)	0.39 (0.29, 0.52)	0.95 (0.65, 1.57)	2.48 (1.45, 3.60)	3.46 (2.60, 4.70)	715
30–44 years	0.50 (0.42, 0.59)	0.47 (0.40, 0.56)	1.03 (0.80, 1.39)	2.63 (1.78, 3.43)	3.71 (2.86, 5.22)	664
Males (all)	0.51 (0.43, 0.60)	0.49 (0.40, 0.59)	1.30 (0.99, 1.62)	2.84, (2.29, 3.68)	4.77 (3.44, 6.74)	3925
Race/ethnicity						
Non-Hispanic White	0.48 (0.39, 0.58)	0.47 (0.36, 0.58)	1.25 (0.87, 1.69)	2.76 (2.06, 3.69)	4.24 (2.92, 6.38)	2477
Non-Hispanic Black	0.52 (0.39, 0.68)	0.51 (0.38, 0.66)	1.13 (0.75, 1.61)	2.37 (1.66, 3.08)	3.63 (2.57, 5.16)	2170
Mexican American	0.32 (0.27, 0.39)	0.33 (0.26, 0.41)	0.61 (0.51, 0.77)	1.23 (0.92, 1.40)	1.66 (1.33, 2.02)	1058
All Hispanic	0.43 (0.35, 0.53)	0.42 (0.34, 0.52)	0.89 (0.70, 1.17)	1.81 (1.39, 2.46)	2.94 (2.19, 3.71)	1902
Asian	1.58 (1.29, 1.93)	2.13 (1.68, 2.53)	4.35 (3.64, 5.13)	7.57 (6.21, 8.61)	10.49 (8.48, 12.51)	997
Education (adults 20+ years)						
<9th grade	0.48 (0.39, 0.60)	0.45 (0.35, 0.56)	1.06 (0.76, 1.37)	2.36 (1.58, 3.54)	4.29 (2.65, 7.42)	472
9–11th grade	0.43 (0.32, 0.54)	0.39 (0.30, 0.53)	0.94 (0.68, 1.16)	1.96 (1.42, 2.66)	2.83 (2.02, 4.89)	685
[a] HS grad or GED	0.50 (0.41, 0.62)	0.48 (0.36, 0.63)	1.20 (0.84, 1.59)	2.65 (1.74, 3.66)	3.75 (2.80, 5.73)	1056

Category	Geometric mean (95% CI)	50th %tile (95% CI)	75th %tile (95% CI)	90th %tile (95% CI)	95th %tile (95% CI)	Unweighted Sample Size
some college or associate degree	0.56 (0.49, 0.65)	0.57 (0.50, 0.65)	1.29 (1.02, 1.61)	2.67 (2.20, 3.44)	3.91 (2.87, 5.35)	1521
college grad and more	0.96 (0.76, 1.21)	1.00 (0.74, 1.45)	2.44 (1.84, 3.10)	4.77 (3.63, 6.59)	7.46 (5.01, 9.72)	1283

[a]High School graduate or equivalent (GED, General Educational Development tests); Limit of detection = 0.12 μg/L

Table 2

Sample-weighted Total Mercury Whole Blood Concentrations (μg/L) in the U.S. Population, NHANES 2011–2012

Category	Geometric mean (95% CI)	50th %tile (95% CI)	75th %tile (95% CI)	90th %tile (95% CI)	95th %tile (95% CI)	Unweighted Sample Size
All ages (in years)	0.70 (0.62, 0.80)	0.64 (0.58, 0.73)	1.38 (1.14, 1.72)	2.87 (2.39, 3.62)	4.40 (3.50, 5.71)	7920
1–5	0.26 (0.24, 0.29)	0.25 (0.22, 0.27)	0.39 (0.34, 0.45)	0.68 (0.54, 0.88)	0.99 (0.79, 1.21)	713
6–11	0.33 (0.29, 0.38)	0.32 (0.28, 0.36)	0.53 (0.48, 0.60)	0.93 (0.78, 1.20)	1.40 (1.02, 2.17)	1048
12–19	0.41 (0.36, 0.48)	0.37 (0.32, 0.45)	0.68 (0.59, 0.80)	1.32 (1.08, 1.75)	2.25 (1.46, 2.87)	1129
20–29	0.66 (0.52, 0.83)	0.63 (0.52, 0.78)	1.23 (0.91, 1.86)	2.61 (1.87, 3.68)	3.86 (2.75, 4.94)	892
30–39	0.76 (0.66, 0.87)	0.68 (0.58, 0.85)	1.38 (1.13, 1.66)	2.86 (2.16, 3.81)	4.40 (3.59, 5.84)	880
40–49	0.89 (0.75, 1.05)	0.80 (0.64, 1.01)	1.63 (1.25, 2.12)	2.98 (2.24, 4.31)	4.83 (3.63, 6.81)	835
50–59	1.00 (0.85, 1.18)	1.00 (0.78, 1.21)	2.17 (1.64, 2.73)	3.38 (2.88, 4.33)	4.78 (3.87, 6.37)	833
60–69	1.13 (0.82, 1.56)	1.00 (0.73, 1.52)	2.21 (1.44, 3.91)	5.23 (2.48, 11.30)	9.28 (3.52, 13.51)	824
70+	0.87 (0.75, 1.02)	0.87 (0.71, 1.03)	1.89 (1.36, 2.41)	3.79 (2.89, 4.50)	5.19 (4.20, 6.12)	766
Females						
All females	0.69 (0.61, 0.79)	0.64 (0.58, 0.74)	1.36 (1.09, 1.75)	2.81 (2.28, 3.50)	4.03 (3.32, 5.08)	3952
1–5 years	0.26 (0.23, 0.29)	0.24 (0.21, 0.29)	0.38 (0.32, 0.47)	0.76 (0.56, 0.93)	1.05 (0.70, 1.65)	345
16–29 years	0.59 (0.48, 0.73)	0.56 (0.44, 0.66)	1.06 (0.78, 1.72)	2.54 (1.64, 3.28)	3.47 (2.72, 4.36)	716
30–44 years	0.73 (0.64, 0.82)	0.64 (0.57, 0.76)	1.21 (0.97, 1.55)	2.70 (1.91, 3.21)	4.08 (2.88, 5.29)	665
Males (all)	0.71 (0.62, 0.81)	0.65 (0.57, 0.73)	1.40 (1.17, 1.72)	3.00 (2.44, 3.91)	4.94 (3.50, 6.79)	3968
Race/ethnicity						
Non-Hispanic White	0.69 (0.58, 0.81)	0.63 (0.55, 0.75)	1.38 (1.09, 1.82)	2.83 (2.18, 3.82)	4.25 (3.02, 6.24)	2493
Non-Hispanic Black	0.68 (0.54, 0.85)	0.63 (0.51, 0.79)	1.24 (0.89, 1.72)	2.45 (1.84, 3.10)	3.80 (2.70, 5.37)	2195
Mexican American	0.48 (0.43, 0.55)	0.48 (0.40, 0.56)	0.81 (0.72, 0.90)	1.44 (1.18, 1.63)	1.90 (1.57, 2.19)	1077
All Hispanic	0.61 (0.53, 0.71)	0.59 (0.49, 0.71)	1.08 (0.89, 1.33)	1.96 (1.60, 2.68)	3.03 (2.37, 3.86)	1931
Asian	1.86 (1.58, 2.19)	2.30 (1.84, 2.64)	4.32 (3.71, 5.21)	7.71 (6.38, 8.79)	10.32 (8.85, 11.97)	1005
Education (adults 20+ years)						
<9th grade	0.69 (0.57, 0.82)	0.63 (0.49, 0.80)	1.24 (0.96, 1.41)	2.54 (1.78, 3.49)	4.71 (2.87, 7.93)	475
9–11th grade	0.64 (0.55, 0.74)	0.58 (0.47, 0.69)	1.18 (0.92, 1.35)	2.23 (1.90, 2.75)	3.04 (2.42, 4.80)	686
[a] HS grad or GED	0.72 (0.61, 0.84)	0.64 (0.55, 0.76)	1.38 (1.09, 1.74)	2.77 (2.00, 3.70)	3.89 (2.86, 6.69)	1057

Category	Geometric mean (95% CI)	50th %tile (95% CI)	75th %tile (95% CI)	90th %tile (95% CI)	95th %tile (95% CI)	Unweighted Sample Size
some college or associate degree	0.79 (0.70, 0.88)	0.75 (0.65, 0.83)	1.39 (1.16, 1.74)	2.81 (2.41, 3.39)	3.98 (3.23, 5.21)	1525
college grad or more	1.24 (1.01, 1.52)	1.18 (0.96, 1.62)	2.56 (1.98, 3.16)	4.97 (3.63, 6.56)	7.52 (5.14, 10.24)	1285

[a]High School graduate or equivalent (GED, General Educational Development tests); Limit of detection = 0.16 μg/L

Table 3

Adjusted Estimates[a] for Methyl and Total Mercury (μg/L) at Selected Ages By Racial/Ethnic Category, NHANES 2011–2012

		Methyl Mercury		Total Mercury	
Category	**Age in Years**	**LSGM**	**95% CI**	**LSGM**	**95% CI**
Males	39.7	0.66[b]	0.56, 0.78	0.90[c]	0.78, 1.04
Females	39.7	0.61	0.51, 0.73	0.84	0.73, 0.97
Non-Hispanic White:					
	2.5	0.13	0.10, 0.16	0.21	0.16, 0.26
	5	0.15	0.12, 0.18	0.24	0.19, 0.30
	10	0.19	0.16, 0.24	0.30	0.25, 0.37
	15	0.25	0.21, 0.30	0.38	0.31, 0.45
	25	0.38	0.31, 0.45	0.55	0.46, 0.65
	40	0.58	0.47, 0.71	0.81	0.68, 0.96
	50	0.68	0.54, 0.85	0.94	0.79, 1.13
	60	0.72	0.55, 0.93	1.00	0.82, 1.22
	80	0.60	0.42, 0.85	0.87	0.66, 1.13
Non-Hispanic Black:					
	2.5	0.19	0.15, 0.24	0.29	0.24, 0.35
	5	0.22	0.17, 0.27	0.32	0.27, 0.39
	10	0.28	0.22, 0.35	0.40	0.33, 0.48
	15	0.35	0.27, 0.44	0.48	0.40, 0.58
	25	0.50	0.39, 0.64	0.66	0.54, 0.81
	40	0.71	0.55, 0.93	0.90	0.73, 1.12
	50	0.80	0.61, 1.05	0.99	0.79, 1.24
	60	0.80	0.61, 1.07	0.99	0.79, 1.25
	80	0.61	0.43, 0.85	0.76	0.57, 1.00
Mexican American[d]:					

Category	Age in Years	Methyl Mercury		Total Mercury	
		LSGM	95% CI	LSGM	95% CI
	2.5	0.15	0.12, 0.19	0.26	0.21, 0.30
	5	0.17	0.14, 0.22	0.28	0.24, 0.33
	10	0.21	0.17, 0.26	0.34	0.29, 0.39
	15	0.26	0.21, 0.31	0.40	0.35, 0.46
	25	0.35	0.29, 0.41	0.52	0.46, 0.59
	40	0.44	0.38, 0.53	0.65	0.58, 0.72
	50	0.46	0.39, 0.54	0.67	0.60, 0.74
	60	0.43	0.36, 0.51	0.63	0.55, 0.71
	80	0.28	0.21, 0.36	0.42	0.33, 0.53
All Hispanic:					
	2.5	0.17	0.14, 0.20	0.27	0.23, 0.32
	5	0.19	0.16, 0.23	0.30	0.26, 0.35
	10	0.24	0.20, 0.29	0.38	0.33, 0.43
	15	0.30	0.25, 0.36	0.46	0.40, 0.52
	25	0.43	0.36, 0.52	0.62	0.55, 0.71
	40	0.62	0.50, 0.76	0.85	0.73, 0.98
	50	0.69	0.55, 0.85	0.93	0.79, 1.09
	60	0.69	0.54, 0.88	0.93	0.78, 1.11
	80	0.52	0.37, 0.71	0.71	0.55, 0.91
Asian:					
	2.5	0.40	0.29, 0.56	0.55	0.43, 0.70
	5	0.47	0.34, 0.64	0.62	0.50, 0.79
	10	0.62	0.47, 0.82	0.81	0.66, 0.99
	15	0.81	0.62, 1.04	1.02	0.84, 1.23
	25	1.25	1.00, 1.57	1.51	1.27, 1.79
	40	2.00	1.62, 2.48	2.31	1.95, 2.73

		Methyl Mercury		Total Mercury	
Category	**Age in Years**	**LSGM**	**95% CI**	**LSGM**	**95% CI**
	50	2.41	1.95, 2.99	2.73	2.30, 3.25
	60	2.63	2.11, 3.28	2.96	2.48, 3.54
	80	2.30	1.75, 3.03	2.66	2.12, 3.33

[a]Model based least squares geometric means (LSGM) and 95% confidence intervals (CIs) for males and females were computed at a weighted mean age of 39.7 years and were adjusted for race/ethnicity, age, age2, and the interaction between race/ethnicity and age. LSGMs and their 95% CIs for various racial groups at selected ages were adjusted for sex using a multiple regression model that included the non-Hispanic White, non-Hispanic Black, Asian, and All Hispanic groups, except where noted (see footnote [d] with regard to Mexican Americans).

[b]The methyl Mercury LSGM for males was significantly higher (p=0.0119) by 8.9% (95% CI: 2.2%–16.1%) than that for females.

[c]The total Mercury LSGM for males was significantly higher (p=0.0037) by 7.4% (95% CI: 2.72%–12.3%) than that for females.

[d]Because Mexican Americans represent a subset of the All Hispanic group, the LSGM estimates are based on a separate multiple regression model that included the non-Hispanic White, non-Hispanic Black, Asian, and Mexican American groups only.

Table 4

Estimated Ratio[a] of Methyl Mercury to Total Mercury at 39.7 Years of Age by Racial/Ethnic Category, NHANES 2011–2012

Race/ethnicity	Estimated Ratio	(95% Confidence Interval)
Non-Hispanic White	0.69	(0.64, 0.74)
Non-Hispanic Black	0.76	(0.72, 0.81)
Mexican American	0.67	(0.63, 0.72)
All Hispanic	0.71	(0.66, 0.76)
Asian	**0.85**	(0.80, 0.90)

[a]Estimated ratios were adjusted to the weighted mean age of 39.7 years.

Table 5

Estimated Prevalence of Persons with Methyl Mercury Greater than 5.8 ug/L in the U.S. population, NHANES 2011–2012

Category	[a] Percent > 5.8 μg/L (95% Conf. Interval)	Unweighted Sample Size
Total Population	3.05 (1.77–4.87)	7841
Age Group (years)		
1–5	0.05 (0.00–0.65)	657
1–9	0.05 (0.00–0.37)	1357
10–19	0.18 (0.03–0.57)	1465
20–29	1.76 (0.75–3.46)	892
30–39	3.32 (2.04–5.07)	880
40–49	3.28 (1.66–5.77)	834
50–59	3.43 (1.77–5.95)	831
60–69	9.26 (3.03–20.42)	822
70+	3.65 (1.79–6.56)	760
Sex		
All Females	2.42 (1.45–3.78)	3916
Females 16–44 years	1.76 (0.82–3.28)	1379
All Males	3.71 (2.01–6.22)	3925
Race/ethnicity		
Non-Hispanic White	2.80 (1.15–5.63)	2477
Non-Hispanic Black	2.11 (1.21–3.40)	2170
Mexican American	0.36 (0.07–1.08)	1058
All Hispanics	1.26 (0.79–1.92)	1902
Asian	15.85 (11.85–20.56)	997

[a]Percent of weighted sample