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## Inorganic and Organic Mercury Levels in the United States National Health and Nutrition Examination Survey (NHANES) 2005-2010

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#### Authors' contributions

This work was carried out in collaboration between all authors. Author BMYC designed the study and was responsible for the data analysis. Author AJC managed the literature searches, wrote the first draft of the manuscript and performed preliminary data analysis. All authors read and approved the final manuscript.

#### Article Information

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**Original Research Article** 

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#### ABSTRACT

**Aims:** Mercury is an environmental hazard. Therefore, we studied recent trends in the blood level of organic and inorganic mercury in the United States (US).

**Methodology:** We analyzed newly available data on blood inorganic mercury levels in NHANES 2005-2010. Organic mercury level was calculated by subtracting inorganic mercury level from the total mercury level. As complex sampling was used in NHANES, appropriate weights were used to adjust for oversampling of minorities and sampling from the same location.

**Results:** There were 8364, 8161 and 8727 participants in NHANES 2005-6, 2007-8 and 2009-10 respectively. Inorganic mercury levels (geometric mean [95% confidence intervals]) were 0.31 [0.30-0.32], 0.30 [0.30-0.31], 0.28 [0.27-0.28]  $\mu$ g/L and organic mercury levels were 0.24 [0.19-0.30], 0.19 [0.14-0.25], 0.27 [0.22-0.33]  $\mu$ g/L in 2005-6, in 2007-8, in 2009-10, respectively. Inorganic mercury levels showed a significant decreasing trend (*P*<.05). Organic mercury levels were significantly lower in participants aged <20 compared to those ≥20 years. The adjusted proportion (mean±SE) of participants with a total mercury level ≥5.8 $\mu$ g/L was 3.0±0.2%, 3.5±0.6%,



and 4.0±0.4% (*P*<.05) in NHANES 2005-6, 2007-8, and 2009-10, respectively. **Conclusions:** Inorganic mercury level has been decreasing during the study period. Organic mercury level was lower in 2007-2008 but increased in 2009-10. The significant increase in organic mercury level in the US general population in 2009-10 is of concern, suggesting that continual monitoring of mercury levels is needed.

Keywords: Mercury; methylmercury; United States National Health and Nutrition Examination Survey; NHANES.

#### 1. INTRODUCTION

Mercury is naturally found in the environment, including the soil, rivers, lakes and the sea [1]. In the environment, mercury exists in three forms: organic mercury, inorganic mercury and elemental mercury. Bacteria convert elemental mercury to organic mercury, mainly in the form of methylmercury, which binds to proteins and accumulates in the food chain. Therefore, mercury concentration is highest in large fish [2,3]. Unlike elemental mercury, methylmercury is absorbed in the gut. It can also cross membranes including the blood-brain barrier. It is a neurotoxicant and has caused outbreaks of mass poisoning, such as in Minamata, Japan and Irag [4,5]. Elemental mercury is present in thermometers, sphygmomanometers and other instruments. Humans may be exposed to it because of accidental spillage or industrial pollution [4,5].

Everyone is exposed to mercury in the environment to some extent, but there is a large variation in exposure [6]. Previous studies showed that seafood and mercury amalgam [7] were major sources of mercury in the general population, whilst people in certain occupations or localities could be exposed to particularly high levels [8]. Children and fetuses are especially sensitive to the toxic effects [9-11]. Mercury affects mental development in children. It can cross the placenta and is also found in breast milk. Thus, national authorities such as the US Food and Drugs Administration and the Environmental Protection Agency, issue advice to pregnant women and women of childbearing age to avoid consumption of shark, swordfish and King mackerel [12]. The US Environmental Protection Agency also issues advisories related to mercury concentrations in fish [13].

Although total blood mercury level was reported to be decreasing in the US [14,15], it is not clear if the levels of organic and inorganic mercury both decreased. The levels of inorganic mercury in the National Health and Nutrition Examination Survey (NHANES) in 2005-2010 have recently become available. Therefore, we set out to analyze the trends in blood inorganic and organic mercury levels in the US population in 2005-2010.

#### 2. METHODS

#### 2.1 Study Subjects

NHANES 2005-2010 is a continuous crosssectional survey of the health and nutritional status of the civilian, non-institutionalized population in the US conducted by the National Center for Health Statistics of the Centers for Disease Control and Prevention. Data are released for each two-year cycle. Detailed methods and protocols are described on its website [16]. All participants gave informed consent and the study received approval from the Centers for Disease Control and Prevention Institutional Review Board. Participants of NHANES 2005-2010 whose mercury levels were available were included in the analysis. In NHANES 2005-2010, total and inorganic mercury levels were available. The organic mercury level was calculated by subtracting the inorganic mercury level from the total mercury level. Measurement of mercury has been described previously [17]. Briefly, total and inorganic mercury were determined in whole blood samples by the Division of Laboratory Sciences, National Center for Environmental Health of the Centers of Disease Control and Prevention, using automated cold vapor atomic absorption spectrophotometry [18]. The detection limits for total and inorganic mercury were 0.33 µg/dL and 0.4 µg/dL respectively. A mercury concentration of 1 µg/L equals 4.99 nmol/L. Stannous chloride and sodium borohydride were the reduction agents for inorganic and total mercury analysis, respectively. All blood collection materials were free of mercury contamination.

#### 2.2 Statistical Analysis

Data analysis was performed using the complex sampling function of SPSS version 22 (IBM).

Data are expressed as mean or percent (SE). Variables with a skewed distribution were logtransformed and are expressed as geometric with 95% confidence intervals. means Examination sampling weights were used to adjust for non-response bias and the oversampling of children, the elderly, blacks, and Mexican Americans [16]. Estimates with a coefficient of variation >0.3 were considered unreliable. For mercury levels below the detection limit, a value equal to the detection limit divided by 1.414 was used. Subtraction of inorganic mercury levels from total mercury levels may give rise to small negative values for organic mercury. As negative concentrations biologically implausible, are organic mercury concentrations in µg/L were transformed

to eliminate negative values:  $M' = \{M+\sqrt{(M^2+0.0004)}\}/2$ . To analyze the trends over time, multiple regression was used, in which survey year (2005-6, 2007-8, 2009-2010) was included as an independent continuous variable.

#### 3. RESULTS

In NHANES 2005-6, 2007-8 and 2009-10, there were, respectively, 10348, 10149 and 10537 participants, of whom 8364, 8161 and 8727 participants respectively had measurements of inorganic and total mercury levels. Their characteristics are shown in Table 1. Table 2 shows the total, inorganic and adjusted organic

Table 1. Characteristics of participants with mercury	measurements in NHANES 2005-2010
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Please insert row title	NHANES 2005-6	NHANES 2007-8	NHANES 2009-10
Number of participants	8364	8161	8727
Male:female	4074:4290	4092:4069	4334:4393
Age ± SE (years)	38.0±0.8	38.7±0.4	38.8±0.5
Number of participants aged <20 years	3862	2805	2975
Number of pregnant participants	351	50	64
Race/Ethnicity			
Mexican American	2223	1684	1944
Other hispanic	277	966	944
Non-hispanic white	3295	3421	3738
Non-hispanic black	2181	1729	1580
Other	388	361	521
Adjusted proportion of participants whose annual household income is under \$20,000	14.8%	15.6%	14.5%

# Table 2. Geometric means and 95% confidence intervals of total, inorganic and organic blood mercury concentrations (µg/L) in NHANES 2005-2010

Blood mercury concentration (µg/L)	NHANES 2005-6	NHANES 2007-8	NHANES 2009-10
All subjects			
Ν	8364	8161	8727
Total	0.87 (0.79-0.95)	0.78 (0.70-0.88)	0.87 (0.79-0.94)
Inorganic	0.31 (0.30-0.32)	0.30 (0.30-0.31)	0.28 (0.27-0.28)
Organic	0.24 (0.19-0.30)	0.19 (0.14-0.25)	0.27 (0.22-0.33)
Age <20 years			
N	3862	2805	2975
Total	0.47 (0.43-0.51)	0.42 (0.39-0.45)	0.46 (0.42-0.50)
Inorganic	0.28 (0.28-0.29)	0.27 (0.27-0.27)	0.26 (0.26-0.27)
Organic	0.05 (0.04-0.06)	0.04 (0.03-0.05)	0.05 (0.04-0.07)
Pregnant women	· · · ·	, , , , , , , , , , , , , , , , , , ,	
N	351	50	64
Total	0.70 (0.60-0.81)	0.74 (0.58-0.96)	0.75 (0.59-0.96)
Inorganic	0.32 (0.29-0.37)	0.29 (0.26-0.33)	0.27 (0.26-0.29)
Organic	0.13 (0.07-0.23)	0.20 (0.11-0.35)	0.25 (0.14-0.47)

blood mercury concentrations in NHANES 2005-2010. Inorganic mercury levels (geometric mean [95% confidence intervals]) were 0.31 [0.30-0.32], 0.30 [0.30-0.31], 0.28 [0.27-0.28] µg/L and organic mercury levels were 0.24 [0.19-0.30], 0.19 [0.14-0.25], 0.27 [0.22-0.33] µg/L in 2005-6, in 2007-8, in 2009-10, respectively (Fig. 1A). Inorganic mercury levels showed a significant

decreasing trend (P<.05). Organic mercury level decreased non-significantly in 2007-2008 but increased significantly in 2009-10. Among pregnant women, inorganic mercury level also showed a significant decreasing trend (P<.05), while there was a significant increasing trend (P<.05) in organic mercury level over the period 2005-200 (Table 2).

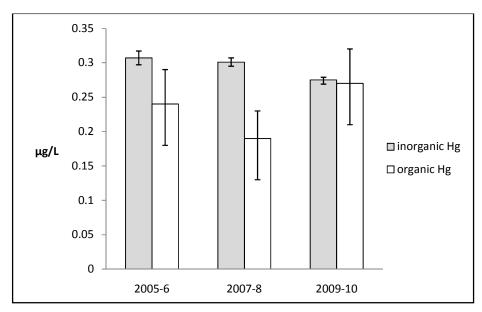
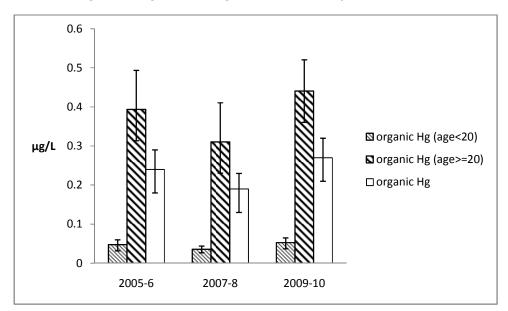


Fig. 1A. Inorganic and organic blood mercury concentration







There were no upper and lower age limits for participation in NHANES as it aimed to be representative of the US population. Organic mercury levels were significantly lower in participants aged <20 compared to those  $\geq$ 20 years (Fig. 1B). The adjusted proportion (mean±SE) of participants with a total mercury level  $\geq$ 5.8µg/L was 3.0±0.2%, 3.5±0.6%, and 4.0±0.4% (*P*<.05) in NHANES 2005-6, 2007-8, and 2009-10, respectively.

#### 4. DISCUSSION

The NHANES sample population is representative of the US population and is arguably the largest database on blood mercury levels in the general population at present. The recurrent survey of the US population in NHANES using standardized protocols allows the survey of trends in mercury levels.

The analysis of data from NHANES 1999-2000 by Schober et al. [17] showed a relationship between dietary consumption of fish and blood mercury concentration. In subsequent years, there were significant falls in blood mercury concentration in US women of reproductive age, in pregnant women and in children [14,15]. Publicity and action at the national level undertaken by multiple governmental departments helped to bring about significant changes in the population [19-21]. Such interventions appear to be effective in reducing the exposure to mercury.

In the period 2005-2010, the proportion of participants with a total mercury level  $\geq$  5.8µg/L increased significantly, illustrating that a small shift in the population mean can result in large changes in prevalence at the extreme ends of the distribution. As the inorganic mercury level has been decreasing during this period, any increase in total mercury level was likely to be due to increase in organic mercury level. Indeed, we found an increase in the blood concentration of organic mercury in 2009-2010. This might only be a temporary anomaly, but if it were the start of an upward trend, or even if the trend of falling blood mercury concentration has become static, there is a case for concern.

The trend of decreasing inorganic mercury and increasing organic mercury was clearer in the subgroup of pregnant women. Although the small sample size of this subgroup prevents firm conclusions to be drawn, this is a subgroup that is more vulnerable to the effects of mercury and may therefore need more careful monitoring.

Although fish is a major source of mercury, particularly organic mercury, in the blood [22, 23], eating fish is recommended nowadays for the prevention of coronary heart disease [24]. It is important to reconcile the health effects of eating fish with the exposure to mercury [25]. Eating large fish higher up on the food chain may be more hazardous than smaller fish. Shellfish is usually relatively low in methylmercury and thus its contribution to blood mercury concentration is less [26]. Unfortunately, it is easier to reduce mercury intake by eating less fish than to modify the types of fish eaten [27]. Therefore, more research is needed to provide a sound scientific basis for public health advice regarding fish in the diet.

There are limitations to the mercury data in NHANES. Although the survey populations were large, only a few percent of participants had elevated mercury levels (≥5.8µg/L) [17] that could be regarded as potentially hazardous. Blood samples were taken once in each participant, so they might reflect accurately the average blood mercury concentration in that individual. Also, the study population is not a cohort and different individuals are randomly selected in successive surveys, so that the trends in individual participants could not be discerned. The source of the mercury in the blood of the participants could not be identified because even when food intake information was available, the mercury concentration in such foods had not been measured. A recent study in England highlighted the mercury contained in wine and herbal tea [28]. Calcium supplements, depending on the source of the calcium, can contain mercury [29].

#### 5. CONCLUSION

In conclusion, inorganic mercury level has been decreasing during the study period. Organic mercury level was lower in 2007-2008 but increased in 2009-10. The increase in organic mercury level in the US general population in 2009-10 is of concern, suggesting that continual monitoring of mercury levels is needed.

#### **COMPETING INTERESTS**

Authors have declared that no competing interests exist.

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