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A national statistical survey assessment of mercury concentrations in fillets of fish collected in the U.S. EPA national rivers and streams assessment of the continental USA

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HIGHLIGHTS

- Hg fish tissue results from 541 U.S. sites on =>5th order streams in 2008–2009.
- Site selection was probabilistic and nationally-representative.
- 25.4% (137 sites, 13 071 river miles, (21 036 km)) > 300 ug kg^{-1} HH and WV thresholds.
- No difference observed between results from urban vs. non-urban sites or among three eco-regions.
- Most exceedances in top predator species.

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ABSTRACT

The U.S. EPA conducted a national statistical survey of fish fillet tissue with a sample size of 541 sites on boatable rivers =>5th order in 2008–2009. This is the first such study of mercury (Hg) in fish tissue from river sites focused on potential impacts to human health from fish consumption to also address wildlife impacts. Sample sites were identified as being urban or non-urban. All sample mercury concentrations were above the 3.33 ug kg^{-1} (ppb) quantitation limit, and an estimated 25.4% (±4.4%) of the 51 663 river miles assessed exceeded the U.S. EPA 300 ug kg^{-1} fish-tissue based water quality criterion for mercury, representing 13 144 ± 181.8 river miles. Estimates of river miles exceeding comparable aquatic life thresholds (translated from fillet concentrations to whole fish equivalents) in avian species were similar to the number of river miles exceeding the human health threshold, whereas some mammalian species were more at risk than human from lower mercury concentrations. A comparison of means from the non-urban and urban data and among three ecoregions did not indicate a statistically significant difference in fish tissue Hg concentrations at $p < 0.05$.

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1. Introduction

Mercury (Hg) is a metal that is persistent, bioaccumulative, and toxic in the environment. It is a constituent of minerals in rocks and soil that enters the atmosphere by the natural degassing of the earth's crust and recycling by volatilization of atmospherically deposited mercury, as well as being mobilized to the environment from anthropogenic sources. Human activities have enriched the atmospheric content of mercury by a factor of 7.5 times compared to levels predating emissions from most human activities (Amos et al., 2013). These activities include mining (including gold extraction), manufacturing, Portland cement production, medical waste incineration, and most prominently, the combustion of fossil fuels, especially coal (UNEP, 2008). As of 2005, however, artisan and small scale gold mining were second only to fossil fuel combustion as sources of Hg emissions to the atmosphere (Pacyna et al., 2010). When Hg is deposited in water, a large portion is converted, by chemical and biochemical processes either in sediments or in the water column, from inorganic Hg to toxic methylmercury (MeHg) that bioaccumulates in fish muscle (Sunderland et al., 2009). Despite very effective efforts to reduce emissions of Hg in many countries, including the U.S., Hg emissions from Asia continue to

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increase (Wilson et al., 2010) with potential implications for increased human exposure in the U.S. and elsewhere (Sunderland et al., 2009; Driscoll et al., 2013). Future levels of Hg emissions will depend on the degree of implementation of measures to remove Hg from fossil fuel combustion emissions in the context of rising demand for electricity (Streets et al., 2009).

Research has shown that exposure to Hg and its compounds through the consumption of fish containing Hg can cause a range of toxic effects in humans, as well as in piscivorous mammalian and avian wildlife (U.S. EPA, 1997). At doses that result from moderate rates of maternal fish consumption, exposure of fetuses to Hg *in utero* can later manifest in children as deficits in subtle neurological end points such as lowered IQ, decrements in motor function, attention, and visuospatial performance (NAS, 2000). Evidence of early childhood effects at low levels of prenatal exposure is clear, and other health effects are likely, including cardiovascular effects and, to a lesser extent, immune system suppression (Karagas et al., 2012). Recent analyses also suggest an association between toenail Hg levels in young adults and the incidence of diabetes in later life (He et al., 2013). The thresholds for neurodevelopmental effects of mercury exposure discussed in this manuscript are based on U.S. EPA reference dose of 0.1 ug kg^{-1} of body weight/day for exposure to Hg (U.S. EPA, 2001b) relative to fish tissue concentrations detected in samples collected by the U.S. EPA. A reference dose (RfD) is an estimate of a daily exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime.

Wildlife are exposed to Hg and MeHg from a variety of environmental sources, mine tailings, industrial effluents, agricultural drain water, impoundments and atmospheric deposition from the electric power generation industry. At the higher levels of the food chain, piscivorous birds and mammals are among the highest risk receptors of MeHg contamination (Wolfe et al., 1998). Toxicity of Hg to piscivorous birds and mammals includes damage to the nervous, excretory, and reproductive systems. (Wolfe et al., 1998). In 1997, EPA identified wet deposition as the primary mechanism by which atmospheric Hg is transported to surface waters and land, although dry deposition may also contribute substantially (U.S. EPA, 1997). More recently, Driscoll et al. (2013) provide a full description of Hg sources, transport and deposition mechanisms, both human health and ecological effects, and cite atmospheric deposition as the primary mechanism of redistribution of mercury.

Large-scale studies of contaminants in fish tissue have found widespread evidence of Hg contamination. In a survey of lakes in the northeastern United States in 1992–1994, the U.S. EPA's Environmental Monitoring and Assessment Program (EMAP) analyzed whole fish composite samples for inorganic and organic contaminants, including Hg. It was found that 26% of northeastern lakes contained fish with Hg levels that exceeded the critical value for humans (200 ug kg^{-1} at that time), 54% of lakes exceeded wildlife critical values for piscivorous mammals (100 ug kg^{-1}), while 98% exceeded wildlife critical values for birds (20 ug kg^{-1}) (Yeardley et al., 1998). During the period 1993–1994, an EMAP survey of fish in the Mid-Atlantic Region found that small fish tissue concentrations exceeded the kingfisher Wildlife Value (30 ug kg^{-1}) over 72% of the stream length. Wildlife Values (WVs,) have been defined as the concentration in fish at which chronic effects to piscivorous wildlife occur (Lazorchak et al., 2003). In 2004 and 2005, the EMAP Great Rivers Assessment measured Hg concentrations in whole fish from the Upper Mississippi, Missouri, and Ohio Rivers to characterize the extent and magnitude of Hg contamination and to identify environmental factors influencing Hg accumulation (Walters et al., 2010). Across all three large river systems, Hg levels exceeded the WV for belted kingfisher in 33–75% of river length and exceeded the human health criteria in 1–7% of river length.

More recently, the U.S. EPA conducted the National Lake Fish Tissue Study (Stahl et al., 2008), which employed a national-scale statistical survey design and targeted human exposure to Hg by focusing on the fillet, the edible portion of collected species. This national lakes study included 486 composite predator fillet samples collected from 500 randomly-selected lakes during 2000–2003. In this nationally representative study, fish tissue from 48.9% of the lake population assessed exceeded the U.S. EPA's 300 ug kg^{-1} human health fish tissue-based water quality criteria (WQC) for Hg (U.S. EPA, 2001a).

The U.S. Geological Survey (USGS) has conducted extensive studies of national and regional fish-tissue contaminant data by combining results of several separate targeted studies conducted over time, analyzing the data and characterizing contaminant levels on the basis of region and watershed type, including propensity for methylation and fish characteristics (Schmitt, 2002; Scudder et al., 2009; Chalmers et al., 2011). These studies have led to identification of patterns and mechanisms of Hg accumulation in fish including the propensity for methylation to occur in watersheds with abundant wetlands (Brigham et al., 2009; Scudder et al., 2009). USGS continues to emphasize locally-focused studies which are national in their distribution (Brigham et al., 2014; Feaster et al., 2014; Chalmers et al., 2014) and explore causative mechanisms for the observed variations in Hg concentrations in a variety of media and settings.

The present work reports the results of a 2008–2009 National Rivers and Streams Assessment (NRSA) survey of rivers in the 48 conterminous United States to investigate Hg concentrations in composited fillet samples (Fig. 1). The survey is nationally-representative of the class of waters (5th order and greater rivers that are boatable) from which the sample of 541 sites was drawn. The fish tissue sampling efforts of this study are focused on providing information for assessments of human health impacts of fish consumption. Results were also used to estimate contaminant exposures to wildlife and aquatic species that consume fish and to compare urban and non-urban sub-groups of sites as well as sites located in each of three National Aquatic Resource Assessment (NARS) ecoregions: West and Mountains (WMTS), Plains and Lowlands (PLNLOW), and Eastern Highlands (EHIGH) (Fig. 1). The design of this study produces assessment results nationally, for three regions, and by urban and non-urban subgroups and relate those results to both human health and wildlife effects. The statistical survey design requires estimates for the assessments to incorporate weights that reflect the stratification and unequal probability of selection. This ensures that the inference (results) apply to all boatable, 5th order and greater rivers in the conterminous states. Results from this study may differ from those employing other non-survey design methods for selection of sites (Peterson et al., 1998).

2. Materials and methods

2.1. NRSA design and site selection

The NRSA included 1924 sites within the conterminous United States that were sampled for a range of environmental indicators, including concentrations of toxic chemicals in fish tissue (U.S. EPA, 2013). Fish tissue samples were collected at a subset of 541 sites that met the three criteria for the river population of interest for the fish tissue contaminant study: (1) boatable U.S. rivers of 5th order and greater (Strahler, 1957); (2) having a permanent fish population; and (3) flowing water during the study period (including the Great Rivers and run-of-the-river ponds, but excluding portions of tidal rivers up to head of salt, and reservoirs). The sample frame was derived from the National Hydrography Dataset (NHD)