Mercury in the National Parks

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Unpredictable.

Or is it?

One thing is certain: Even for trained researchers, predicting mercury's behavior in the environment is challenging. Fundamentally it is one of 98 naturally occurring elements, with natural sources, such as volcanoes, and concentrated ore deposits, such as cinnabar. Yet there are also human-caused sources, such as emissions from both coal-burning power plants and mining operations for gold and silver. There are elemental forms, inorganic or organic forms, reactive and unreactive species. Mercury is emitted, then deposited, then re-emitted—thus earning its mercurial reputation. Most importantly, however, it is ultimately transferred into food chains through processes fueled by tiny microscopic creatures: bacteria.

Mercury is ephemeral, but enduring and pervasive. It poses serious risks to environmental and human health. So, can we predict exactly where? What areas are most at risk?

Mercury is a highly toxic pollutant that has been both extensively utilized and widely distributed across the globe by humankind's activities. Because of its ubiquity and considerable toxicity, the US Geological Survey (USGS) has conducted several decades of research to unravel mercury's complex and seemingly mysterious behavior. One product of this work has been the collection of an unprecedented amount of information to better understand the story of mercury in the national parks, and other areas.

Once mercury enters an aquatic or terrestrial food web, trouble begins to brew. Mercury can harm all forms of life; it is one of very few elements for which there is no known biochemical or biophysical need. In wildlife, high mercury concentrations can result in altered behavior and reduced foraging efficiency, reproductive success, and even survival. Exposure to high levels of mercury in humans may cause damage to the brain, kidneys, and the developing fetus. Pregnant women and young children are particularly sensitive to mercury exposure.

Mercury contamination is evident everywhere. More than 16 million lake acres and 1 million river miles are under fish consumption advisories due to mercury in the United States, and 81% of all fish consumption advisories issued by the US Environmental Protection Agency are because of mercury contamination (USEPA 2013).

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Mercury contamination can be a substantial water quality issue for national parks in even the most remote, relatively pristine locations far removed from point-source emissions, because many of the landscape characteristics of aquatic ecosystems in protected lands—abundant wetlands, full forest canopies, and naturally fluctuating water levels—are associated with the production of the most toxic and bioaccumulative form of mercury: methylmercury. Undeveloped landscapes with dense wetlands and forests generally yield highly favorable settings for converting inorganic mercury, which is relatively unavailable for biological uptake, to methylmercury. Wetlands are commonly home to anaerobic sediments, which in turn serve as ideal landscape settings to host sulfate-reducing bacteria, commonly implicated as the primary methylating agent in the environment. Tall and dense forested canopies act as air filters, collecting and concentrating mercury from the air onto foliage, which later drops to the ground in rain, snow, or litterfall. Mercury then collects in the soil and eventually moves into streams and lakes. Drying and rewetting cycles resulting from seasonal fluctuations of the hydrocycle also promote mercury methylation.

The importance of the methylation process on how mercury manifests itself as a serious environmental concern cannot be overstated. In fact, were it not for mercury's relatively uncommon susceptibility to become methylated, it would certainly be of little or no consequence to living organisms except under the most extreme of contamination conditions.

Further, many national park ecosystems are largely intact. With this, there are complex food webs and long food chains that also promote high concentrations of mercury at top levels. Predatory fish, which commonly include those sought after for sport and human consumption, are more likely to have elevated mercury concentrations due to biomagnification within the food web. Older fish are particularly at risk given the increased susceptibility for contaminant bioaccumulation over their longer life spans. Other factors that may further exacerbate mercury accumulation and heighten ecosystem sensitivity, specifically in high-elevation and high-latitude areas, include the shorter growing season and slower growth of aquatic species.

A major concern about mercury in national parks is the fact that much of the mercury found in these remote areas is largely the result of air pollution from outside the parks. Although there are natural sources of mercury such as volcanoes and mercury-enriched geogenic deposits, much of the mercury that affects parks comes from burning fossil fuels, like coal, in power plants. Waste incinerators, industrial boilers, cement manufacturing, and mining operations are other human-related mercury emission sources. Once emitted to the air, mercury can travel great distances before it returns to the earth with rain, snow, dust, and fog, or via passive uptake by photosynthetically active plants. Upon conversion to methylmercury, a transformation that easily occurs in the ideal environmental conditions provided by many national parks, mercury both bioaccumulates (builds up) and biomagnifies (increases in concentration with each successive step up the food chain) in organisms (Figure 1). Organisms that live at the top of food chain (e.g., bald eagles, common loons, bears, lake trout, humans, etc.) are most at risk for exposure to high levels of mercury through fish consumption.

Mercury threatens the very resources that the National Park Service (NPS) is mandated to protect. The NPS Organic Act (16 USC 1 [1997]) directs the agency to promote and reg-

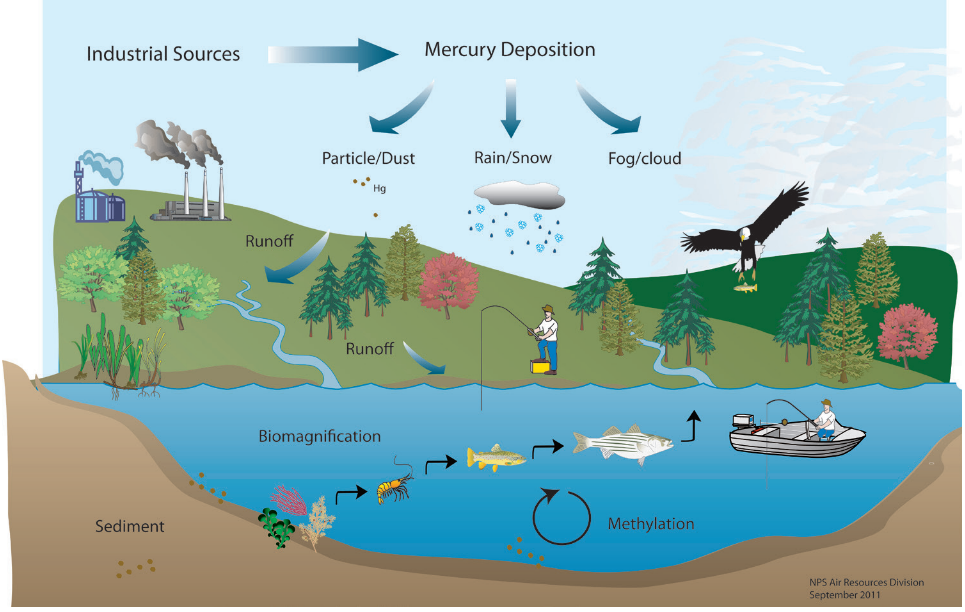

Figure 1. Sources and paths of mercury in the environment.

ulate the use of the national parks, whose "purpose is to conserve the scenery and the natural and historic objects and the wild life therein, and to provide for the enjoyment of the same … as will leave them unimpaired for the enjoyment of future generations." Additionally, under the Clean Air Act (42 USC 7470 [2]), NPS is mandated to "preserve, protect and enhance the air quality in national parks … and other areas of special national or regional natural, recreational, scenic or historic value."

NPS and USGS are mutually interested in studies concerning the effects of mercury, as well as other air pollutants, on natural resources. These studies tie into National Park Service goals to lead America in preserving and restoring treasured resources, demonstrate environmental leadership, offer superior recreational experiences, foster exceptional learning opportunities that connect people to parks, and be managed with excellence. The focus of USGS research on mercury in the national parks centers on ecosystem and human health risk, both predicted and actual. The projects highlighted herein specifically outline work contributed by the USGS Mercury Research Lab (MRL) and USGS Forest and Rangeland Ecosystem Science Center (FRESC), in collaboration with the NPS Air Resources Division (ARD), across parks, regions, and networks (Figure 2).

Where we have been

Everglades National Park (FL): Investigating the mercury cycle. Due to an increased public concern for wildlife and human health resulting from mercury toxicity in the Florida Everglades, USGS MRL initiated the Aquatic Cycling of Mercury in the Everglades (ACME) project in 1995. The overall objective of the ACME project was to conduct intensive, pro-

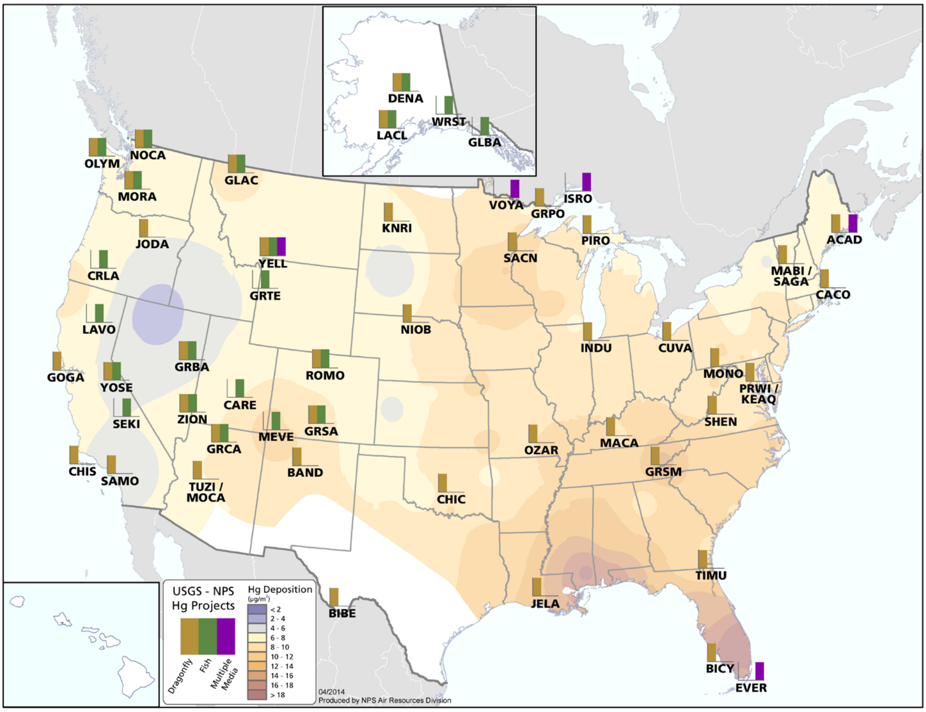

Figure 2. Recent USGS–NPS collaborative studies on mercury in the national parks: fish (2008–2012), dragonfly larvae (2011–2015), and multiple media (1995–present; intensive research on mercury dynamics and distribution at hotspot parks). The background layer illustrates 2012 mercury wet deposition estimates. Data from National Atmospheric Program Mercury Deposition Network; http://nadp.sws.uiuc.edu/mdn/.

cess-oriented research that focuses on the primary mercury sources, cycling pathways, and bioaccumulation in the Everglades, and to provide an anticipatory understanding of how the ecosystem restoration program may affect mercury in the future. ACME made several key contributions toward improving understanding of the mercury cycle in the Everglades, including basic information on the relationship between methylmercury production and biogeochemical variables, such as nutrients, sulfate, sulfide, and dissolved organic matter (Krabbenhoft et al. 2000). Key findings included: (1) sulfate loading to the Everglades increases microbial sulfate reduction in soils and ultimately enhances methylmercury production and bioaccumulation in some parts of the Everglades ecosystem; and (2) the large gradients in sulfur, methylmercury and dissolved organic matter across the Greater Everglades Ecosystem are driven in part by agricultural drainage and water management practices. Dvonch et al. (1995) attributed elevated mercury concentrations in several Everglades rain event samples to local sources. At Everglades National Park, elevated concentrations of mercury in invertebrates, frogs, fish, wading birds, pythons, alligators, and Florida panthers have been documented; some at levels known to cause neurologic and reproductive impairment (NPS 2011a).

Yellowstone National Park (MT/WY): Studying the source, pathways, and fate of mercury. Yellowstone National Park is saturated with spectacular geothermal features. These geysers and hot springs also happen to be natural sources of mercury. Given that, many scientists speculated that the park might be one of the largest natural mercury emission sources on the planet. USGS scientists tested that assumption and set up the Mobile Atmospheric Mercury Laboratory to assess the relative importance of sources from within and outside Yellowstone National Park (Hall et al. 2006). Results indicate that Yellowstone is not as large a source of mercury to the atmosphere as was once thought. In fact, scientists found that wildfires burning near or in the park released appreciably more mercury to the atmosphere than the park's geothermal sources. USGS also studied the dynamics in the park's thermal features, using the ratio of naturally occurring mercury isotopes present in the geothermal waters to trace or identify sources of mercury to the environment. Although mercury occurs naturally in hot springs, its most toxic form, methylmercury, appears to be entering the food chain largely by accumulating in slimy microbial mats (King et al. 2006; Boyd et al. 2009; Sherman et al. 2009). The results of these studies have increased our understanding of the origins, transport, and fate of mercury from Yellowstone's geothermal areas. In addition, new insights have been gained on the relative contributions of natural versus human mercury sources and local versus regional mercury sources.

Where we are now

NPS Inventory & Monitoring networks: Predicting mercury risk. To reiterate: the toxicological risk of mercury contamination is strongly tied to factors that facilitate the conversion of inorganic mercury to the more toxic and bioavailable form, methylmercury. Methylmercury production is driven by multiple biogeochemical factors such as organic carbon availability and quality, redox fluctuations, inorganic mercury speciation and adsorption, and sulfur chemistry (Figure 3). These factors influence both the activity of mercury-methylating microbial groups, as well as the availability of inorganic mercury. However, these factors and their relative importance can vary in time and across the landscape, making predictions of risk difficult. Understanding how these local drivers influence mercury cycling across ecosystem types is a critical component to developing robust predictions of potential mercury impacts to aquatic ecosystems in national parks and other sensitive areas.

USGS research summarized in Lubick (2009) and Wentz et al. (2014) showed that mercury deposition, primarily atmospheric deposition from industrial emissions, is just one factor that influences the levels of bioavailable methylmercury. However, "variations in ecosystem properties that govern methylmercury production in an ecosystem are probably much more important [in determining which ecosystems have fish with high methylmercury] than the variation of mercury deposition across the country" (Lubick 2009, citing M.E. Brigham). That said, while variability in ecological conditions tend to drive spatial trends in methylmercury abundance, swings in deposition drive the majority of temporal methylmercury variability.

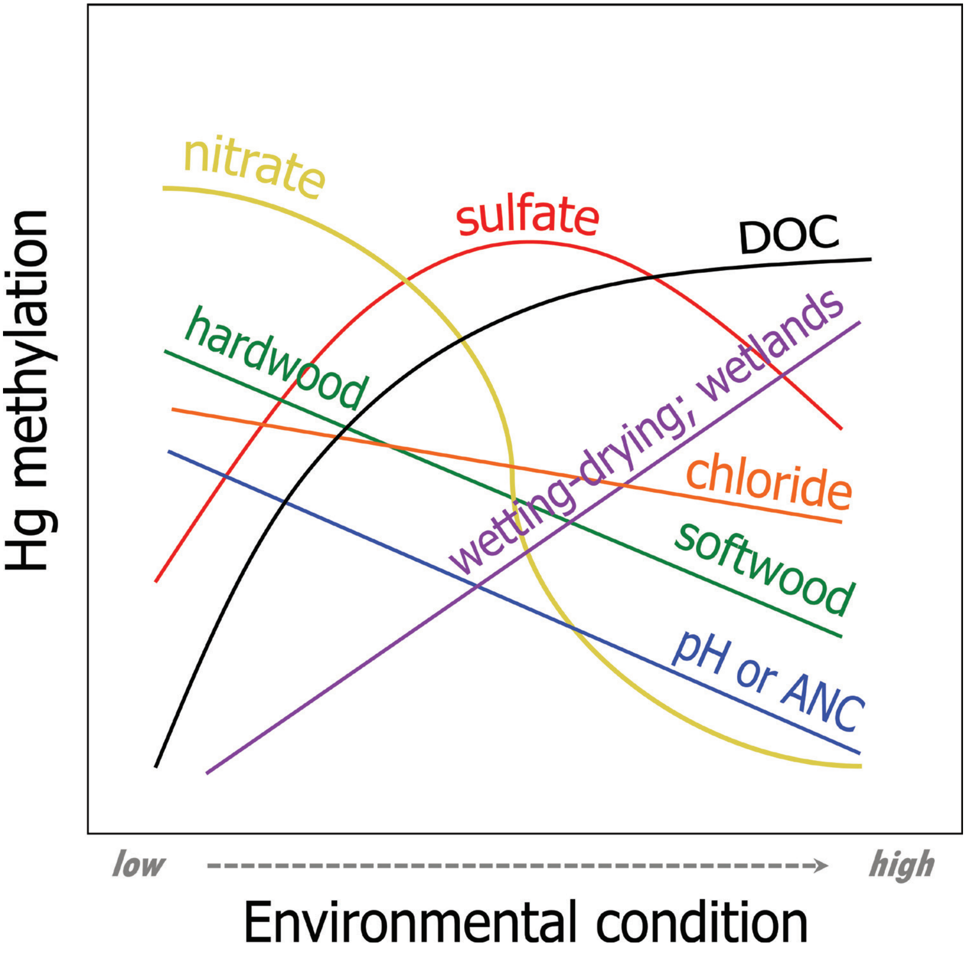

Figure 3. Emerging relationships between mercury (Hg) methylation and environmental conditions or gradients. Water chemistry and site characteristics measured in USGS studies in national parks allow for testing of these responses. Figure adapted by Eagles-Smith et al. 2013.

Wetland abundance within a watershed, carbon levels within soils, dissolved organic carbon (DOC) in surface waters, suspended sediment concentrations in streams, and streamflow have all been shown to be key factors in determining the levels of methylmercury in waters, and mercury concentrations in aquatic fauna, within a given watershed (Brigham et al. 2009; Chasar et al. 2009; Marvin-DiPasquale et al. 2009).

Although our understanding of mercury cycling and distribution is well understood at national parks such as Everglades and Yellowstone, these sites are the exceptions, not the rule. Comparatively few parks nationwide have received substantial study of mercury cycling and ecosystem risk. In order to assess potential risk of mercury contamination at other national parks, USGS MRL is working in collaboration with NPS ARD to refine a sensitivity model for predicting methylmercury concentrations in surface waters of the 270 parks in the Inventory & Monitoring networks. Based upon common water chemistry data (i.e., DOC, sulfate, pH) and landcover criteria (wetland coverage), the model predicts aqueous methylmercury concentrations and identifies park units that are likely to have conditions most conducive to methylmercury production (Krabbenhoft et al. 2011).

An associated NPS-mapping-specific application for national parks is currently under development. The tool will allow users to select a particular park, drill down to the mega-watershed level (HUCs; hydrologic unit codes) within each park, and assess the predictive variables and the estimated methylmercury concentration. For each respective layer, the parameters were classified by quintiles, relative to all other NPS units included in the model: the top two upper quintiles (4th and 5th) represent the highest risk, the middle two quintiles (2nd and 3rd) represent moderate risk, and the lower quintile (1st) the lowest risk. For example, watersheds that contain the highest percentage of wetland land cover (the 5th quintile) fall into the high-risk category.

The methylmercury risk mapper provides direction for *in situ* studies of biota in national parks, providing resource managers with insight on potential hotspots, or areas at particular risk for elevated mercury levels. The interactive tool was made publicly available in the summer of 2014.

Western national parks: Assessing fish and ecosystem health. Fish tissue from freshwater environments represents an important component for evaluating mercury cycling, bioaccumulation, and ecological risk, including the potential risk to humans consuming fish. Fish are the fulcrum on which the story of mercury pivots. The public identifies with fish; people eat them. Fish provide recreational enjoyment through sport fishing; they also offer spiritual and cultural benefits, particularly for tribes who depend on them to sustain life. The dietary benefits of consuming fish include improved cardiac health from increased omega-3 fatty acid consumption or potential reduced intake of unhealthy fats due to food substitutions. The risk of elevated mercury in fish is not only a concern for people who eat fish, but for land managers who manage other fish-eating organisms, such as birds and mammals, and the fish themselves.

While previous studies identified regional patterns in mercury deposition (Krabbenhoft et al. 2002) and elevated mercury concentrations in some fish from remote, high-elevation water bodies in a few western national parks (Schwindt et al. 2008), there was a lack of a systematic characterization and assessment of mercury risk across remote areas of the West. In addition, for many years there was an assumption among researchers that generally drier western US areas experience less mercury loading due to lower rainfall amounts. However, more recent information has revealed that eastern versus western atmospheric loading differences are largely minimized by a better appreciation for the importance of mercury loading from dry deposition (e.g., dust), and thus there is a better appreciation for more mutually susceptibility along longitudinal gradients.

Given the significant role that atmospheric mercury deposition plays in these areas, USGS FRESC worked in collaboration with NPS ARD to study mercury in freshwater fish across 21 western national parks, from Alaska to Arizona. Between 2008 and 2012, NPS resource managers collected more than 1,400 individual fish from 86 lakes and rivers extending over a distance of 4,000 km. USGS scientists measured mercury concentrations in fish muscle tissue. Sixteen fish species were sampled, with a focus on commonly consumed sport fish found across the study area such as brook, rainbow, cutthroat, and lake trout. Smaller prey fish consumed by birds and wildlife were also sampled. The primary objectives included: (1) comparing fish mercury concentrations between parks and among sites within parks, (2) determining at what spatial scale variation in fish mercury concentrations is attributed, and (3) evaluating fish mercury concentrations in parks with respect to a range of wildlife and human health benchmarks (Eagles-Smith et al. 2014).

Findings indicate that mercury levels varied greatly, both from park to park and among sites within each park (Figure 4). Although fish mercury concentrations were elevated in some sites, the majority of fish across the region had concentrations that were below most benchmarks associated with impaired health of fish, wildlife, and humans. In most parks, mercury concentrations in fish were moderate to low in comparison with similar fish species from other locations in the western states. Mercury concentrations were below the US Environmental Protection Agency's (USEPA's) fish tissue criterion for safe human consumption in 96% of the sport fish sampled. There were, however, particular areas identified that had elevated fish mercury concentrations, including levels that exceed human consumption and/or wildlife health benchmarks. The average concentration of mercury in sport fish from Lake Clark and Wrangell–St. Elias (AK) national parks exceeded USEPA's human health criterion. Mercury levels in individual fish at Lassen Volcanic (CA), Mount Rainier (WA), Rocky Mountain (CO), Yellowstone (WY), and Yosemite (CA) national parks also exceeded the human health criterion (Eagles-Smith et al. 2014).

Mercury concentrations in individual fish also exceeded the most conservative fish toxicity benchmark at Capitol Reef (UT), Lake Clark (AK), Lassen Volcanic (CA), Mount Rainier (WA), Rocky Mountain (CO), Wrangell–St. Elias (AK), Yosemite (CA), and Zion (UT) national parks, and levels in some fish exceeded the most sensitive health threshold for fish-eating birds at all parks except Crater Lake (OR), Denali (AK), Grand Teton (WY), Great Basin (NV), Great Sand Dunes (CO), Mesa Verde (CO), and Sequoia–Kings Canyon (CA) national parks. Other national parks in this study were Glacier (MT), Glacier Bay (AK), Grand Canyon (AZ), North Cascades (WA), Olympic (WA), and Yellowstone (WY) (Eagles-Smith et al. 2014).

Where we are going: Evaluating mercury risk using dragonfly larvae

Given the complexities associated with local drivers of mercury cycling and the development of robust predictions of potential impacts to aquatic ecosystems in national parks, the application of biosentinel organisms has emerged as an important monitoring and research tool. Biosentinels can provide a better-integrated indicator of mercury variation among locations than water, and are more appropriate proxies for human and wildlife risk (Knights et al. 2005; Simonin et al. 2008; Sackett et al. 2009). Biosentinels are similar to the "canary in the coal mine," a surrogate for environmental health, that can be used to detect the potential risk

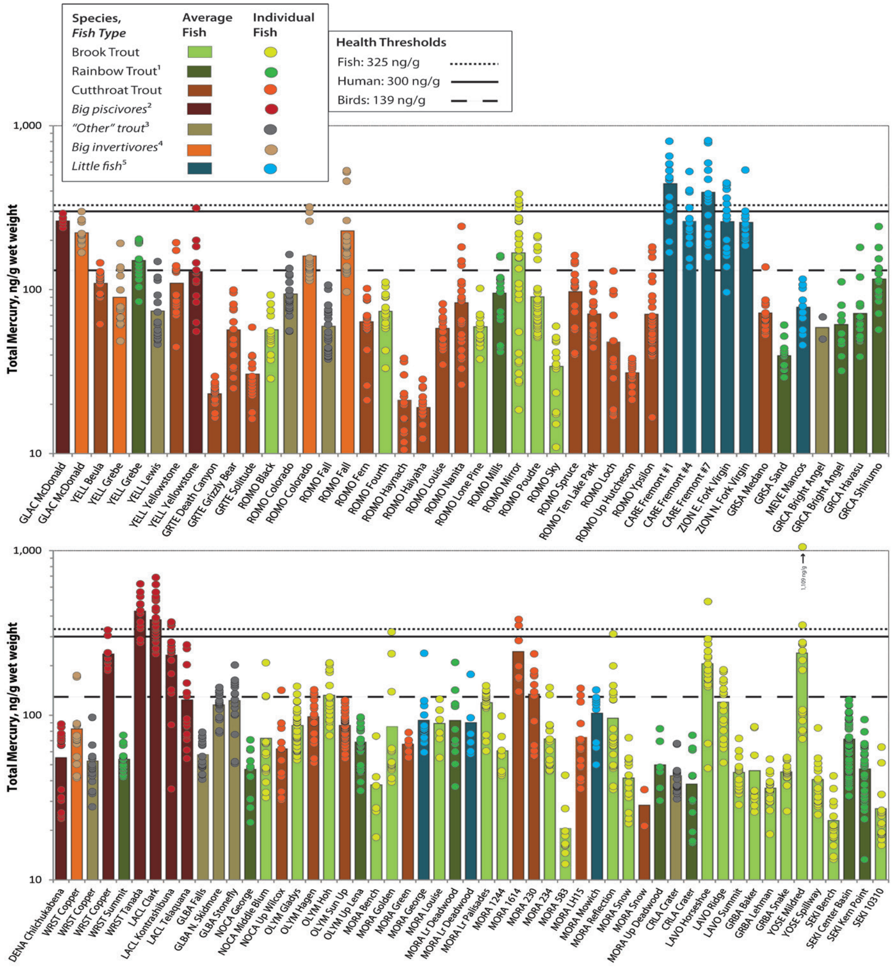

Figure 4. Total mercury in average fish muscle tissue (bars) and individual fish (circles), by species in wet weight (ww), compared with health benchmarks established for fish toxicity (325 ng/g ww), highly sensitive fish-eating birds (139 ng/g ww), and human consumers (300 ng/g ww; USEPA criterion). Data are plotted on a log10 scale. Figure from NPS 2014.

to humans and wildlife by providing advance warning of a danger. Effective biosentinels of ecosystem risk for mercury are widespread and ubiquitous, relatively easy to sample, linked to key energetic processes within ecosystems, ecologically well studied, and responsive to localized changes in methylmercury availability and cycling.

Dragonfly larvae (*Odonata: Anisoptera*) are aquatic macroinvertebrates that meet those criteria, and are being collected in at least 50 parks across the nation over five years (2011–2015) for analysis of mercury. USGS FRESC and MRL recently teamed up with NPS ARD, University of Maine, participating parks, citizen scientists, and other partners to build upon a successful pilot effort to evaluate and establish dragonfly larvae as robust biosentinels of aquatic mercury contamination in NPS units across the country (Flanagan et al. 2012; Wiener et al. 2013). This project is the first of its kind to validate a common and abundant biosentinel in national parks across the US, sample freshwaters in parks in a single coordinated study to determine mercury risk, and engage citizen scientists in the process.

Dragonfly larvae are shedding light on the risk of mercury contamination throughout the national park system. While fish are perhaps the most commonly used indicators because they occur across a wide geography and provide strong linkages to human and wildlife health, dragonfly larvae are relatively easier to collect, and represent the risk to mercury in fishless ecosystems like shallow ponds, ephemeral pools, and marshes—some of the most productive and ecologically important aquatic habitats. Preliminary results from the pilot study in dragonfly larvae indicate that mercury concentrations are greater at parks in the eastern US than those in the western US, and site differences within parks reveal that dragonfly larvae can reveal fine-scale differences in mercury risk. Related research shows that mercury in dragonfly larvae was correlated with both methylmercury in water and mercury in fish in the same water bodies (Haro et al. 2013), confirming their utility as an effective indicator of ecosystem risk.

Twenty-two national parks have participated in the project to date, from Denali (AK) and Big Cypress (FL), to Acadia (ME) and Golden Gate (CA), collecting over 700 dragonfly larvae at 50-plus sampling sites. Close to 300 citizen scientists, including students, Youth Conservation Corps members, and bioblitz participants have thus far contributed approximately 1,700 hours of volunteer time. Up to an additional 28 parks will participate in 2014–2015. Public engagement in this project directly implements the NPS Call to Action Items #7, "Next Generation Stewards," and #16, "Live and Learn," by enlightening a new generation of citizen scientists about the connection of all living things and the influence humans have upon natural systems, and how environmentally responsible decisions can protect our parks and the planet (NPS 2011b).

This project links chemical and habitat parameters with food web bioaccumulation and ecological risk. The main objectives are to: (1) use an established citizen scientist program network to collect samples for assessing variation in mercury and methylmercury in freshwaters and biosentinels across US national parks, and (2) determine how temporal variation, site characteristics, water chemistry, and biological drivers affect freshwater and biosentinel mercury accumulation. Habitat variables, developmental stages, and genus/species-specific traits of dragonfly larvae will also be considered.

Furthermore, this project contributes to the refinement and expansion of the methylmercury prediction model and mapper (Krabbenhoft et al. 2011) by providing both water chemistry predictor data (e.g., DOC, sulfate, pH) and measured total and methylmercury in surface waters for each participating park, and new data for previously unmodeled parks. This biosentinel project also provides the opportunity to compare predicted methylmercury vulnerability from the geospatial model to observed mercury in a single taxon—the dragonfly—across all participating parks. Validation data such as these, which confirm relative mercury burdens in biota, are sparse, and rarely are the same biota sampled across multiple parks or regions in a standardized way.

Where does that leave us?

NPS safeguards nearly 400 highly valued places for the protection of unique natural and cultural resources and scenic beauty. Research and monitoring efforts across the 84 million acres represented by national parks include assessments of mercury in insects, amphibians, fish, birds, water, sediment, snow, air, vegetation, and wildlife.

Variation in site-specific mercury concentrations within individual parks suggests that more intensive sampling in some parks will be required to effectively characterize mercury contamination at these locations. Future targeted research and monitoring across park habitats would help identify patterns of mercury distribution across the landscape and ultimately facilitate informed management decisions aimed at reducing the ecological risk posed by mercury contamination in sensitive ecosystems protected by NPS. Other investigations on source attribution and actual effects on park resources will further our understanding of this complex issue.

Continued coordination with other entities will build awareness of the issue of mercury contamination in the national parks. For instance, NPS and USGS FRESC are working together on developing a mercury benchmark to assess the condition for park planning processes. NPS is also working with state officials on potential fish consumption advisories, as is the NPS Office of Public Health to communicate advisories. Results are also related to USEPA efforts, including nationwide monitoring programs.

Further, the data collected herein serves as a baseline by which responses to anticipated future decreases in mercury emissions under USEPA's mercury and air toxics standards (MATS) can be assessed for effectiveness in removing mercury from food webs. The MATS final rule requires an approximate 90% reduction in mercury emissions from 1,400 of the largest coal- and oil-fired utilities by 2015. There are also implications for the international arena and global mercury treaties, and the myriad aspects of global change, which will affect the behavior and distribution of mercury worldwide (Krabbenhoft and Sunderland 2013).

Mercury is lively, complicated, and mercurial. It challenges the very mission of the national parks to leave wildlife unimpaired for future generations. Thanks to the working partnership between USGS and NPS, society is gaining a better understanding of the risk to national parks. Ultimately, NPS would like to see less contaminants in park ecosystems, especially those such as mercury where concentrations exceed thresholds for potential negative health effects on wildlife, and in some cases, people.

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