# Draft Guidance for Implementing the January 2001 Methylmercury Water Quality Criterion

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United States Environmental Protection Agency
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# DISCLAIMER

This guidance provides advice on how to implement the water quality criterion recommendation for methylmercury that the U.S. Environmental Protection Agency (EPA) published in January 2001. This guidance does not impose legally binding requirements on EPA, states, tribes, other regulatory authorities, or the regulated community, and may not apply to a particular situation based upon the circumstances. EPA, state, tribal, and other decision makers retain the discretion to adopt approaches on a case-by-case basis that differ from those in the guidance where appropriate. EPA may update this guidance in the future as better information becomes available.

The Office of Science and Technology, Office of Water, U.S. Environmental Protection Agency has approved this guidance for publication. Mention of trade names, products, or services does not convey and should not be interpreted as conveying official EPA approval, endorsement, or recommendation for use

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# 1 Executive Summary

In January 2001, EPA published ambient water quality criteria (AWQC) recommendations for methylmercury for the protection of people who eat fish and shellfish. This criterion, 0.3 mg methylmercury/kg fish tissue wet weight, marks EPA's first issuance of a water quality criterion expressed as a fish and shellfish tissue value rather than as an ambient water column value.

Research shows that exposure to mercury and its compounds can cause certain toxic effects in humans and wildlife (USEPA 1997c). As of 2004, 44 states, 1 territory, and 2 tribes have issued fish consumption advisories for mercury covering 13.2 million lake acres and 765,000 river miles (USEPA 2005a). Mercury is widely distributed in the environment and originates from both natural and anthropogenic processes, including combustion and volcanoes. Methylmercury is highly bioaccumulative and is the form of mercury that bioaccumulates most efficiently in the food web.

Under section 303(c) of the Clean Water Act (CWA), states and authorized tribes must adopt water quality criteria that protect designated uses. This document provides technical guidance to states and authorized tribes exercising responsibility under section CWA 303(c) on how to use the new fish tissue-based criterion recommendation as they develop their own water quality standards for methylmercury. One approach that States and authorized tribes may decide to use is to translate the tissue residue value to a water column value through use of methylmercury bioaccumulation factors (BAFs). If a state or authorized tribe decides to use this approach, EPA recommends three potential approaches for relating a concentration of methylmercury in fish tissue to a concentration of mercury in ambient water. The approaches are:

- Deriving site-specific methylmercury BAFs
- Using bioaccumulation models
- Using EPA's draft default methylmercury BAFs

All three approaches have limitations, especially in the amount of data necessary to develop a BAF. This guidance discusses the advantages and limitations of each approach. States and authorized tribes may also consider calculating their own fish tissue criteria or adopting site-specific criteria for methylmercury to reflect local or regional fish consumption rates or relative source contributions. EPA encourages states and authorized tribes to develop a water quality criterion for methylmercury using local or regional data rather than the default values if they believe that such a water quality criterion would be more appropriate for their target population. This guidance also discusses variances and use attainability analyses (UAAs) relating to methylmercury.

This document describes methods for measuring mercury and methylmercury in both tissue and water. These methods can analyze mercury and methylmercury in tissue and water at very low levels—well below the previous criterion for mercury in water and the current criterion of methylmercury in fish tissue. This document also provides guidance for field sampling plans, laboratory analysis protocols, and data interpretation on the basis of previously published EPA guidance on sampling strategies for contaminant

monitoring. This document also describes how states can assess the attainment of water quality criteria and protection of designated uses by comparing sampling data to water quality criteria.

EPA expects that, as states and authorized tribes adopt the methylmercury criterion, the number of waterbodies states report as impaired due to mercury contamination might increase. EPA expects this to occur because the number of river miles and lake acres under fish consumption advisories due to methylmercury in fish tissue greatly exceeds the number of waters listed by states as impaired. EPA expects that, as a result of this revised methylmercury water quality criterion, together with a more sensitive method for detecting mercury in effluent and the water column, and increased monitoring of previously unmonitored waterbodies, the number of waterbodies that states report on CWA section 303(d) lists as impaired due to mercury contamination may increase. Thus, this guidance also discusses approaches for managing the development of Total Maximum Daily Loads (TMDLs) for waterbodies impaired by mercury. This includes approaches for addressing waterbodies where much of the mercury is from atmospheric sources and how TMDLs can take into account ongoing efforts to address sources of mercury, such as programs under the Clean Air Act (CAA) and pollution prevention activities. This guidance also includes a recommended approach for directly incorporating the methylmercury tissue criterion in National Pollutant Discharge Elimination System (NPDES) permits.

# 2 Introduction

## 2.1 What is the interest in mercury?

Mercury occurs naturally in the earth's crust and cycles in the environment as part of both natural and human-induced activities. The amount of mercury mobilized and released into the biosphere has increased since the beginning of the industrial age. Most of the mercury in the atmosphere is elemental mercury vapor, which circulates in the atmosphere for up to a year, and hence can be widely dispersed and transported thousands of miles from sources of emission. Most of the mercury in water, soil, sediments, plants, and animals is in the form of inorganic mercury salts and organic forms of mercury (e.g., methylmercury). Divalent mercury, when bound to airborne particles, is readily removed from the atmosphere by precipitation and is also dry deposited. Even after it deposits, mercury commonly returns to the atmosphere either as a gas or associated with particles, and redeposits elsewhere. As it cycles between the atmosphere, land, and water, mercury undergoes a series of complex chemical and physical transformations, many of which are not completely understood.

This guidance focuses on an organic mercury compound known as methylmercury. Methylmercury most often results from microbial activity in wetlands, the water column, and sediments and is the form of mercury that presents the greatest risks to human health. The methylation process and methylmercury bioaccumulative patterns are discussed in more detail in section 2.3.

### *2.1.1 What are the health effects of mercury?*

Exposure to methylmercury can result in a variety of health effects in humans. Children who are exposed to low concentrations of methylmercury prenatally might be at risk of poor performance on neurobehavioral tests, such as those measuring attention, fine motor function, language skills, visual-spatial abilities, and verbal memory. (NRC 2000, USEPA 2002e, USEPA 2005b). In 2000, the National Academy of Sciences (NAS)/National Research Council (NRC) reviewed the health studies on mercury (NRC 2000). EPA's current assessment of the methylmercury reference dose (RfD) relied on the quantitative analyses performed by the NRC (USEPA 2002e). The RfD is an estimate (with uncertainty spanning perhaps an order of magnitude) 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 (USEPA 2002e). In its review of the literature, NRC found neurodevelopmental effects to be the most sensitive endpoints and appropriate for establishing a methylmercury RfD (NRC 2000). On the basis of the NRC report, EPA established an RfD of 0.0001 mg/kg per day (0.1 microgram of methylmercury per day for each kilogram of a person's body mass) in 2001 (USEPA 2002e). EPA believes that exposures at or below the RfD are unlikely to be associated with appreciable risk of deleterious effects. It is important to note, however, that the RfD does not define an exposure level corresponding to zero risk; mercury exposure near or below the RfD could pose a very low level of risk that EPA deems to be non-appreciable. It is also important to note that the RfD does not define a bright line, above which individuals are at risk of adverse effects (USEPA 2005b).

The primary route by which the U.S. population is exposed to methylmercury is through the consumption of fish containing methylmercury. The exposure levels at which neurological effects have been observed in children can occur via maternal consumption of fish (rather than high-dose poisoning episodes) (USEPA 2005b). In 2005, the National Health and Nutrition Examination Survey (NHANES) published results of a study of blood mercury levels in a representative sample of U.S. women of childbearing age (CDC 2005). The report data for the period 1999–2002 show that all women of childbearing age had blood mercury levels below 58 µg/L, a concentration associated with neurologic effects in the fetus. These data show that 5.7 percent of women of childbearing age had blood mercury levels between 5.8 and 58 µg/L; that is, levels within an order of magnitude of those associated with neurological effects. Typical exposures for women of childbearing age were generally within two orders of magnitude of exposures associated with these effects, according to data from NHANES (CDC 2005, USEPA 2005b).

With regard to other health effects of methylmercury, some recent epidemiological studies in men suggest that methylmercury is associated with a higher risk of acute myocardial infarction, coronary heart disease, and cardiovascular disease in some populations. Other recent studies have not observed this association. The studies that have observed an association suggest that the exposure to methylmercury might attenuate the beneficial effects of fish consumption (USEPA 2005b). There also is some recent evidence that exposures of methylmercury might result in genotoxic or immunotoxic effects. Other research with less corroboration suggests that reproductive, renal, and hematological impacts could be of concern. There are insufficient human data to evaluate whether these effects are consistent with methylmercury exposure levels in the U.S. population (USEPA 2005b).

Deposition of mercury to waterbodies can also have an adverse impact on ecosystems and wildlife. Plant and aquatic life, as well as fish, birds, and mammalian wildlife, can be affected by mercury exposure; however, overarching conclusions about ecosystem health and population effects are difficult to make. Mercury contamination is present in all environmental media with aquatic systems experiencing the greatest exposures due to bioaccumulation. Bioaccumulation refers to the net uptake of a contaminant from all possible pathways and includes the accumulation that might occur by direct exposure to contaminated media as well as uptake from food. Elimination of methylmercury from fish is so slow that long-term reductions of mercury concentrations in fish are often due to growth of the fish ("growth dilution"), whereas other mercury compounds are eliminated relatively quickly. Piscivorous avian and mammalian wildlife are exposed to mercury mainly through the consumption of contaminated fish and, as a result, accumulate mercury to levels greater than those in their prey (USEPA 1997c). The *Regulatory Impact Analysis of the Clean Air Mercury Rule* (USEPA 2005b) provides a full discussion of potential ecosystem effects updated since publication of the 1997 *Mercury Study Report to Congress* (USEPA 1997c). Thus, the approach outlined in the Clean Air Mercury Rule provides states an alternative methodology for designing their site-specific TMDL analyses.

### 2.1.2 How frequent are the environmental problems?

As of 2004, 42 states reported at least one waterbody as being impaired due to mercury, and over 8,500 specific waterbodies were listed as being impaired due to mercury, either solely or in combination with other pollutants. In 2001, EPA mapped concentrations of mercury in fish tissue from fish collected from waterbodies all over the country (i.e., not limited to the 595 waters identified by the states) and compared these to the 2001 national recommended water quality criterion of 0.3 mg methylmercury/kg fish tissue wet weight (see Figure 1). These data were not randomly or systematically collected, but rather reflect fish tissue information that states had collected as part of their fish consumption advisory programs. Approximately 40 percent of the watershed-averaged fish tissue concentrations exceeded 0.3 mg methylmercury/kg fish tissue wet weight (USEPA 2001d).

A statistical comparison of the data presented in Figure 1 (from the National Listing of Fish Advisories (NLFA) fish tissue database), versus data from the National Lake Fish Tissue Study (NLFTS), a national random sample of fish tissue in 500 lakes and reservoirs throughout the United States, showed the NLFA data to be biased high (USEPA 2005b). The bias was found to be the result of sampling bias in the NLFA toward fish of species and sizes that tended to bioaccumulate more mercury. When data from the NLFA and NLFTS were normalized to a set of standard species and lengths, the bias was removed. (See USEPA 2005b, Figure 4-11, page 5-16 which shows fish tissue data averaged by watershed (i.e., hydrologic unit codes, or HUCs.) As a result, the NLFA data suggest that fewer watersheds contain fish with methylmercury that exceed the criterion.

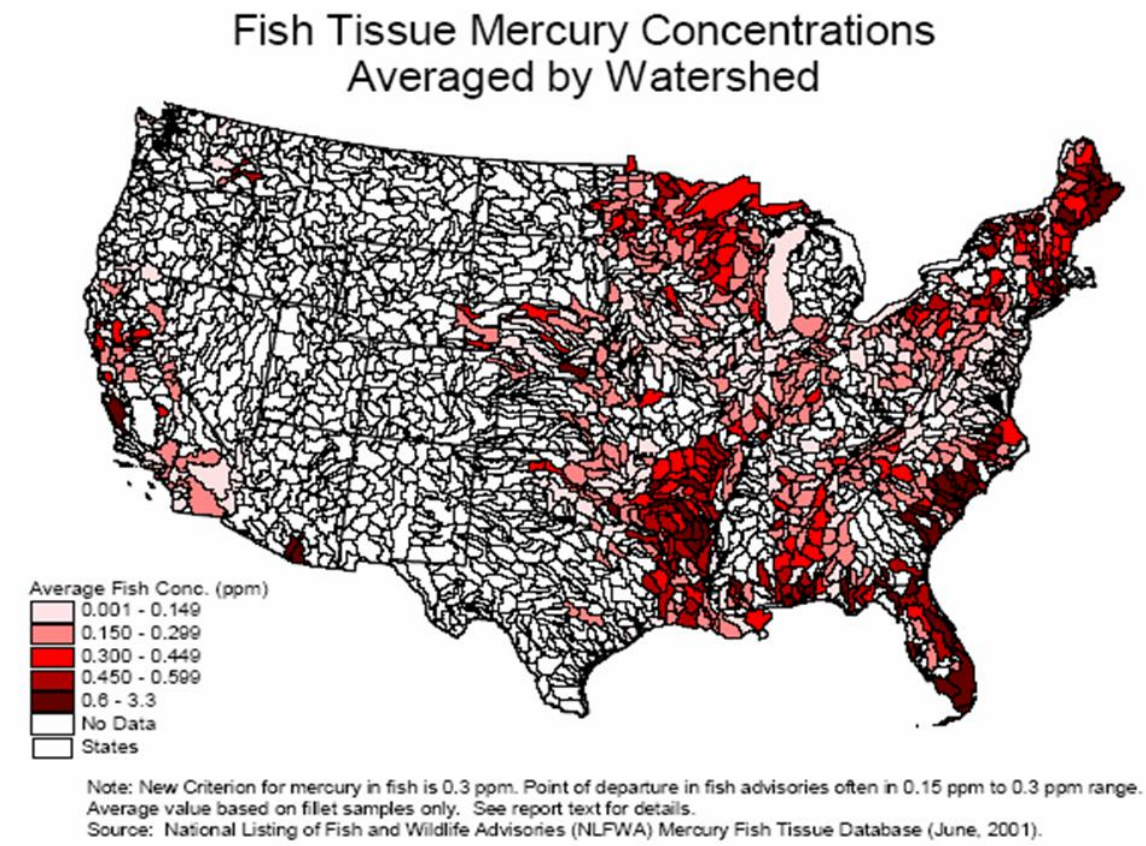


**Figure 1. Fish Tissue Mercury Concentrations Averaged by Watershed (USEPA 2001d)**

As of December 2004, 44 states, 1 territory, and 2 tribes have issued fish consumption advisories[1] for mercury covering 13.2 million lake acres and 765,000 river miles (see Figure 2). Twenty-one states have issued advisories for mercury in all freshwater lakes and rivers in their state, and 12 states have statewide advisories for mercury in their coastal waters (USEPA 2005a). EPA believes that the increase in advisories is primarily due to increased sampling of previously untested waters and not necessarily due to increased levels or frequency of contamination. Although states, territories, tribes, and local governments also continue to issue new fish advisories, most new fish advisories involve mercury and are a result of increased monitoring and assessment rather than increased domestic releases of mercury. In fact, U.S. mercury emissions have declined by more than 45 percent since 1990 (USEPA 2005a).

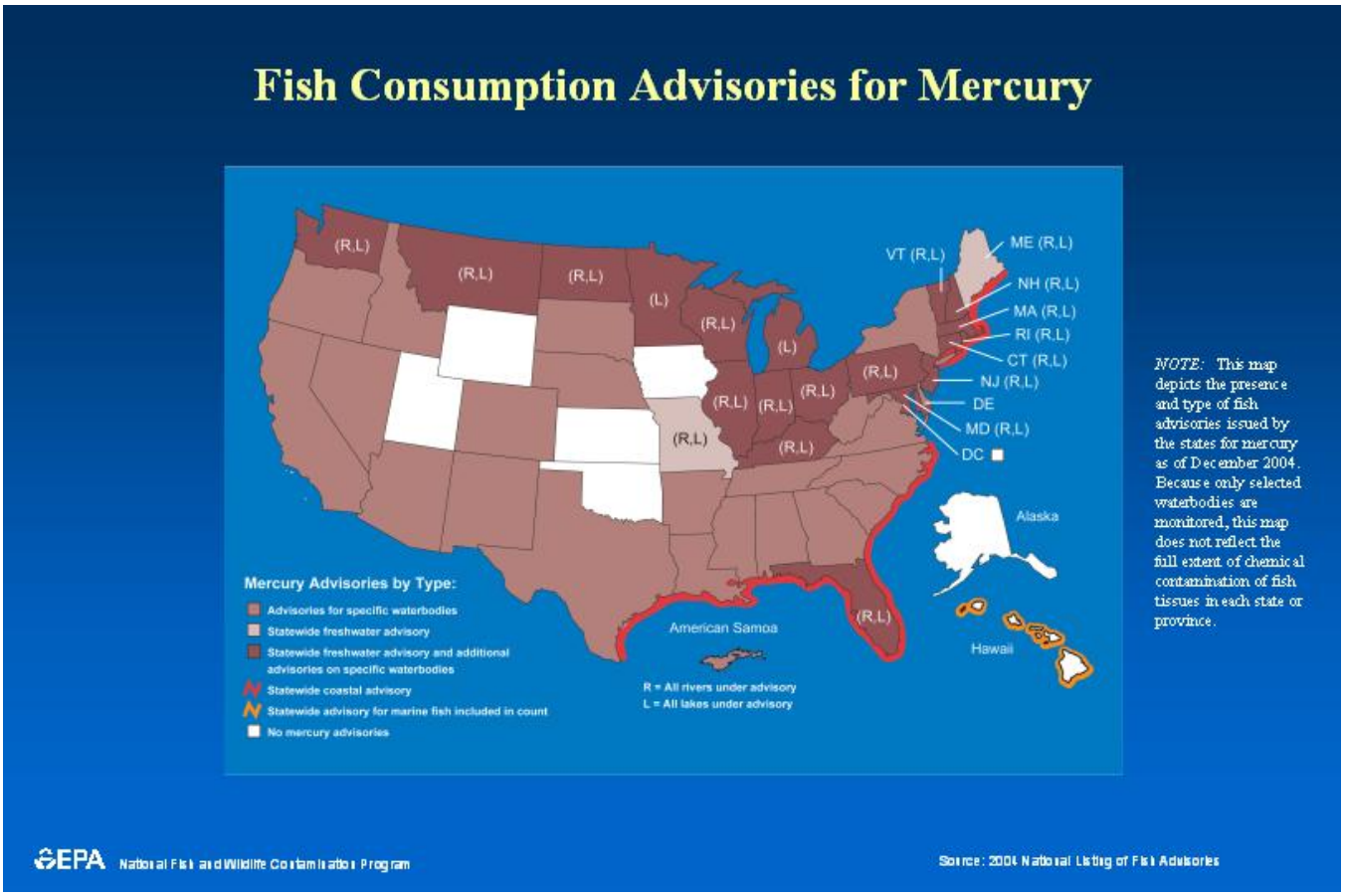


**Figure 2. Total Number of State Mercury Fish Consumption Advisories 2004**

## 2.2 What are the sources of mercury in fish?

Mercury is emitted from both natural and anthropogenic sources. Mercury's residence time in the atmosphere is much longer than that of most metals, because mercury can

[1] States issue their advisories and guidelines voluntarily and have flexibility in what criteria they use and how the data are collected. As a result, there are significant variations in the numbers of waters tested, the pollutants tested for, and the threshold for issuing advisories. Based on self-reporting, the national trend is for states to monitor different waters each year, generally without retesting waters monitored in previous years.

circulate for up to a year (USEPA 1997a). Such mobility enables elemental mercury to disperse and be transported over thousands of miles from likely sources of emission, across regions, and around the globe. As a result, the mercury detected in fish in U.S. surface waters is derived from both U.S. and international sources. EPA estimates that approximately 83 percent of the atmospheric mercury deposited on land and water in the country is from a combination of sources outside the United States and Canada, as well as natural and re-emitted sources. EPA's current air quality modeling does indicate a substantial variation across the country, with domestic sources influencing mercury deposition much more in the east and global sources being a more significant contributor to mercury deposition in the west, where relatively few domestic sources exist. This estimate was based on the advanced, state-of-the-science modeling assessment of the atmospheric fate, transport, and deposition of mercury conducted by EPA for the Clean Air Mercury Rule (CAMR) (USEPA 2005d).

Natural sources of mercury include geothermal emissions from volcanoes and crustal degassing in the deep ocean, as well as dissolution of mercury from other geologic sources (Rasmussen 1994). Anthropogenic sources of mercury in the United States include combustion (e.g., utility boilers, municipal waste combustors, commercial/industrial boilers, MWIs), manufacturing sources (e.g., chlor-alkali, cement, pulp and paper manufacturing), and mining (USEPA 1997a).

U.S. anthropogenic emissions of mercury to the air have declined more than 45 percent since passage of the 1990 CAA Amendments. These amendments provided new authority to EPA to reduce emissions of mercury and other toxic pollutants to the air. In 1990, more than two-thirds of U.S. human-caused mercury emissions came from just three source categories: coal-fired power plants, municipal waste combustion, and medical waste incineration (see Figure 4). Regulations were issued in the 1990s to control mercury emissions from waste combustion. In addition, actions to limit the use of mercury, most notably congressional action to limit the use of mercury in batteries and EPA regulatory limits on the use of mercury in paint, contributed to the reduction of mercury emissions from waste combustion during the 1990s by reducing the mercury content of waste. More recent regulations, including regulation of mercury emissions from chlorine production facilities that use mercury cells and regulation of industrial boilers, will further reduce emissions of mercury.[2]

The largest single source of anthropogenic mercury emissions in the country currently is coal-fired power plants. Mercury emissions from U.S. power plants are estimated to account for about one percent of total global mercury emissions. In March 2005, EPA signed the CAMR to permanently cap and reduce mercury emissions from coal-fired power plants (USEPA 2005e). This rule makes the United States the first country in the world to regulate mercury emissions from utilities. CAMR builds on EPA's Clean Air

[2] EPA has issued several regulations pursuant to the CAA to address these air emissions, including recent regulations covering coal-fired power plants. For example, see Title 40 of the *Code of Federal Regulations* (CFR) Part Cb (standards for municipal waste combustors); 40 CFR Part 60, subpart Ce (standards for MWIs); 40 CFR Part 63 subpart IIIII (standards for chlor-alkali plants); 40 CFR 63.1203 (a)(2) and (b)(2) (standards for existing and new hazardous waste-burning incinerators), 40 CFR 63.1204 (a)(2) and (b)(2) (standards for existing and new hazardous waste-burning cement kilns), and § 63.1205 (a)(2) and (b)(2) (standards for existing and new hazardous waste-burning lightweight aggregate kilns); 40 CFR Part 63, Subpart DDDDD (standards for industrial boilers); and 70 *Federal Register* 28,606 (May 18, 2005) (codified at 40 CFR Parts 60, 72 and 75) (standards for power plants). See also section 8.2 of this document.

Interstate Rule (CAIR) to significantly reduce emissions from coal-fired power plants. When fully implemented, these rules will reduce utility emissions of mercury nearly 70 percent.

Point sources of mercury discharging into waters are also regulated by NPDES permits. Chlor-alkali facilities are subject to effluent guidelines that impose treatment levels reflective of the Best Available Technology Economically Achievable (40 CFR Part 415). All NPDES permits must assure that permitted discharges achieve water quality standards (40 CFR 122.42(d)). Nonpoint source discharges are not regulated under federal regulations, but to the extent that these sources cause a water to exceed its water quality standards, states will develop TMDLs that identify the necessary reductions in these sources for achieving the water quality standards.

Anthropogenic emissions are only one part of the mercury cycle, however. Releases from human activities today add to the mercury reservoirs that already exist in land, water, and air, both naturally and as a result of previous human activity.

## 2.3 How does methylmercury get into fish and shellfish?

Mercury is widely distributed in the environment. Understanding the distribution and cycling of mercury among the abiotic (nonliving) and biotic (living) compartments of aquatic ecosystems is essential to understanding the factors governing methylmercury uptake in fish and shellfish tissue. The following is a synopsis of the current understanding of mercury cycling in the environment as described in the *Regulatory Impact Analysis of the Clean Air Mercury Rule* (USEPA 2005b).

Mercury occurs naturally in the environment as several different chemical species. The majority of mercury in the atmosphere (95–97 percent) is present in a neutral, elemental state ($Hg^0$) (Lin and Pehkonen 1999), while in water, sediments, and soils, the majority of mercury is found in the oxidized, divalent state (Hg(II)) (Morel et al. 1998). A small fraction of this pool of divalent mercury is transformed by microbes into methylmercury ($CH_3Hg(II)$) (Jackson 1998). Methylmercury is retained in fish tissue and is the only form of mercury that biomagnifies in aquatic food webs (Kidd et al. 1995). Transformations among mercury species within and between environmental media result in a complicated chemical cycle.

The relative contributions of local, regional, and long-range sources of mercury to fish mercury levels in a given waterbody are strongly affected by the speciation of natural and anthropogenic emissions sources. Elemental mercury is oxidized in the atmosphere to form the more soluble mercuric ion (Hg(II)) (Schroeder et al. 1989). Particulate and reactive gaseous phases of Hg(II) are the principle forms of mercury deposited onto terrestrial and aquatic systems because they are more efficiently scavenged from the atmosphere through wet and dry deposition than $Hg^0$ (Lindberg and Stratton 1998). Because Hg(II) species or reactive gaseous mercury (RGM) and particulate mercury (Hg(p)) in the atmosphere tend to be deposited more locally than $Hg^0$, differences in the species of mercury emitted affect whether it is deposited locally or travels longer distances in the atmosphere (Landis et al. 2004).

A portion of the mercury deposited in terrestrial systems is re-emitted to the atmosphere. On soil surfaces, sunlight might reduce deposited Hg(II) to $Hg^0$, which might then evade back to the atmosphere (Carpi and Lindberg 1997, Frescholtz and Gustin 2004, Scholtz et al. 2003). Significant amounts of mercury can be codeposited to soil surfaces in throughfall and litterfall of forested ecosystems (St. Louis et al. 2001), and exchange of gaseous $Hg^0$ by vegetation has been observed (e.g., Gustin et al. 2004). Hg(II) has a strong affinity for organic compounds such that inorganic mercury in soils and wetlands is predominantly bound to dissolved organic matter (Mierle and Ingram 1991). Concentrations of methylmercury in soils are generally very low. In contrast, wetlands are areas of enhanced methylmercury production and account for a significant fraction of the external methylmercury inputs to surface waters that have watersheds with a large portion of wetland coverage (e.g., St. Louis et al. 2001).

In the water column and sediments, Hg(II) partitions strongly to silts and biotic solids, sorbs weakly to sands, and complexes strongly with dissolved and particulate organic material. Hg(II) and methylmercury sorbed to solids settle out of the water column and accumulate on the surface of the benthic sediment layer. Surficial sediments interact with the water column via resuspension and bioturbation. The amount of bioavailable methylmercury in water and sediments of aquatic systems is a function of the relative rates of mercury methylation and demethylation. In the water, methylmercury is degraded by two microbial processes and sunlight (Barkay et al. 2003, Sellers et al. 1996). Mass balances for a variety of lakes and coastal ecosystems show that *in situ* production of methylmercury is often one of the main sources of methylmercury in the water and sediments (Benoit et al. 1998, Bigham and Vandal 1994, Gbundgo-Tugbawa and Driscoll 1998, Gilmour et al. 1998, Mason et al. 1999). Changes in the bioavailability of inorganic mercury and the activity of methylating microbes as a function of sulfur, carbon, and ecosystem specific characteristics mean that ecosystem changes and anthropogenic "stresses" that do not result in a direct increase in mercury loading to the ecosystem, but alter the rate of methylmercury formation, might also affect mercury levels in organisms (e.g., Grieb et al. 1990).

Dissolved Hg(II) and methylmercury accumulate in aquatic vegetation, phytoplankton, and benthic invertebrates. Unlike Hg(II), methylmercury biomagnifies through each successive trophic level in both benthic and pelagic food chains such that mercury in predatory, freshwater fish is found almost exclusively as methylmercury (Bloom 1992, Watras et al. 1998). In fish, methylmercury bioaccumulation is a function of several uptake (diet, gills) and elimination pathways (excretion, growth dilution) (Gilmour et al. 1998, Greenfield et al. 2001). Factors such as pH, length of the aquatic food chain, temperature, and dissolved organic carbon (DOC) can affect bioaccumulation (Ullrich et al. 2001). As a result, the highest mercury concentrations for a given fish species correspond to smaller, long-lived fish that accumulate methylmercury over their life span with minimal growth dilution (e.g., Doyon et al. 1998). In general, higher mercury concentrations are expected in top predators, which are often large fish relative to other species in a waterbody.

## 2.4 Why is EPA publishing this document?

In a January 8, 2001, *Federal Register* notice (66 FR 1344), EPA announced the availability of its recommended water quality criterion for methylmercury. In that notice, EPA also stated that development of the associated implementation procedures and guidance documents would begin by the end of 2001. As such, EPA makes this guidance available to fulfill that commitment to enable states and authorized tribes to adopt the recommendations set forth in *Water Quality Criterion for the Protection of Human Health: Methylmercury* (USEPA 2001c), or other water quality criteria for methylmercury on the basis of scientifically defensible methods, into their water quality standards.

This nontraditional approach in developing a water quality criterion as a fish and shellfish tissue value raises several implementation questions on both technical and programmatic fronts. Development of water quality standards, NPDES permits, and TMDLs present many challenges because these activities have usually been based on a water concentration (e.g., as a measure of mercury levels in effluent). This guidance addresses issues associated with states and authorized tribes adopting the new water quality criterion into their water quality standards programs and implementation of the revised water quality criterion in TMDLs and NPDES permits. Further, because atmospheric deposition serves as a large source of mercury for many waterbodies, implementation of this criterion involves coordination across various media and program areas.

EPA expects that, as a result of this revised methylmercury water quality criterion, together with a more sensitive method for detecting mercury in effluent and the water column, and increased monitoring of previously unmonitored waterbodies, the number of waterbodies that states report on CWA section 303(d) lists as impaired due to mercury contamination might increase. This guidance discusses approaches for managing the development of TMDLs for waterbodies impaired by mercury. This includes approaches for addressing waterbodies where much of the mercury comes from atmospheric sources and how TMDLs can take into account ongoing efforts to address sources of mercury, such as programs under the CAA and pollution prevention activities. This guidance also includes a recommended approach for directly incorporating the methylmercury tissue criterion in NPDES permits.

## 2.5 What is the effect of this document?

This guidance document presents suggested approaches, but not the only technically defensible approaches, to criteria adoption and implementation. The guidance does not substitute for applicable sections of the CWA or EPA's regulations; nor is it a regulation itself. Thus, it cannot impose legally binding requirements on EPA, states, authorized tribes, or the regulated community and may not apply to a particular situation. EPA, state, territorial, and tribal decision makers retain the discretion to adopt approaches on a case-by-case basis that differ from this guidance where appropriate. EPA may change this guidance in the future.