6.0 METHYLMERCURY BIOACCUMULATION

6.1 INTRODUCTION

Aquatic organisms can accumulate and retain certain chemicals in their bodies when exposed to these chemicals through water, their diet and other sources. This process is called bioaccumulation. In order to prevent harmful exposures to waterborne pollutants through the consumption of contaminated fish and shellfish, national 304(a) water quality criteria for the protection of human health must address the process of chemical bioaccumulation in aquatic organisms. For deriving national 304(a) ambient water column criteria to protect human health, EPA accounts for potential bioaccumulation of pollutants in fish and shellfish through the use of national bioaccumulation factors (BAFs). A national BAF is a ratio (in L/kg) which relates the concentration of a chemical in water to its expected concentration in commonly consumed aquatic organisms in a specified trophic level. The magnitude of bioaccumulation by aquatic organisms varies widely depending on the chemical but can be extremely high for some highly persistent and hydrophobic chemicals. For such highly bioaccumulative chemicals, concentrations in aquatic organisms may pose unacceptable human health risks from fish and shellfish consumption even when concentrations in water are too low to cause unacceptable health risks from drinking water consumption alone. These chemicals may also biomagnify in aquatic food webs, a process whereby chemical concentrations increase in aquatic organisms of each successive trophic level due to increasing dietary exposures (e.g., increasing concentrations from algae, to zooplankton, to forage fish, to predator fish). Methylmercury is a chemical that bioaccumulates and biomagnifies to a relatively high extent. Methylmercury BAFs for upper trophic level freshwater and estuarine fish and shellfish typically consumed by humans generally range between 500,000 and 10,000,000 (Glass et al., 1999; Lores et al., 1998; Miles and Fink, 1998; Monson and Brezonik, 1998; Watras et al., 1998; Mason and Sullivan, 1997).

6.2 ISSUES IN DEVELOPING METHYLMERCURY BAFS

The fates of mercury and methylmercury in the environment are complex processes affected by numerous biotic and abiotic factors that are subjects of ongoing research by various government, private, and academic groups around the world. Methylation of mercury is a key step in the entrance of mercury into food chains. The biotransformation of inorganic mercury species to methylated organic species in water bodies can occur in the sediment and the water column. Inorganic mercury can be absorbed by aquatic organisms but is generally taken up at a slower rate and with lower efficiency than is methylmercury. Methylmercury continues to accumulate in fish as they age. Predatory organisms at the top of aquatic and terrestrial food webs generally have higher methylmercury concentrations because methylmercury is typically not completely eliminated by organisms and is transferred up the food chain when predators feed on prey; for example, when a largemouth bass feeds on a bluegill sunfish, which fed on aquatic insects and smaller fish, all of the prey could contain some amount of methylmercury that gets transferred to the predator. Nearly 100% of the mercury that bioaccumulates in upper trophic level fish (predator) tissue is methylmercury (Bloom, 1992; Akagi, 1995; Kim, 1995; Becker and Bigham, 1995).

Numerous factors can influence the bioaccumulation of mercury in aquatic biota. These include, but are not limited to, the acidity (pH) of the water, length of the aquatic food chain, temperature, and dissolved organic material. Physical and chemical characteristics of a watershed, such as soil type and erosion or proportion of area that is wetlands, can affect the amount of mercury that is transported from soils to water bodies. Interrelationships among these factors are poorly understood and are likely to be site-specific. No single factor (including pH) has been correlated with extent of mercury bioaccumulation in all cases examined. Two lakes that are similar biologically, physically, and chemically can have different methylmercury concentrations in water, fish, and other aquatic organisms (Cope et al., 1990; Grieb et al., 1990; Jackson, 1991; Lange et al., 1993). For more in-depth discussions about the chemical, physical, and biological interactions affecting methylmercury bioaccumulation in aquatic organisms see the *Mercury Study Report to Congress* (MSRC), Volume III and Volume III Appendix D (U.S. EPA, 1997c), and the compilation of papers in *Mercury Pollution: Integration and Synthesis* (Watras and Huckabee, 1994).

To derive section 304(a) water quality criteria for the protection of human health, EPA needs to conduct a human health risk assessment on the pollutant in question and to gather information on the target population's exposure to the pollutant. Traditionally, EPA has expressed its section 304(a) water quality criteria guidance to protect human health in the form of pollutant concentrations in ambient surface water. To account for human exposure through the aquatic food pathway when deriving a water column-based water quality criterion, EPA uses national BAFs (U.S. EPA 2000). A BAF is a ratio (in L/kg) that relates the concentration of a chemical in water to its expected concentration in commonly consumed aquatic organisms in a specified trophic level (U.S. EPA 2000). A national BAF is meant to be broadly applicable to all waters in the United States, whereas a site-specific BAF is based on local data and integrates local spacial and temporal factors that can influence bioaccumulation. For pollutants that biomagnify, such as methylmercury, EPA's preferred approach for deriving national BAFs for use in deriving section 304(a) water quality criteria is to use empirical field data collected in the natural environment. EPA prefers this approach because BAFs derived with field data integrate the chemical, biological, and physical factors that can affect bioaccumulation in fish and shellfish. With this preference in mind, EPA explored the feasibility of developing field-derived national methylmercury BAFs for each trophic level of the aquatic food chain consumed by humans (i.e., trophic levels 2-4). Using Agency guidance on BAFs contained in the 2000 Human Health Methodology and procedures outlined in Volume III, Appendix D of the peer-reviewed MSRC (U.S. EPA, 1997c), EPA empirically derived draft national methylmercury BAFs for each trophic level of the aquatic food chain. The draft national BAFs were single value trophic level-specific BAFs calculated as the geometric mean of field data collected across the United States and reported in the open literature as well as other publically available reports. These draft methylmercury BAFs were compiled in a draft internal report and submitted to a panel of external scientific experts for peer review. The Appendix contains a summary of the internal BAF report and BAF peer review report. The entire internal draft methylmercury BAF report and peer review report can be obtained from the Water Docket W-00-20.

Within any given trophic level, the individual empirically derived draft methylmercury BAFs generally ranged up to two orders of magnitude. This range in BAFs reflects the various biotic factors (such as food chain interactions and fish age/size) and abiotic factors (such as pH and dissolved organic carbon). The large range in the individual empirically derived draft methylmercury BAFs results in uncertainty as to the ability of single trophic level-specific national methylmercury BAFs to accurately predict bioaccumulation of methylmercury in general across the waters of the United States. Presently, it is EPA's understanding that the mechanisms that underlie many of the influencing factors are not well understood and can not be accurately predicted. As the science of methylmercury improves, in the future it may be possible predict or model these processes and use such information to more accurately predict bioaccumulation. Until such time, EPA is unable to improve the predictive power of the methylmercury BAFs by universally accounting for influencing factors. This is not the case for other highly bioaccumulative pollutants; for example polychlorinated biphenyls (PCBs). For such pollutants, EPA has methods that improve the predictive capability of empirically derived or model predicted BAFs (such as normalizing fish tissue concentrations to lipid and normalizing ambient water concentrations to dissolved and particulate organic carbon). EPA is actively involved in, and will continue to support, various types of research aimed at better understanding the fate of mercury in the environment and the processes that underlie methylmercury bioaccumulation. EPA hopes that results of new research will enable better predictions of methylmercury bioaccumulation.

The BAF peer reviewers recognized the need for methylmercury BAFs and were supportive of most aspects of the methodology used to derive the draft national methylmercury BAFs. The peer reviewers did have issues with certain data used to derive the methylmercury BAFs and certain assumptions about food chain relationships. Overall, most of the peer reviewers believed that derivation of single-value trophic level-specific national BAFs for methylmercury that would be generally applicable to all waters of the United States under all conditions is difficult at best, and perhaps impossible. This opinion was based on consideration of the highly site-specific nature of methylmercury bioaccumulation in aquatic environments and the large range in the empirically derived draft methylmercury BAFs. These peer reviewers recommended developing methylmercury BAFs on a more local or regional scale, if not on a site-specific basis. Although EPA generally agrees with this suggestion, the data needed to derive BAFs at more localized scales across the U.S. are not available. See Appendix A for a summary of the internal BAF report and the BAF peer review report.

6.3 CONSIDERATION OF A FISH TISSUE RESIDUE CRITERION

After considering the various issues about mercury fate in the environment, the recent report by the National Research Council (NRC, 2000) on the toxicological effects of mercury, and the methylmercury BAF peer review comments, EPA concluded that it is more appropriate at this time to derive a fish tissue (including shellfish) residue water quality criterion for methylmercury rather than a water column-based water quality criterion. EPA believes a fish tissue residue water quality criterion for methylmercury is appropriate for many reasons. A fish tissue residue water quality criterion integrates spatial and temporal complexity that occurs in aquatic systems and that affect methylmercury bioaccumulation. A fish tissue residue water quality criterion in this instance is more closely tied to the CWA goal of protecting the public health because it is based directly on the dominant human exposure route for methylmercury. The concentration of methylmercury is also generally easier to quantify in fish tissue than in water and is less variable in fish and shellfish tissue over the time periods in which water quality standards are typically implemented in water quality-based controls, such as NPDES permits. Thus, the data used in permitting activities can be based on a more consistent and measurable endpoint. Finally, this approach is consistent with the way in which fish advisories are issued. Fish advisories for mercury are also based on the amount of methylmercury in fish tissue that is considered acceptable, although such advisories are usually issued for a certain fish or shellfish species in terms of a meal size. A fish tissue residue water quality criterion should enhance harmonization between these two approaches for protecting the public health.

Because EPA did not use national, empirically derived methylmercury BAFs to establish today's section 304(a) recommended methylmercury water quality criterion, EPA has deferred further efforts to derive national BAFs for methylmercury at this time. EPA notes, however, that there may be adequate field data for some waterbodies or geographical regions on which to base accurate predictive, site-specific methylmercury BAFs. EPA may reconsider developing national methylmercury BAFs in the future once more field data is available for a broader range of species and aquatic ecosystems, or once more information is available describing the mechanisms that affect bioaccumulation. Such information could enable EPA to more accurately predict methylmercury bioaccumulation on a broader scale given a certain total mercury concentration in water.

7.0 WATER QUALITY CRITERION CALCULATION

7.1 EQUATION FOR TISSUE RESIDUE CONCENTRATION AND PARAMETERS USED

The equation for calculating the methylmercury fish tissue residue criterion is:

$$TRC = \frac{BW \times (RfD - RSC)}{\sum_{i=2}^{4} FI_i}$$

Where:

TRC	=	Fish tissue residue criterion (mg methylmercury/kg fish) for freshwater and estuarine fish
RfD	=	Reference dose (based on noncancer human health effects) of 0.0001 mg methylmercury/kg body weight-day
RSC	=	Relative source contribution (subtracted from the RfD to account for marine fish consumption) estimated to be 2.7 x 10^{-5} mg methylmercury/kg body weight-day
BW	=	Human body weight default value of 70 kg (for adults)
FI	=	Fish intake at trophic level (TL) i (i = 2, 3, 4); total default intake is 0.0175 kg fish/day for general adult population. Trophic level breakouts for the general population are: TL2 = 0.0038 kg fish/day; TL3 = 0.0080 kg fish/day; and TL4 = 0.0057 kg fish/day.

This yields a methylmercury TRC value of 0.3 mg methylmercury/kg fish (rounded to one significant digit from 0.288 mg methylmercury/kg fish).

This equation is essentially the same equation used in the 2000 Human Health Methodology to calculate a water quality criterion, but is rearranged to solve for a protective concentration in fish tissue rather than in water. Thus, it does not include a BAF or drinking water intake value (as discussed above, exposure from drinking water is negligible). The TRC of 0.3 mg methylmercury/kg fish is the concentration in fish tissue that should not be exceeded based on a total consumption of 0.0175 kg fish/day.

7.2 SITE-SPECIFIC OR REGIONAL ADJUSTMENTS TO CRITERIA

Several parameters in the Water Quality Criterion equation can be adjusted on a site-specific or regional basis to reflect regional or local conditions and/or specific populations of concern. These include the fish consumption rates and the RSC estimate. States and authorized Tribes can also choose to apportion an intake rate to the highest trophic level consumed for their population or modify EPA's default intake rate based on local or regional consumption patterns. EPA strongly encourages States and authorized Tribes to consider developing a criterion using local or regional data over the default values if they believe that they would be more appropriate for their target population. States and authorized Tribes are encouraged to make such adjustments using the guidance provided in the 2000 Human Health Methodology (U.S. EPA, 2000a).

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