# Use of Tissue Residue Data in Exposure and Effects Assessments for Aquatic Organisms

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Under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA, or Superfund), the National Oceanic and Atmospheric Administration (NOAA) is responsible for protecting and restoring natural resources that may be injured by releases from Superfund sites in coastal areas. As part of NOAA's role of providing EPA with technical support in the evaluation of natural resource concerns at hazardous waste sites, NOAA's Coastal Resources Coordination Branch often recommends collecting tissue residue data.

Tissue residue data can play an important role at different stages in the process of evaluating hazardous waste sites, including ecological risk assessment, modeling conducted to evaluate different remedial alternatives, and the monitoring necessary to determine the effectiveness of remediation. An ecological risk assessment is a major component of hazardous waste site evaluations. For an ecological risk assessment to provide useful information for remedy selection, exposure should be related both to the source of contamination and to an important toxicity endpoint. This means that it is necessary to link any estimated or observed adverse effects to site-related contamination, which emphasizes the importance of identifying and quantifying major exposure pathways. Because bioaccumulation is often more of a concern for higher-trophic level organisms, which may receive much of their contaminant exposure via the food web, any effort to develop bioaccumulation-based criteria should identify and address significant, sensitive toxicity endpoints in fish and other higher-trophic level organisms. For example, reproductive or early life stage toxicity in fish is both a sensitive toxicity endpoint and an endpoint with potentially significant implications for the health of a fish population.

The appropriate application of tissue residue data depends upon the type of contaminant under consideration. For some contaminants, tissue residue concentrations may be directly linked to toxicity endpoints. Also, the concentrations of contaminants in the tissues of aquatic organisms often provide a direct measure of bioavailability that integrates exposure over time and integrates exposure from water, sediment, and food web pathways. In this presentation, I will be focusing primarily on the use of field-collected tissue residue data in exposure and effects assessments for aquatic ecological risk assessment. Because of the importance of bioaccumulation for higher-trophic level organisms, I will focus my discussion on the use of tissue residue data in assessing toxicity and exposure in fish. Addressing these issues is particularly important since currently available criteria for sediment explicitly exclude any consideration of contaminant exposure through the food web. "These [sediment] criteria do not address the question of possible contamination of upper trophic level organisms..." (USEPA, 1993a). And, "...a site-specific investigation appears to be the only available method for performing an evaluation of the effect of contaminated sediments on the body burdens of upper-trophic level organisms" (USEPA, 1993b).

Aquatic risk assessment requires information on the bioavailable fraction of the contaminants in the system. Accurate and meaningful assessments of exposure from water and sediment are difficult due to a potentially high degree of temporal and/or spatial variability. Because of high bioconcentration factors, particularly for hydrophobic organic contaminants, very low analytical detection limits are required to ensure that concentrations below analytical detection limit values could not contribute to significant exposure concentrations. Determining the bioavailable contaminant fraction is also complicated by partitioning suspended particulates and dissolved organic carbon. Modeling food web exposure in fish requires making many assumptions for which there is frequently little site-specific or reliable information available. Collection of site-specific tissue residue concentrations can be used to reduce some of this uncertainty in the application of the exposure models used in the assessment. In



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many cases, tissue residue data can be used to provide time-integrated and location-specific information on bioavailable contaminant concentrations.

## Metals

The bioaccumulation of metals in benthic macroinvertebrates can provide a useful measure of the extent and magnitude of contamination that temporally integrates exposure via the water column and sediment. Because benthic invertebrates represent an important source of food for fish, the accumulation of metals in invertebrates, which may have some tolerance to chronic exposure to metals, may also serve as a significant source of exposure to fish. Exposure via the food web may be particularly important for fish early life stages, which are also very sensitive to waterborne exposure to metals.

The combined effects of reduced survival and growth in fish early life history stages, which may have important implications for adverse effects on the health of a local fish population, is one of the most sensitive toxicity endpoints for waterborne exposure to a variety of metals (such as cadmium, copper, lead, mercury, and zinc). Laboratory studies on the effects of metals contamination on young of the year fish in the Clark Fork River in Montana demonstrated that feeding metals-contaminated invertebrates (at concentrations comparable to those found in the river) resulted in reduced growth and survival in rainbow trout (Woodward et al., 1994) and reduced growth and physiological abnormalities in rainbow trout and brown trout (Woodward et al., 1995). These studies also indicated that the exposure via dietary intake could be at least as important as aqueous exposure in age-0 fish. Another study, however, found no effects on survival or growth in age-0 rainbow trout that were fed metals-contaminated brine shrimp at similar concentrations (Mount et al., 1994). Additional studies are needed to more clearly resolve the relative importance of additional dietary exposure and to develop a method for determining the total effective dose that includes all exposure pathways.

In conducting a risk assessment for metals-contaminated sites, tissue residue concentrations in invertebrate prey organisms may reduce the uncertainty in estimating exposure via the water column and sediment (by integrating exposure over time at specific locations) as well as also providing information on exposure via the food web. This may be particularly important in assessments involving vulnerable receptor populations (for example, the early life history stages of threatened and endangered salmonid populations).

## Polycyclic Aromatic Hydrocarbons (PAHs)

Because fish rapidly metabolize and excrete PAHs, fish tissue residue concentrations of parent PAH compounds do not provide a useful measure of exposure to fish (Varanasi et al., 1989). However, exposure to PAHs has been linked to reproductive impairment, immune dysfunction, increased incidence of liver lesions, and other histopathological endpoints in fish (Malins et al., 1987; Johnson et al., 1988; Varanasi et al., 1992). High concentrations of PAHs in stomach contents of fish from a PAH-contaminated harbor indicate that the consumption of contaminated benthic invertebrates can be an important exposure pathway (Malins et al., 1985).

Measuring tissue concentrations in invertebrates that have minimal capacity for metabolism of PAHs, such as bivalve shellfish, can provide location-specific and temporally integrated information on bioavailable PAH concentrations, which can be used in food web models of exposure to fish and other higher trophic level organisms. Using tissue residue concentrations from caged bivalves allows for more control of specific location and other variables such as organism size or age. Other methods for determining the relative differences in PAH exposure include measuring concentrations of PAH metabolites in fish bile (Krahn et al., 1984, 1986), which can provide useful information on recent exposure to PAHs, and semipermeable membrane devices (SPMDs), which provide temporally integrated measurements of dissolved and presumably bioavailable PAHs in the water column at specific locations (Lebo et al., 1992; Huckins et al., 1996). The patterns of PAH concentrations in field-collected or caged bivalves and SPMDs can also play an important role in linking exposure to the source of PAHs using various fingerprinting methods.

## Polychlorinated Biphenyls (PCBs)

Fish tissue residue PCB concentrations, unlike PAHs, can provide useful information on both exposure and toxicity. Reproductive and early life stage effects (e.g., reduced larval growth or survival), where the primary exposure to the developing larvae results from the maternal transfer of accumulated PCBs to the eggs, is one of the most sensitive toxicity endpoints for PCB effects in fish. Consequently, PCB concentrations in the ovaries of mature female fish immediately prior to spawning may be the most useful measurement for estimating potential reproductive effects in species of concern. Higher-trophic level species, which tend to accumulate the highest concentrations of PCBs, may be at the greatest risk.

Field-collected tissue residue PCB concentrations in fish can play an integral role in exposure assessments, since they provide a measure of exposure via all exposure pathways. This is particularly important since fish generally receive much of their PCB exposure via the food web. The composition of the PCBs found in environmental samples often differs substantially from the original PCB source (e.g., commercial Aroclor mixtures). This difference results from physicochemical and biological processes that differentially affect individual PCB congeners (Safe et al., 1987). For example, the solubility and volatility of individual PCB congeners range over several orders of magnitude and greatly affect partitioning between sediment-water and water-air interfaces. Thus, less chlorinated congeners tend to be more soluble in water and are more quickly removed from sediment into water.

Biological processes also alter the pattern of congener distributions in tissue residue samples. Differences in uptake, depuration, metabolism, and efficiency of assimilation among individual congeners may result in large differences between the PCB mixtures in samples and in commercial Aroclor mixtures, and these differences would be expected to increase with increasing trophic level.

Because of the major differences between the PCB congener distributions in environmental samples and in commercial mixtures, analysis methods based on Aroclor standards can provide only a semi-quantitative estimate of total PCBs (Safe et al., 1987). Modeling PCB behavior in the environment, monitoring trends in PCB concentrations, and relating fish tissue PCB residues to their sources can be considerably enhanced by congener-specific analysis. Samples of several species of fish collected from multiple locations over 170 miles of the Hudson River were analyzed for PCB congeners as part of the ecological risk assessment for a Superfund Remedial Investigation. Samples of the same species at a given location had very similar congener compositions, although their total PCB concentrations sometimes varied by a factor of 2 or more on either a wet weight or lipid-normalized basis. The PCB congener compositions and concentrations in fish tissue are being used in the ecological risk assessment to help clarify the relationship between the PCB sources in the river and the fish body burdens.

Two general screening approaches are available to evaluate the potential toxicity of PCBs to fish based on PCB body burdens: comparison to total PCB concentrations associated with adverse effects, or estimation of dioxin toxic equivalent concentrations and comparison to No Observable Adverse Effect Levels (NOAELs) for 2,3,7,8-tetrachloro-dibenzo-p-dioxin (TCDD). A variety of field and laboratory studies provide evidence of reproductive and other adverse effects in different fish species at low total PCB tissue residue concentrations. The total PCB concentrations in fish tissue residue samples associated with adverse effects range over several orders of magnitude, which may reflect the differences in toxicity among different PCB mixtures and/or a high degree of inter-species difference in sensitivity to the effects of PCBs. The combination of these factors results in considerable uncertainty in estimating the risk of reproductive toxicity to selected fish species based on total PCB tissue residue concentrations.

The coplanar PCBs—a group of PCB congeners that is similar in structure and biological activity to the highly toxic polychlorinated dioxins and dibenzofurans—are considered to be the most toxic PCBs (Safe, 1984). Although most of these coplanar PCBs are found at relatively low concentrations in the commercial PCB mixtures, several have been identified as important components of PCB tissue residues in aquatic biota and may be preferentially accumulated, particularly by higher-trophic level organisms (Safe, 1984; Hansen, 1987; Smith et al., 1990; Schwartz et al., 1993). Recent studies suggest that only the non-ortho substituted PCB congeners exhibit dioxin-like activity in fish (Walker and Peterson, 1991; Newsted et al., 1995; Zabel et al., 1995). The early life stage toxicity of these individual PCB congeners has been investigated in trout and compared to the toxicity of dioxin to develop fish-specific toxic equivalent factors , or TEFs (Walker and Peterson, 1991; Zabel et al., 1995). The TEFs for the non-ortho congeners calculated for trout are 1 to 2 orders of magnitude less than the TEFs determined for mammalian systems. Thus, using mammalian-based TEFs for PCBs may overestimate the potential for dioxin-like reproductive toxicity in fish.

## Conclusions

Tissue residue data can play an important role in aquatic ecological risk assessments at hazardous waste sites, in part by linking exposure to the source of contamination and to important toxicity endpoints.

The development of bioaccumulation-based criteria should identify and address toxicity endpoints in fish that are both sensitive and ecologically significant. This is particularly important for higher-trophic level species, which may receive much of their contaminant exposure via the food web.

For some contaminants, such as PAHs and possibly metals, dose-response models that include all exposure pathways need to be developed. Tissue residue concentrations in macroinvertebrates may provide key input into exposure models for these types of contaminants.

Tissue residue PCB concentrations in fish have important applications to both exposure and toxicity assessments. Because the PCB congener distributions in environmental samples often differ considerably from the original commercial PCB mixtures, congener-specific PCB analysis of fish tissue samples provides more useful information on exposure than a standard Aroclor analysis. PCB concentrations in the ovaries of mature female fish immediately prior to spawning may be the most useful measurement for estimating potential reproductive toxicity in fish species of concern.

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