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Comparison of Predicted and Observed Dioxin Levels in Fish: Implications for Risk Assessment*

Judy S. LaKind & Daniel Q. Naiman**

Introduction

Dioxin¹ is an ubiquitous environmental contaminant, inadvertently created during industrial processes such as incineration and pulp bleaching. In 1984, the U.S. Environmental Protection Agency (EPA) announced a water quality criterion or allowable water concentration, for dioxin. The water quality criterion calculation included specific values indicating dioxin's cancer potency and ability to accumulate in fish tissue. Both of those values are highly controversial, as is the degree of dioxin's impact on aquatic life and wildlife.

Since 1984, new information has become available, prompting EPA to revisit the dioxin issue.² Despite controversy surrounding dioxin, it is clear that it bioaccumulates in fish, creating a potential route of human exposure. Data on dioxin concentrations in fish may be used by state health agencies to issue fish consumption advisories and bans. In addition, data on dioxin levels in fish have prompted legal action against parties thought to be responsible. It is therefore important to consider whether the data base is adequate for regulatory and other purposes.

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¹ 2,3,7,8-tetrachlorodibenzo-p-dioxin, also commonly referred to as TCDD.

² See, e.g., *Reilly Launches Precedent-Setting EPA Look at Dioxin Risk Assessment*, Inside EPA Weekly Report, April 12, 1991.

Data on levels of dioxin accumulation in fish come principally from two major EPA studies.³ In these studies, particular attention was paid to waters receiving chlorine-bleaching pulp and paper mill discharge known to contain dioxin. The methodologies of the two EPA studies were distinctly different: In one study, dioxin levels in fish were determined by sampling and analysis, as part of EPA's National Study of Chemical Residues in Fish (EPANS);⁴ in the second study, fish dioxin levels from U.S. waters receiving pulp and paper mill discharges were modelled, or predicted, based on levels of dioxin entering the receiving water (EPARA).⁵ Both observed fish dioxin levels (EPANS) and predicted dioxin levels (EPARA), in conjunction with standard EPA health risk assessment factors for dioxin,⁶ yield information on carcinogenic and noncarcinogenic risks to people consuming fish containing dioxin. If the predicted and observed fish dioxin levels are comparable, then the use of either modelled fish dioxin levels or observed fish dioxin levels for calculating human health risks would be acceptable for regulatory and advisory purposes. Alternatively, disparity between the predicted and observed fish dioxin levels should compel regulatory agencies to carefully examine the two data sets in order to select the more reliable data as the basis for decision-making.

While comparison of predicted and observed dioxin data sets appears to be important in providing the public with optimal risk information associated with the consumption of fish containing dioxin, this type of analysis has not yet been performed. Here we present 1) a description of the EPA methodology used in the EPARA for modelling fish tissue dioxin levels near pulp and paper mill discharges, 2) a

³ U.S. ENVIRONMENTAL PROTECTION AGENCY (EPA), OFFICE OF WATER REGULATIONS AND STANDARDS, RISK ASSESSMENT FOR 2,3,7,8-TCDD AND 2,3,7,8-TCDF CONTAMINATED RECEIVING WATERS FROM U.S. CHLORINE-BLEACHING PULP AND PAPER MILLS, (1990) (hereafter EPARA); EPA, OFFICE OF SCIENCE AND TECHNOLOGY, NATIONAL STUDY OF CHEMICAL RESIDUES IN FISH, (1992) (hereafter EPANS); EPA, OFFICE OF WATER, AMBIENT WATER QUALITY CRITERIA FOR 2,3,7,8-TETRACHLORODIBENZO-P-DIOXIN (1984) (hereafter EPA 1984).

⁴ EPANS, *supra*.

⁵ *Supra* note 3.

⁶ *Id.*; EPA 1984, *supra*, note 3.

comparison of the modelled results with observed dioxin levels in fish, 3) an analysis of the discrepancies between the modelled and observed data and 4) a discussion of potential shortcomings associated with both data sets. We believe that the EPARA methodology used to predict fish uptake of contaminants is inappropriate for dioxin, as well as other hydrophobic organic chemicals. Therefore, despite the drawbacks of both modelled and observed fish dioxin data sets, we support the use of sampling and analysis data for determination of human health risks associated with the consumption of dioxin-contaminated fish, until scientifically defensible bioaccumulation models are incorporated into the environmental regulatory process.

Modelling Fish Dioxin Levels

The EPA methodology for predicting bioaccumulation of dioxin by fish near pulp and paper mill effluent is briefly reviewed here.⁷ Pulp and paper mill effluent dioxin concentrations were used to model fish dioxin levels via the following paradigm:

| | | | | | | |
|--------------------|---|------------------------|---|------------------|---|-------------|
| dilution factor | → | receiving water dioxin | → | uptake factor | → | fish dioxin |
|--------------------|---|------------------------|---|------------------|---|-------------|

Effluent dioxin concentrations from 104 bleach kraft pulp and paper mills in the U.S. were determined as part of a joint EPA/paper industry study known as the 104-Mill Study.⁸ These data, in conjunction with a simple dilution model, provided estimates of dioxin levels in receiving waters. It should be noted that the simple dilution model assumes that 100% of the instream dioxin (dissolved dioxin as well as dioxin sorbed to organic matter) is bioavailable to aquatic organisms. Since dioxin, a hydrophobic compound, sorbs to organic matter in the water column and sediment, an alternative model which accounts for the partitioning of dioxin between sorbed and dissolved phases (the Exposure Assessment Modelling System, or EXAMS II) was also used. EXAMS II assumes that only the dissolved dioxin is bioavailable; thus, this

⁷ EPARA, *supra* note 3.

⁸ EPA, OFFICE OF WATER REGULATIONS AND STANDARDS, US EPA PAPER INDUSTRY COOPERATIVE DIOXIN SCREENING STUDY (1988).

model predicts a lower bioavailable instream dioxin concentration than the simple dilution model. Instream dioxin concentrations determined by the simple dilution model will be considered in this paper, since 1) the results of simple dilution calculations provide an upper bound on fish tissue dioxin levels⁹.and 2) the simple dilution model is traditionally used for setting pollutant limits as part of the permitting process under the Clean Water Act.

One of a variety of factors may be used to predict fish uptake of dioxin, based on instream concentrations. EPA selected a water-to-fish factor, called a bioconcentration factor, or BCF. (BCFs are experimentally and theoretically derived ratios between fish tissue contaminant levels and water contaminant levels.) While a wide range of values for dioxin BCFs have been determined,¹⁰ to date, the most widely used BCF value for regulatory purposes is 5,000;¹¹ this value was also used in the EPARA model. This means that the model would predict fish dioxin levels to be 5,000 times the estimated total instream dioxin concentrations.

Observed Fish Dioxin Levels

From 1985 to 1987, as part of its National Study,¹² EPA conducted a sampling and analysis program to determine concentrations of dioxins and furans (as well as several other pollutants) in fish tissue from water bodies around the country. Analyses were performed on 1) bottom-feeding fish composites (of 3 to 5 adult fish) analyzed whole, as an indicator of pollutant levels at each site and 2) game fish fillet composites, to provide an indication of potential human health risks from consumption of fish.¹³ Data on fish fillet dioxin concentrations

⁹ EPANS, *supra* note 3.

¹⁰ EPA, OFFICE OF RESEARCH AND DEVELOPMENT, INTERIM REPORT ON THE ASSESSMENT OF 2,3,7,8-TETRACHLORODIBENZO-P-DIOXIN RISK TO AQUATIC LIFE AND ASSOCIATED WILDLIFE (1993).

¹¹ EPA 1984, *supra* note 3; EPA, Office of Water, Memorandum: Updated Tracking Report on State Water Quality Criteria for dioxin (2,3,7,8-dioxin) (1992).

¹² EPARA, *supra* note 3.

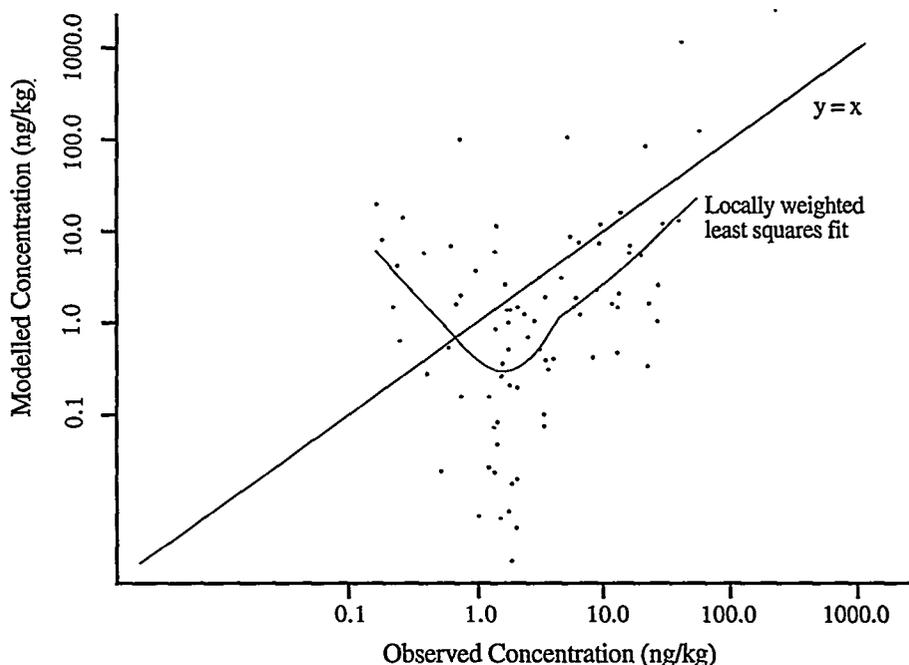
¹³ *Id.*

(ng/kg) specific to pulp and paper mill receiving waters were included as an Appendix in the EPARA.

Comparison of Modelled and Observed Fish Dioxin Levels

Predicted fish dioxin levels (derived with the simple dilution model and a BCF of 5,000) were compared to observed fish dioxin levels from as many of the 104 pulp and paper mill sites as possible (i.e. sites where both modelled and observed data were available), in order to ascertain the extent of the difference between modelled concentrations and observed concentrations. (Non-detect values were omitted, leaving 82 sites in the data set.)

Figure 1¹⁴
Plot of Concentration of Dioxin in Fish



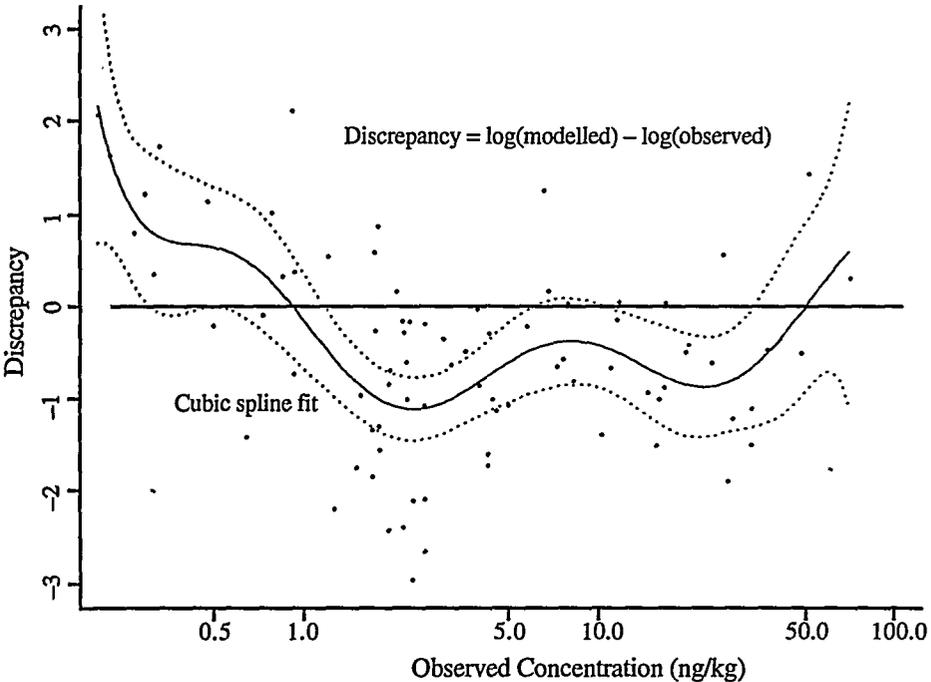
Data for observed dioxin levels are plotted in ng/kg, equivalent to parts per trillion, against data for modelled levels above. If an ideal model were used to predict fish tissue dioxin levels from known effluent

¹⁴ Both modelled and observed values are from EPARA, *supra* note 3.

concentrations and ideal sampling and analysis data were obtainable, the data should cluster about the $y = x$ line. That this is not true is illustrated in Figure 1 where a standard nonparametric smoothing technique (locally weighted least squares)¹⁵ is used to fit a curve indicating average modelled concentration for a given observed concentration. Figure 1 clearly indicates that, for low values of observed concentrations, the modelled concentrations tend to lie above the $y = x$ line, i.e. for the model to overpredict. For medium to high concentrations, the model tends to underpredict, as indicated by the fact that data points tend to lie below the $y = x$ line.

Figure 2¹⁶

Plot of Discrepancies between Modelled and Observed Concentrations vs. Observed Concentrations of Dioxin in Fish

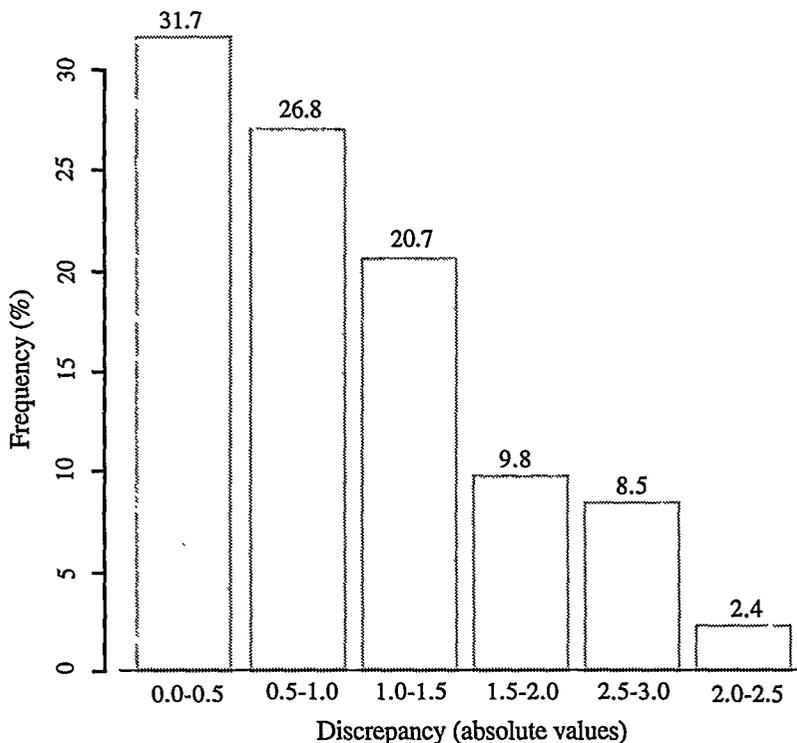


¹⁵ William S. Cleveland, *Robust locally weighted regression and smoothing scatterplots*, 24 J. AM. STATISTICAL ASSN. 829 (1979).

¹⁶ A fitted curve with a 95% confidence band is indicated. Discrepancies are differences between log concentrations, so that one unit corresponds to an order of magnitude.

To better describe these qualitative features, discrepancies between observed and predicted concentrations (the difference between the two concentrations) were plotted against the observed concentration for each of the sites. For an ideal model (and ideal observed data), the data points would be expected to congregate about $y = 0$, at least on the average. A standard curve fitting technique (cubic spline fit)¹⁷ was applied and appears as the solid curve in Figure 2. As seen there, for observed concentrations below 1 ng/kg, the model tends to overpredict dioxin levels as compared to observed levels. For values above 1 ng/kg, the model tends to underpredict fish tissue dioxin levels as compared to observed levels.

Figure 3
Distribution of Discrepancies between Modelled and Observed Log Dioxin Concentration in Fish Near 82 Pulp and Paper Mills



¹⁷ EDWARD K. BLUM, NUMERICAL ANALYSIS AND COMPUTATION THEORY AND PRACTICE (1972).

A histogram of the absolute values of discrepancies appears above. The absolute value of the discrepancy indicates the order of magnitude of difference between observed and predicted concentrations. For 41.4% of the sites, the predicted and modelled concentrations differ by at least one order of magnitude, and, for 10.9% of the sites, the difference is at least two orders of magnitude. Note particularly that for one site, the model underpredicts by three orders of magnitude relative to the observed concentration.

Discussion

Several factors are likely to contribute to the inconsistencies between predicted fish dioxin levels and observed dioxin levels in fish.

Observed data

Since only two composites were taken from each site, it is difficult to ascertain the "representativeness" of the fish sampled. For example, variations in fish dioxin levels would be expected depending on the species of the fish (interspecies differences affecting dioxin accumulation include physiology, metabolic rates, enzyme activity and lipid content), and the sex, age and habitat of the fish (e.g., proximity to effluent, migratory behavior). Thus, data on one fish composite may over- or underestimate "average" fish dioxin levels.

Modelled data

Traditionally, to predict fish dioxin levels for regulatory purposes, an extremely complex process has been modelled using very simple assumptions. For example, the EPARA utilizes a model to predict aquatic organism accumulation of dioxin which does not consider: the species of organisms, varying dioxin concentrations depending on fish size, age, lipid content, seasonal effects, the concentration of organic matter in the effluent and receiving water, as well as river flow (the latter two impacting the instream bioavailable dioxin concentration). The Food and Drug Administration (FDA) has stated:¹⁸

the inability to estimate water levels downstream from the mills or to describe the location or movement of the fish

¹⁸ FOOD AND DRUG ADMINISTRATION, QUANTITATIVE RISK ASSESSMENT COMMITTEE, REPORT (1990).

prevents any satisfactory attempt at estimating the dioxin and furan levels in fish based on the levels of dioxin and furan congeners in mill effluent.

In addition, the EPARA uses a bioconcentration factor to predict fish tissue dioxin levels. Yet, there are limitations associated with the use of bioconcentration factors outside of the laboratory.¹⁹ In particular, due to its extreme hydrophobicity, most dioxin bioaccumulation results from ingestion of dioxin-contaminated food and sediment, rather than uptake of dissolved dioxin across the gills (bioconcentration).²⁰ In fact, model underprediction at high dioxin concentrations, as shown in Figures 1 and 2, is consistent with the inadequacy of the BCF approach, which does not take into account biomagnification of dioxin.²¹ Since bioconcentration is not the primary uptake route for dioxin in fish, other uptake routes, such as ingestion, and associated accumulation factors, will have to be explored and included in the risk assessment process in order to better predict fish dioxin levels and human health risks.²² Improved bioaccumulation models²³ incorporate site specific information such as food chain structure, sediment effects, age classes of fish, and differing assimilation and depuration rates in various

¹⁹ Judy LaKind & Erik Rifkin, *Current Methods for Setting Dioxin Limits in Water Requires Reexamination*, 24 ENV'L SCI. & TECH. 963 (1990); Erik Rifkin & Judy LaKind, *Dioxin Bioaccumulation: Key to a Sound Risk Assessment Methodology*, 33 J. TOX. & ENV'L HEALTH 103 (1991); Philip M. Cook et al., *Laboratory Study of Dioxin Bioaccumulation by Lake Trout from Lake Ontario Sediments, Food Chain and Water*, in LAKE ONTARIO BIOACCUMULATION STUDY — FINAL REPORT (1990); William R. Sherman, Russell E. Keenan & Donald G. Gunster, *Reevaluation of dioxin Bioconcentration and Bioaccumulation Factors for Regulatory Purposes*, 37 J. TOX. & ENV'L HEALTH 211 (1992); Derek C.G. Muir, Wayne L. Fairchild & D. Michael Whittle, *Predicting Bioaccumulation of Chlorinated Dioxins and Furans in Fish Near Canadian Bleached Kraft Mills*, WATER POLLUTION RESH. J. CANADA, in press (1993).

²⁰ Cook et al., *supra*.

²¹ Derek Muir, personal communication.

²² Cook et al., *supra* note 19; Helen M. Goeden & Allan H. Smith, *Estimation of Human Exposure from Fish Contaminated with Dioxins and Furans Emitted by a Resource-Recovery Facility*, 9 RISK ANAL. 377 (1989).

²³ John P. Connolly, *Application of a Food Chain Model to Polychlorinated Biphenyl Contamination of the Lobster and Winter Flounder Food Chains in New Bedford Harbor*, 25 ENV'L SCI. & TECH. 760 (1991); Robert V. Thomann, John P. Connolly & Thomas F. Parkerton, *An Equilibrium Model of Organic Chemical Accumulation in Aquatic Food Webs with Sediment Interaction*, 11 ENV'L TOX. & CHEM. 615 (1992).

species and size/age classes. These site-specific models are not routinely used, however, for regulatory or advisory purposes.

Conclusions

Given the high public anxiety associated with potential dioxin exposure, the disparities between the observed and modelled fish dioxin data sets should be examined to select the superior data set for risk assessment purposes. EPA has used modelled fish dioxin levels to determine human health risks.²⁴ For example, an integral part of the EPARA was the calculation of carcinogenic and non-carcinogenic risks to human consumers of fish contaminated with dioxin. While the stated purpose of the EPARA was to estimate the "risk potential posed by the entire chlorine-bleaching pulp and paper industry," and not to rank specific mills according to risks,²⁵ this was not the outcome. In fact, the results of the EPARA were used as the basis for recommendations to state health agencies to issue fish consumption advisories, as well as to provide information to the public on the risks associated with the consumption of fish caught near specific pulp and paper mill discharges.²⁶ Thus, the EPARA's conclusions are more far-reaching than merely an overall indication of industry-wide impacts; the results have served as an important source of risk communication information.

Alternatively, the EPANS data provide the public with site-specific information on fish dioxin levels that could be translated into carcinogenic/noncarcinogenic risk information. FDA has stated that:²⁷

...the levels of dioxin and furan congeners in fish samples determined as part of the National... Study, although limited, offer the best available estimate of likely levels of dioxin congeners in fish near pulp mills.

Because of problems associated with the models used in EPARA, we believe that until an improved bioaccumulation model is incorporated into the risk assessment process, determination of human health risks associated with the consumption of fish containing dioxin should be based on sampling and analysis data.



²⁴ EPARA, *supra* note 3.

²⁵ *Id.*

²⁶ EPA, Environmental News, Sept. 24, 1990.

²⁷ *Supra* note 18.