

Prediction of PCB Concentrations in Two Species of Fish on the
Kalamazoo River

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Introduction

The Kalamazoo River drains an approximately 2000-square-mile watershed including nearly 400 miles of tributaries in Southwest Michigan. The lower approximately 80 miles of the river are part of the Allied Paper, Inc./Portage Creek/Kalamazoo River Superfund Site. Portage Creek is a tributary joining the Kalamazoo River at Kalamazoo, Michigan, the lower three miles of which are also included in the Site. The presence of polychlorinated biphenyls (PCB) was first reported in the Kalamazoo River and biota of the river in 1971. This consequently resulted in consumption advisories for fish from the Kalamazoo River and Portage Creek. Several subsequent studies have documented the presence of PCB within the surface water, sediments, floodplain soil, and biota of both the Kalamazoo River and Portage Creek, as well as in landfills adjacent to both surface water bodies. In an effort to monitor human-health and ecological risk on the river system, samples of carp and smallmouth bass were collected at several sites within the Kalamazoo River and Portage Creek. Among these sites, the greatest sampling effort occurred at Plainwell Impoundment and Lake Allegan, the most upstream- and downstream-impoundments, respectively, within the superfund site.

Assessment of the efficacy of remedial alternatives on the Kalamazoo River system requires evaluation of future risks to human and ecological health, and quantification of uncertainty in those predictions. Risks result from contact between ecological and human receptors that are of sufficient duration and intensity to elicit adverse effects (EPA, 1992). In this region, human health risks from chlorinated organic compounds such as PCB are primarily associated with ingestion of contaminated fish tissue (Birmingham et al., 1989; Newhook et al., 1988; Fitzgerald et al., 1996). Quantification of human health risks requires prediction of future fish-tissue PCB concentrations and quantification of uncertainty in those predictions.

Temporal trends of the mean or median PCB concentration in fish tissue are typically nonlinear and often modeled as a first order decay process. Stow et al. (1999) pointed out that the first order assumption requires that concentrations decay to zero, thereby precluding the possibility that contaminant concentrations may ultimately reach some steady state nonzero equilibrium or that decay rates may vary temporally. In an effort to correct this weakness, they considered two models for median PCB concentration in fish tissue; a first order decay model with nonzero asymptote (NZA) and a mixed order model (MO).

$$C_{NZA}(t) = C_F + C_0 \cdot e^{-k(t-t_0)}$$
$$C_{MO}(t) = \left[C_P^{1-q} - k \cdot (t - t_p) \cdot (1 - q) \right]^{\frac{1}{1-q}}$$

Following Stow, et al. (1999) we use a mixed-order model for the decay rate of PCB concentrations in fish tissue samples taken from the Kalamazoo River. Although this model offers more flexibility than first

order decay, it comes at a statistical cost. The most straightforward methods for prediction and quantification of uncertainty cannot be applied to these models because neither can be transformed into a linear model and analyzed (Neter et al., 1996), nor are they in the class of generalized linear models (McCullagh and Nelder, 1989) for which a significant amount of theory has been developed. Stow, et al. (1999) used non-linear least squares methods to fit the model to their data, but we found this method to be inadequate for quantifying the uncertainty of our predictions because there was no effective way to generate confidence limits. In an effort to avoid these difficulties, we used profile likelihood methods (Venzon and Moolgavkar, 1988) and performed simulations to evaluate the robustness of these methods.

Methods

Carp and smallmouth bass filets were collected at Plainwell Impoundment and Lake Allegan from 1983 to 1999 and weight, length, lipid content, and total PCB concentration were measured. PCB concentration in fish tissue is often associated with lipid content and length or weight, so we investigated the appropriateness of adjusting tissue PCB concentrations for covariation with lipid, length, and/or weight. Weight was highly correlated with length and added almost nothing to the fit when included in the presence of length, so we excluded it from models to avoid problems with multicollinearity. For this reason, we will discuss only lipid and length.

We assumed a model of the form:

$$PCB(t) = e^{X \cdot \mathbf{b}} \cdot MO(t) \cdot LN(1, \mathbf{S}),$$

$X \cdot \mathbf{b}$ is a linear model representing the relationship of log-PCB with log-length and log-lipid, $MO(t)$ is the mixed-order model, and $LN(1, \mathbf{S})$ is a lognormal error distribution. We fit this model using a two-stage process. In order to account for the effect of lipid and length, we fit a linear model with log-PCB, log-lipid and log-length treating year as a categorical variable. The factors and interactions included in the model were chosen separately for each species in order to simplify the model fitting procedure and the comparison of results within species across sites. Years with fewer than four data points were insufficient for modeling the interaction terms and so were collapsed into the nearest year (if this occurred, it was in the first year of sample data for a given site and species). Using the residuals from the linear model, we calculated the adjusted concentrations as:

$$PCB_{Adjusted} = \exp(RESID + X_{rep} \cdot \hat{\mathbf{b}}).$$

X_{rep} is a vector of the measurements for a representative fish, which we chose to be the overall average lipid and length within each species. We fit the MO temporal trend model to the adjusted data.

Due to differences between the two species and two locations, we modeled each species-location combination separately. We fit the model to the data using the maximum likelihood estimators for the parameters.

The choice of t_p can be made to coincide with some initial time t_0 (as in Stow et al., 1999), but the choice is arbitrary as long as it is within a certain (possibly infinite) interval. This interval is dependent on the parameters. The details of this relationship are provided in the appendix. We chose t_p to be 1990 because it seemed to result in better convergence for the optimization routines (see appendix). We constrained the parameters such that the model had positive concavity and was real-valued between 1975 and 2030. A more detailed account of these choices is provided in the appendix. As noted in Stow, et al (1999), as q approaches 1, the mixed-order model approaches first order decay. We assumed the errors were lognormally distributed. Rather than transforming the data and working in log-scale with normal error, we modeled the mean directly using lognormal error. The second derivative matrix of the likelihood function was ill-conditioned so we used a derivative free algorithm called the downhill simplex method developed by Nelder and Mead (1965) to maximize the likelihood function in Matlab©. We checked our assumption of lognormal error by performing a test for normality given by Looney and Gullledge (1985) on the log of the standardized residuals.

Profile likelihood approaches have been developed as a way to make inferences about a particular parameter of interest when there are a number of other parameters that are necessary for the model, but uninteresting apart from that (i.e. nuisance parameters). We were interested in predicting mean PCB concentration in fish at 2010. To estimate approximate confidence intervals for the future mean concentration, the other parameters were treated as nuisance parameters. The likelihood was calculated for fixed values of the mean parameter by maximizing over the other parameters. In order to accomplish this, we re-parameterized the model with m_{2010} (C_{2010}) as a parameter rather than C_{1990} (consider that $C_{1990}=m_{1990}$). Using the asymptotic χ^2 distribution of the generalized likelihood ratio test (Bain and Engelhardt, 1992), we generated profile-likelihood based confidence intervals for the mean in 2010.

Simulations

Using Lake Allegan carp and Plainwell Impoundment carp as the base datasets, we performed three pairs of simulations to evaluate three different aspects of our procedure. These were the coverage of the likelihood methods (lognormal error), robustness to faulty error assumptions (empirical distribution), and the failure to account for variability from the initial linear model (full procedure empirical). In each simulation we needed a ‘true’ model to use as the basis for the simulation. We chose to use the fitted models for the base datasets as the true model of the mean. These simulations differed only in how the data for the non-linear regression was generated.

The data for the lognormal error simulations was model based. Each dataset was generated having the same within year sample sizes as the base dataset. The generated data was lognormal with the mean in a given year equal to the model and shape parameter equal to the fitted value of the shape parameter for the base dataset. In the empirical distribution simulations, we used a re-sampling approach and sampled with replacement by year from the length and lipid *adjusted values* of the base dataset. Yearly sample sizes in the generated data were kept consistent with those found in the base dataset. The data for the full procedure empirical simulations was also re-sampled, except this time it was from the *unadjusted values* of the base dataset. We sampled with replacement by year from the unadjusted values of the base dataset and then adjusted these data for length and lipid with two-way interactions as described above. Yearly sample sizes in the generated data were kept consistent with those found in the base dataset.

Once the data had been generated, we followed the profile likelihood procedure described above to generate a confidence interval for the predicted mean PCB concentration in 2010. If the optimization routines did not converge for a given dataset, it was noted. After generating 1000 samples and their corresponding confidence intervals, we tabulated the percentage of generated confidence intervals that contained the true value. We used this Monte Carlo estimate of our coverage probability to evaluate how well our procedure performed under the assumptions of the simulation.

Results

At both sites for carp, we found there was at least one significant two-way interaction, so we used a linear model with two-way interactions to adjust the data. Two-way interactions were not significant for the bass at either site, nor was length. Bass at each site were adjusted using a linear model which included only time and lipid as factors. The model fitting results (including p-values) are summarized in Table 1.

Trend analyses were done on adjusted PCB concentrations for each site-species combination, with estimated parameters, predictions, and confidence limits on the predictions given in Table 2. Plots of the data along with the fitted model and prediction intervals are given in Figure 1.

We tested log standardized residuals for normality and found that for both species at Lake Allegan and carp at Plainwell Impoundment, the data were significantly different than normal ($p < 0.005$). The distribution of transformed residuals for smallmouth bass at Plainwell Impoundment was similar to a normal distribution ($p > 0.1$). Probability plots are given in Figure 2.

The simulations to evaluate whether the asymptotic coverage probability using the generalized likelihood ratio resulted in coverage probabilities of 96% and 99% for Lake Allegan and Plainwell Impoundment carp

datasets respectively. Coverage probabilities generated with the empirical distribution simulations were 99.8% and 99.7% for Lake Allegan and Plainwell Impoundment carp, respectively. The full procedure empirical simulations had coverage of 73.9% for Lake Allegan carp and 77.8% for Plainwell Impoundment carp. The simulation results are summarized in Table 3.

Discussion

Adjustments to fish concentration based on lipid and/or length are necessary to accurately interpret temporal trends in PCB concentration in fish tissue. We initially hoped that modeling on the log-log scale would untangle the time-lipid and time-length interactions. However, this was not generally the case (Table 1). Although each site-species combination varied in the terms that were found to be significant, we felt justified in using the same linear model for each species, regardless of the site. We were not interested in interpreting this linear model and so the significance of any given term was not as important to us as trying to make sure that the adjusted data would be as free as possible from the effects of lipid and length in order to isolate the temporal trend. The adjusted data are our best attempt to show what PCB concentrations would have been if all the fish within a species were identical in length and/or lipid content.

It is important to understand that our predicted PCB concentrations for 2010 are scaled to a historically representative fish. The actual PCB concentrations that are found in future fish will almost certainly continue to vary with lipid content and length. Although it may be reasonable to say that we are 95% confident that mean PCB concentrations will be within certain limits for fish similar to our representative fish, if the fish are exceptionally different from this representative, we cannot conclude that measured PCB concentrations should be within these limits. It is also important to note that we made predictions only about the *mean of the distribution* of PCB concentrations for a fish with representative length and lipid content in 2010. The profile likelihood based confidence intervals we generated are not for an individual fish. Individual fish could be expected to have greater variability, though we cannot say how much more based only on our results.

As noted in the introduction, the mixed-order model asymptotically goes to first order decay as λ goes to 1. For all of the site-species combinations we analyzed, first order decay may be a reasonable model over the study period. It should be noted that in the case of Lake Allegan carp, this approximately first-order model is strongly driven by the exceptionally large sample from 1986 with relatively high levels of PCB. Nearly half of the total data for that site-species combination comes from that year alone. When one looks at post 1990 data only, there appears to be little or no trend (Figure 3). We suspect that if PCB concentrations start to level off, it will take some time before there are enough years of data to start forcing the fitted mixed order model to differ from first order decay.

Although commonly assumed in the analysis of this type of data, lognormal error does not appear to be a statistically justifiable assumption for two of the four site-species combinations we analyzed. The carp data at both sites were significantly different from lognormal. The normal probability plot (Figure 2) for Plainwell Impoundment carp data indicates that this significance is probably due primarily to one or two outliers. The normal probability plot for Lake Allegan Carp indicates the deviation from lognormality is not due to the presence of one or two outlying data points. Given the necessity of an error distribution for likelihood based methods and the lack of commonly used alternative error distributions, we continued with the assumption of lognormal error despite the deviations we found. Recognizing the questionable nature of this assumption, we performed the simulation studies to check the robustness of our likelihood-based method and found that, with these data, the method was robust to deviations from lognormal error.

A problem with the two-step procedure using adjusted data is that residuals from the linear model may be dependent. It may also be possible to use likelihood methods to incorporate the dependencies (since they are a function of the design matrix and do not depend on the data values) explicitly. However, the increased amount of computation and coding that would be required to implement this method are probably not justified because the residuals are expected to be nearly independent for sample size much larger than the number of parameters (Graybill, 1976).

There is a second problem with the two-step procedure as we have implemented it; our results do not take into account the variability of parameter estimates in fitting the linear model. The coverage estimates from the third set of simulations indicating actual coverage may be closer to 75% than 95%. This indicates that our intervals are likely to be extremely conservative. This problem needs to be addressed before making any firm claims about what PCB concentrations are likely to be seen in the future. One possible alternative would be to use an a priori adjustment such as a lipid ratio. However, it is not clear that this would accurately account for the effect of lipid and/or length, in general. Another approach, which we will investigate in future research, is to use a reparameterization of the mixed-order model (Ratkowsky, 1990). It is hoped that this reparameterization of the mixed-order model will have better convergence properties that would allow a bootstrap approach using Newton-Raphson algorithms to fit the model.

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Appendix 1: Analytical and Numerical Issues in Fitting the Mixed-Order Model

In Stow et.al.(1999), the mixed-order model was given as a generalization of first-order exponential decay (growth). Our initial assumption was that the mixed-order model had qualitative behavior similar to that of exponential decay. It seemed reasonable to think that for different parameter regimes the behavior of the model would be such that it was decreasing (increasing) for all time. Instead, we found that the model has a variety of behaviors that can give nonsensical predictions.

Looking at the model given by Stow et al., we have:

$$C(t) = \left[C_0^{1-q} - k \cdot (t - t_0) \cdot (1 - q) \right]^{\frac{1}{1-q}},$$

where t_0 and C_0 are taken to be some initial time and concentration respectively.

For fixed δ , k , t_0 , and C_0 ,

$$[C_0^{1-q} - k \cdot (t - t_0) \cdot (1 - q)]$$

must be non-negative for a given t in order for a modeled value at that time to be real (except for special cases of δ). If $\delta = 1$ and $k = 0$, this cannot be true for all time. To see this, note that in order for it to be true, it must be the case that

$$C_0^{1-q} \geq k \cdot (t - t_0) \cdot (1 - q). \quad (1)$$

With fixed δ , k , t_0 , and C_0 , as $t \rightarrow \infty$, the right hand side of (1) is unbounded in either the positive or negative direction depending on the sign of $k(1-\delta)$. As $t \rightarrow -\infty$, the right hand side of (1) is unbounded in the opposite direction, so it follows that (1) holds on only one of the two intervals $(-\infty, t_c)$ and (t_c, ∞) , with

$$t_c = t_0 + \frac{C_0^{1-q}}{k \cdot (1 - q)}.$$

$C(t_c) = 0$ if $\delta < 1$ and is unbounded if $\delta > 1$.

In order to fit the model and use it for prediction, it was necessary to constrain the parameters in such a way that predictions have real values. The simplest constraint is to make $k(1-\delta) = 0$. This constraint guarantees the model is real valued for all time greater than t_c . Constraining the model so that it is real valued for all time greater than t_c is more than necessary. We generally found a significantly better model (as measured by the maximum likelihood) when we only constrained the model to be real valued on a specified finite interval (that is, t_c was forced to be outside the finite interval of interest, but the type of interval was not forced as with the previous constraint). A consequence of this relaxation is that it is not generally possible to make predictions for all time. However, given the ad hoc nature of this model and the time scale on which we would expect it to be accurate, we think this limitation is acceptable. The key to generating these constraints is the fact that the right hand side of (1) is monotone with respect to t . This monotonicity implies that constraining the endpoints to satisfy (1) will guarantee that the entire interval satisfies (1).

There were numerical issues that complicated the process of fitting the model to the data. A Newton-Raphson based approach to maximize the likelihood function was not reliable because the second derivative matrix was ill-conditioned with our data. Instead, we used a much slower derivative free algorithm called the downhill simplex method developed by Nelder and Mead (1965). Even this algorithm failed to converge to the optimal solution when maximizing simultaneously over all the parameters (though

it was usually not far off). This fact was discovered when the profiling approach we used in prediction converged to better fitting models.

In order to find a profile-likelihood based confidence interval for the mean in a given year t_p , we re-parameterized the model to include C_p as a parameter rather than C_0 . It is possible to show that

$$C(t) = \left[C_0^{1-q} - k \cdot (t - t_0) \cdot (1 - \mathbf{q}) \right]^{\frac{1}{(1-q)}} = \left[C_p^{1-q} - k \cdot (t - t_p) \cdot (1 - \mathbf{q}) \right]^{\frac{1}{(1-q)}}$$

as long as t_p is in the interval where the model is real valued. Although we chose t_0 to be 1975, we found that in practice, we had better convergence when we used the second form of the equation with t_p taken to be 1990. We speculate that this could be because 1975 was relatively close to t_c and the unbounded behavior of the model as it approaches t_c could effect the convergence of the optimization routines.

TABLE 1: FITTING THE LINEAR MODEL

		CARP									
		LAKE ALLEGAN					PLAINWELL IMPOUNDMENT				
	Source	DF	Type III SS	MSE	F Value	Pr > F	DF	Type III SS	MSE	F Value	Pr > F
Full Model	log(lipid)	1	0.203	0.203	0.76	0.3862	1	0.207	0.207	0.96	0.3310
	log(length)	1	0.745	0.745	2.77	0.0983	1	1.245	1.245	5.76	0.0188
	yr	8	7.854	0.982	3.65	0.0007	6	1.116	0.186	0.86	0.5281
	log(length)*yr	8	7.061	0.883	3.28	0.0018	6	1.056	0.176	0.81	0.5620
	log(length)*log(lipid)	1	0.236	0.236	0.88	0.3511	1	0.130	0.130	0.60	0.4397
	log(lipid)*yr	8	2.629	0.329	1.22	0.2905	6	1.119	0.186	0.86	0.5263
	log(lipid)*log(length)*yr	8	2.692	0.337	1.25	0.274	6	1.072	0.179	0.83	0.5532
No Three-way Interaction	log(lipid)	1	4.788	4.788	17.56	<.0001	1	0.055	0.055	0.26	0.6135
	log(length)	1	1.773	1.773	6.50	0.0117	1	0.854	0.854	4.00	0.0487
	yr	9	7.879	0.875	3.21	0.0014	6	2.932	0.489	2.29	0.0429
	log(length)*yr	9	7.251	0.806	2.96	0.0029	6	2.879	0.480	2.25	0.0465
	log(lipid)*log(length)	1	4.221	4.221	15.49	0.0001	1	0.020	0.020	0.09	0.7593
log(lipid)*yr	9	5.203	0.578	2.12	0.0308	6	2.239	0.373	1.75	0.1200	
		SMALLMOUTH BASS									
		LAKE ALLEGAN					PLAINWELL IMPOUNDMENT				
	Source	DF	Type III SS	MSE	F value	Pr>F	DF	Type III SS	MSE	F Value	Pr > F
Full Model	log(lipid)	1	0.020	0.020	0.1	0.7576	1	0.001	0.001	0.01	0.9425
	log(length)	1	0.012	0.012	0.06	0.8081	1	0.011	0.011	0.05	0.8311
	yr	3	0.257	0.086	0.41	0.7453	2	0.036	0.018	0.07	0.9288
	log(length)*yr	3	0.302	0.101	0.49	0.6946	2	0.029	0.014	0.06	0.9423
	log(length)*log(lipid)	1	0.031	0.031	0.15	0.7004	1	0.000	0.000	0.00	0.9955
	log(lipid)*yr	3	0.120	0.040	0.19	0.9008	2	0.172	0.086	0.36	0.7013
	log(lipid)*log(length)*yr	3	0.124	0.041	0.2	0.8969	2	0.171	0.086	0.36	0.7031
No Three-way Interaction	log(lipid)	1	0.418	0.418	2.14	0.1515	1	0.062	0.062	0.27	0.6064
	log(length)	1	0.019	0.019	0.10	0.7555	1	0.640	0.640	2.77	0.1052
	yr	4	0.552	0.138	0.71	0.5920	2	1.052	0.526	2.28	0.1179
	log(length)*yr	4	0.590	0.147	0.75	0.5607	2	1.110	0.555	2.40	0.1056
	log(lipid)*log(length)	1	0.473	0.473	2.42	0.1273	1	0.091	0.091	0.39	0.5355
log(lipid)*yr	4	0.755	0.189	0.97	0.4366	2	0.158	0.079	0.34	0.7124	
No Interactions	log(lipid)	1	1.159	1.159	5.58	0.0221	1	4.187	4.187	17.06	0.0002
	log(length)	1	0.031	0.031	0.15	0.7029	1	0.058	0.058	0.24	0.6283
	yr	4	21.076	5.269	25.37	<.0001	3	2.023	0.674	2.75	0.0557

TABLE 2: FITTED TIME TREND AND PREDICTED MEAN FOR 2010					
		SMALLMOUTH BASS		CARP	
		LAKE ALLEGAN	PLAINWELL IMPOUNDMENT	LAKE ALLEGAN	PLAINWELL IMPOUNDMENT
Adjustment	Length (cm)	34.8115	34.8115	50.5159	50.5159
Values	Lipid (g/kg)	0.9753	0.9753	3.4218	3.4218
Model Parameter Estimates	C_{1990}	2.3739	2.1187	6.1205	4.3600
	q	0.8096	0.9222	1.3120	1.1799
	k	0.1548	0.0827	0.0960	0.0538
	s	0.5200	0.4626	0.7304	0.4288
Predictions	lcl	0.0353	0.1312	0.2397	0.5362
	m_{2010}	0.0623	0.4017	0.6088	1.2493
	ucl	0.2077	0.9353	0.9658	1.9887

Adjustment values refer to the length and lipid values used to adjust the residuals from the initial linear models and put all fish from each species on the same scale. The model parameter estimates are the maximum likelihood estimates for the parameters of the mixed order model and the lognormal shape parameter s . Lcl and ucl are the lower and upper 95% profile likelihood based confidence limits for the maximum likelihood estimate of m_{2010} , the mean PCB concentrations in 2010.

TABLE 3: SIMULATION RESULTS						
Simulation Type	Base Dataset	Failed	Covered	Missed	%Coverage	
Lognormal Error	Lake Allegan	4	958	38	96.2	
	ainwell Impoundme	5	988	7	99.3	
Empirical Distribution	Lake Allegan	4	994	2	99.8	
	ainwell Impoundme	7	990	3	99.7	
Full Procedure Empirical Distribution	Lake Allegan	13	729	258	73.9	
	ainwell Impoundme	3	776	221	77.8	

1000 datasets were generated for each simulation. For each generated dataset, it was noted whether the procedures failed to converge (Failed), the generated confidence interval contained the true value (Covered), or did not contain the true mean (Missed). A Monte Carlo estimate for the coverage of confidence interval (%Coverage) is given by Covered/[Missed+Covered].

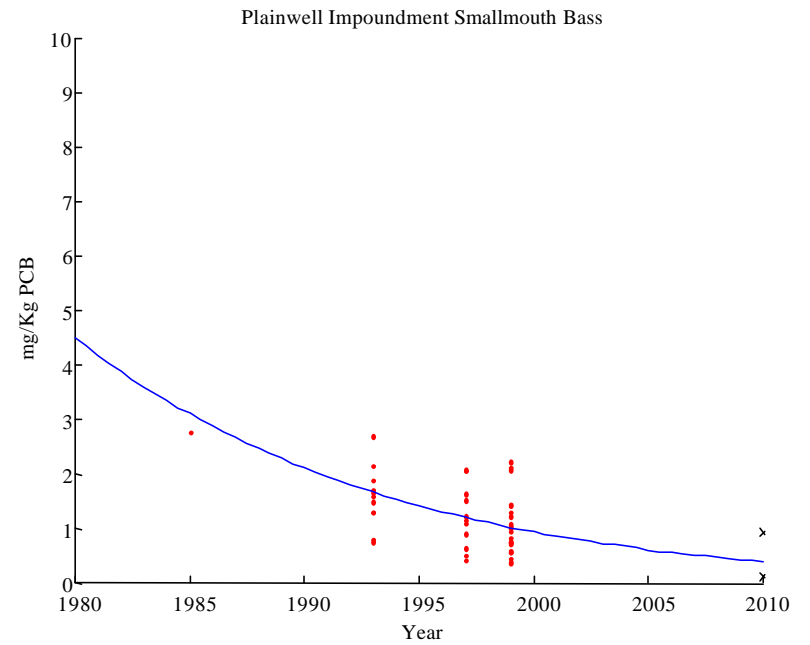
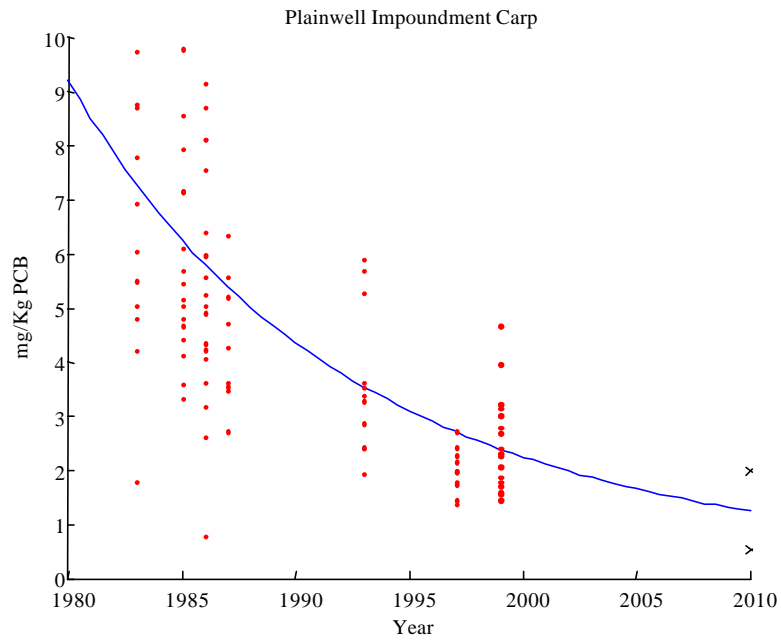
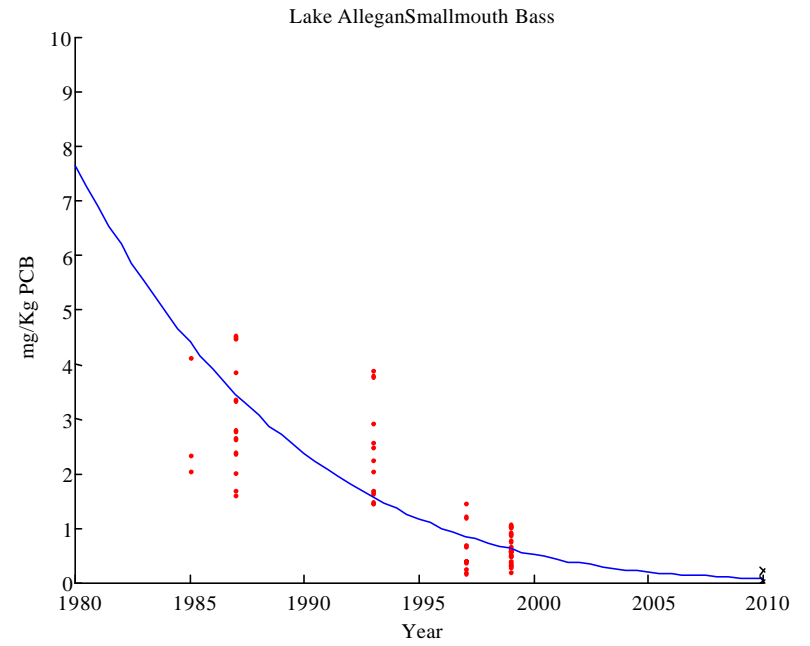
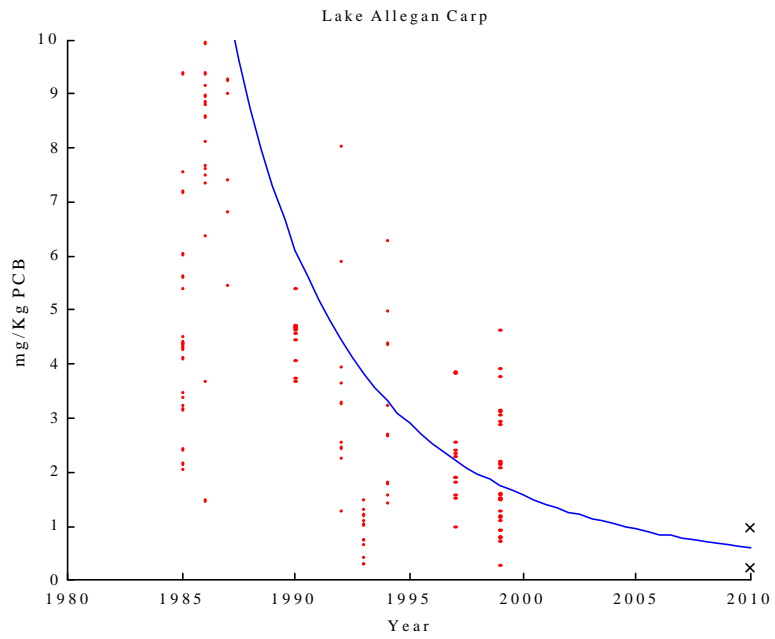


Figure 1: Fitted Model and Confidence Limits for Predicted 2010 Mean

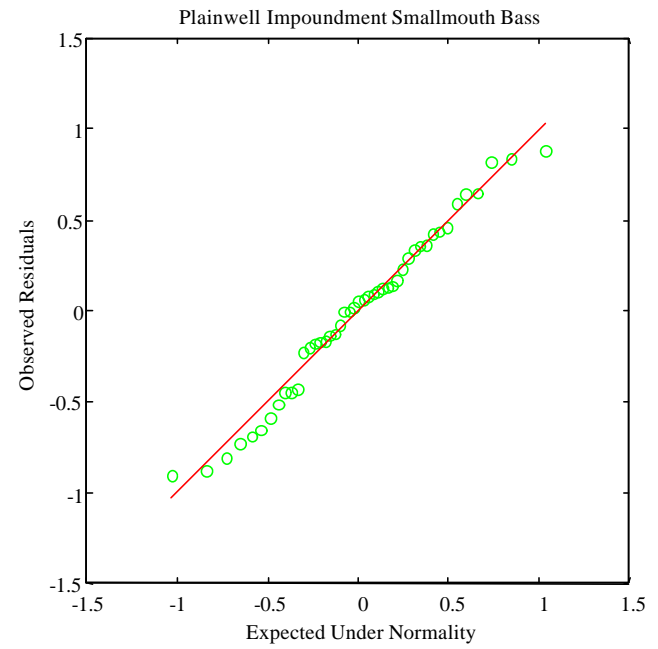
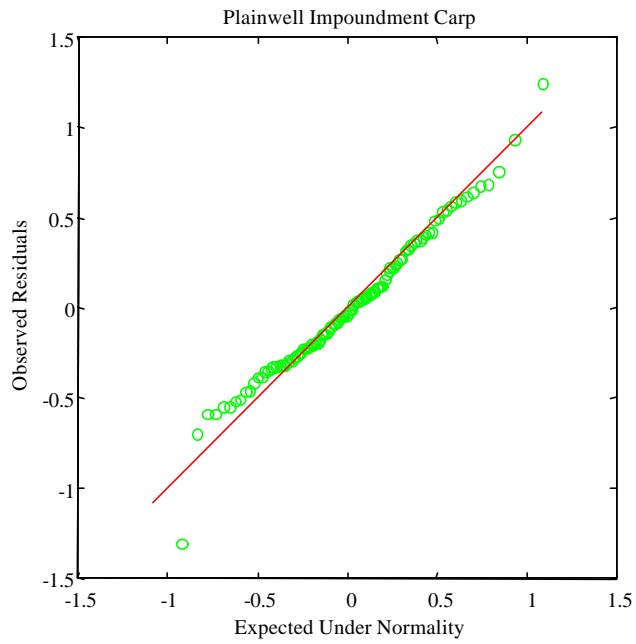
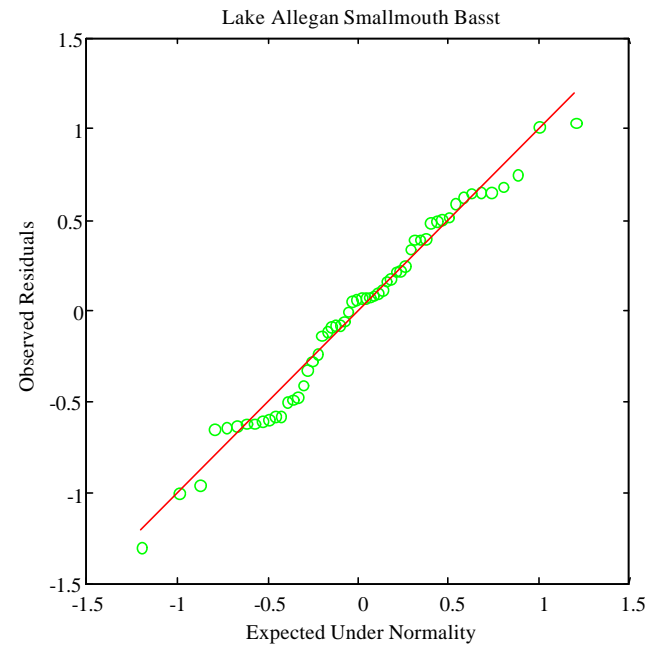
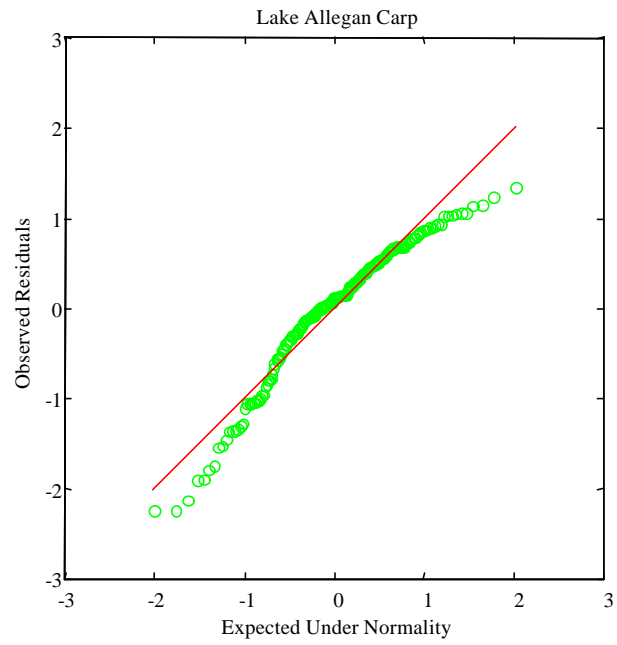


Figure 2: Normal Probability Plots

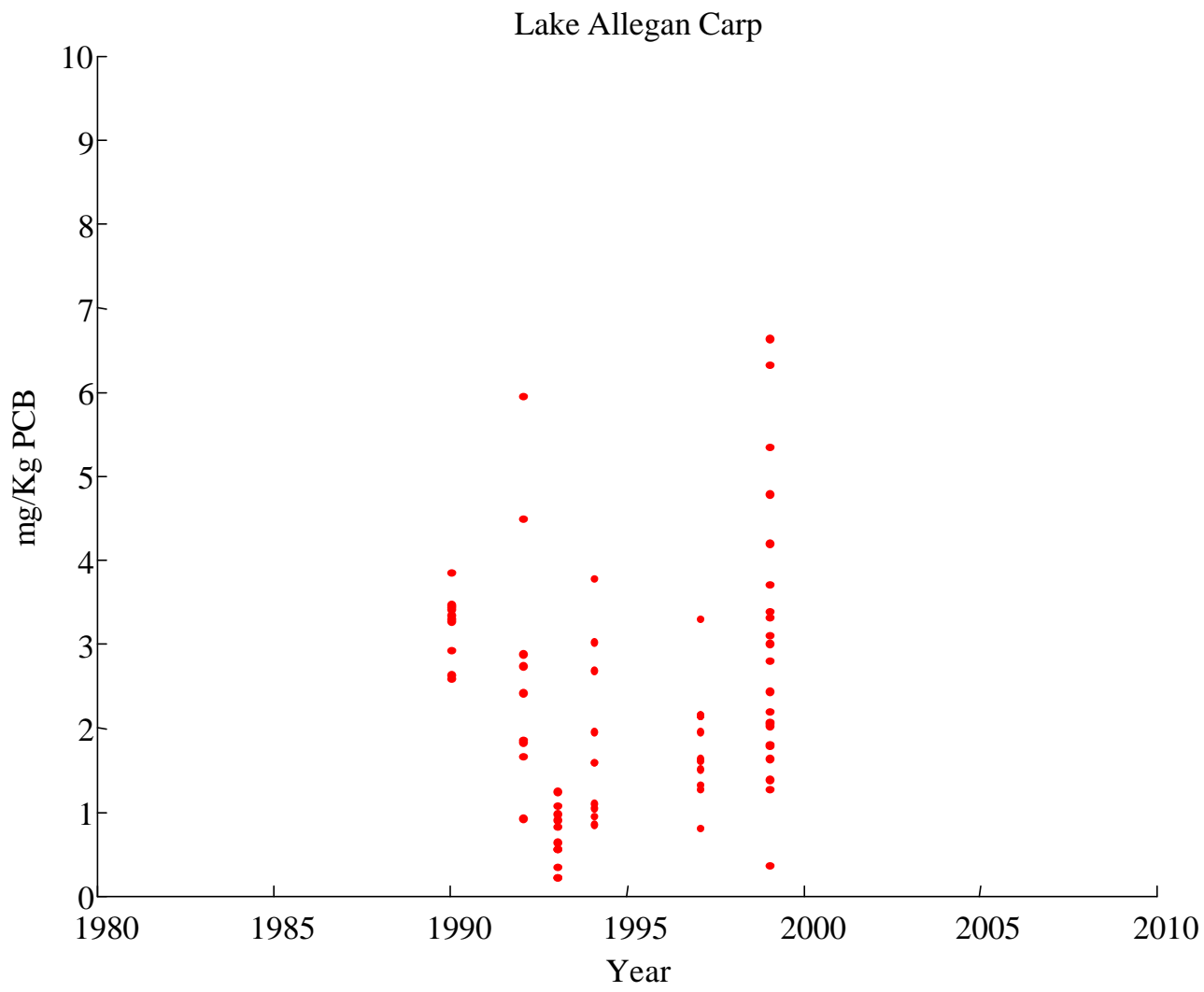


Figure 3: Lake Allegan Carp Post 1990 Data