

# Patterns of co-contamination in freshwater and marine fish of the northeastern United States

Kimberly Bourne (✉ [Kimberly.bourne@duke.edu](mailto:Kimberly.bourne@duke.edu))

Duke University

Amanda Curtis

University of Illinois at Urbana-Champaign

Jonathan Chipman

Dartmouth College

Celia Y. Chen

Dartmouth College

Mark E. Borsuk

Duke University

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## Research Article

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# Abstract

Persistent, bioaccumulative, and toxic contaminants (PBTs) are known to co-occur in fish tissue, yet this covariance has not been explicitly incorporated into model-based risk assessments that inform fish consumption advisories. We utilize available US EPA datasets to statistically model the covariance among PBT concentrations in fish tissue and the dependence of this covariance on waterbody and watershed conditions. We find that most PBTs positively covary, whether fish were collected in rivers, lakes, or coastal waters. Mercury in lakes, and mercury, PFCs, and heptachlor in rivers, covary negatively with the other PBTs. While much of the variance and covariance in PBTs can be statistically related to fish characteristics and watershed and waterbody conditions, a large amount remains in model residuals. This implies that single contaminant models, even if highly precise, can misestimate total health risk by neglecting the substantial covariance with other PBTs that is left unmodelled.

## 1.0 Introduction

Human activities involving the use of chemicals and fossil fuels in industrial, residential, and agricultural settings have led to the presence of contaminants throughout the natural environment [1–3]. Persistent, bioaccumulative and toxic substances (PBTs) continue to pose human health risk despite efforts to ban their use or curb their emissions [4]. Of particular concern are mercury and organic compounds including pesticides, fungicides, industrial chemicals, and their metabolites. The concentrations of these environmental contaminants are particularly high in the northeastern U.S. due to dense urbanization, a history of agriculture, and proximity to coal fired power plants [5].

The primary route of human exposure to PBTs is through the consumption of fish [6]. These substances bioaccumulate in food chains [7, 8], leading to concentrations in fish tissue that can cause a variety of health problems, especially to vulnerable populations such as children, the elderly, pregnant women, and their unborn children [9]. The northeastern U.S. has one of the highest consumption rates of fish in the country, so its residents are particularly at risk of contaminant exposure via fish tissue [10].

In the U.S., the health risks posed by contaminated fish are managed using consumption advisories in the form of recommended meal restrictions. However, advisories vary from state to state in how they are calculated and the number of contaminants included. In a comparative analysis of state consumption advisories across the US, it was found that about a third of states based advisories on a single contaminant either statewide or for each waterbody. Almost half of states include advisories based on multiple contaminants but offer no explanation as to how this advice is developed [11]. There is evidence that many of the common contaminants included in these advisories have additive or synergistic health effects when consumed simultaneously, but many chemical mixtures have yet to be tested. The toxicity of chemical mixtures is therefore a critical area of health research [11–14], and the CDC has an ongoing effort to build a database of interaction profiles under the Agency for Toxic Substances and Disease Registry [15]. As the nature of most interactions is currently unknown, the EPA recommends that, as a

precaution, additivity should be assumed for carcinogens and contaminants with similar mechanisms of toxicity [16].

Even if the health impacts of chemical mixtures were to be well-characterized, a separate but related question is, “What are the particular mixtures to which various populations are being exposed?” Answering this question requires analysis of the covariance of contaminants in fish tissue and the dependence of this covariance on local watershed and waterbody conditions. Most studies of fish tissue concentrations to date have only considered contaminants individually or in small groups [4, 17–20]. However, because we expect many contaminants to have common sources and similar environmental fate and transport mechanisms. A complete assessment of health risk should consider many contaminants simultaneously and with an increased focus on measuring many contaminants at once, the data required to do such an assessment is more available than ever [21].

In this paper, we apply a novel, multivariate Bayesian modeling method to analyze publicly available fish tissue data from the EPA to address the following three questions: 1) How do measured PBT concentrations in fish tissue covary across different waterbodies and how does this covariance differ between lakes, rivers, and coastal zones? 2) How much of the observed PBT covariance can be explained by models that account for local watershed and waterbody factors? 3) What are the practical implications of PBT covariance that cannot be addressed by models and therefore remains a component of model predictive uncertainty?

## 2.0 Methods

### 2.1 Data Description

Our analysis employed three datasets that are publicly available from the U.S. EPA: the Environmental Monitoring and Assessment Program (EMAP) Northeast Lakes data (1991–1994), the National Rivers and Streams Assessment (NRSA) data (2008–2009), and the National Coastal Assessment (NCA) northeast region data (2000–2006) [22–24]. The sample sites included in this study are mapped in Fig. 1. Measured PBTs are listed in Table 1, and fish, waterbody, and watershed variables are listed in Table 2. Most data come from the original datasets, however land use characteristics were derived from the National Land Cover Database (NLCD) for the years 2000 for the EMAP data and 2006 for the NRSA data [25]. A combination of the 2000 and 2006 NLCD data was used for NCA based on which year was closest in time to the fish tissue samples. Because watersheds of the NCA coastal locations are not well-defined, land use characteristics were derived from any land area within 200 km of the point at which the fish tissue samples were collected. Fish species trophic levels were determined through literature review (see Appendix A for details).

### 2.2 Data Analysis

Data were analyzed using the R package GJAM [26]. This package is designed to model multivariate response variables that can be both discrete and continuously valued. Importantly for our purpose, all

response variables representing PBT concentrations can be modeled simultaneously, with explicit representation of their covariance structure. Also importantly, GJAM appropriately handles data which are variously censored, a common trait of fish tissue measurements that may fall below method detection limits (MDLs) that differ by contaminant. GJAM treats values reported to be below the MDL as being unknown on the interval between zero and the MDL, then simulates these values from a latent scale derived from the truncated distribution of measured values above the MDL. This approach avoids biases that are incurred when censored values are replaced by zeros, the MDL, or any other specific value [27–29].

Our GJAM model structure is depicted graphically in Fig. 2. In this model, the measured (possibly censored and transformed) concentrations  $Y_{ik}$  of contaminant  $k$  in fish tissue sample  $i$  are assumed to be the manifestation of the true (uncensored) concentrations  $W_{ik}$  with mean  $\mu_{ik}$  (each modeled as a linear function of  $j$  measured environmental and fish attribute predictors  $X_{ij}$  and parameters  $B_{jk}$ ) and model error described by a common multivariate distribution with variance-covariance matrix  $\Sigma_{kk}$ . Thus, the covariance of predicted mean concentrations  $\mu_{ik}$  represents the portion of the overall contaminant covariance that is explainable by differences in environmental and fish attributes, while the matrix  $\Sigma_{kk}$  represents the portion of the covariance that cannot be explained by these factors and is based on the joint distribution of model residuals. This remaining covariation should not be ignored, but rather explicitly considered in waterbody assessments, as it represents the residual risk of simultaneous exposure to multiple contaminants.

GJAM employs a Bayesian approach to model fitting. Parameter and variance-covariance estimates are represented by the posterior distribution  $P[B, \Sigma | X, Y]$ , which is derived from Bayes theorem as the product of a likelihood function of the fish tissue concentration measurements,  $P[Y | X, B, \Sigma]$ , and a joint prior distribution of the parameters and model error variance-covariance matrix,  $P[B, \Sigma]$ :

$$P[B, \Sigma | X, Y] = \frac{P[Y | X, B, \Sigma] \times P[B, \Sigma]}{\iint P[Y | X, B, \Sigma] \times P[B, \Sigma] dB d\Sigma} \# (1)$$

We used a multivariate normal likelihood function, after a square-root transformation of concentration measurements to counter skewness and maintain strictly positive values. To minimize the influence of the prior, we used a widely-dispersed multivariate normal distribution on the parameters with mean of zero and a Jeffrey's prior on the variances and covariances.

We simulated the posterior distribution using Gibbs sampling, a version of Markov Chain Monte Carlo (MCMC) simulation implemented in GJAM. This allows for estimation of missing and censored values, which were a common occurrence in our data. At each iteration of the Gibbs sampler, missing and censored data are replaced by inferred values derived from that iteration's posterior parameter estimates.

Each of the three EPA datasets was analyzed separately using the variables available. Environmental predictor variables (Table 2) included those used in previous analyses of individual contaminants [18, 30–32]. Fish length, trophic level, lipid content, and species were used to control for fish characteristics,

except for EMAP which does not include fish length and NRSA which does not include lipid concentration. Further, in the NRSA and NCA datasets, trophic level and species are redundant, so only species data were considered. Species data is included as a categorical variable in the model with GJAM selecting the species with the most data points as the reference species. Contaminants included all PBTs reported by each dataset (Table 1), with efforts to achieve consistency when possible. For example, not all datasets report specific PCB congeners, so we considered only total PCBs. Contaminant properties (e.g., octanol-water partition coefficient) were not considered explicitly, as their effects are presumed to be captured by the estimated model parameters. If only one observation existed for a species in a dataset, that observation was removed. Our analysis thus employed 222 (EMAP), 1001 (NRSA), and 118 (NCA) fish tissue samples across 158, 679, and 118 locations, respectively.

## 2.3 Model Selection

We present results for the model of each dataset that includes the combination of environmental predictor variables leading to the minimal value of the deviance information criterion (DIC). The DIC is a measure of goodness of fit, penalizing for model complexity [33, 34]. Unlike other criteria such as the Akaike Information Criterion (AIC), the DIC is easily calculated from MCMC samples. We explored the space of possible models using a two-way stepwise algorithm, initiated with a minimal model including only tissue type and species as predictors, and proceeding to alternately add and remove additional predictors to reduce the DIC value.

## 3.0 Results

Covariation among contaminants is presented for each dataset in the form of three normalized variance-covariance matrices of (square-root transformed) fish tissue concentrations representing, respectively: (a) variation in the original measured data, (b) variation attributable to modeled factors, and (c) variation left unexplained by modeled factors. Normalization involved dividing each entry of each variance-covariance matrix by the product of standard deviations of the measured values of the two respective contaminants. Thus, in matrix (a) all diagonal elements equal one and off-diagonal elements represent the correlations between measured contaminant values. In matrix (b), the (off-)diagonal elements represent the normalized (co-) variance explained by the model (equivalent to model  $R^2$  values in the case of the diagonal elements). The elements of matrix (c), therefore, represent the (co-)variance not explained by the model.

Parameter estimates for each data set are presented as matrices of the normalized sensitivity of (square-root transformed) fish tissue contaminant concentrations to each fish attribute and environmental predictor variable contained in the final model. Sensitivity values were calculated by multiplying each mean model parameter estimate by the average of its corresponding predictor variable and dividing by the average of the corresponding (square-root transformed) contaminant concentration. To better compare these values across all predictor-contaminant combinations, they are displayed as the signed quantiles across all sensitivity values, and thus range from -1 to 1.

## 3.1 EMAP Lakes Data

The covariance structure of the EMAP data (Fig. 3, matrix a) reveals that in the tissues of lake fish, mercury is negatively correlated with all other PBTs, which are positively correlated with one another. A linear model accounting for environmental and fish attributes is able to explain a moderate proportion of the variance in individual contaminants, with  $R^2$  values ranging from 0.29 to 0.72 (diagonal elements in matrix b, with specific values given in Appendix B), and a similar proportion of the covariance among contaminants (ratio of the off-diagonal elements in matrix a to those in matrix b) including the negative correlations between mercury and other contaminants. Overall, the chlordanes and related chemicals are predicted most accurately, both individually and in concert, while aldrin, mercury, hexachlorobenzene, and DDT and its breakdown products, as well as their covariances, are predicted least well. A moderate amount of (co-)variance remains unexplained (matrix c), as well as a mild negative covariance between hexachlorobenzene and both aldrin and oxychlordanes that was induced by the model's slight overestimate of the covariance between these contaminants.

The model best accounting for the variances and covariances among fish tissue contaminants in the EMAP data is one that includes, in addition to trophic level, lipid content, and fish species, the environmental variables percent agriculture, alkalinity, latitude, urban land, and zooplankton abundance (Fig. 4). Significant parameters in this model indicate that the measured contaminants generally increase with greater trophic level and lipid content and, relative to yellow perch, are generally greater in lake trout and white perch and lower in Allegheny pearl dace, blue gill, brown bulkhead, and chain pickerel, with a few exceptions. Both lake trout and white perch have higher concentrations of all contaminants. Contaminant concentrations are generally lower at sites from higher latitudes, except for aldrin and mercury which are higher. Greater levels of urban land and zooplankton abundance are each generally associated with higher contaminant concentrations. Fish tissue concentrations are generally most sensitive to trophic level, species, and latitude.

## 3.2 NRSA River Data

The NRSA data (Fig. 5, matrix a) reveal that mercury, heptachlor, and total PFC concentrations negatively covary with most other PBTs in the tissue of fish from rivers and streams, and that these other PBTs positively covary with each other. Only a fraction of this covariance structure was captured in the modeled mean concentrations (matrix b) with proportions of explained co-variance ranging from zero to 0.59. Endosulfan II was predicted especially poorly, as the model only captured 16% of the variation in this contaminant, and a comparably low proportion of its covariance with other contaminants. Mercury, heptachlorepoxyde, the nonachlors, the chlordanes, and their covariances were predicted reasonably well, with  $R^2$  values all above 0.3. A substantial amount of variance and covariance in the NRSA data remains unattributable to modeled factors (matrix c).

The model best accounting for the variances and covariances of fish tissue contaminants in the NRSA data is one that includes, in addition to lipid content, total length, and species, the environmental

variables: riparian vegetation, instream dissolved organic carbon (DOC) and pH (Fig. 6). Riparian vegetation is a positive and significant predictor for total PFCs but a significant negative predictor for heptachlor. DOC is a significant positive predictor for mercury, total PFCs, endrin ketone, and heptachlorepoxyde. For nearly all contaminants, pH is a significant and positive predictor of fish tissue contaminant concentration. Heptachlor and mercury generally show quite different relationships across species (relative to smallmouth bass) than the other contaminants.

### 3.3 NCA Data

In coastal waterways, all contaminants positively covary (Fig. 7, matrix a). This dataset is limited, however, in that many contaminants were not measured concurrently. DDT and related metabolites covary particularly strongly with each other and with transnonachlor, with normalized covariance (correlation) values close to 1. While the best fitting model yields  $R^2$  values for 4,4-DDT, 4,4-DDD, 4,4-DDE, total PCBs, and transnonachlor from 0.43 to 0.66, most others are quite low. Additionally, only small portion of the observed covariance among contaminants is reproduced by the model (matrix b), with the vast majority remaining unexplained (matrix c).

For the NCA data, the best fitting model (Fig. 8) includes the fish characteristics: tissue type (offal generally lower than filet and whole fish generally greater), lipid content (weak positive and negative relations with contaminant concentration), fish length (positive when significantly different from zero), and species (mixed sign and significance, but perch and catfish species notably positive). Significant environmental variables include: the percentages of agricultural, forested, and urban land, as well as the total nitrogen concentration, all of mixed sign and significance depending on the contaminant. Comparisons between species show channel catfish and white perch having relatively high concentrations of all contaminants in their tissues.

## 4.0 Discussion

The data on contaminant concentrations in fish tissue show that PBTs generally covary positively across all types of waterbodies, with the exception of mercury against other contaminants in both lakes and rivers, and total PFCs and heptachlor against other contaminants in rivers. In coastal waters, these particular contaminants were not measured concurrently with others. The positive covariance among most contaminants implies that, lacking any more detailed information on fish characteristics or environmental conditions, one should assume that if one contaminant is elevated at a site, most others will be as well. However, in lakes and rivers, mercury is low when other contaminants are high in fish tissue. Gandhi et al. (2017) show empirically that the covariance patterns present in fish tissue data from the Great Lakes imply that the current practice of basing fish consumption advisories on the single most restrictive contaminant is not adequately protective. Gandhi et al. note, however, that both the magnitude and sign of the covariance among contaminants can differ according to location and fish species. They thus call for a more generalizable statistical approach for performing chemical mixture assessments

than the simulation-based method they employ. In particular, they call not for multiple single-contaminant models, but rather for single multi-contaminant models such as that employed in the current study.

The performance of our multivariate modelling approach differed across the three data sets. In lakes, the joint distribution of multiple contaminants was captured reasonably well, with  $R^2$  values as high as 0.72 and never below 0.29, and similar proportions of covariance among contaminants explained by the model. This is a level of explanatory power that is similar to previously published, single-contaminant models [5,30, 35]. The model performed less well in rivers, with captured proportions of variances and covariances ranging from 0.16 to 0.59. This may be because riverine fish may occupy a variety of locations over the course of their lifetimes and therefore land use variables are not predictive fish tissue contaminant concentrations. The model of coastal marine systems was able to account for only a small proportion of the variance and covariance among fish tissue contaminant concentrations ( $R^2$  values ranging from 0.04 to 0.66, with most in the lower half of this range). This may be because of the wider variety of tissue types, species, and corresponding life cycles of fish included in this data set. Also, as for rivers, fish living in coastal areas are highly mobile and may not spend the majority of their time near the land used to calculate land use percentages. In general across all waterbodies, the contaminants best fit by the models were mercury, total PCBs, DDT and its metabolites, the nonachlors, and oxychlordanes and their covariances.

The majority of the (co-)variation accounted for by the models could be attributed to fish characteristics, such as tissue type, lipid content, length, trophic level, and species. However, watershed land uses and waterbody characteristics were also significant predictors – again in line with the results of single-contaminant modeling [5, 30, 36]. In the model of the EMAP lake and river data, for example, lakes with watersheds having more urban land showed consistently higher fish tissue contaminant concentrations (except for mercury). For coastal fish, agricultural and forested land were significant predictors of contaminant concentrations, although the sign of these relationships varied by contaminant.

Water chemistry has been shown to be important in the bioaccumulation of Hg but is less well explored for organic contaminants [30, 37–39]. We found that fish tissue mercury concentrations decreased with greater alkalinity in lakes and pH in rivers. This is consistent with the finding that acidity is associated with enhanced mercury methylation and uptake in lower alkalinity and pH systems [38]. Concentrations of contaminants other than mercury were positively associated with pH levels in rivers. While metal contaminants are more bioavailable at lower pH, there is little research on the relationship of pH with organic contaminant bioavailability. Mercury, PFCs, and some other contaminants were found to be positively associated with DOC in riverine fish from the NRSA dataset. Like pH, the influence of DOC on metal transport and bioavailability are known but not commonly studied for organic contaminants. Mercury concentrations in water are positively related to DOC, but DOC appears to reduce the bioaccumulation of mercury particularly at high DOC ranges [40, 41]. In the few studies that exist for organic contaminants, the influence of DOC does not show consistent patterns [42–45].

Our intent with this study was to demonstrate a method of modeling multiple contaminants simultaneously, using a Bayesian approach that can account for both missing and censored values (i.e. below detection limit). With such a model, one can assess how much of the covariance present among fish tissue contaminants can be accounted for by similar responses to modeled factors, and how much remains unaccounted for. Because we used a simple linear approach to model many contaminants, in many species, across many aquatic environments, each with particular fish characteristics, water chemistry attributes, and land use variables, our model results may not be as precise or mechanistically interpretable as more focused analyses. Yet, the  $R^2$  values we were able to obtain are comparable to those of previously-published statistical models of single contaminants.

A more novel contribution of this study is our demonstration that, while a multivariate model can explain some of the covariance inherently present among contaminants, the proportion of covariance that remains *unexplained* is at least as large as the proportion of *variance* left unexplained for individual contaminants. This means that the application of multiple single-contaminant models – even those that fully account for predictive uncertainty – misestimate true human health risk. This is because unexplained covariance implies that, even after all modeled factors are taken into account, the likelihood of unusually high (or low) levels of one contaminant at a site are not independent of the likelihood of unusually high (or low) levels of another contaminant. The fact that most contaminants (except mercury and heptachlor in rivers) display unexplained variation with positive covariance means that unexpectedly high levels of multiple contaminants are likely to occur simultaneously, exacerbating human health risk.

It is important to emphasize that this underestimation of health risk that results from ignorance of positive error covariance among contaminants is a systematic bias and not just an overlooked contribution to predictive uncertainty. This is clear from the fact that the variance of the sum of two correlated random variables  $X_1$  and  $X_2$ , for example, is computed as:

$$\sigma_{X_1+X_2}^2 = \sigma_{X_1}^2 + \sigma_{X_2}^2 + 2 \cdot Cov(X_1, X_2) = \sigma_{X_1}^2 + \sigma_{X_2}^2 + 2 \cdot \rho \cdot \sigma_{X_1} \cdot \sigma_{X_2} \quad (2)$$

where  $\sigma^2$  represents variance,  $Cov()$  represents covariance, and  $\rho$  represents correlation between  $X_1$  and  $X_2$ . Thus, with positive covariance/correlation between two variables, the variance of the sum is always greater than the sum of the variances. Thus, the probability of high concentrations will be larger than would be calculated by assuming independence between the variables. This result can easily be extended to more than two random variables and to any linear combination of variables, making it applicable to the analysis of contaminants assumed to have additive health effects [16].

Models can be useful tools for predicting fish tissue contaminant concentrations at unsampled sites or under new conditions as a function of watershed attributes, water quality measures, and fish characteristics. However, model-based estimates of human dietary exposure for the purposes of setting fish consumption advisories need to account for the full distribution of possible contaminant concentrations, not just single point estimates. Several studies have demonstrated the value of a Bayesian approach in this context, as these methods allow for the characterization of both parametric

and structural error [35, 46, 47]. This approach has long been advocated in the academic literature [48, 49] and is starting to make its way into fish consumption advisory decisions, at least for individual contaminants [50]. The present study is the logical extension of that approach to multiple contaminants, acknowledging that, if one contaminant happens to be unexpectedly elevated at a site, others are likely to be as well.

While our study demonstrates a moderate ability to predict contaminant concentrations with a multivariate model, much of the covariance among contaminants remains in the residuals. This indicates that the presence and composition of mixtures are relatively unpredictable, even knowing watershed attributes, water quality measures, and fish characteristics. Further analyses might reveal the importance of including additional predictor variables or formulating nonlinear models, and thus more precisely characterize the patterns of suites of contaminants. In the meantime, uncertainty in both single contaminants and their co-occurrence needs to be reflected in fish consumption advisories and in more holistic assessments of risk-risk and risk-benefit tradeoffs [51, 52].

## **Declarations**

### **6.1 Acknowledgements**

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### **6.2 Author Contributions**

K.B. performed the analysis and created the figures. A.C., J.C. and K.B. collected the data. M.E.B. and K.B. wrote the manuscript. All authors reviewed the manuscript.

### **6.3 Declaration of interests**

The authors of this manuscript declare no competing financial or non-financial interests.

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### **6.5 Ethics Declaration Statement**

As this study utilizes previously collected data, there are no ethics standard or forms that apply.

### **6.6 Consent to Participate**

Not Applicable

### **6.7 Consent for Publication**

All authors of this manuscript have reviewed it and consent to its publication.

## 6.8 Data Availability

Data analyzed in this study can be found at the following sources. Additional data taken from multiple publications is described in the appendices.

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## Tables

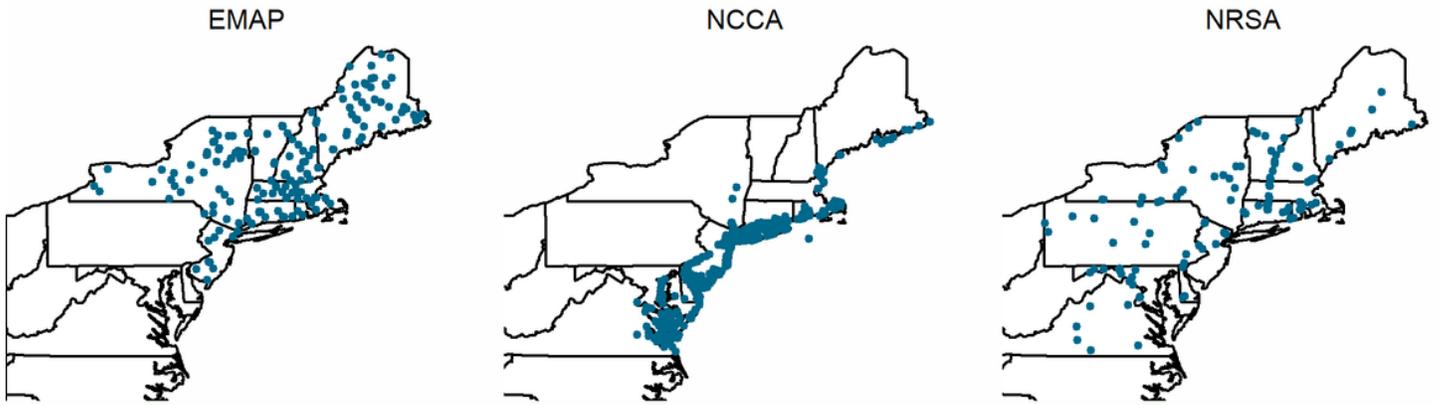
Table 1  
PBTs modelled for each data source.

EMAP	NRSA	NCA
2,4'-DDE	Alpha Chlordane	2,4'-DDE
2,4'-DDD	Cis-Nonachlor	2,4'-DDT
2,4'-DDT	Dieldrin	4,4'-DDD
4,4'-DDD	Endosulfan II	4,4'-DDE
Aldrin	Endrin	4,4'-DDT
Alpha Chlordane	Endrin Ketone	2,6'-Dimethylnaphthalene
Cis-Nonachlor	Gamma Chlordane	Acenaphthylene
Dieldrin	Heptachlor	Anthracene
Gamma Chlordane	Heptachlorepoxyde	Benzo(b)fluoranthene
Hexachlorobenzene	Mercury	Biphenyl
Mercury	Oxychlordane	Chrysene
Oxychlordane	Total PCBs	Fluoranthene
Total PCBs	Total PFCs	Mercury
Trans-Nonachlor	Trans-Nonachlor	Methylnaphthalene
		Naphthalene
		Total PCBs
		Pyrene
		Trans-Nonachlor

Table 2  
Fish, waterbody, and watershed variables considered by data source. (See Appendix A for definitions and units.)

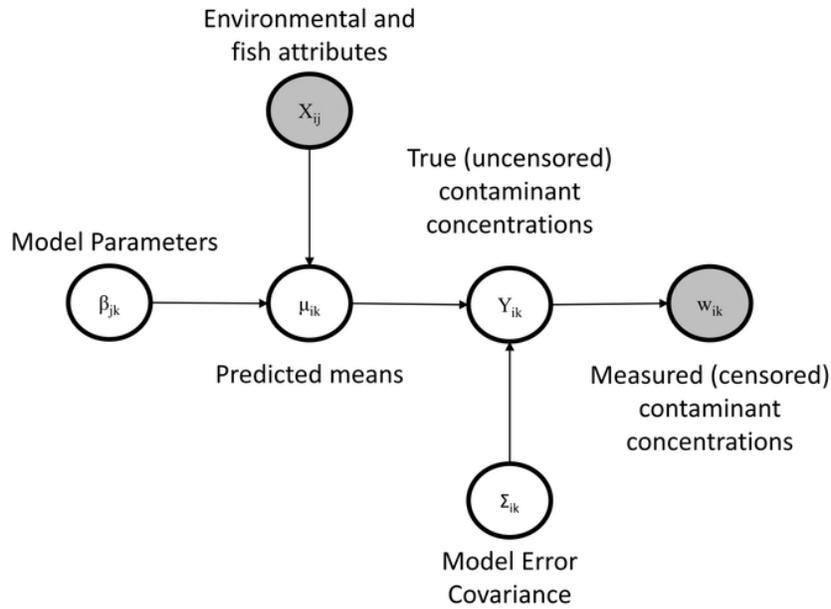
<b>EMAP</b>	<b>NRSA</b>	<b>NCA</b>
Estimated Trophic Level	Fish Length	Chlorophyll a
Species	Species	Total Nitrogen
Lipids	Percent Ephemeroptera	Sediment Toxicity
Total Zooplankton Abundance	Total Nitrogen	Abundance of Tolerant Benthic Organisms
Alkalinity	pH	Abundance of Sensitive Benthic Organisms
DOC	DOC	Tissue Type
Chlorophyll a	Watershed Area	Percent Agriculture
Percent Human Disturbance	Percent Agriculture	Percent Forest
Percent Mining	Percent Urban	Percent Impervious
Percent Urban	Percent Shrubgrass	Percent Urban
Watershed Area	Percent Wetland	Percent Wetlands
Percent Agriculture	Percent Impervious	Latitude
Latitude	Barrier Disturbances	Longitude
Longitude	Riparian Vegetation	Lipids
	Longitude	Length
	Latitude	Species

## Figures



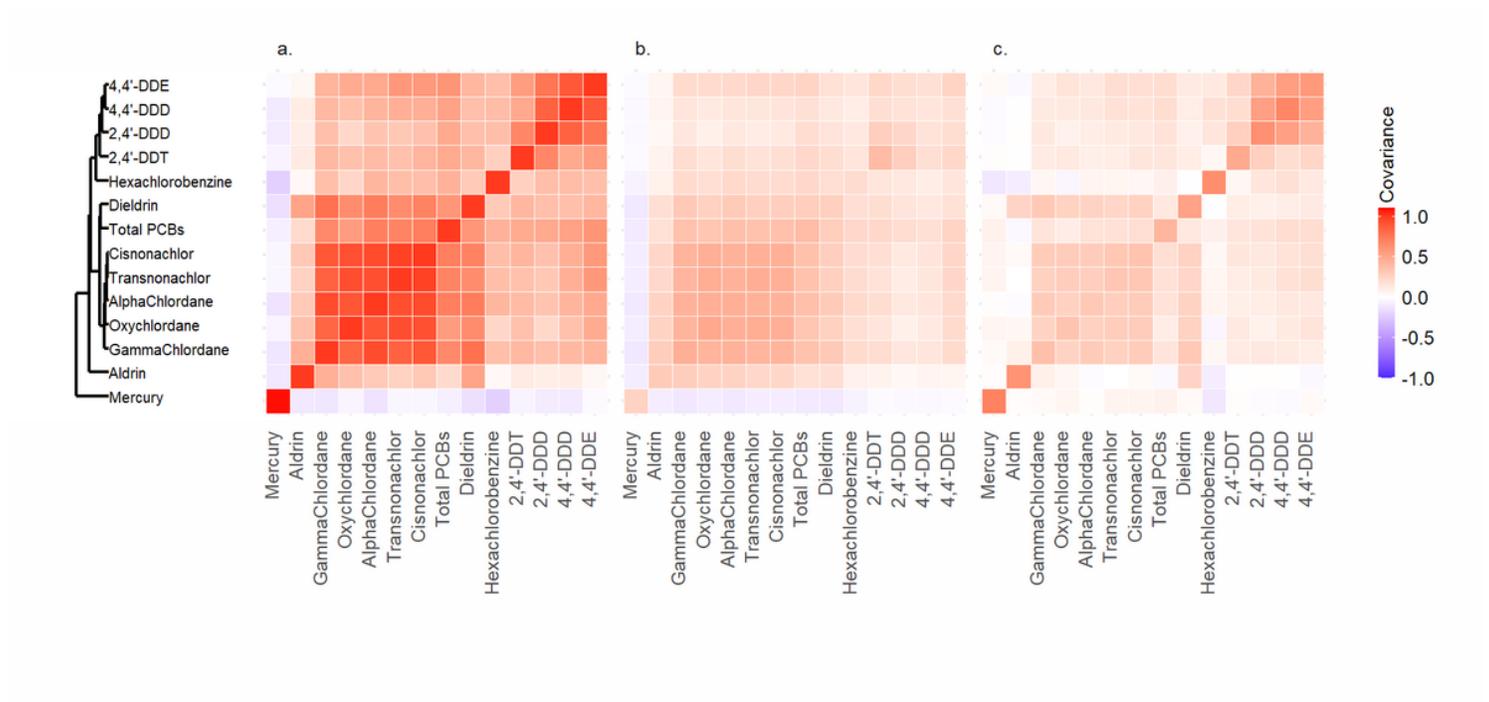
**Figure 1**

Spatial representation of the sample locations included in this study for each dataset.



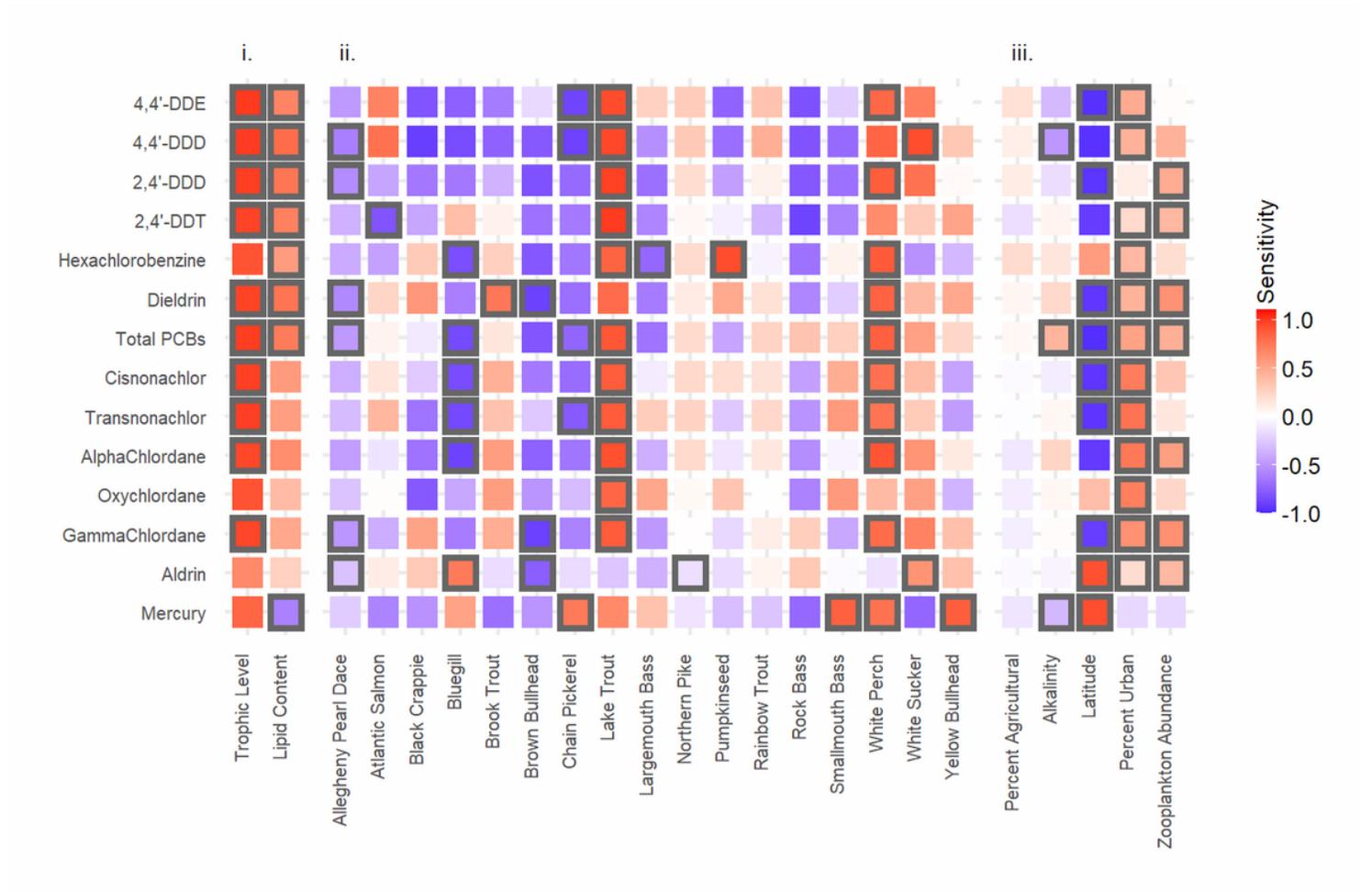
**Figure 2**

Graphical representation of our GJAM model structure. Shaded nodes represent observed variables, others are unobserved. Subscript  $i$  represents individual fish tissue samples,  $k$  represents the various contaminants, and  $j$  represents environmental and fish attributes.



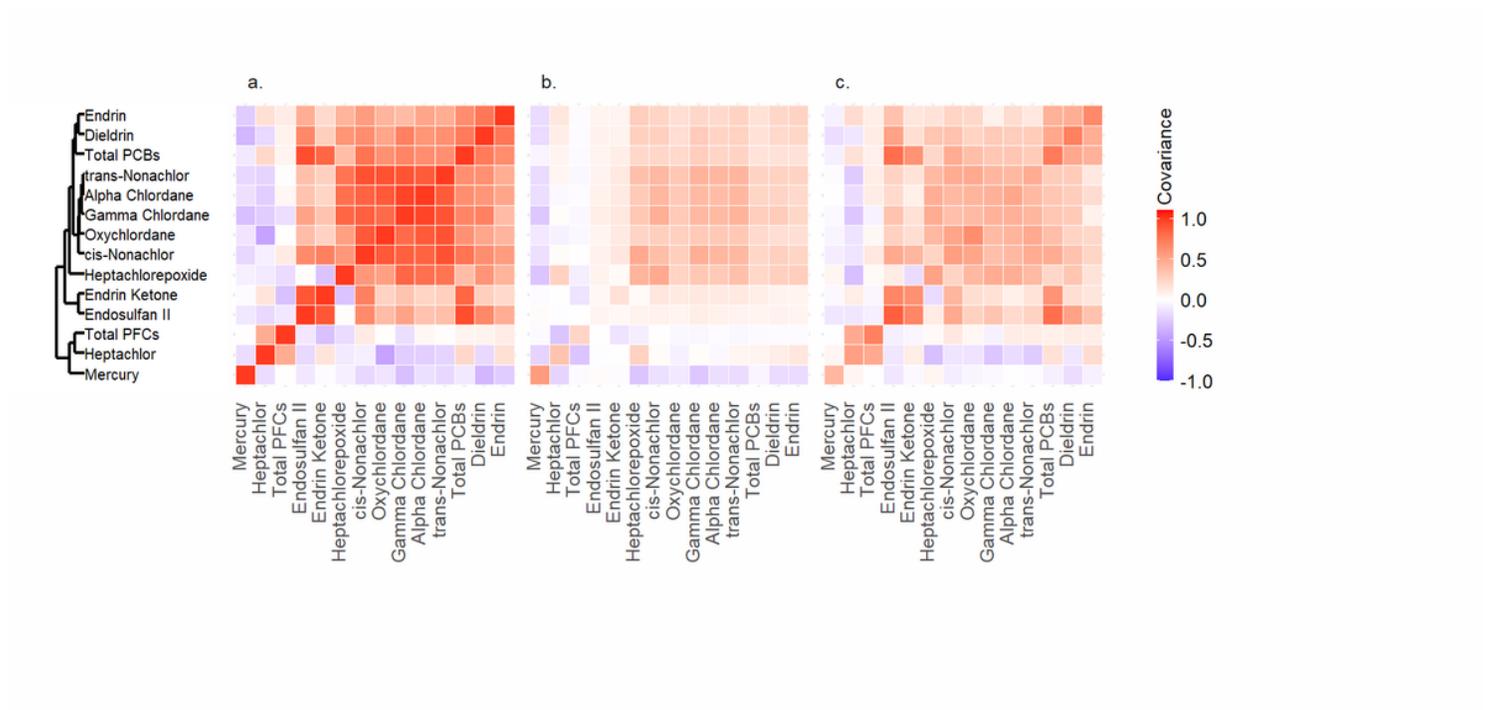
**Figure 3**

Heat maps of normalized variance-covariance matrices of (square-root transformed) fish tissue contaminant concentrations from the EMAP data, corresponding to: (a) measured data variation, (b) model-predicted variation, and (c) unexplained variation. Contaminants are ordered based on a hierarchical cluster analysis of original fish tissue concentrations shown on the left-hand side of the figure.



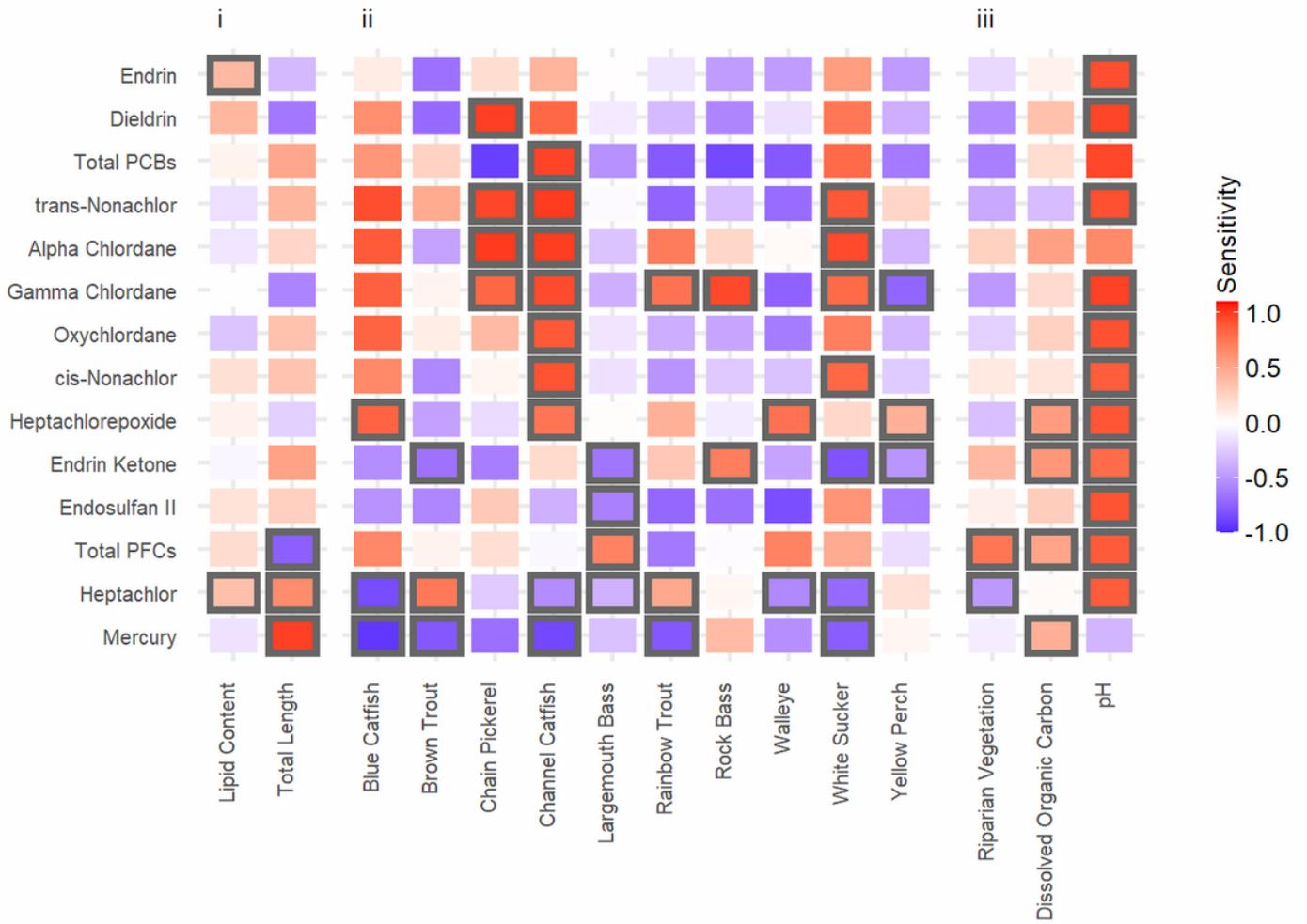
**Figure 4**

Heat map of the sensitivity of fish tissue contaminant concentrations from the EMAP dataset to (i) fish attribute, (ii) species, and (iii) environmental predictor variables. Solid cell borders indicate parameters with a 95% Bayesian posterior credible interval that does not contain 0. Species effects are shown with reference to Yellow Perch.



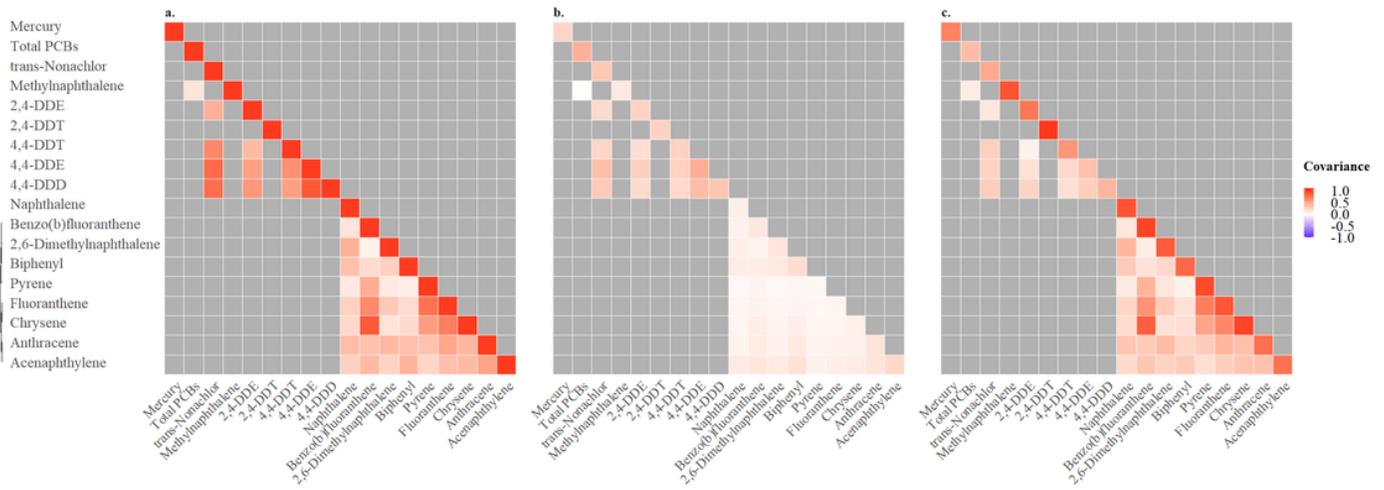
**Figure 5**

Heat maps of normalized variance-covariance matrices of (square-root transformed) fish tissue contaminant concentrations from the NRSA data, corresponding to: (a) measured data variation, (b) model-predicted variation, and (c) unexplained variation. Contaminants are ordered based on a hierarchical cluster analysis of original fish tissue concentrations shown on the left-hand side of the figure.



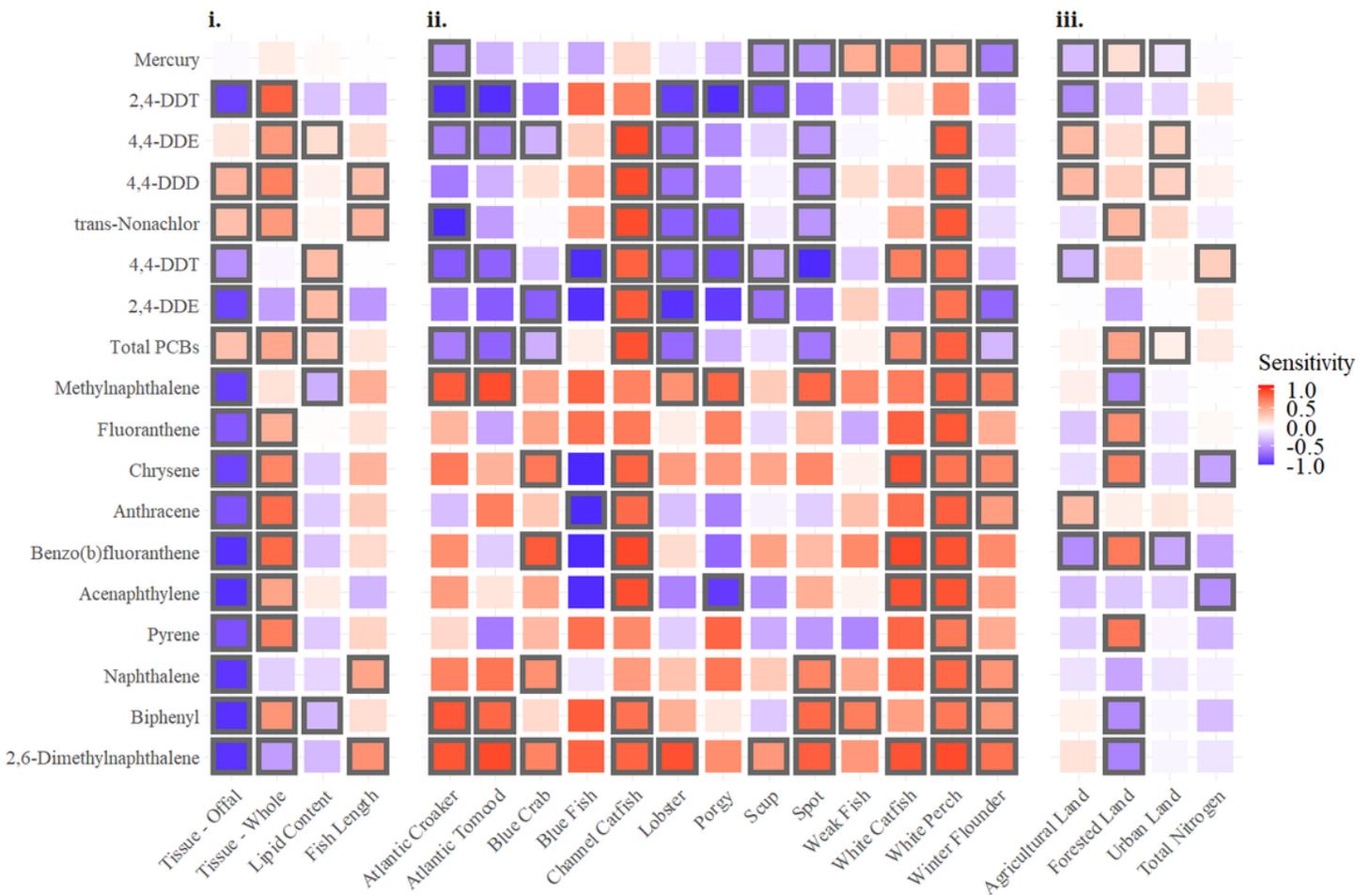
**Figure 6**

Heat map of the sensitivity of fish tissue contaminant concentrations from the NRSA dataset to (i) fish attribute, (ii) species, and (iii) environmental predictor variables. Solid cell borders indicate parameters with a 95% Bayesian posterior credible interval that does not contain 0. Species effects are shown with reference to Smallmouth Bass.



**Figure 7**

Heat maps of normalized variance-covariance matrices of (square-root transformed) fish tissue contaminant concentrations from the NCA data, corresponding to: (a) measured data variation, (b) model-predicted variation, and (c) unexplained variation. Contaminants are ordered based on a hierarchical cluster analysis of original fish tissue concentrations shown on the left-hand side of the figure.



## Figure 8

Heat map of the sensitivity of fish tissue contaminant concentrations from the NCA dataset to (i) fish attribute, (ii) species, and (iii) environmental predictor variables. Solid cell borders indicate parameters with a 95% Bayesian posterior credible interval that does not contain 0. Species effects are shown with reference to Summer Flounder, and tissue type effects are shown with respect to filet.

## Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [Appendix.docx](#)