SPATIAL PATTERNS OF MERCURY CONTAMINATION

OF FISH IN THE SOUTH CENTRAL UNITED STATES

by

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Introduction

Mercury (Hg) contamination is hazardous to human and wildlife health (Mergler et al. 2007, Scheuhammer et al. 2007). Most mercury contamination in the environment comes from anthropogenic sources that release inorganic mercury into the atmosphere (Selin 2009). After being deposited in a water body, inorganic Hg may undergo methylation by aquatic microbes, primarily sulfate-reducing bacteria, resulting in the production of toxic methyl mercury (MeHg) (Selin 2009). Methyl mercury can enter food webs and biomagnify as it moves through the food chain to higher trophic levels, reaching very high concentrations in top predators such as piscivorous fish (Weiner et al. 2003). Consumption of Hg-contaminated fish is the primary pathway for Hg contamination in humans (Mergler et al. 2007). Mercury exposure is a risk for juvenile and adult humans but it is of particular concern during fetal development when it can lead to developmental and cognitive deficits (Mergler et al. 2007).

Because of the adverse effects of Hg on biotic systems, there has been considerable research focused on the prediction of Hg contamination of water bodies (Driscoll et al. 2006, Evers et al. 2007, Sackett et al. 2009, Ward et al. 2010, Yu et al. 2011). An emerging conceptual model predicts that areas of the landscape with elevated Hg and sulfate (SO_4) deposition, high coverage of forests and wetlands, and low coverage of agriculture are most likely to contain food webs with elevated Hg concentrations (Driscoll et al. 2007, Evers et al. 2007, Drenner et al. 2011) (Fig. 1). Atmospheric deposition of Hg is the primary source of Hg to aquatic environments (Wiener et al. 2003) and has been correlated with contamination of fish (Hammerschmidt and Fitzgerald 2006). Sulfate deposition also affects Hg in fish, because SO_4 availability enhances

Hg methylation by sulfate-reducing bacteria (Drevnick et al. 2007). Some land cover types such as forests and wetlands promote Hg contamination of food webs while other land cover types such as agricultural areas reduce Hg contamination of food webs (Driscoll et al. 2007). In this study, I tested this conceptual model in ecoregions of six states in the south central United States.

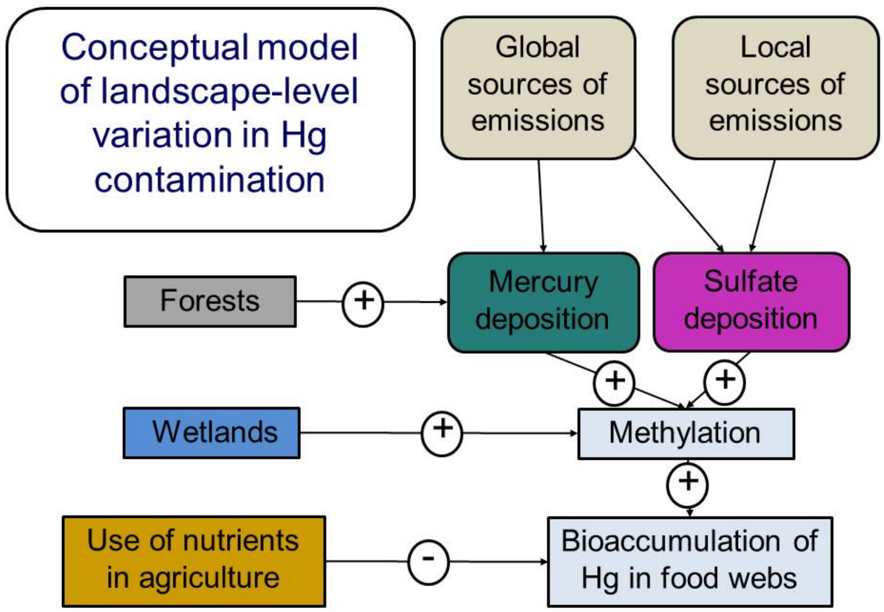

Figure 1. Conceptual model predicting the effects of atmospheric deposition and environmental landscape factors on mercury contamination of fish in the South Central United States (Wiener et al. 2003, Hammerschmidt and Fitzgerald 2006, Drevnick et al. 2007, Driscoll et al. 2007, Evers et al. 2007).

Methods

I examined spatial patterns of Hg in largemouth bass (*Micropterus salmoides*) in 14 of the United States Environmental Protection Agency (USEPA) Level III ecoregions in Oklahoma, Texas, Arkansas, Louisiana, Mississippi and western Tennessee. Ecoregions provide a pragmatic way to investigate Hg accumulation in fish, especially when site-specific data are not available (Sackett et al. 2009). Ecoregions are well suited for spatial studies, because they denote areas of general similarity in the environment and the probable response to disturbance (Bryce et al. 1999). Additionally, they are designed to serve as a spatial framework for monitoring and management of ecosystems (McMahon et al. 2001).

I focused on largemouth bass because it is widely distributed and an economically important species of freshwater game fish that is commonly included in fish tissues contaminant databases. Adult largemouth bass are piscivorous top predators, often having high Hg concentrations relative to other fish species (Kamman et al. 2005, Chumchal and Hambright 2009). Mercury concentration data for largemouth bass was obtained from a published paper (Ray Drenner, personal communication), state databases from the Oklahoma Department of Environmental Quality (Jay Wright, personal communication), Arkansas Department of Environmental Quality (Alan Price, personal communication), Mississippi Department of Environmental Quality (Al Gibson, personal communication), Tennessee Department of Environment and Conservation (Deborah Arnwine, personal communication), Tennessee Valley Authority (Jason Yarbrough, personal communication), Louisiana Department of Environmental Quality and the EPA National Study of Chemical Residues in Lake Fish Tissue. Because mercury concentrations in fish vary by length, and consistent length samples are difficult to obtain, the National Descriptive Model of Mercury in Fish (NDMMF) was utilized to estimate concentration of Hg in largemouth bass by location and year for each site location. A total length (TL) of 35.5-cm was chosen for length standardization because it is a size of largemouth bass commonly-caught by anglers. The 2006 NDMMF fish database, including all fish species, was used to estimate mercury concentrations in 35.5 cm TL largemouth bass equivalents. The final data set consisted of 40,564 largemouth bass and largemouth bass equivalents at 893 sites (Fig. 2). All data were collected for the period 1969-2010. For site locations where fish were sampled multiple years, Hg concentrations were averaged for all sample years to obtain a single site average.

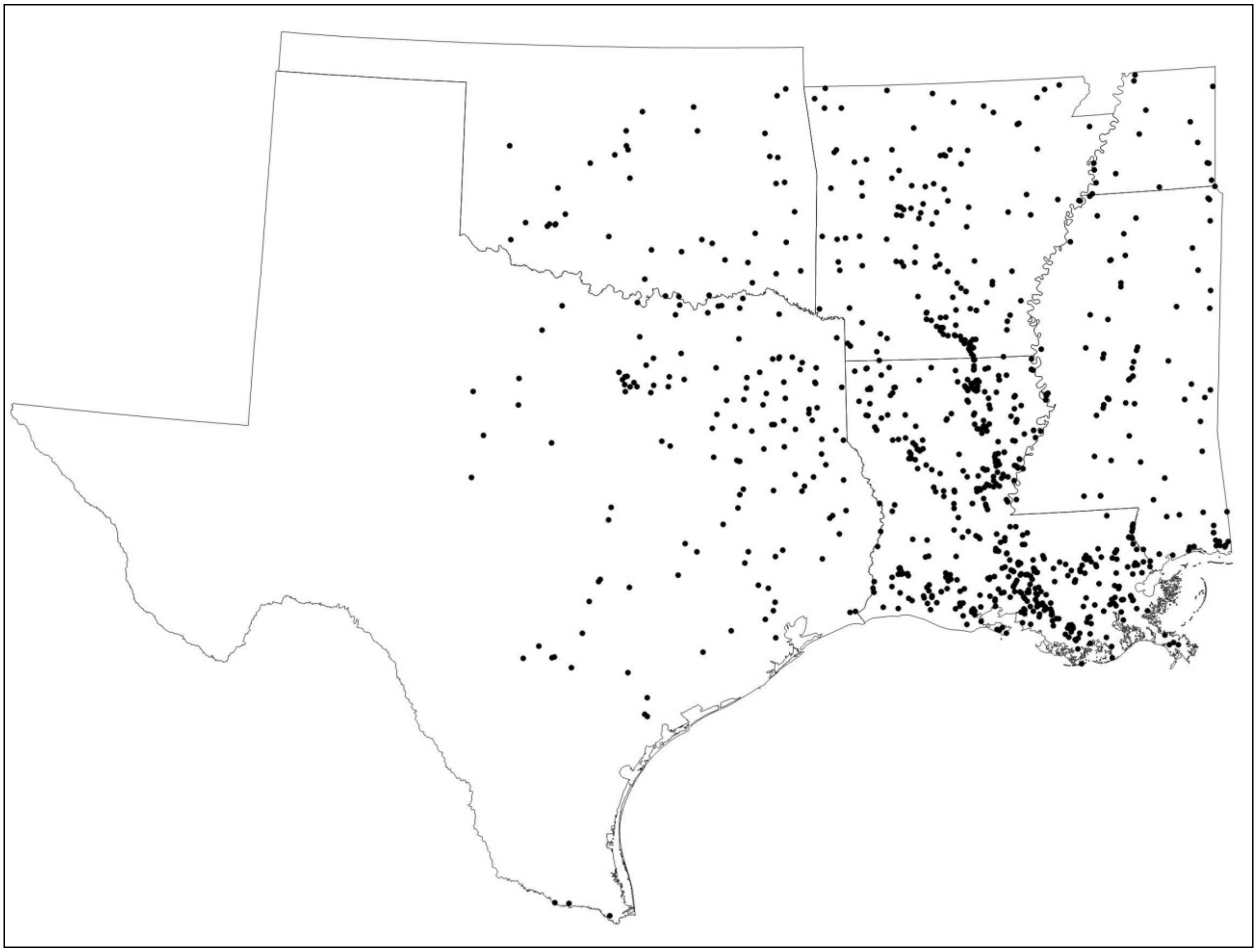

Figure 2. Map of the South Central States area showing the locations of fish collection sites.

Using latitude and longitude coordinates, sample sites were located in Google Earth® to verify location and place names. The resulting fish dataset was mapped in ArcMap 10 (Environmental Systems Research Institute, Redlands, California) along with the National Land Cover Database 2006 (NLCD2006) (Fry et al. 2011) and the US EPA Level III Ecoregions of the Conterminous United States (USEPA 2011). The NLCD2006 is a 16-class land cover classification scheme that has been applied consistently across the conterminous United States at a spatial resolution of 30 meters (Fry et al. 2011). The land cover classes used in this study are described in Table 1. The USEPA ecoregion vector data was converted to a 30 m^2 raster to overlay and tabulate totals for each land cover classification in each ecoregion area. GIS raster data mapping the average annual SO_4 ion wet deposition (kg/ha) for 1990 to 2008 and average annual Hg wet deposition ($\mu g/m^2$) for 2006 to 2009 for the contiguous United States was obtained from the National Deposition and Atmospheric Program (NADP) (Christopher Lehman and David Gay, personal communication). Zonal statistics for average annual SO_4 ion wet deposition and average annual Hg wet deposition were calculated for each ecoregion. Using a spatial join, the ecoregion was identified for each fish site data point and average Hg in largemouth bass was summarized by ecoregion. Only ecoregions containing ten or more sampling sites were used for analysis resulting in 14 ecoregions within the study area.

Table 1. Descriptions of land cover classes modified from NCLD 2006 (Fry et al. 2011).

Classification	Classification Description
Deciduous Forest	Areas dominated by trees generally greater than 5 meters tall, and greater than 20% of total vegetation cover. More than 75% of the tree species shed foliage simultaneously in response to seasonal change.
Evergreen Forest	Areas dominated by trees generally greater than 5 meters tall, and greater than 20% of total vegetation cover. More than 75% of the tree species maintain their leaves all year. Canopy is never without green foliage.
Agricultural Areas	Areas characterized by herbaceous vegetation that has been planted or is intensively managed for the production of food, feed, or fiber; or is maintained in developed settings for specific purposes. Herbaceous vegetation accounts for 75% to 100% of the cover. **Pasture/Hay** – areas of grasses, legumes, or grass-legume mixtures planted for livestock grazing or the production of seed or hay crops, typically on a perennial cycle. Pasture/hay vegetation accounts for greater than 20% of total vegetation. **Cultivated Crops** – areas used for the production of annual crops, such as corn, soybeans, vegetables, tobacco, and cotton, and also perennial woody crops such as orchards and vineyards. Crop vegetation accounts for greater than 20% of total vegetation. This class also includes all land being actively tilled.
Woody Wetlands	Areas where forest or shrubland vegetation accounts for greater than 20% of vegetative cover and the soil or substrate is periodically saturated with or covered with water.

I tested for differences between mean Hg concentrations of largemouth bass in ecoregions using ANOVA followed by the Games-Howell post-hoc test. Linear regression analysis was performed for each ecoregion's land cover and deposition statistic to determine variable correlation with average Hg in largemouth bass. Analysis was performed in SPSS (Version 19.0.0, Chicago, IL, USA). Statistical significance was inferred at $P<0.10$ for all analyses to avoid Type II errors.

Results

Mercury contamination of fish varied with ecoregion (Fig. 3). The Texas Blackland Prairies had the lowest average Hg concentration (210 ng/g), while the Southern Coastal Plains had the highest average Hg concentration (648 ng/g). Three ecoregions (Texas Blackland Prairies, East Central Texas Plains and Cross Timbers) had average Hg concentrations less than 300 ng/g. Six ecoregions (Mississippi Alluvial Plain, Ozark Highlands, Central Great Plains, Boston Mountains, Western Gulf Central Plains and Arkansas Valley) had average concentrations of Hg ranging from 300 to 500 ng/g. Five ecoregions (Mississippi Valley Loess Plains, Southeastern Plains, Ouachita Mountains , South Central Plains and Southern Coastal Plain) had average Hg concentrations above 500 ng/g.

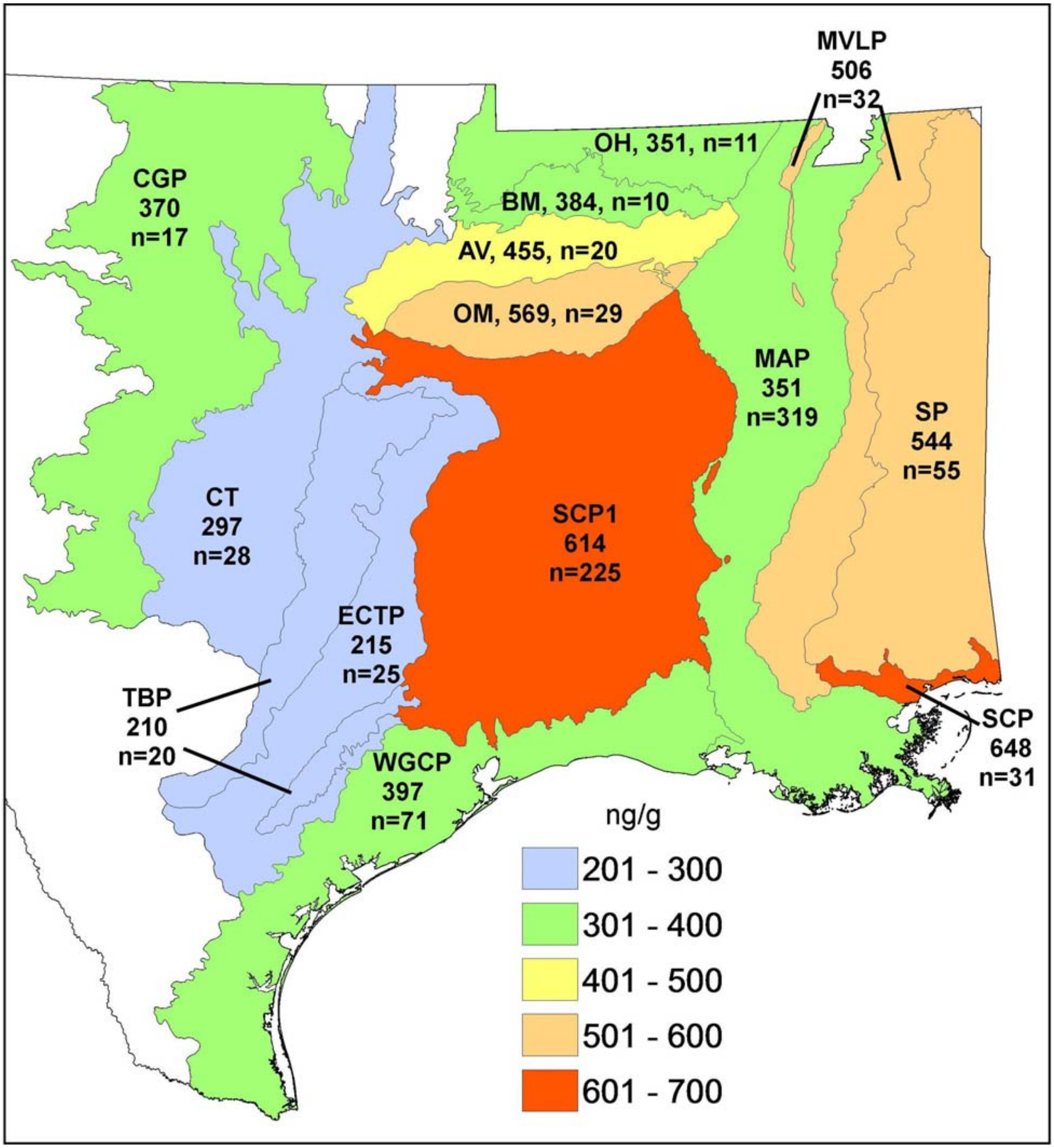

Figure 3. Average Hg concentrations in largemouth bass in 14 ecoregions. Abbreviations for the ecoregions are: AV – Arkansas Valley, CGP – Central Great Plains, CT – Cross Timbers, BM – Boston Mountains, ECTP – East Central Texas Plains, MAP – Mississippi Alluvial Plain, MVLP – Mississippi Valley Loess Plain, OH – Ozark Highlands , OM – Ouachita Mountains, SCP – Southern Coastal Plains, SCP1 – South Central Plains, SP – Southeastern Plain, TBP – Texas Blackland Prairies, WGCP – Western Gulf Coastal Plain. Average Hg concentrations in the ecoregion and number of sites are given below the ecoregion label.

I detected significant differences in mercury concentrations in fish between ecoregions (Table 1). Mercury concentrations of the three ecoregions with average Hg concentration less than 300 ng/g (Texas Blackland Prairies, East Central Texas Plains, and Cross Timbers) were significantly lower than Hg concentrations in the five ecoregions with average Hg concentrations greater than 500 ng/g (Mississippi Valley Loess Plain, Southeastern Plains, Ouachita Mountains, South Central Plains and Southern Coastal Plains). The mercury concentration in the Mississippi Alluvial Plain was significantly higher than Texas Blackland Prairies and East Central Texas Prairie but significantly lower than four of the surrounding ecoregions, the Southeastern Plain, Ouachita Mountains, South Central Plains and Southern Coastal Plains. Mercury concentrations in the Ozark Highlands, Central Great Plains, Boston Mountains and Western Gulf Coastal Plains were significantly lower than South Central Plains and Southern Coastal Plains.

Table 2. Probability values from paired comparisons of ecoregions using the Games-Howell post hoc test. Bold values are statistically significant, $p<0.10$. Ecoregions are listed from lowest to highest average Hg concentrations (Texas Blackland Prairies and Southern Coastal Plain, respectively).

	Texas Blackland Prairies	East Central Texas Plains	Cross Timbers	Mississippi Alluvial Plains	Ozark Highlands	Central Great Plains	Boston Mountains	Western Gulf Coastal Plains	Arkansas Valley	Mississippi Valley Loess Plains	Southeastern Plains	Ouachita Mountains	South Central Plains	Southern Coastal Plain
Texas Blackland Prairies		1.000	0.758	**0.003**	0.327	0.500	0.266	**0.000**	0.337	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**
East Central Texas Plains	1.000		0.722	**0.001**	0.337	0.519	0.278	**0.000**	0.353	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**
Cross Timbers	0.758	0.722		0.970	0.999	0.988	0.972	0.500	0.898	**0.031**	**0.000**	**0.005**	**0.000**	**0.000**
Mississippi Alluvial Plain	**0.003**	**0.001**	0.970		1.000	1.000	1.000	0.945	0.993	0.111	**0.000**	**0.021**	**0.000**	**0.000**
Ozark Highlands	0.327	0.337	0.999	1.000		1.000	1.000	0.999	0.997	0.471	**0.090**	0.133	**0.004**	**0.004**
Central Great Plains	0.500	0.519	0.988	1.000	1.000		1.000	1.000	1.000	0.865	0.457	0.453	**0.065**	**0.048**
Boston Mountains	0.266	0.278	0.972	1.000	1.000	1.000		1.000	1.000	0.877	0.446	0.458	**0.066**	**0.047**
Western Gulf Coastal Plains	**0.000**	**0.000**	0.500	0.945	0.999	1.000	1.000		1.000	0.682	**0.049**	0.194	**0.000**	**0.001**
Arkansas Valley	0.337	0.353	0.898	0.993	0.997	1.000	1.000	1.000		1.000	0.999	0.996	0.889	0.780
Mississippi Valley Loess Plains	**0.000**	**0.000**	**0.031**	0.111	0.471	0.865	0.877	0.682	1.000		1.000	1.000	0.774	0.634
Southeastern Plains	**0.000**	**0.000**	**0.000**	**0.000**	**0.090**	0.457	0.446	**0.049**	0.999	1.000		1.000	0.968	0.877
Ouachita Mountains	**0.000**	**0.000**	**0.005**	**0.021**	0.133	0.453	0.458	0.194	0.996	1.000	1.000		1.000	0.997
South Central Plains	**0.000**	**0.000**	**0.000**	**0.000**	**0.004**	**0.065**	**0.066**	**0.000**	0.999	0.774	0.968	1.000		1.000
Southern Coastal Plains	**0.000**	**0.000**	**0.000**	**0.000**	**0.004**	**0.048**	**0.047**	**0.001**	0.780	0.634	0.877	0.997	1.000	

Mean concentrations of Hg in largemouth bass within ecoregions were positively correlated with Hg deposition, SO_4 deposition, evergreen forests and woody wetlands coverage, and inversely correlated with agriculture (Figs. 4 and 5). Coverage by evergreen forests was the best predictor of mercury contamination of fish, explaining 73% of the variance. I did not detect a significant correlation between Hg concentrations in fish and coverage by deciduous forests.

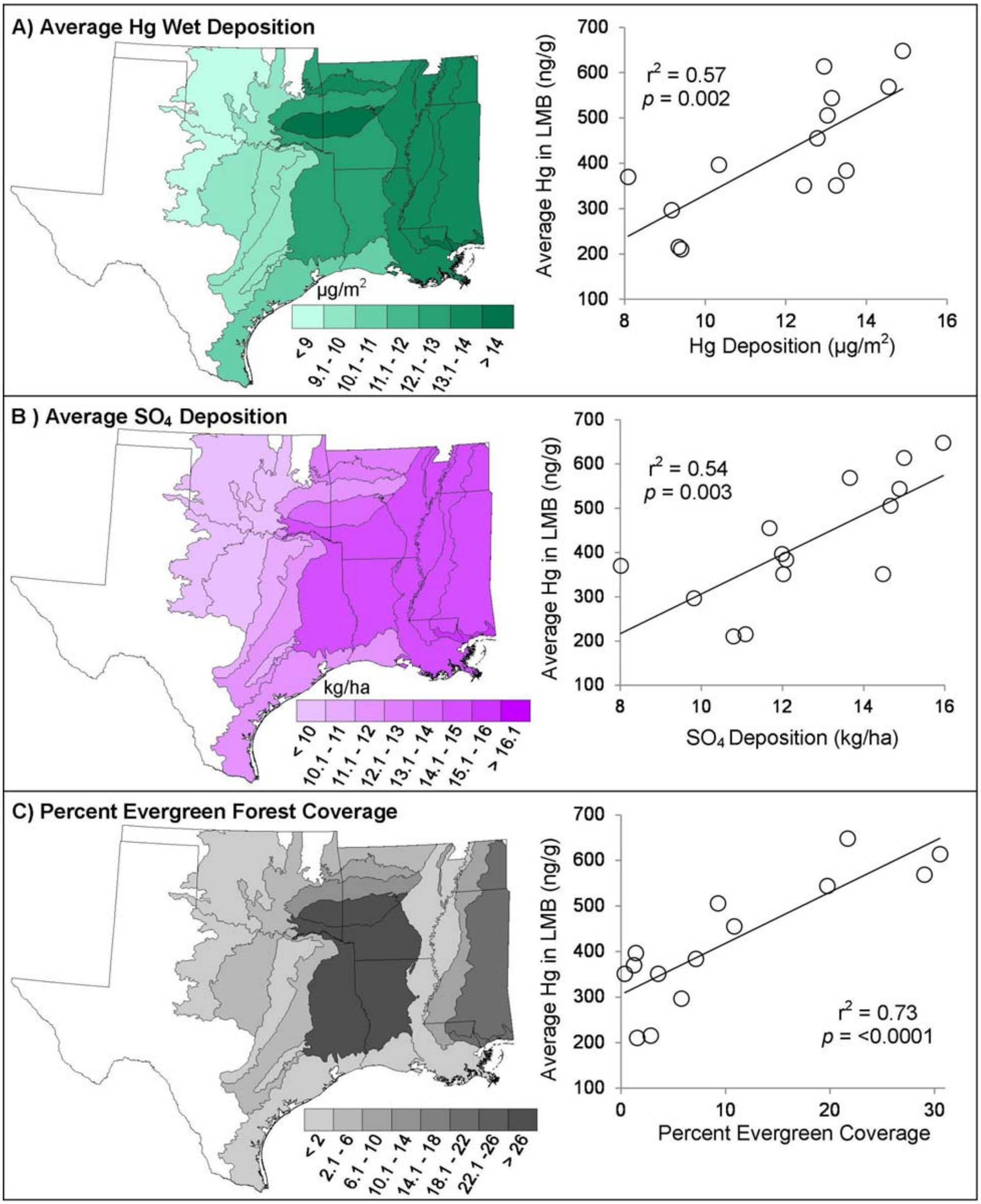

Figure 4. Conceptual model variable maps and regression analysis. A) Average Hg wet deposition (μg/m^2), B) Average SO_4 deposition (kg/ha), C) Percent coverage of evergreen forests

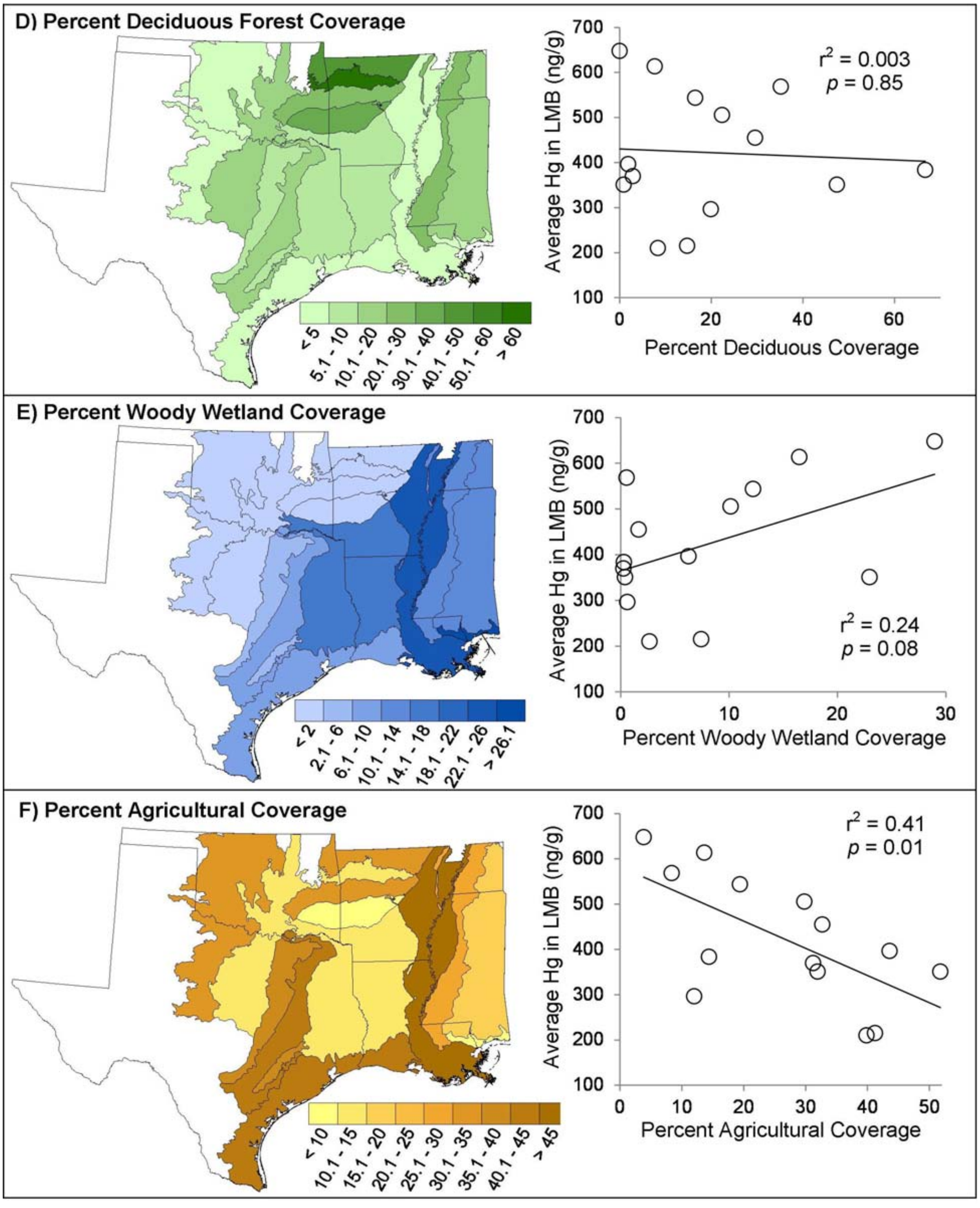

Figure 5. Conceptual model variable maps and regression analysis. D) Percent coverage of deciduous forests, E) Percent coverage of woody wetlands, F) Percent coverage of agricultural areas

Discussion

Most studies on Hg contamination of fish and wildlife have been in the northeastern United States and the Great Lakes area (Kamman et al. 2005, Driscoll et al. 2007, Evers et al. 2007). These studies have identified large geographic areas with aquatic food webs that are heavily contaminated by Hg. My study is the first multi-state study conducted in the southern United States showing the extensive nature of Hg contamination in fish. Of the 893 sites analyzed in this study, 58% have Hg concentrations in fish above 300 ng/g, the USEPA screening level for fish consumption advisories (EPA 2001). Over 70% of sites in ecoregions with evergreen forest coverage above 20% had Hg concentrations in fish above the EPA screening level. This is one of the most highly Hg contaminated areas in the U.S. and the contamination may extend further to the east. Humans and wildlife using fish resources in this area are at risk.

The conceptual model used in this study was based on the studies conducted in the northeast and the relationships they revealed between fish Hg contamination levels and landscape environmental variables (Driscoll et al. 2007, Evers et al. 2007). Drenner et al. (2011) was first to test this conceptual model in the southern US by examining mercury contamination of largemouth bass from 145 reservoirs in four ecoregions of North Texas. The highest level of Hg contamination in fish was in the South Central Plains ecoregion which receives high levels of Hg and SO_4 deposition and contains extensive forest and wetland coverage and little agriculture. The study had important implications for other areas of the United States outside of Texas, because the South Central Plains extended into parts of Oklahoma, Louisiana and Arkansas.

In the present study, I expanded the ecoregion-level analyses of mercury contamination of fish to 14 ecoregions across six states in the south central U.S. The highest levels of Hg contamination in fish were in four ecoregions: the Southern Coastal Plains (648 ng/g), South Central Plains (614 ng/g), Ouachita Mountains (569 ng/g), and Southeastern Plains (544 ng/g). These ecoregions receive high levels of Hg and SO_4 deposition and contain extensive evergreen forest coverage but little agriculture. Coverage by evergreen forests explained 73 percent of the variance of average mercury concentrations in largemouth bass in the 14 ecoregions.

Importance of forest coverage for Hg deposition

Forests have been found to increase Hg deposition, potentially leading to increased contamination of aquatic food webs. Atmospheric deposition of Hg is greater under forest canopies than deposition occurring in open areas (Kolka et al. 1999, Witt et al. 2009, Mowat et al. 2011). The elevated Hg deposition in forest systems is caused by the capture of particulates (dry deposition) by the forest canopy (Kolka et al. 1999). The canopy enhances atmospheric Hg deposition via throughfall and litterfall. Throughfall occurs when precipitation passes through the forest canopy and washes dry deposition off the leaves resulting in higher concentrations of Hg than precipitation in open areas (Kolka et al. 1999, Rea et al. 2000, St Louis et al. 2001). Some particulate Hg adsorbed to leaves is also transported to the ground as litterfall along with Hg contained in leaves from stomatal uptake (Rea et al. 2000). Litterfall generally represents the largest input of THg and MeHg to the landscape on an annual basis (St. Louis et al. 2001, Rea et al. 2002, Graydon et al. 2008).

Hg deposition in forests is influenced by forest canopy type (Witt et al. 2009). Conifer species capture more Hg deposition than deciduous species, partly due to greater leaf and branch surface area (Kolka et al. 1999, St. Louis et al. 2001). Leaves and needles may scavenge Hg from the atmosphere as dry particulate Hg or reactive gaseous mercury (RGM) species (St. Louis et al. 2001). Conifer canopies are more efficient filters of airborne particles than are deciduous canopies as indicated by much higher Hg concentrations and total deposition in throughfall and stemflow waters under conifers (Kolka et al. 1999). While the evergreen forests of the southern United States contain non-coniferous species (magnolias, live oaks, etc.) the dominant coverage is coniferous pines (Schultz 1999). I found that average Hg concentrations in largemouth bass in ecoregions was significantly correlated with percent coniferous coverage whereas deciduous forest coverage was not correlated with Hg concentrations in largemouth bass.

Forest management practices may affect mercury contamination of water bodies. Today coniferous forests in this region of the south central United States are primarily loblolly pine (*Pinus taeda*), which replaced the native forests after they were logged prior to the 1930s (Schultz 1999). Commercial logging is still practiced across this area and logging can be associated with Hg release in forested areas and contamination of aquatic systems (Povari et al. 2003). Disturbances such as logging and burning of forests have been shown to increase Hg concentrations in fish over undisturbed areas (Garcia and Carignan 2005). After logging or soil preparation, significant increases in the total Hg and MeHg load from catchments to lakes can be expected for a number of years (Povari et al. 2003). These forest management practices may contribute to variability of Hg contamination of fish within watersheds having the same land cover. Fish in water bodies in areas where forests are being logged would be expected to have higher Hg contamination levels than in unlogged areas.

Implications for Fish Consumption Advisories

The issuance of fish consumption advisories is an important and widely used management tool to reduce the risk of adverse health effects in humans caused by consumption of Hg-contaminated fish (Jakus et al. 1998). Though issuing fish consumption advisories may impact revenue generated from recreation fishing, it is more cost-effective than the potentially large costs associated with reduction of contaminants in the environment (Jakus et al. 1998). Fish consumption advisories are typically issued by states for individual water bodies, or in some cases, on a statewide basis. Mercury contamination is the primary reason (89%) of all state fish consumption advisories in the United States (Katner et al. 2010). In the current study I found that only three of the 14 ecoregions had average Hg concentrations in largemouth bass below the EPA screening level for fish consumption advisories. Additionally I found that over 70 percent of the water bodies within ecoregions with evergreen coverage of 20 percent or greater have Hg concentrations in largemouth bass above the EPA screening level for fish consumption advisories.

I recommend that states consider issuing fish consumption advisories for mercury by ecoregions with high Hg concentrations in fish. My study shows that risk of consuming fish with Hg concentrations above EPA level is dependent on ecoregions and especially high in areas with high Hg deposition and high coverage by evergreens. The public should be informed that fish from ecoregions receiving high levels of Hg deposition and with high levels of evergreen coverage may constitute significant health hazard. Advisories issued on an ecoregion specific basis would provide the additional benefit of including small man-made ponds that numerically dominate this region of the United States (Smith et al. 2002) but are rarely sampled by government agencies for mercury contamination in fish.

Conclusions

It is commonly recognized that Hg contamination in fish is widespread in the Great Lake States, the northeastern United States, Canada, northern Europe, as well as remote wilderness areas (Kolka et al. 1999). My study shows a large geographic area in the south central United States is being severely impacted by Hg contamination. I found a strong link between land cover types and Hg contamination in largemouth bass at an ecoregion scale. Mercury contamination in fish was highest in ecoregions with high Hg deposition and increased evergreen coverage. Since it is logistically impossible, state agencies should prioritize their monitoring efforts to water bodies that are at a higher risk for elevated Hg contamination in fish (Rypel 2010). The public should be warned that fish in water bodies from ecoregions with high mercury deposition and evergreen coverage may constitute a significant hazard to human health through increased exposure to Hg from fish.

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ABSTRACT

SPATIAL PATTERS OF MERCURY CONTAMINATION
OF FISH IN THE SOUTH CENTRAL UNITED STATES

by Christina Michelle Jones, 2012
Environmental Science
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Mercury (Hg) is a toxic metal that is found in aquatic food webs and is hazardous to humans. An emerging conceptual model predicts areas with the potential to contain food webs with elevated concentrations of Hg receive high amounts of Hg and sulfate deposition, have high coverage of forests and wetlands and low coverage of agriculture. The objective of this study was to test this conceptual model using concentrations of Hg in fish in the south central United States. Coverage by evergreen forests explained 73 percent of the variance of average mercury concentrations in the 14 ecoregions. Over 70% of the water bodies in ecoregions with evergreen forest coverage of 20% or greater have Hg concentrations in largemouth bass above the EPA criterion level of 300 ng/g. Evergreen forests in states in the southern ecoregions may constitute a significant hazard to human health through increased exposure to Hg from fish.