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Spatial Patterns of Mercury Contamination and Associated Risk to Piscivorous Wading Birds of the South Central United States

Christopher T. Gerstle, Ray W. Drenner, and Matthew M. Chumchal*

Department of Biology, Texas Christian University, Fort Worth, Texas, USA

Abstract: Piscivorous birds are top predators in aquatic ecosystems and are vulnerable to mercury (Hg) exposure and associated adverse health effects. In some areas of North America, the health risk posed to piscivorous birds by Hg contamination has not been characterized because concentrations of Hg in bird tissues have not been extensively monitored. When data on Hg in tissues of piscivorous birds are not available, the concentration of Hg in the blood of piscivorous birds can be estimated from the concentration of Hg in prey fish. We used concentrations of Hg in different lengths of a proxy prey fish, bluegill (*Lepomis macrochirus*), to estimate the concentration of Hg in the blood of 4 species of adult piscivorous wading birds (little blue herons [*Egretta caerulea*], green herons [*Butorides virescens*], great egrets [*Ardea albus*], and great blue herons [*Ardea herodias*]) in 14 ecoregions of the south central United States. The 4 species of birds consume different sizes of fish with different concentrations of Hg and were predicted to have different concentrations of Hg in their blood, with little blue herons < green herons < great egrets < great blue herons. For each species of bird, there were significant differences in average estimated concentrations of Hg in blood between ecoregions, with estimated concentrations of Hg in blood increasing with Hg deposition. The level of predicted risk varied with ecoregion and bird species and was highest for great blue herons. We recommend that future studies of Hg contamination of piscivorous wading birds of the southern United States focus on great blue herons in water bodies within ecoregions that have high Hg deposition. *Environ Toxicol Chem* 2019;38:160–166. © 2018 SETAC

Keywords: Piscivorous wading birds; Mercury; Risk; Spatial patterns

INTRODUCTION

Piscivorous birds are top predators in aquatic ecosystems and are vulnerable to mercury (Hg) exposure and associated adverse health effects (Schuehammer et al. 2007; Ackerman et al. 2015). Piscivorous birds are primarily exposed to Hg through their diet (Ackerman et al. 2015). Because the concentration of Hg in the tissues of fish increases with fish length (Chumchal and Hambright 2009), piscivorous birds that feed on large fish accumulate higher concentrations of Hg than piscivorous birds that feed on small fish (Jackson et al. 2016). Therefore, piscivorous birds that feed on large fish are at greater risk from negative health effects associated with Hg exposure. Depending on the level of exposure, Hg can cause physiological and behavioral abnormalities and reproductive impairment in birds (Jackson et al. 2016).

In some areas of North America, the risk posed by Hg contamination to the health of piscivorous birds is unknown because concentrations of Hg in the tissues of birds have not been comprehensively monitored (Ackerman et al. 2015; Jackson et al. 2016). Ackerman et al. (2015) proposed that in regions where data on concentrations of Hg in the tissues of piscivorous birds are not available, the concentrations of Hg in the blood of piscivorous birds can be estimated from the concentrations of Hg in prey fish. Using data on concentrations of Hg in fish to estimate concentrations of Hg in piscivorous birds capitalizes on the large databases that have been assembled by government agencies because of the risk posed by Hg-contaminated fish to human health (Ackerman et al. 2015). These estimates of Hg concentrations in bird blood can be followed up with direct monitoring of Hg concentrations in species of piscivorous birds from regions predicted to be at risk from Hg contamination.

In the present study, we use a large database of concentrations of Hg in fish tissues assembled by Drenner et al. (2013) to estimate the concentrations of Hg in the blood of adult piscivorous wading

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* Address correspondence to m.m.chumchal@tcu.edu
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birds and associated risk to bird health, in ecoregions of the south central United States (all or part of Texas, Oklahoma, Louisiana, Arkansas, Mississippi, and Tennessee). Although Hg contamination of piscivorous wading birds has been studied in other areas of North America including Canada, the Great Lakes region, and the western, northeastern, and southeastern United States (Sundlof et al. 1994; Gariboldi et al. 1998; Evers et al. 2005; Bryan et al. 2015; Ackerman et al. 2016; Jackson et al. 2016; Champoux and Boily 2017), there is little information about the Hg contamination of piscivorous wading birds in the south central United States (Schulwitz et al. 2015; US Fish and Wildlife Service 2018). This area is well suited for studying spatial patterns of Hg contamination and associated risk to piscivorous wading birds because Hg deposition and concentrations of Hg in fish vary across ecoregions of the south central United States (Drenner et al. 2013).

MATERIALS AND METHODS

We estimated concentrations of Hg in blood and associated risk to adults of 4 species of piscivorous wading birds that are found throughout the south central United States: little blue herons (*Egretta caerulea*; Rodgers and Smith 2012), green herons (*Butorides virescens*; Davis and Kushlan 1994), great egrets (*Ardea albus*; McCrimmon et al. 2011), and great blue herons (Vennesland and Butler 2011). We selected these 4 species of wading birds because as adults they consume different sizes of fish prey (Davis and Kushlan 1994; McCrimmon et al. 2011; Vennesland and Butler 2011; Rodgers and Smith 2012; Supplemental Data, Table S1), potentially placing them at different levels of risk from Hg exposure. Although piscivorous wading birds can feed primarily on fish, they exhibit opportunistic foraging behavior and consume other aquatic and terrestrial biota, which can comprise a significant proportion of the diet (Davis and Kushlan 1994; McCrimmon et al. 2011; Vennesland and Butler 2011; Rodgers and Smith 2012). To estimate risk to birds from Hg exposure, we assumed their diets were composed of 100% fish. This assumption will result in an overestimate of risk to birds feeding on terrestrial food items, which typically have low concentrations of MeHg (Speir et al. 2014).

We used the approach developed by Ackerman et al. (2015) to estimate concentrations of Hg in blood of piscivorous wading birds from the concentrations of Hg in whole bodies of prey fish. Using data compiled from field studies, Ackerman et al. (2015) found that the concentration of Hg in blood of piscivorous birds was related to the concentration of Hg in whole bodies of prey fish according to the equation:

$$\ln(FemaleBirdBloodHg_{WW}) = 1.788 + 0.6182 \times (\ln(PreyFishHg_{ww}))$$

where $FemaleBirdBloodHg_{ww}$ is the estimated concentration of Hg in the blood of female piscivorous birds in $\mu g/g_{ww}$ and $PreyFishHg_{ww}$ is the concentration of Hg in the whole bodies of prey fish in $\mu g/g_{ww}$. The inverse log of the model output (ln-estimated concentration of Hg in the blood of female piscivorous birds) is presented in the figures and tables. Ackerman et al.'s (2015) model was developed using a robust dataset that included data from 3 piscivorous bird species (common loon [*Gavia immer*], western grebe [*Aechmophorus occidentalis*], and Clark's grebe [*Aechmophorus clarkia*]) and prey fish collected from multiple sites across North America. A potential limitation of Ackerman et al.'s (2015) model is that it is not known how well it predicts blood Hg concentrations in other species of piscivorous birds, including herons. Therefore, estimates of Hg concentrations in bird blood and associated risk in the present study should be viewed as hypotheses.

In assessments of Hg risk to birds, proxy species of prey fish can be used as indicators of Hg exposure (Evers et al. 2011; Depew et al. 2013). In the present study, we used bluegill (*Lepomis macrochirus*) as a proxy species of prey fish in the diets of wading birds. Sunfish (*Lepomis* spp.) are a component of the diets of wading birds (Niethammer and Kaiser 1983; Stickley et al. 1995; Smith 1997; Glahn et al. 1999). Bluegill are commonly found in both lentic and lotic systems of the south central United States (Lee et al. 1980) and have concentrations of Hg that are intermediate among fish in the region (Chumchal et al. 2010; Fry and Chumchal 2012).

For the present study, we used a database of concentrations of Hg in fish from Drenner et al. (2013) to estimate concentrations of Hg in the blood of adult piscivorous wading birds. This database includes information on Hg concentrations in fish from the National Fish Data Base (Wente 2004), supplemental data from state agencies (Drenner et al. 2013), and additional analyses of fish from Texas (Drenner et al. 2011); it consists of 21 739 fish samples collected between 1969 and 2010 (95% of the fish samples were collected between 1990 and 2010). Fish were collected from a variety of water body types that included lentic (e.g., oxbow lakes, reservoirs, human-made ponds) and lotic (e.g., creeks, rivers, streams, bayous) sites.

It is not possible to collect fish of the same size and species from different sites over large regions (Wente 2004). To allow for the assessment of Hg in fish of similar species and lengths, the US Geological Survey, in cooperation with the US National Institute of Environmental Health Sciences, developed a statistical model using data on concentrations of Hg in fish from the National Listing of Fish and Wildlife Advisories dataset (Wente 2004; Supplemental Data). The model can be used to estimate concentrations of Hg in sizes and species of fish that were not sampled from a given site (Wente 2004). In the present study, we used the model (now called the National Descriptive Model of Mercury in Fish [NDMMF; Wente 2004]) to estimate concentrations of Hg in whole bodies of bluegill at 728 lentic and lotic sampling sites across the south central United States (Supplemental Data, Figure S1).

We predicted the concentration of Hg in bluegill at each sampling event (a location and time at which fish samples were collected). If multiple sampling events occurred in the same lentic waterbody, the events were merged by calculating the arithmetic mean of the estimated concentrations of Hg. If multiple sampling events occurred in the same lotic waterbody and were within 1 km of each other, they were merged in the same manner. We excluded from our final dataset any sampling event for which we could not verify the coordinates or that had occurred outside the states of Arkansas, Louisiana, Mississippi,

Oklahoma, Tennessee, or Texas or the 14 US Environmental Protection Agency (USEPA) level III ecoregions we examined. This resulted in 1 to 37 sampling events/site (mean = 2.11 sampling events/site).

The model output is concentrations of total Hg. We assumed that 100% of the total Hg in bluegill was MeHg because Bloom (1992) has estimated that MeHg accounts for at least 95% of the total Hg in several species of fish. The USEPA (US Environmental Protection Agency 2000) recommends analyzing the concentration of total Hg in fish tissues as a proxy for the concentration of MeHg.

We used the NDMMF to estimate concentrations of Hg for 4 total lengths of bluegill (4.3, 5.5, 6.5, and 17.5 cm) that correspond to the midpoint of the total length ranges of fish in diets of little blue herons, green herons, great egrets, and great blue herons, respectively (Supplemental Data, Table S1). Estimated concentrations of Hg in bluegill (Supplemental Data, Figure S2) were used to estimate concentrations of Hg in the blood of little blue herons, green herons, great egrets, and great blue herons according to the model of Ackerman et al. (2015) described above.

We averaged the estimated concentrations of Hg in blood from each sampling site by ecoregion. Our study area included 14 USEPA level III ecoregions (US Environmental Protection Agency 2013), which contained from 10 to 255 sampling sites (mean = 52 sampling sites/ecoregion; Supplemental Data, Figure S1). We used ecoregions as the unit of analysis because they are well suited for spatial studies. Ecoregions denote areas of the environment with similar landscapes (Bryce et al. 1999), and therefore can serve as a spatial framework for monitoring and management of ecosystems (McMahon et al. 2001).

Because Hg deposition adjusted for the presence of conifers (hereafter termed conifer-adjusted Hg deposition) accounts for 80% of the variation in Hg contamination of fish among

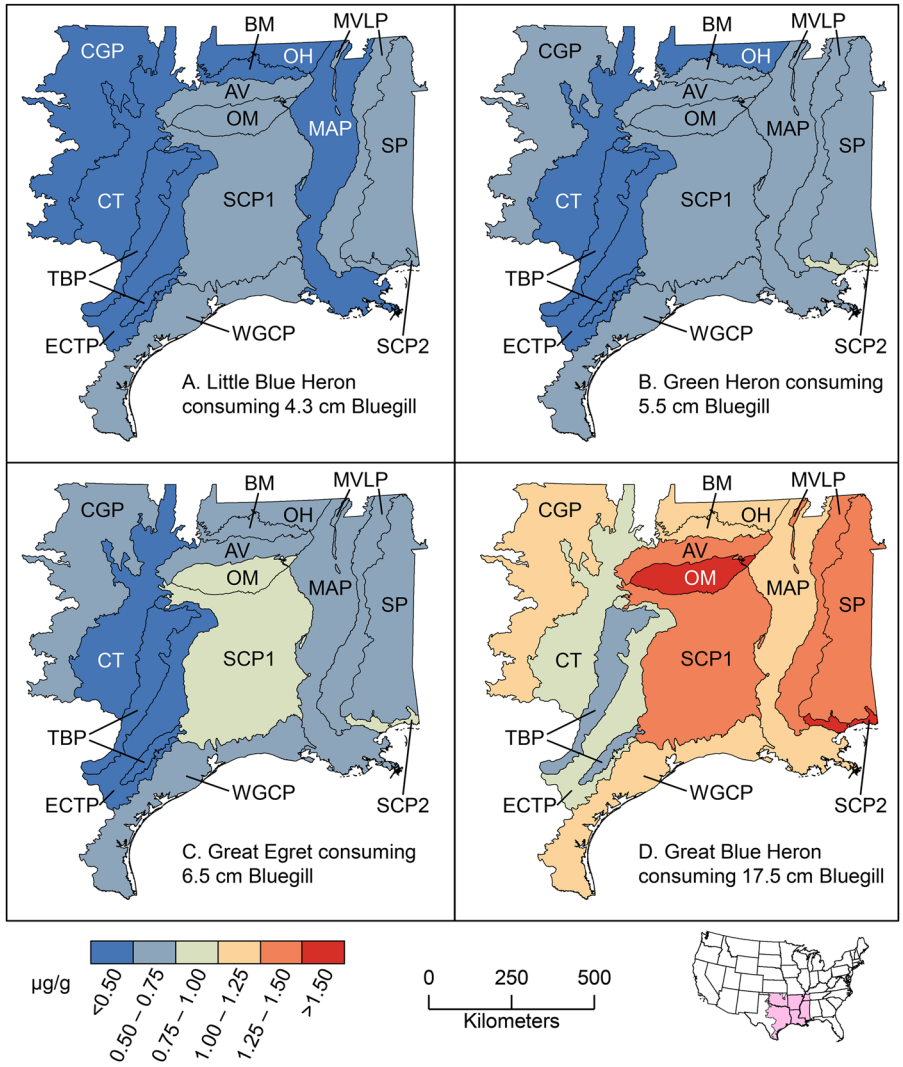

FIGURE 1: Estimated concentrations of mercury (Hg) in blood of (**A**) little blue herons, (**B**) green herons, (**C**) great egrets, and (**D**) great blue herons from 14 ecoregions in the south central United States. Ecoregions are: Arkansas Valley (AV), Boston Mountains (BM), Central Great Plains (CGP), Cross Timbers (CT), East Central Texas Plains (ECTP), Mississippi Alluvial Plain (MAP), Mississippi Valley Loess Plains (MVLP), Ozark Highlands (OH), Ouachita Mountains (OM), South Central Plains (SCP1), Southeastern Plains (SP), Southern Coastal Plain (SCP2), Texas Blackland Prairies (TBP), and Western Gulf Coastal Plain (WGCP).

TABLE 1: Average (± standard error) conifer-adjusted Hg deposition and average estimated concentration of Hg in the blood of 4 species of piscivorous wading birds in 14 US Environmental Protection Agency (USEPA) level III ecoregions[a]

USEPA level III ecoregion	Conifer-adjusted Hg deposition (μg/m^2/y)	Little blue heron estimated blood Hg (μg/g)	Green heron estimated blood Hg (μg/g)	Great egret estimated blood Hg (μg/g)	Great blue heron estimated blood Hg (μg/g)
Arkansas Valley (AV)	18.4	0.54 ± 0.07	0.62 ± 0.08	0.68 ± 0.09	1.26 ± 0.16
Boston Mountains (BM)	17.4	0.48 ± 0.05	0.55 ± 0.05	0.60 ±0.06	1.12 ± 0.10
Central Great Plains (CGP)	8.5	0.49 ± 0.06	0.55 ± 0.07	0.61 ± 0.08	1.13 ± 0.14
Cross Timbers (CT)	11.3	0.38 ± 0.03	0.43 ± 0.03	0.48 ± 0.03	0.90 ± 0.06
East Central Texas Plains (ECTP)	10.4	0.33 ± 0.03	0.38 ± 0.03	0.42 ± 0.03	0.78 ± 0.06
Mississippi Alluvial Plain (MAP)	13.4	0.45 ± 0.01	0.52 ± 0.01	0.57 ± 0.02	1.06 ± 0.03
Mississippi Valley Loess Plains (MVLP)	17.9	0.59 ± 0.05	0.67 ± 0.06	0.73 ± 0.06	1.36 ± 0.11
Ouachita Mountains (OM)	31.6	0.66 ± 0.04	0.75 ± 0.05	0.82 ± 0.05	1.52 ± 0.09
Ozark Highlands (OH)	14.2	0.43 ± 0.04	0.49 ± 0.05	0.53 ± 0.05	1.00 ± 0.09
South Central Plains (SCP1)	28.9	0.63 ± 0.02	0.72 ± 0.02	0.78 ± 0.03	1.45 ± 0.05
Southeastern Plains (SP)	23.6	0.60 ± 0.03	0.68 ± 0.03	0.75 ± 0.04	1.38 ± 0.07
Southern Coastal Plain (SCP2)	28.0	0.69 ± 0.04	0.78 ± 0.04	0.86 ± 0.05	1.59 ± 0.09
Texas Blackland Prairies (TBP)	10.0	0.27 ± 0.04	0.32 ± 0.04	0.35 ± 0.05	0.67 ± 0.08
Western Gulf Coastal Plain (WGCP)	10.9	0.50 ± 0.02	0.57 ± 0.03	0.62 ± 0.03	1.16 ± 0.05

[a] Data from Drenner et al. (2013). Ecoregion abbreviations shown parenthetically correspond to abbreviations in Figures 1 and 3.

ecoregions of the south central United States (Drenner et al. 2013), we examined the relationship between average estimated concentration of Hg in the blood of piscivorous wading birds and conifer-adjusted Hg deposition. Atmospheric Hg adheres to components of conifers (i.e., needles and other material such as bark, branches, and reproductive structures) and can be incorporated into needles when Hg enters through the stomata (Graydon et al. 2008). Mercury is then transported to the ground via throughfall and litterfall (Graydon et al. 2008). Mercury deposition can be 5 times higher under the canopies of conifer forests compared with open areas (Drenner et al. 2013). In the present study, we used conifer-adjusted Hg deposition determined by Drenner et al. (2013) for the 14 ecoregions (Table 1). These authors computed conifer-adjusted Hg deposition data using conifer coverage from the 2006 National Land Cover Database and annualized precipitation-weighted average Hg concentration between 2006 and 2009 from weekly observations in the US National Atmospheric Deposition Program/Mercury Deposition Network.

We assessed the potential level of risk posed by Hg to the health of piscivorous wading birds using risk categories described in Jackson et al. (2016) that were based on a literature review in Ackermann et al. (2016). We acknowledge that selection of risk benchmarks for avian piscivores is complicated by variation in sensitivity among taxa and limited data on effects (Jackson et al. 2016), which can result in alternative interpretations of risk (e.g., Fuchsman et al. 2017). In the present study, estimated concentrations of Hg in blood were classified into 5 risk categories: 1) background (<0.5 μg/g) or below adverse effect thresholds, 2) low risk (0.5–1.0 μg/g) or elevated above background but below most

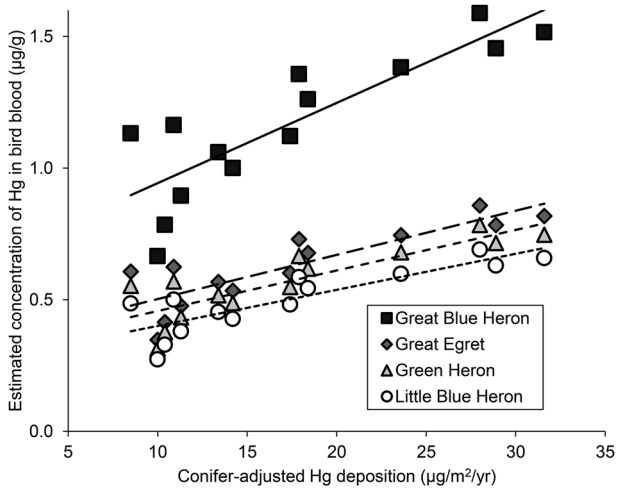

FIGURE 2: Relationship between the average estimated concentrations of mercury (Hg) in the blood of 4 piscivorous wading birds and conifer-adjusted Hg deposition in ecoregions of the south central United States. Linear relationships were $y = 0.031x + 0.64$, $y = 0.017x + 0.33$, $y = 0.015x + 0.30$, and $y = 0.014x + 0.26$ for great blue herons, great egrets, green herons, and little blue herons, respectively.

effect thresholds, 3) moderate risk (1.0–2.0 μg/g) or adult physiological and behavioral abnormalities likely to occur, 4) high risk (2.0–3.0 μg/g) or reproductive impairment likely to occur, and 5) extra high risk (>3.0 μg/g) or a high likelihood of significant reproductive failure (Jackson et al. 2016). Within each ecoregion, we determined the percentage of sampling sites in each risk category.

For each of the 4 species of wading birds, we tested for differences in the average estimated concentrations of Hg in blood between the 14 ecoregions using univariate analysis of variance (ANOVA). Data were log-transformed prior to statistical analysis because they did not meet the assumption of normality, but the figures depict non–log-transformed data for ease of interpretation. We examined main effects of wading bird species and conifer-adjusted Hg deposition and their interaction effects on the estimated concentration of Hg in blood using analysis of covariance (ANCOVA). Data met the assumptions of ANCOVA. Statistical significance was inferred at $p < 0.05$, and all analyses were conducted with IBM SPSS Statistics 22 software.

RESULTS AND DISCUSSION

For each species of wading bird, we detected significant differences in average estimated concentrations of Hg in blood between ecoregions (ANOVA, $F_{13,714} = 11.1$, $p < 0.001$; Table 1 and Figure 1). For each species, the lowest and highest estimated concentrations of Hg occurred in the Texas Blackland Prairies and the Southern Coastal Plain, respectively, with a >2-fold difference in estimated concentrations of Hg in blood between these 2 ecoregions. For little blue herons, green herons, great egrets, and great blue herons, average estimated concentrations of Hg in blood ranged from 0.27 to 0.69, 0.32 to 0.78, 0.35 to 0.86, and 0.67 to 1.59 μg/g, respectively (Table 1).

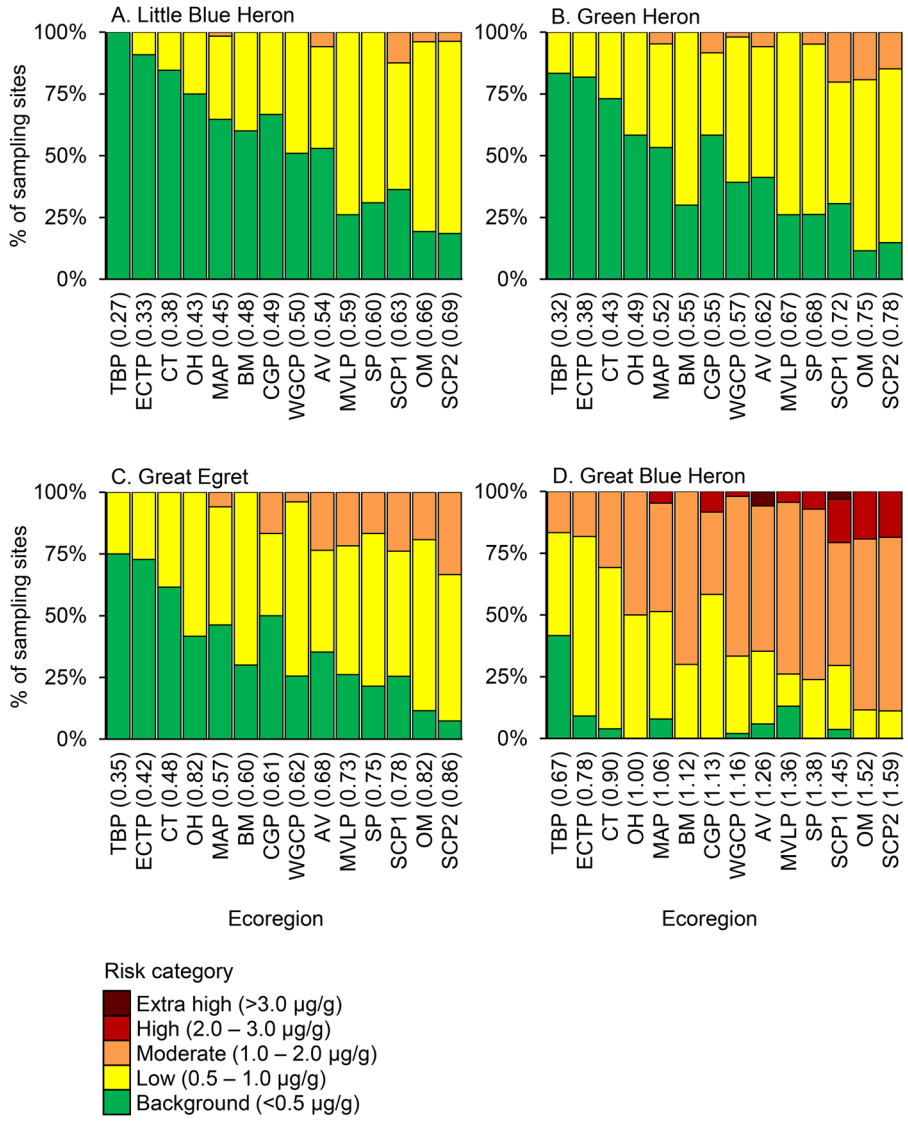

FIGURE 3: Percentage of sampling sites within ecoregions with estimated concentrations of mercury (Hg) in bird blood that correspond to Hg risk categories. (**A**) Little blue herons, (**B**) green herons, (**C**) great egrets, and (**D**) great blue herons. Risk categories are from Jackson et al. (2016). Ecoregions are arranged according to average estimated concentration of Hg in blood (μg/g; shown in parentheses on the *x*-axis). Ecoregion abbreviations are defined in the legend of Figure 1.

We detected a main effect of wading bird species (ANCOVA, $F_{3,48} = 6.4$, $p = 0.001$) on average estimated concentrations of Hg (Figure 2). Average (± standard error) estimated concentrations of Hg were 0.50 ± 0.03, 0.57 ± 0.04, 0.63 ± 0.04, and 1.17 ± 0.07 μg/g for little blue herons, green herons, great egrets, and great blue herons, respectively (Table 1). In a study of wading birds in south Florida, Sundlof et al. (1994) found that liver concentrations of Hg varied with species of wading birds with little blue herons < great egrets < great blue herons. Sundlof et al. (1994) concluded that wading birds that consume large fish as a major component of the diet (e.g., great blue herons and great egrets) accumulate greater concentrations of Hg than those species that primarily eat smaller fish or arthropods (e.g., little blue herons).

We detected a main effect of conifer-adjusted Hg deposition (ANCOVA, $F_{1,48} = 114$, $p < 0.001$) on average estimated concentrations of Hg in bird blood (Figure 2). For each bird species, the estimated concentrations of Hg increased with conifer-adjusted Hg deposition. However, the magnitude of the increase in the estimated concentration of Hg with increasing conifer-adjusted deposition varied with species, which resulted in a significant species × conifer-adjusted Hg deposition interaction effect (ANCOVA, $F_{3,48} = 4.6$, $p = 0.006$). The slope of the relationship between average estimated concentrations of Hg in bird blood and conifer-adjusted Hg deposition was greatest for great blue herons, suggesting that this species would be at greater risk of Hg exposure in response to elevated Hg deposition.

The level of risk to wading birds in the present study varied with species and ecoregion (Figure 3). Within ecoregions, the percentage of sampling sites with risk above background increased with the average estimated concentration of Hg in blood. Little blue herons, green herons, and great egrets were predicted to be at less risk than great blue herons. Within individual ecoregions, great blue herons were predicted to be at low risk to extra high risk at 58 to 100% of the sampling sites. Relative to the other 3 species, great blue herons were predicted to be at moderate risk at a higher percentage of sampling sites. Only great blue herons were predicted to be at high or extra high risk at any site.

Although the present study is the first spatial analysis of Hg contamination and associated risk to adults of 3 of the species examined (little blue herons, green herons, and great egrets), adult great blue herons have been included in other studies of Hg risk to piscivorous birds in North America (Ackerman et al. 2016; Jackson et al. 2016). Studies from western North America found average concentrations of Hg in great blue heron blood of 0.21 μg/g (Ackerman et al. 2016). More than 90% of the great blue herons that were sampled were categorized as having no to low risk from Hg in western North America (Ackerman et al. 2016). Conversely, in the south central United States, we estimated that great blue herons would have average concentrations of Hg in their blood of 0.67 to 1.59 μg/g, which is 3 to 8 times that found by Ackerman et al. (2016) in great blue herons in some areas of western North America. These results indicate that great blue herons may be at greater risk in the south central United States than in western North America, possibly due to elevated Hg deposition in the south central United States compared with the western United States (National Atmospheric Deposition Program 2018). The risk to great blue herons may extend from the south central United States throughout the southeastern United States. The Southeastern Plains and Southern Coastal Plain, 2 ecoregions with high conifer-adjusted Hg deposition and high predicted concentrations of Hg in bird blood, extend from our study area across the southeastern United States to the Atlantic coast. We recommend that future studies of Hg contamination of piscivorous wading birds of the southern United States directly monitor Hg concentrations and should focus on great blue herons in waterbodies within ecoregions that have high conifer-adjusted Hg deposition.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at DOI: 10.1002/etc.4299.

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Data Accessibility—Data are available on request from the corresponding author (m.m.chumchal@tcu.edu).

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