Pathways and consequences of contaminant flux to Acadian flycatchers (*Empidonax virescens*) in urbanizing landscapes of Ohio, USA

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HIGHLIGHTS

- We studied Hg levels in sediment, water, aquatic insects, and Acadian Flycatchers.
- One goal was to explore Hg effects on flycatcher reproduction at riparian sites.
- We also evaluated if the exposure pathways differed by landscape context.
- Low Hg levels in the flycatchers were negatively related to reproductive success.
- Flycatcher Hg levels were negatively related to sediment Hg in rural landscapes.

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ABSTRACT

A prevalent environmental contaminant, mercury (Hg) is mobile and persistent in aquatic systems, where it often occurs in its bioavailable form meth mercury. Because meth mercury can bioaccumulate in aquatic insects and then transfer to terrestrial food webs, riparian consumers reliant upon aquatic emergent insects, should be disproportionately affected. Using the aerial insectivore Acadian flycatcher (*Empidonax virescens*) as a focal species, we examined (1) the extent to which total Hg loads in breeding flycatchers affected body condition and reproductive output and (2) potential pathways of contaminant flux in 19 riparian forest fragments distributed across an urban-to-rural landscape gradient in Ohio, USA. From April–August 2011–2012, we collected blood samples from adult (n = 76) and nestling (n = 17 from 7 nests) flycatchers, monitored their annual reproductive success (i.e., total number of fledglings), and sampled water, sediment, and aquatic emergent insects at each site. Hg concentrations in adult flycatcher blood (47 to 584 μg/kg, x = 211.8, SD = 95.5) were low relative to published advisory levels and not related to body condition. However, even at low concentrations, blood Hg was negatively related to reproductive success, with a 0.83 decline in the number of fledglings per μg/kg (log x) increase of blood Hg. Adult flycatchers had 11x greater concentrations of blood Hg than their offspring. Hg levels in flycatcher blood were not predicted by Hg concentrations in sediment, water, or aquatic emergent insects, with the exception of rural landscapes alone, in which flycatcher Hg was negatively related to sediment Hg. In addition to illustrating the difficulty of predicting exposure pathways that may vary among landscape contexts, our study provides evidence that even trace levels of contaminants may impair reproductive success of free-living songbirds.

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1. Introduction

Aside from mortality, mercury as an environmental contaminant can elicit a wide range of sub-lethal effects, including altered behavior, depressed immune function, diminished reproductive productivity, and decreased body condition (Brasso and Cristol, 2008; Cristol et al., 2008; Hargreaves et al., 2010; Jackson et al., 2011a). Among contaminants likely to adversely affect wildlife, mercury (Hg) is of particular concern because it is geographically widespread due to atmospheric deposition (Swain et al., 1992; Fitzgerald and Engstrom, 1998; Blais, 2005; Pacyna et al., 2010), often remains in aquatic sediment for long periods (>50 years), and can be redistributed over large areas with flood events (Jackson et al., 2011b). Of significant concern, Hg emissions have increased over the past 100 to 150 years, and atmospheric transport and deposition of Hg at locations far from point sources have increasingly been observed (Swain et al., 1992; Fitzgerald and Engstrom, 1998; Weiss, 2005). Hg may be particularly problematic in urban landscapes due to both point-source inputs and high atmospheric concentrations associated with industrial activity (VanMetre, 2012). As such, rivers
moving through cities tend to have elevated contaminant levels in part because their sediment can act as long-term reservoirs of contaminants (Chang and Cockerham, 1994).

Mercury can enter and biomagnify in aquatic ecosystems through multiple pathways (Leadley and Gottgens, 2001; Evers, 2005; Blais et al., 2007; Walters et al., 2008). Aquatic insect larvae can bioaccumulate Hg either through food intake or via passive facilitated diffusion from the water itself (Rainbow, 1996). As these insects emerge as adults (hereafter ‘aquatic emergent insects’), they may serve as a vehicle for the transport of Hg from aquatic to terrestrial systems, through consumption by terrestrial consumers (Menzie, 1980; Cristol et al., 2008; Sullivan and Rodewald, 2012). Nakano and Murakami (2001), for example, observed that aquatic emergent insects can provide up to 35% of the diet for insectivorous summer resident birds in the riparian zone. In fact, insectivorous songbirds foraging in riparian habitats can concentrate contaminants at levels comparable to those detected in piscivorous bird species (Brasso and Cristol, 2008; Cristol et al., 2008). Thus, trophic pathways can lead to Hg biomagnification across aquatic and terrestrial systems (Menzie, 1980; Cristol et al., 2008; Walters et al., 2008; Sullivan and Rodewald, 2012).

Many aerial insectivorous bird species, including the Acadian flycatcher (Empidonax virescens; hereafter “flycatcher”; Bisson et al., 2000; Whitehead and Taylor, 2002) breed in riparian forests and consume a variety of insect prey, including aquatic emergent insects, which could make them susceptible to contaminant exposure from both aquatic and terrestrial environments. Aerial insectivorous birds are thought to be threatened by widespread declines in insect populations as well as potential mismatches in reproductive timing with peak insect abundance resulting from climate change (Nebel et al., 2010; North American Bird Conservation Initiative, U.S. Committee, 2010). As a member of the declining aerial insectivore guild (Nebel et al., 2010), the flycatcher also is of conservation interest and is ranked as a species of “Medium” vulnerability in the 2010 State of the Birds Report (North American Bird Conservation Initiative, U.S. Committee, 2010).

Previous studies have documented adverse reproductive effects of elevated Hg levels in insectivorous songbirds (Evers et al., 2012). For example, in an area with point-source contamination of Hg, tree swallows (Tachycineta bicolor) produced fewer fledglings than in an uncontaminated reference area, and second-year females produced smaller eggs than older females within the contaminated area (Brasso and Cristol, 2008). In the Scioto River system of central Ohio, where adverse effects on aquatic life is a serious concern due to sediment contamination (U.S. Environmental Protection Agency, 2004), Hg concentrations in multiple species of riparian swallows have recently been found to be higher at rural vs. urban reaches and at least in part linked to dietary reliance on aquatic emergent insects (Alberts et al., 2013). As is the case with many other rivers of the US Midwest, the Scioto River flows through a mixture of urban, agricultural, and forested landscapes, and thus human use has created a legacy of contamination of aquatic biota (e.g., Blockson et al., 2010). However, potential relationships between aquatic system contamination and reproductive success of aerial insectivorous birds in different landscape contexts (i.e., urban and rural) remain unresolved.

Because of a shared reliance of riparian swallows and flycatchers on a mixture of aquatic and terrestrial flying insect prey, we anticipated that flycatcher Hg loads would also be linked to sediment and aquatic emergent insect contaminant loads in the Scioto River system, and that this exposure would relate to flycatcher reproductive success. Using linear mixed models, we examined the extent to which total Hg (hereafter, Hg) loads in breeding flycatchers from 19 riparian study sites distributed in urban and forested landscapes of the Scioto River system in and around Columbus, OH affected body condition and reproductive output. Using weighted linear regression, we also explored potential aquatic-to-terrestrial pathways of contaminant flux in river–riparian systems of Ohio, USA. Our results introduce initial evidence that even trace levels of Hg may be related to flycatcher reproductive success and that pathways of Hg exposure may vary between urban and rural landscapes.

2. Materials and methods

2.1. Study system

Our study system was located in the Upper Scioto River basin in central Ohio, USA. Sites (19) consisting of both a mature riparian forest fragment (>100 m wide and ≥250 m long) and an adjacent river reach were located along Alum Creek, Big Darby Creek, Big Walnut Creek, Blacklick Creek, the Olentangy River, and the Scioto River mainstem (Appendix A, Fig. 1). Dominant tree species at study sites consisted of American elm (Ulmus americana), American hackberry (Celtis occidentalis), black walnut (Juglans nigra), boxelder (Acer negundo), Eastern cottonwood (Populus deltoides), honey locust (Gleditsia triacanthos), silver maple (Acer saccharum), sycamore (Plantanus occidentalis), sugar maple (Acer saccharorum), and white ash (Fraxinus americana). Woody understory vegetation included honeysuckle (Lonicera spp.), multiflora rose (Rosa multiflora), common spicebush (Lindera benzoin), dogwood (Cornus spp.), hawthorn (Crataegus spp.), Ohio buckeye (Aesculus octandra), tall paw paw (Asimina triloba), and saplings of overstory trees (Borgmann and Rodewald, 2004; Leston and Rodewald, 2006).

2.2. Avian sampling and reproductive monitoring

Territorial adult flycatchers were captured in mist nets on average 39 days after settlement at our study sites, between 14 June 2011 to 13 August 2011 and 29 May 2012 to 11 August 2012. This ensured that sampled blood reflected a bird’s diet on the breeding habitat rather than from the migratory period. Blood reflects an organism’s diet within the short term (9–15 d; Hobson and Clark, 1992; Reapoh et al., 2002; Evers et al., 2005). Blood samples were collected from nesting flycatchers when feasible (i.e., nest height < 3 m) at 6 to 9 days after hatching to assess potential age-related differences in Hg concentrations. Acadian flycatcher nests are typically located at the tips of thin tree branches above 3 m in height (Rowse, personal observation), and as such we were unable to sample nestlings from many of the monitored nests without risk of destroying nests. Our nesting sample size was also restricted due to the depredation of many nests prior to hatching or while the nestlings were too small for blood sampling. We captured a total of 76 adult flycatchers (57 males, 17 females, and 2 of unknown sex; 16 total in 2011, 60 in 2012, with 1–7 individuals per site). We sampled 17 nestlings from 6 territories (7 total nests, with 2 nests averaged as 1 for a single territorial male), all in 2012. Nestling samples from each nest were averaged into a single blood Hg concentration for that nest, with Hg values below the detection limit treated as “0” for the mean (Table 1).

Each individual flycatcher was banded with a numbered aluminum USGS size 0 band and a unique combination of three color bands. Blood was sampled immediately following banding and the collection of morphological measurements (i.e., mass, tarsus length, fat), following the procedures described by the Ornithological Council (Fair et al., 2010). We calculated size-adjusted mass based on these measurements, as described in Section 2.6. Approximately 50 to 100 μl of blood was collected (~1% of the individual’s body mass) from the subcutaneous ulnar (brachial) vein punctured with a disposable 26G needle (BD PrecisionGlide intradermal bevel needle). Blood samples were immediately transferred to dried blood spot (DBS, PerkinElmer, Waltham, MA, USA) cards. DBS cards are useful for sampling blood from small-bodied birds (<20 g; adult birds in our study had a mean weight of 12.8 g, SD = 0.7 g, nesting mean weight = 9.7 g, SD = 1.4 g), in that a small volume of blood is sufficient for analysis (Shlosberg et al., 2011; Stove et al., 2012). All flycatcher sampling was conducted in accordance with the Institutional Animal Care and Use Committee.
Acadian flycatcher territories within study sites were intensively monitored at 2–5 day intervals, with shorter intervals as time of expected fledging approached, from May through August 2011 and 2012. Nest searching was conducted both through observing behavior and systematic searching of vegetation within territories. All nests were monitored until failure or fledging. We tracked the total number of young produced by individual flycatchers for each year (i.e., across all nesting attempts) by monitoring color-banded individuals throughout the season.

### Sediment, water, and aquatic emergent insect sampling

In late summer and early fall 2012, we collected river sediment at each study site reach following Alberts et al. (2013). We collected samples at 5 locations evenly distributed along the river reach.
(approximately 200 m along the study site), using a push corer and sampling 0–10 cm in depth. Sediment samples were stored in plastic bags and subsequently homogenized and dried by reach. Along with sediment, we collected samples of river water at one location per reach. Water samples were stored in 250 mL polyethylene bottles and refrigerated until analysis. Sediment and water samples were analyzed for a panel of toxic elements (Table 2) at the Michigan State University Diagnostic Center for Animal and Population Health (DCPAH, methods described below). All contaminant concentrations in water samples were below detection limits, and thus were not included in statistical analyses.

Aquatic emergent insects were collected from each reach between 25 August 2012 and 9 October 2012, using floating, 1-m² pyramidal emergent insect traps similar to the style of Aubin et al. (1973). Invertebrates were collected at 10 days after traps were set, were preserved in 70% ethanol, and later identified to order in the laboratory with the use of Triplehorn and Johnson (2005). Aquatic emergent insects were dried at 60 °C for 48 h, and numerically dominant families were homogenized by reach for analysis at DCPAH. Analysis for total Hg (Table 2) was conducted using inductively coupled plasma-atomic emission spectrometry (methods described below).

2.4. Toxicology analysis

All samples were analyzed at the Diagnostic Center for Animal and Population Health, Toxicology Section, Michigan State University, for a panel of toxic elements, including Hg (µg/kg, dry weight). Instrument calibration [ICP-AES (Varian Vista, currently with Agilent, Santa Clara, CA) and ICP-MS (Agilent 7500ce)] was performed with NIST-traceable stock solution standards for each element (GFS Chemicals, Inc., Cincinnati, OH). Lab reagent blanks [NIST (National Institute of Standards and Technology, Gaithersburg, MD) – traceable Multi-mix (Alfa Aesar Speckpure, Ward Hill, MA)] and NIST Standard Reference Materials (SRM) digests (Mussel Tissue 2976, Montana II Soil 2711a, and/or Trace Elements in Water 1643e) were analyzed for quality assurance in each sequence. Cold vapor atomic absorption spectrometry (CECIA CVAA, Omaha, NE) was used for Hg analysis, with similar standards, instrument calibration, and quality control. Although total Hg was measured rather than methylmercury, concentrations of Hg in blood of insectivorous birds have been shown to be primarily methylmercury (>95%; Evers et al., 2005; Rimmer et al., 2005).

2.5. Landscape gradient

The amount of urbanization within 1 km of each forested site was previously quantified using a principal component analysis (PCA) on five variables (percent cover of pavement, roads, buildings, agriculture, lawn, and number of buildings), with the first component explaining >79% of variation among sites (eigenvalue = 3.99), and referred to as the “urban index” (Rodewald and Shustack, 2008). This urban index represents a gradient of landscape composition change. Positive urban indices (≥0) represent more developed landscapes with greater numbers of buildings (>800) and higher percentages of road cover, pavement, and lawn, whereas negative indices (<0) are more rural with higher percentages of agriculture (Rodewald and Shustack, 2008, Appendix A).

2.6. Statistical methods

Statistical analyses were performed using R 3.0.0 software (R Core Team, 2013). Contaminant data were loge transformed to meet the assumption of normality prior to statistical analyses. Every flycatcher for which we had blood tested was included in the analysis (i.e., including those for which no Hg was detected).

We used linear mixed-effects models fit by maximum likelihood estimates (R package “nlme” (Pinheiro et al., 2012)), to evaluate the potential relationship between Hg concentrations in blood and flycatcher productivity. Models included blood concentrations of Hg as a fixed effect and site as a random effect variable. Site was treated as a random effect variable because (1) Hg exposure might have varied among sites as a consequence of landscape position, (2) Hg was measured for multiple individual birds at each site, and (3) Hg levels of individuals within each site were unlikely to be independent. Samples from both years were used in analyses because Hg concentrations in blood were not significantly different between years (F1,74 = 0.665, P = 0.418). Because males and females did not differ significantly in blood concentrations of Hg (F2,73 = 1.045, P = 0.357), we used only samples from males in the productivity models to avoid multiple samples for a given territory. Adult contaminant concentrations were compared to nesting concentrations using a paired t-test.

The relationship between blood contaminant concentration and size-adjusted body condition in adult flycatchers was evaluated using linear mixed models with blood contaminant concentration as fixed effects and site as a random effect. The size-adjusted measure of body condition was calculated from a PCA (R package “lattice” (Sarkar, 2008)) on wing, tarsus, and tail, resulting in a frame size PC. Separately for each sex, we then ran a linear regression of the first principle component on mass (regression was significant for males: F1,55 = 9.149, P = 0.004, but not for females: F1,15 = 0.012, P = 0.920). We used the residuals from this regression (i.e., deviations of each bird from their expected mass given frame size) as our index of body condition for males (Rowse, 2013).

To explore potential relationships between Hg in flycatchers and in the aquatic system (e.g., sediment and aquatic emergent insects), we used weighted linear regressions which accounted for differing numbers of flycatcher blood samples at each site, (i.e., site was the unit of measure). We assessed relationships between (1) urbanization and Hg in sediment, insects, and flycatcher blood, (2) sediment and insect Hg concentrations, (3) insect and flycatcher blood Hg concentrations, and (4) sediment and flycatcher blood Hg concentrations.

Given our small sample size and the high variability among individual birds, we used α ≤ 0.05 to indicate statistical significance, and 0.05 < P ≤ 0.10 to indicate a trend (as per Bocharova et al., 2013). Overall fit of the models was checked visually with plots of residuals and by examining parameter estimates.

![Fig. 2.](image-url) Weighted regression between sediment mercury in rural sites on blood mercury (µg/kg, loge) in Acadian flycatchers was significant (R² = 0.216, F1,30 = 8.286, P = 0.007). Samples collected from study sites in central Ohio, USA.
3. Results

Hg concentrations in adult flycatcher blood ranged from 47 to 584 μg/kg (X = 211.8, SD = 95.5). Hg in blood was not significantly related to male body condition (F1,35 = 0.003, P = 0.955). Hg concentrations in 17 nestlings ranged from below detection limits (<20 μg/kg) to 38 μg/kg (X = 19.1, SD = 13.5).

Blood concentrations of Hg were negatively related to the number of fledglings produced by males (F1,26 = 3.021, P = 0.094, Fig. 2) and females (Pearson’s r = −0.521, df = 14, P = 0.039). Blood Hg concentrations were 11 × higher for adult males than for nestlings (paired t-test: t = 4.595, df = 5, P = 0.006) and were negatively correlated with nestling blood Hg concentrations (Pearson’s r = −0.857, P = 0.029).

Hg concentrations in water were below detection limits (i.e., Hg < 10 ng/L, Table 2). Hg concentrations (μg/kg, dry weight) varied from 7 to 99 μg/kg (X = 26.3, SD = 22.7, Table 2) in river sediment and from 36 to 238 μg/kg (X = 95.3, SD = 58.4, Table 2) in aquatic emergent insects. Dipteran families were typically the most numerically-dominant families collected from the study reaches, with small-bodied Chironomidae exhibiting the greatest relative abundance at many sites. Thus, the majority of insects collected were <10 mm in length.

Weighted linear regression showed that Hg concentrations in sediment were not significantly related to aquatic emergent insect (F1,9 = 2.988, P = 0.122) or flycatcher blood (F1,52 = 0.454, P = 0.504) Hg loads. Likewise, Hg in insects did not predict Hg concentrations in flycatcher blood (F1,40 = 2.145, P = 0.148).

Urbanization was not related to Hg concentrations in insects (F1,14 = 1.722, P = 0.211), sediment (F1,13 = 0.3376, P = 0.571), or blood (F1,17 = 0.002, P = 0.968). However, when urban and rural sites were analyzed separately, we found evidence of a landscape influence, such that there was a significant negative relationship between sediment and flycatcher Hg in rural sites but not at urban sites (Fig. 2).

4. Discussion

Despite finding relatively low levels of Hg (47 to 584 μg/kg, X = 211.8, SD = 95.5) in the blood of adult Acadian flycatchers, we found that reproductive success declined with increased Hg loads. A growing body of literature suggests that terrestrial insectivorous birds in riparian habitats can be exposed to Hg and other contaminants via prey-mediated contaminant flux from aquatic to terrestrial systems (Menzie, 1980; Cristol et al., 2008; Walters et al., 2008; Sullivan and Rodewald, 2012), yet we found that Hg concentrations from aquatic sediment were not related to blood concentrations of Hg in flycatchers across the urban–rural landscape gradient. Given that body condition was unrelated to Hg levels, reduced reproductive success in birds with higher Hg levels in blood might instead be explained by changes in adult behavior (e.g., nest abandonment, altered provisioning or incubation), or by egg hatching success (not documented in our study due to nest heights). Although brood parasitism and predation are common in our system and known to depress reproduction (Rodewald et al., 2013), neither of these interspecific interactions seem to drive the reduced reproductive output associated with Hg. Even after excluding nests that were parasitized or depredated (partially or fully), the number of fledglings and blood Hg were still negatively correlated (r = −0.44, n = 21, P = 0.04).

Diminished reproductive success due to Hg contamination has been documented in many avian species (Evers et al., 2008; Wolfe et al., 1998), concordant with our results. Reduced reproductive success of Carolina wrens at Hg contaminated sites was attributed primarily to nest abandonment (Jackson et al., 2011a). Likewise, Brasso and Cristol (2008) found that tree swallows produced fewer fledglings in a highly contaminated area compared to a reference area, but this pattern emerged only for second-year breeding females and could be attributed to other factors besides Hg contamination. Longcore et al. (2007) found that fledging success for tree swallows was not affected by Hg loads in nestlings, thus it is more likely that adult Hg loads affect overall reproductive success rather than nestling Hg loads. Although many studies have documented the negative reproductive consequences of Hg in blood in birds (e.g., Spalding et al., 2000; Brasso and Cristol, 2008; Jackson et al., 2011a), our study presents initial evidence that even trace levels of Hg may be related to depressed reproductive output of free-living songbirds. In fact, birds in our study were on average 70% below the Hg concentration threshold (~700 μg/kg) expected to depress nest success by 10% in several passerine species in the northeast U.S. (Evers et al., 2012). Moreover, our observation of reduced reproductive output is likely a conservative estimate, as we did not account for potential sublethal effects of Hg such as egg hatching success (often not measurable due to nest heights) or altered behavior in adults provisioning nestlings or fledglings (Jackson et al., 2011a). However, trace level effects of Hg on reproduction in this system may suggest that higher levels of Hg would have a greater negative effect. Thus, our findings present a cautionary note given the prevalence and persistence of Hg in the environment (Evers, 2005).

Differences in Hg levels between adults and nestlings illustrate the difficulty of generalizing patterns of exposure among individuals even within the same species and family group. The higher blood levels of Hg in adults likely were not a consequence of diet, as nestlings are reported to consume food items similar to adults (Whitehead and Taylor, 2002). However, the possibility remains that adults consume larger-bodied insects (e.g., odonates) that may disperse farther from water and might biomagnify more contaminants than smaller-bodied insects (e.g., midges) that typically occupy lower trophic positions.

Likewise, the relationship between Hg levels and nestling age (Rowse, 2013) might be related to provisioning biases, as Mumford (1964) observed adults bringing large-bodied prey (e.g., damselflies and dragonflies) only to large nestlings (only after a few days post-hatching). Another probable cause of low Hg levels in nesting blood is the depuration of Hg into feathers during the period of rapid feather growth (Spalding et al., 2000; Condon and Cristol, 2009). The low Hg levels in nestlings we found is consistent with companion studies in the same study system, where Alberts et al. (2013) observed that Hg concentrations in adult riparian swallows (multiple spp.) were significantly higher than those observed in nestlings. More surprisingly, we found that the concentrations of Hg in adult blood were negatively related to that in the blood of nestlings, though the sample size of nestlings was small (n = 17).

Given that Hg concentrations in sediment did not predict Hg in aquatic emergent insects, and insect Hg did not predict Acadian flycatcher blood Hg concentrations, there is likely a disconnect or missing link in the contaminant flux pathways examined. Alberts et al. (2013) recently found that Hg in sediment predicted blood Hg in four riparian swallow species in the same study system. Interestingly, their study showed that despite similar levels of Hg contamination in river sediments, rural swallows had higher levels of Hg than birds in urban river reaches (Alberts et al., 2013). Although we have little evidence of a strong landscape effect, the fact that Hg concentrations in sediment and flycatcher blood were significantly associated only at rural sites suggests that there may be a change in biogeochemical processes or flycatcher foraging behavior along the urban–rural gradient that mediates the relationship between sediment and flycatcher Hg concentrations. Indeed, among other predator variables including stream discharge and annual precipitation, Shanley et al. (2005) found that percent urban land cover was an important predictor of total Hg in streams. Inclusion of ephemeral pools or flooded areas within the riparian floodplain may also be important to consider as wetlands and other lateral aquatic habitats can increase mercury methylation in redox conditions (Shanley et al., 2005) and could be important foraging areas for flycatchers.

Relationships between sediment and blood Hg levels also may be obscured by differences in the bioavailability of Hg in sediment, which can alter the amount of uptake by aquatic insects (Edmonds et al., 2012). It may be important to carefully consider which aquatic insect taxa to include in analyses of contaminant flux pathways; insects that
more on aquatic autotrophic pathways than detrital-based pathways (e.g., terrestrial leaf inputs) can have greater Hg loads due to differences in uptake (Leady and Gottgens, 2001; Jardine et al., 2012). Because the amount of methylmercury (a bioavailable and toxic form of Hg) was not identified in laboratory analyses, but rather total Hg was measured, the proportion of Hg in sediment that was bioavailable for uptake by insects in this system is not known. Likewise, we lack data on the organic carbon content of sediment or water pH, which can influence bioavailability (Edmonds et al., 2010; Jardine et al., 2012). That said, several studies of trophic transfer pathways and biomagnification of contaminants have not measured or used organic carbon content in analyses, yet were able to detect significant relationships between aquatic prey and terrestrial predators in contaminant biomagnification (e.g., Walters et al. (2008, 2010)), including studies in the same river system (Alberts et al., 2013).

Our inability to strongly link Hg flux from sediment or aquatic emergent insects to Acadian flycatchers in our system suggests that the diet of Acadian flycatchers may either be composed of a greater relative proportion of terrestrial insects (or potentially aquatic insects that rely heavily on detrital-based energy pathways and thus may not as strongly bioaccumulate Hg) than we anticipated (e.g., foraging in upland habitat, farther than aquatic emergent insect dispersal distances) or be highly variable temporarily (e.g., foraging on aquatic emergent insects primarily during large emergence events that may not have been captured during our 10-day sampling periods (Nakano and Murakami, 2001; Iwata et al., 2003)). This latter possibility is of special concern given that insect and flycatcher sampling were separated by 1–2 months. Although bioassay of the aquatic emergent insect flux has been observed to be relatively constant between early summer ($\pi = 11.4$ mg $m^{-2} \cdot$ day$^{-1}$) and late summer/early fall ($\pi = 8.9$ mg $m^{-2} \cdot$ day$^{-1}$) in the Scioto River system, flux density early in the season ($\pi = 5.79$ individuals $m^{-2} \cdot$ day$^{-1}$) can be much higher than later ($\pi = 3.15$ individuals $m^{-2} \cdot$ day$^{-1}$), Kautza and Sullivan, unpublished data). However, how these shifts in the aquatic emergent insect community influence flycatcher reliance on aquatic insects remains unclear. Thus, further study documenting flycatcher foraging activities in river systems, relative to both taxonomic and/or size preferences as well as temporal variability in diet will be important to resolve these questions. Additional research is also needed in our system to determine if biogeochemical factors (e.g., water pH, dissolved organic matter) or forest structure along riverbanks alter the pathways of contaminant flux to terrestrial insectivores along urban–rural gradients.

5. Conclusions

Our study contributes to the growing body of knowledge regarding effects of Hg contamination to passerine reproductive success, and provides insight into effects of trace levels of mercury on a free-living terrestrial insectivore. We found suggestive evidence that environment-to-consumer contaminant pathways may vary even within the same region, likely a consequence of variability in behavioral, biogeochemical, and landscape attributes. Thus, pathways of Hg flux from aquatic to terrestrial systems may be context dependent. For example, the contrasting relationship we found between Hg in sediment and in flycatcher blood at urban and rural sites warrants further investigation, especially in relation to landscape features that we did not examine such as river impediments. Because contamination of watersheds is widespread and persistent (U.S. Environmental Protection Agency, 2004), managers should consider inclusion of an aquatic component to management of terrestrial organisms in order to more fully capture and understand the sources and fates of contaminants and their consequences on wildlife.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.03.095.

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