EXHIBIT C

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Widespread Contamination of Polychlorinated Biphenyls in South Carolina and North Carolina (USA): A Legacy of Malarial Eradication and Mosquito Control

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Abstract. The South Carolina Department of Health and Environmental Control (SCDHEC) has been conducting fish tissue monitoring for Polychlorinated Biphenyls (PCBs) since 1974 and, based on the results, restrictive fish consumption advice has been in place at two reservoirs in South Carolina for several decades. But in 2009, widespread contamination was reported in fish from the Catawba-Wateree and Yadkin-Pee Dee River Basins. Therefore, beginning in 2010, additional monitoring of fish tissue for PCBs in the rivers and reservoirs of these two basins was initiated. Results from a spatial analysis, combined with evidence from historic literature, suggests that the source of the PCB contamination, in part, is from past direct application of used transformer oil on reservoirs located along the two rivers, the origins of which were hydroelectric projects in both basins. The use of used motor oil for mosquito control and malaria eradication was widespread in the first half of the twentieth century, and results suggest that for some utility operations, PCB oil was utilized to augment these programs. The global ramifications of these findings are not yet known, but they should encourage reconsideration of origin, transport, and fate of PCBs in other regions, particularly where a known source of environmental contamination is not obvious.

INTRODUCTION

Polychlorinated Biphenyls (PCBs) were discovered in the late 1880s with commercial production by the company Monsanto beginning in 1927 (Cairns et al. 1986). Because they are chemically stable, are of low flammability, and are poor conductors of electricity they had many industrial applications, especially as a cooling oil for electric transformers and capacitors. They were also widely used in paints and resins, carbon paper production, and hydraulics. The toxicity of PCBs to humans was acutely seen when families in Kyushu, Japan, exhibited disfiguring dermatitis and liver and kidney damage after consuming PCB-contaminated rice oil (Saeki et al. 1971). PCBs are considered a probable human carcinogen by the US Environmental Protection Agency (EPA), and US production ceased in the late 1970s (Eisler 1986). However, because of their chemical stability and widespread use, PCBs remain in the environment and continue to be found in elevated levels in soil and in the tissue of fish and other animals.

Structurally, PCBs were produced by the chlorination of biphenyl rings resulting in 209 possible configurations, which are generally referred to as congeners. These congeners can be grouped based on the number of chlorine atoms, regardless of position, into 10 possible classes called homologs (e.g., tetrachlorobiphenyls). Aroclor was the brand name given to PCB mixtures by the company Monsanto, which produced most of the PCBs in the United States (Cairns et al. 1986). The trade name Aroclor was followed by a four-digit numeric value (e.g., Aroclor 1260), with the first two digits indicating the number of carbon atoms and the last two indicating the approximate percentage, by mass, of the chlorine content.

South Carolina waterways are ecologically diverse, containing mountainous, piedmont, and coastal ecoregions (Griffith et al. 2002). Many waterways are self-contained within the boundaries of the state while some originate in, or are shared by, Georgia, North Carolina, and a small portion of Virginia. Historically, the state of South Carolina has issued fish consumption advisories due to the presence of contaminants in these waterways. There are four distinct regions of South Carolina that contain levels of PCBs in fish tissue that resulted in restrictive consumption advice being issued (SCDHEC 2021) (Figure 1). These regions are Lake

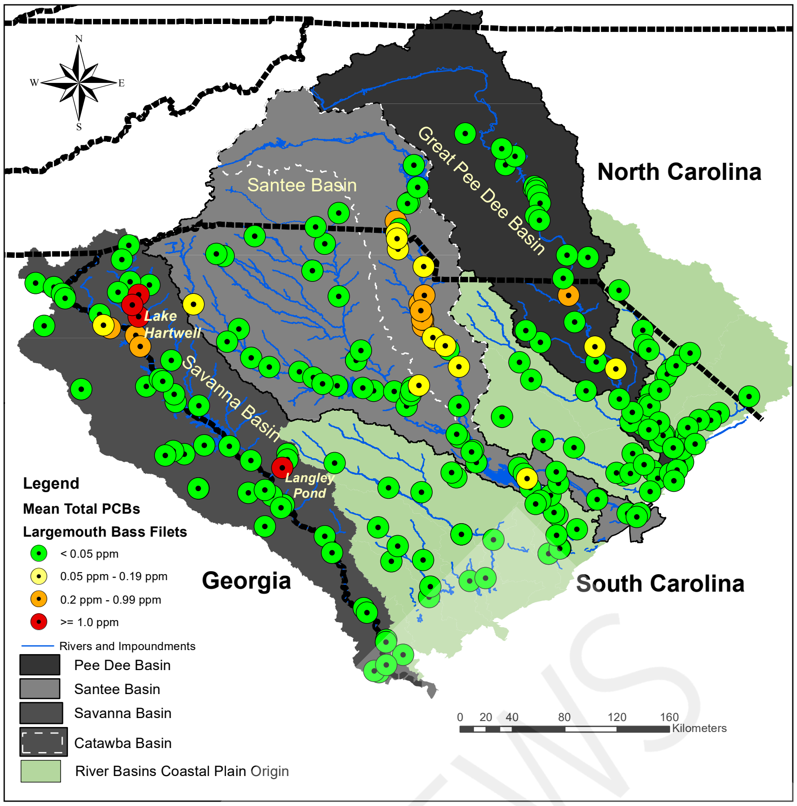

Figure 1. Mean total PCBs (ppm wet weight) in largemouth bass fillets from waters of South Carolina, showing major drainage basins of the state.

Hartwell in northern SC, Langely Pond in Aiken County SC, the Catawba River Basin, and the Great Pee Dee River Basin.

The first report of PCBs in biota of South Carolina was from Lake Hartwell, Pickens County, in 1976 (Bruner and Hill 1977; Aldridge 1978). The source of this contamination was an industrial discharge that emptied into Twelve Mile Creek, which entered Lake Hartwell a short distance downstream. Since then, significant research has been conducted on PCB contamination in this region, and in 1990 the Sangamo Operable Unit Two, the point of contamination, was finalized on the National Priorities List (US EPA Superfund) (Gaymon 1992a). Elevated levels of PCBs in fish tissue were also reported from Langley Pond, Aiken County, South Carolina (Darr 1986). This reservoir was created in the late 1800s and received large amounts of industrial waste, mostly from textile plants, for nearly 100 years. In more recent years, widespread PCB contamination was also discovered in two large river basins of the Carolinas. Fish from the Catawba/Wateree Basin and the Great Pee-Dee/Yadkin Basin have been found to contain levels of PCBs to warrant both South Carolina and North Carolina issuing restrictive consumption advice for certain fish species (SCDHEC 2021; NCDHHS 2021). The rivers and reservoirs of the Catawba/Wateree and the Great Pee-Dee/Yadkin Basins are the focus of this paper.

Low but detectable levels of PCBs were first reported from whole fish samples from the Catawba River Basin in the 1970s (Aldridge 1978). While the Great Pee Dee River was monitored, no detectable levels of PCBs were reported at that time. PCBs were again reported from fish collected in 1986 from a large reservoir, Lake Marion, located downriver from the Catawba River (Marcus 1987). The first large-scale investigation of PCBs and other contaminants in Catawba River fish was conducted by the Duke Power Company in 1993 (Coughlan 1995). Largemouth bass fillets were collected from 26 different locations in both North Carolina and South Carolina, most from reservoirs managed by Duke Power in both states. In this study PCBs were not detected in largemouth bass fillets; however, the detection limits of 0.28 ppm to 0.35 ppm for Aroclor 1260 were much higher than the 0.05 ppm detection limits reported by Aldridge (1978) and Marcus (1987). These detection limits were also much higher than the initial threshold that triggers limited fish consumption advice by the SCHDEC and the North Carolina Department of Health and Human Services (NCDHHS).

Beginning in 1999, the US EPA (2009a) conducted a national probabilistic fish contaminant study of lakes and reservoirs in the US. The results of the study were summarized in a national report in 2009, which revealed PCBs contamination in fish collected from several reservoirs on the Catawba River. This rediscovery of PCBs in South Carolina fish brought renewed awareness of the problem, leading the SCDHEC and the North Carolina Department of Environmental Quality (NCDEQ) to conduct additional monitoring of this basin. The results led to the issuance of restrictive consumption advice for several fish species from the Catawba River chain of reservoirs and certain portions of the river by the SCDHEC and the NCDHHS. Not long after these findings, PCBs were found in fish from the upper Great Pee Dee River in South Carolina. Near this time, the NCDEQ was investigating PCBs in several reservoirs in the Yadkin/Pee Dee Basin, with an Alcoa Plant located on Badin Lake considered a possible source (NCDHHS 2009; Mort 2017). In South Carolina, restrictive fish consumption advisories were already in place for the Great Pee Dee River because of mercury, but the discovery of PCBs prompted an investigation into possible sources for widespread contamination in fish from both the Catawba and Pee Dee Basins. Therefore, the primary objective of the study presented here was to investigate and determine the probable source or sources of PCB contamination in the Great Pee Dee/Yadkin River Basin and the Catawba/Wateree Basin. A secondary outcome of the study was a characterization of the spatial trends of PCBs in fish of South Carolina, including those from waterways that are shared with Georgia and North Carolina.

MATERIALS AND METHODS

DESCRIPTION OF STUDY AREA

Figure 1 shows the area of study as we conceived it, showing a portion of Georgia that shares the Savanna River Basin with South Carolina and the portions of North Carolina. In addition to the Savanna River, the Great Pee Dee River and Santee River Basins are large Atlantic drainages with headwaters that originate in the Blueridge Mountains. There are also many smaller basins self-contained within the political boundaries of South Carolina, all of which originate in or near the coastal plain of the state. Highlighted in Figure 1 is the Catawba River Basin, an upper section of the Santee River Basin. Lake Hartwell and Langley Pond, in which high levels of PCBs have occurred for many years, are labeled and sampling sites are indicated with red points. All the waters of South Carolina, whether self-contained or shared, meander through the landscape of the state and empty into the Atlantic Ocean. Except for the Savanna River, the Atlantic terminus of which is shared with Georgia, the confluence of these waterways with the Atlantic occurs completely within the boundaries of South Carolina.

Like much of the United States below the historic glacial line, South Carolina lacks large lakes and very few, small, natural, lentic waterways. However, over the past 150 years, numerous large and small dams have been erected across rivers and streams, which are referred to by various terms such as lakes, reservoirs, impoundments, and ponds. No large reservoirs, defined as greater than 50 feet in dam height, existed in South Carolina in 1850, but by 1987 there were 92 (US ACE 2021). These dams were mostly erected beginning in the 1900s for various purposes including hydroelectricity production, flood control, drinking water sources, and a source for industrial cooling water. In most cases, over time, they also became important for recreation, with permanent and secondary homes being constructed along their shores. One of the more extensively modified rivers in the Carolinas is the Catawba River, with many hydroelectric reservoirs constructed on the river by what is now Duke Energy. There are 11 such large impoundments on the Catawba, which extends from Lake James in North Carolina to over 400 km downriver to Lake Wateree in South Carolina (Table 1). The Yadkin/Pee Dee River also has several hydroelectric dams that were built in the early twentieth century (Table 1). A series of four reservoirs were constructed beginning in 1917 by the Tallassee Power Company and Alcoa Corporation. For many years these were owned and operated by the Alcoa Corporation, but in 2017 they were acquired by Cube Hydro Carolinas, LLC (Cube Hydro 2021). Between the lowermost point of these reservoirs and the point where the Great Pee Dee River crosses into South Carolina there exists Lake Tillery and Blewett Falls Reservoir. Originally operated by Carolina Power and Light, these two projects are now owned and operated by Duke Energy. Table 1 shows water bodies in the study area where restrictive fish consumption advisories have been issued by the SCDHEC, the NCDHHS, and the GADNR because of PCBs, which were current at the time of this writing.

FISH COLLECTION AND PROCESSING

Glover et al. (2010) and the SCDHEC (2001) described methods utilized in the collection and processing of fish tissue in South Carolina. This involves the collection of fish using standard electroshocking techniques at public water bodies in South Carolina (US EPA 2000a). Largemouth bass were targeted at all sites while certain other species were collected when present, particularly game fish that may be an important part of the local fishery. Specimens were placed on ice and returned to the laboratory where they were processed individually. Standard measures were recorded and a skin-on, scale-off fillet was utilized for most species. For catfish species, skin-off fillets were taken. Using standard US EPA (2000a) methods, fillets were homogenized with dry ice and delivered to the SCDHEC laboratory for analysis.

Fish samples from other agencies, including those from the US EPA, Georgia, and North Carolina, utilized similar collection techniques. More details on specific collection and processing methods can be found in NCDENR (2013) for the state of North Carolina, in Georgia Department of

Table 1. Fish Consumption Advisories for PCBs in Select Basins in South Carolina

Basin	State	Reservoir	Year Built	Fish Consumption Advisory—PCBs*
Catawba	NC	Lake James	1923	No
Catawba	NC	Rhodhiss Res.	1925	No
Catawba	NC	Hickory Res.	1927	No
Catawba	NC	Lookout Shoals Res.	1915	No
Catawba	NC	Lake Norman	1963	STP, HYS (NCDHHS)
Catawba	NC	Mountain Island Res.	1924	BLC, CHC (NCDHHS)
Catawba	NC/SC	Lake Wylie	1904	LMB (NCDHHS); LMB, CHC, BKS (SCDHEC)
Catawba	SC	Fishing Creek Res.	1916	LMB, BLC, CHC, WHB, BKS (SCDHEC)
Catawba	SC	Great Falls Res.	1907	No
Catawba	SC	Cedar Creek Res.	1909	LMB, BLC, CHC, WHB, BKS (SCDHEC)
Catawba	SC	Lake Wateree	1920	LMB, BLC, CHC, WHB, STB, BKS (SCDHEC)
Pee Dee	NC	High Rock Res.	1927	Catfish all species (NCDHHS)
Pee Dee	NC	Tuckertown Res.	1962	No
Pee Dee	NC	Badin Lake	1917	LMB, Catfish all species (NCDHHS)
Pee Dee	NC	Falls Res.	1919	Catfish all species (NCDHHS)
Pee Dee	NC	Lake Tillery	1928	Catfish all species (NCDHHS)
Pee Dee	NC	Blewett Falls Res.	1912	No
Savannah	SC/GA	Lake Hartwell	1959	All Species (SCDHEC); LMB, CHC, STP, HYS (GADNR)
Savannah	SC	Langley Pond	1870	All Species (SCDHEC)

Note. LMB = largemouth bass, HYS = hybrid striped bass, BLC = blue catfish, CHC = channel catfish, BKS = black crappie, WHB = white catfish, STP = striped bass. *No Advisory generally because of lack of data rather than absence of tissue PCBs.

Natural Resources (1992), in US EPA (2000b) for the USEPA National Lakes Study, and in US EPA (2016) for the US EPA Catawba Indian Nation special study.

FISH TISSUE DATASETS

The SCDHEC data served as the primary data source for the state and included fish tissue fillets collected beginning in 1991. Watersheds shared by the two Carolinas include Santee (which contains the Catawba/Wateree), Yadkin/Pee Dee, Little Pee Dee/Lumber, and Waccamaw. For the portions of these watersheds outside of South Carolina, data were provided by the North Carolina Department of Environmental Quality (NCDEQ) in partnership with the North Carolina Department of Health and Human Services (NCDHH). The states of Georgia and South Carolina share the Savannah River Basin, and data collected by the Georgia Department of Natural Resources (GADNR) were also used in this study. A national study on fish tissue contamination was conducted by the US EPA (2009a) in the late 1990s and early 2000s. Of the 500 lakes sampled across the United States, 10 were contained within or flowed into South Carolina. In support of the Catawba Indian Nation, fish tissue PCB data were also obtained by the US EPA (2016) from the Catawba River. Data for largemouth bass fillets from Coughlan (1995) for the Duke Power reservoirs were not used in our analysis because there were high detection limits for Aroclors in that study (0.28 ppm–0.35 ppm). The combined datasets represent 2,075 individual or composite samples analyzed for PCBS and 4,232 fish specimens. There were 44 different species collected and processed from 303 different sampling locations from 1991 to 2016. The dominant species was largemouth bass, with 46% of all samples represented by this species. Channel catfish were the second most dominant species, representing 10.7% of the samples. This large volume of data, over space and time, allowed for a comprehensive assessment of PCBs in fish from the waters of South Carolina and provided insight into possible sources.

For analysis and reporting and display, we used total PCBs reported in parts per million (ppm) wet weight. We report wet weights because most of the data used for this study were only available in this format due to its use for determining fish consumption. To compute total PCB levels for each fish or fish tissue composite sample, individual congeners or Aroclors were summed for each sample. For reporting and display we used the mean of total PCBs for samples from a specific sampling point. EPA methods 8082 and 1656 were utilized for Aroclor analysis of fish tissues, while EPA method 1668 was used for congener-specific analysis of fish tissues. For additional information on standard operating procedures for laboratory tissue analysis, see SCDHEC (2012) for South Carolina, NCDENR (2013) and Mort (2017) for North Carolina, GADNR (1992) for Georgia, US EPA (2016) for Catawba Indian Nation Special Investigation, and US EPA (2000c) for the US EPA national study on lake fish tissue.

To compare the many samples for which Aroclor-only data were available (mostly in SC) with those sites with congener-only data (in NC), PCB congeners were summed and grouped into their homolog class and then converted to a percentage for each homolog for a given sample. The Agency for Toxic Substances and Disease Registry (ATSDR 2000) listed the approximate composition of each Aroclor as expressed in homolog percentages. For example, Aroclor 1260 was shown to be dominated by hexachlorobiphenyls, while pentachlorobiphenyls are the dominant component of Aroclor 1254. We used the percent homologs for each sampling site and a given species of fish to approximate the most probable Aroclor composition, which allowed sites at some North Carolina reservoirs to be compared to those in South Carolina. We did this for the congener data from the EPA national fish tissue study (US EPA 2009a), although both Aroclor and congener data were analyzed for all fish in this project. Having both Aroclors and congeners for the same sample allowed for additional insight into the extrapolation of Aroclors from congener data.

SPATIAL ANALYSIS

Spatial analysis and display were performed with Esri ArcView 10 (2020). Important data layers included the National Hydrography Dataset, the National Landcover Dataset, and in-house layers created by the SCDHEC, such as fish tissue monitoring locations. For many of the figures presented here, sample sites are represented as points on maps, and colors represent levels of PCBs that correspond to the thresholds that trigger a consumption advisory by the SCDHEC (2021). These categories are "no restrictions" of consumption (<0.05 ppm, Green), "eat no more than one meal per week" (0.05 ppm to 0.19 ppm, Yellow), "eat no more than one meal per month" (0.2 ppm to 0.99 ppm, Orange), and "do not eat any" (≥ 1.0 ppm, Red). Because consumption advisories are given for water bodies and not points, these figures are not intended to represent an advisory but rather to represent spatial trends. Further, the states of Georgia and North Carolina have slightly different means and methods for issuing fish consumption advisories than South Carolina, making this scale not as relevant for these states. However, the first trigger for issuing restrictive consumption advisory in all three states is fish tissue PCB levels of 0.05 ppm or above. Fish consumption advisories for South Carolina, North Carolina, and Georgia may be found at SCDHEC (2021), NCDHHS (2021), and GADNR (2021), respectively.

Largemouth bass fillets were well represented in the dataset across space and time and served as a good surrogate for spatial trends. Channel catfish and blue catfish fillets were also used to evaluate trends, especially where largemouth bass were absent. To provide a more robust spatial coverage and increase sample size, these two catfish species were combined to examine trends. We could not confidently assess temporal trends because of insufficient data over time at specific sample locations.

STATISTICAL ANALYSIS

Testing for differences of PCBs for point data was not possible due to small sample sizes at many locations. To increase sample size, multiple sample sites at individual bodies of water were combined. Tests for differences in PCB levels in largemouth bass in the Catawba River Watershed were conducted for 7 waterways (Mountain Island Reservoir, Lake Wylie, Catawba River, Fishing Creek Reservoir, Cedar Creek Reservoir, Lake Wateree, and Wateree River). Lake Norman was not included in this analysis due to small sample size (n = 2). For catfish, 8 groups were evaluated, which included the above water bodies and Lake Norman. For the Great Pee Dee Watershed, 5 groups were evaluated for both largemouth bass and catfish (High Rock Reservoir, Badin Lake, Falls Reservoir, Lake Tillery, and the Great Pee Dee River in South Carolina). To test for assumptions of normality, Q-Q plots were constructed and Shapiro-Wilk Tests for normality were conducted. For the Catawba River Watershed, both for largemouth bass and catfish, the data were not normally distributed. The nonparametric Kruskal-Wallis Test was thus used, and the post hoc Dunn's test was used to determine which groups differed. For both largemouth bass and catfish data in the Great Pee Dee Watershed, data were found to be normally distributed, and ANOVA tests were used to test for differences in groups. For ANOVA, the post hoc Tukey-Kramer test was used to compare groups. The accepted level of significance for all tests was $p < 0.05$. The data analysis for this paper was generated using the Real Statistics Resource Pack software (Release 7.6) (Zaiontz 2020).

RESULTS

Figure 1 shows the spatial distribution of total PCBs in largemouth bass fillets from waters of South Carolina to include those shared with Georgia and North Carolina. Mean total PCB values for largemouth bass fillets ranged from below detection (< 0.05 ppm) to 6.4 ppm at a site on Lake Hartwell in South Carolina. Of the 171 SCDHEC sampling locations with largemouth bass fillet data, 15% had mean total PCB levels greater than or equal to 0.05 ppm. Of the 145 points in Figure 1 shown in green (mean total PCBs <0.05), 94% had no detectable PCBs found in any of the fish samples analyzed for Aroclors. The maximum value

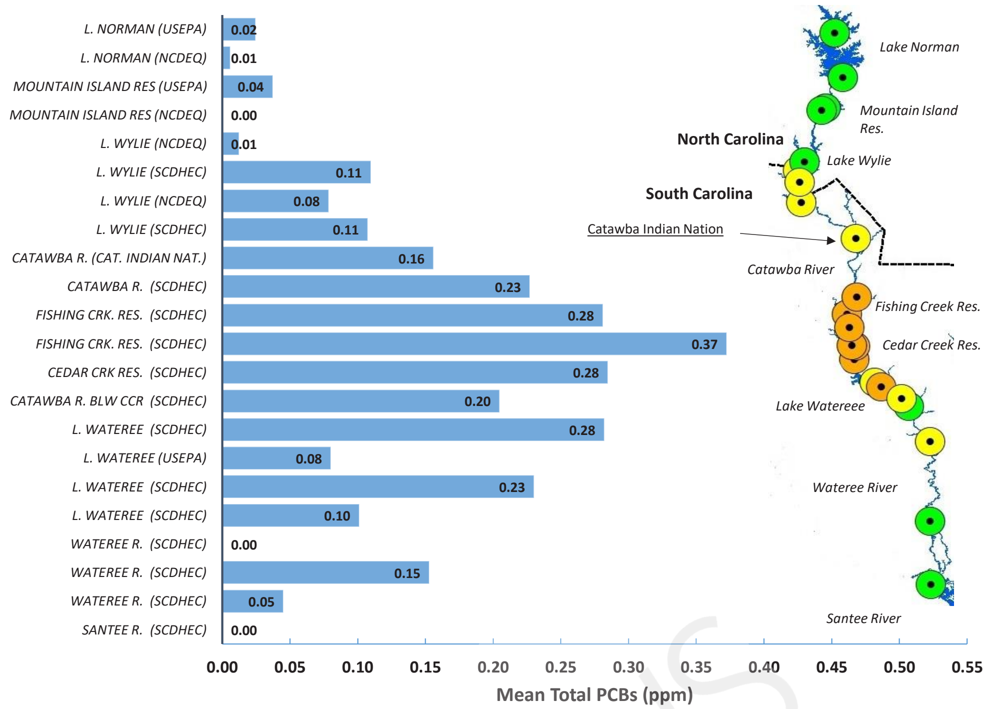

Figure 2. Mean total PCBs (ppm wet weight) in largemouth bass fillets from the Catawba River Basin of the Carolinas (means labeled on each bar). Mean total PCBs for green points <0.05 ppm, yellow points 0.05–0.19 ppm, and orange points 0.20–0.99 ppm.

for a largemouth bass fillet sample was 19.7 ppm, a site on Lake Hartwell in South Carolina, and the maximum mean total PCBs for largemouth bass fillets (n=49 samples) was 6.4 ppm, from a different sampling site on this same reservoir. PCBs in fish tissue at levels that have triggered consumption advisories have been found in four areas: Lake Hartwell (SC/GA), Langley Pond (SC), the Catawba/Wateree River Basin (SC/NC), and the Yadkin/Pee Dee River Basin (SC/NC) (see Table 1 and Figure 1). Shown also as yellow points in Figure 1 are three additional locations where mean PCB levels exceeded 0.05 ppm for largemouth bass fillets, which included Sesquicentennial State Park Lake, a small reservoir in the center of the state, the Saluda River in the western part of South Carolina, and a site toward the coast on the Santee River directly below the dam on Lake Marion. However, these three sites had small sample sizes of individual fish (5, 3, and 2 respectively), and their means were elevated because of a single specimen at each location. Additional data are needed to determine any real trends, and at this time consumption advisories for PCBs have not been issued for these water bodies (SCDHEC 2021).

LAKE HARTWELL AND LANGLEY POND

The highest levels of fish tissue PCBs occurred in Lake Hartwell and Langley Pond, with readings well above 1 ppm mean total PCBs in the fillets of many fish species (Figure 1). Both locations have a long history and record of PCB contamination, and remediation efforts have occurred at both (US EPA 2009b; CH2M Hill Engineers 2016). The source of PCBs in Lake Hartwell was a chemical plant on Twelve Mile Creek, which drains into the lake. Langley Pond was constructed in 1870 as an impoundment of Horse Creek. Large numbers of textile mills occurred in this watershed, releasing industrial waste into the creek for decades, which settled into this downstream impoundment. A complete evaluation of these two sites is beyond the scope of this paper, but further information can be found in other studies (Bruner and Hill 1977; Aldridge 1978; Marcus 1987; Gaymon 1992b; Darr 1986; US EPA 2009b; CH2M Hill Engineers 2016).

CATAWBA/WATEREE WATERSHED

The pattern of PCBs in fish tissue in the Catawba/Wateree Basin is illustrated in Figures 1 through 4. PCBs were present in largemouth bass fillets in Lake Norman and Mountain Island Reservoir, North Carolina, but at relatively low levels (mean total <0.05 ppm). Higher levels (mean total >0.05 ppm) were found in fish from Lake Wylie at the North Carolina-South Carolina border. In South Carolina values were still higher in the Catawba River near the Catawba Indian Nation, which is below the confluence of Sugar Creek. The headwaters of Sugar Creek are in North Carolina, and much of the city of Charlotte occupies this watershed. Levels consistently exceeded 0.2 ppm at Fishing Creek Reservoir and remained elevated in Cedar Creek Reservoir just downriver. The highest mean PCB levels in largemouth bass fillets for the basin were 0.37 ppm at a site on Fishing Creek Reservoir (n=22 samples) (Figure 2). While PCB values were obtained by several agencies utilizing different laboratories and methods (Aroclors vs. Congeners), the patterns were remarkably consistent for total PCB tissue levels throughout the watershed.

The Kruskal-Wallis test indicated that some waterways in the Catawba Basin were significantly different for mean PCB concentrations in both largemouth bass and catfish fillets (H=42.7, $p < 0.001$) and catfish (H=30, $p < 0.001$) (Figures 3 and 4). Figure 3 shows the group differences for largemouth bass, with Cedar Creek Reservoir and Fishing Creek Reservoir having the highest levels of PCBs and Lake Norman, Mountain Island Reservoir, and Lake Wylie having the lowest. The post hoc Dunn's test showed that mean PCB values in largemouth bass were not significantly different in Like Wylie and Mountain Island Reservoir, though these two reservoirs were, in general, significantly different ($p < 0.05$) from the others. An exception was that mean PCB levels in largemouth bass in Lake Wylie and the Wateree River were not significantly different (z=0.87, p=0.38). Figure 4 shows the results for PCBs in catfish from the Catawba River Basin. The patterns were similar between catfish and largemouth bass. The highest levels of PCBs were in fish from waterways beginning at the Catawba River in South Carolina, while they were lower in the 3 upriver reservoirs (Lake Norman, Mountain Island Reservoir, and Lake Wylie). While the patterns were similar for catfish and largemouth bass, results of the Dunn's post hoc test indicated that PCBs in catfish in the Catawba River, Fishing Creek Reservoir, Cedar Creek Reservoir, Lake Wateree, and the Wateree River were not significantly different from each other (Figure 4).

Though evaluations and reporting were conducted only on largemouth bass and catfish, consumption advisories have been issued for several species in these water bodies by the SCDHEC and the NCDHHS (Table 1).

Aroclor 1260 was the dominate PCB mixture in fish from the Catawba/Wateree Basin in both North Carolina and South Carolina (Figure 5). This included samples analyzed by the SCDHEC, the EPA's national fish tissue study (US EPA 2009a), the EPA Catawba Indian Nation special study (US EPA 2016), and the Aroclor samples processed by the NCDEQ. The homolog profile computed from the EPA national fish tissue study (US EPA 2009a) for Lake Norman, Mountain Island Reservoir, and Lake Wateree shows a composition that suggests Aroclor 1260,and strongly resembles that which was given by ATSDR (2000) (Figure 5). The Aroclor analysis for these same three sites and specimens, both for the fillets and whole fish (n=6 samples), showed 100% Aroclor 1260. This homolog profile suggesting Aroclor 1260 was similar to the profile seen in most fish species in the Yadkin River Reservoirs in North Carolina (Mort 2017) but different from the Aroclor analysis processed from the Great Pee Dee River in South Carolina, Lake Hartwell in South Carolina, and Langley Pond in South Carolina, all of which were reported as predominantly Aroclor 1254 (Figure 5).

YADKIN/PEE DEE WATERSHED

Figures 6 through 8 show the pattern of PCB contamination in largemouth bass and catfish fillets from the Yadkin/Pee Dee River Basin in North Carolina and South Carolina. PCBs are relatively low (<0.05 ppm) in fish from a series of reservoirs on the Yadkin River and the Great Pee Dee River in North Carolina near the state line. In the Great Pee Dee River in South Carolina the levels are substantially higher, with mean PCB levels in largemouth bass fillets at 0.26 ppm at station PD-012. There was a decreasing trend from this site moving downriver. Levels were above 0.05 ppm at PD-337 (n=5 samples), which was 74 km downriver of PD-012, but were low or below detection limits beginning at PD-622 (n=5 samples), which is 98 km downriver of PD-012. The ANOVA analysis indicated group differences in PCBs of largemouth bass in the Great Pee Dee River Basin (F=4.7, $p<0.01$). In general, this was driven by the high levels of PCBs in the Great Pee Dee River in South Carolina. The Tukey-Kramer Test indicated that reservoirs in North Carolina were not statistically different from each other ($p>0.05$), with the exception of Falls Reservoir, where significantly different levels were reported from the Great Pee Dee River in South Carolina.

Patterns were similar for catfish fillets as for largemouth bass in the Great Pee Dee Basin (Figure 8). However, PCBs for catfish were sufficiently elevated in some of the Yadkin Reservoirs in North Carolina to the point where the NCDHHS (2021) issued restrictive consumption advice for catfish. Group differences were significant (ANOVA F=6.3, $p<0.001$) and appeared to be driven by the high values in the Great Pee Dee River in South Carolina (Figure 8). However, means were not significantly different for Badin Lake and the Great Pee Dee River in South Carolina (Tukey-Kramer q=2.7, p=0.33).

Most PCBs from fish collected from stations on the upper Pee Dee River in South Carolina were reported as Aroclor 1254 (92%) (Figure 5). This was comparable to Langley Pond (97%). PCBs from fish in Lake Hartwell were also reported as mostly Aroclor 1254 (67%), but 29% were also reported as Aroclor 1260. This contrasts with the Catawba River Basin, where 95% of Aroclors were reported as 1260. However, the homolog profile for sites on the Yadkin Reservoirs in North Carolina was like those on the Catawba and suggests Aroclor 1260 as the dominant mixture in fish tissue from most locations. Mort (2017) reported similar results and provided a comprehensive evaluation of the homolog and congener patterns in the tissue of fish from these Yadkin reservoirs.

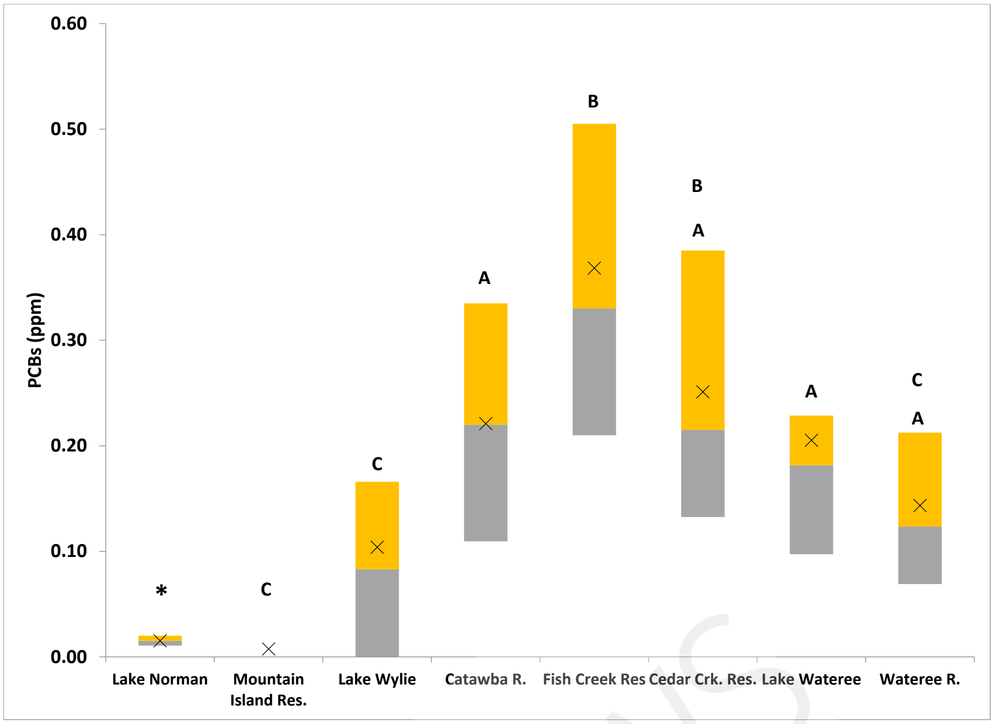

Figure 3. Box plot of PCBs in largemouth bass from water bodies in the Catawba River Watershed showing quartile 1 (bottom of grey columns), median (top of grey columns, bottom of yellow columns), quartile 3 (top of yellow columns), and mean (X). Group means were significantly different (Kruskal-Wallis Test H=42.7, $p < 0.001$). Groups with different letters were significantly different from each other ($p < 0.05$), while groups with the same letter were not significantly different (Dunn's post hoc test). *Not included in model due to low sample size.

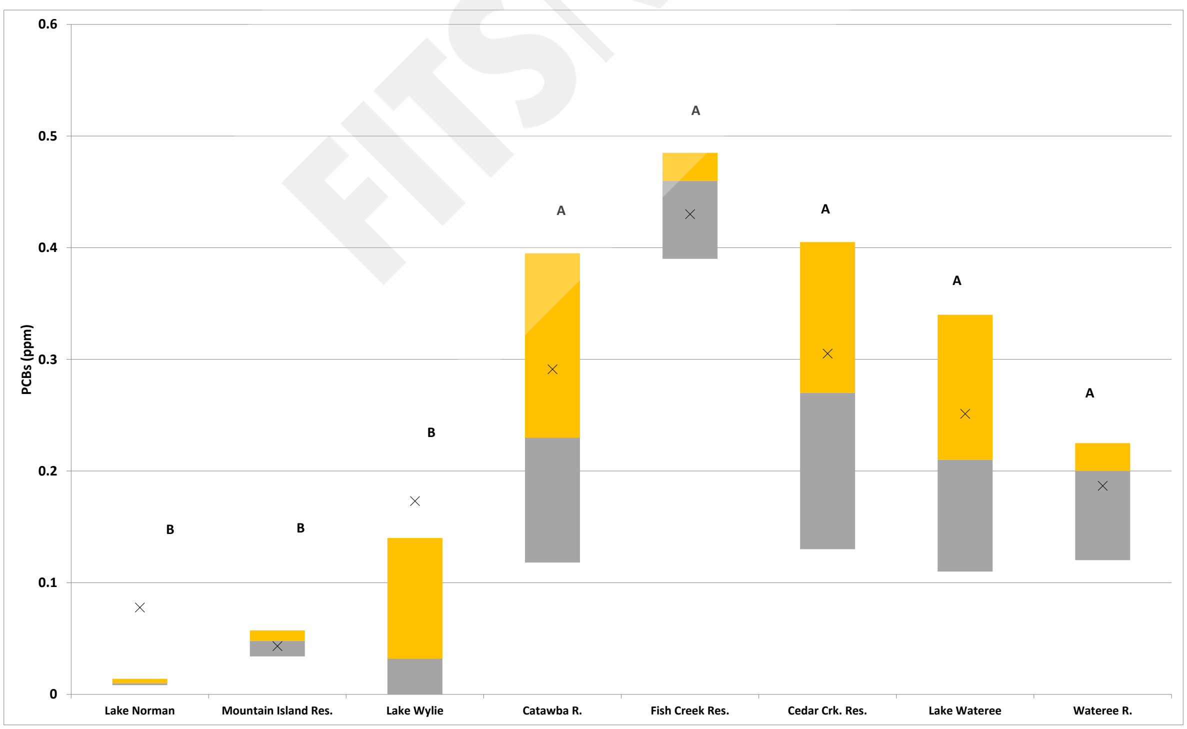

Figure 4. Box plot of PCBs in catfish (blue catfish and channel catfish combined) from water bodies in the Catawba River Watershed showing quartile 1 (bottom of grey columns), median (top of grey columns, bottom of yellow columns), quartile 3 (top of yellow columns), and mean (X). Group means were significantly different (Kruskal-Wallis Test H=30, $p < 0.001$). Groups with different letters were significantly different from each other ($p < 0.05$), while groups with the same letter were not significantly different (Dunn's post hoc test).

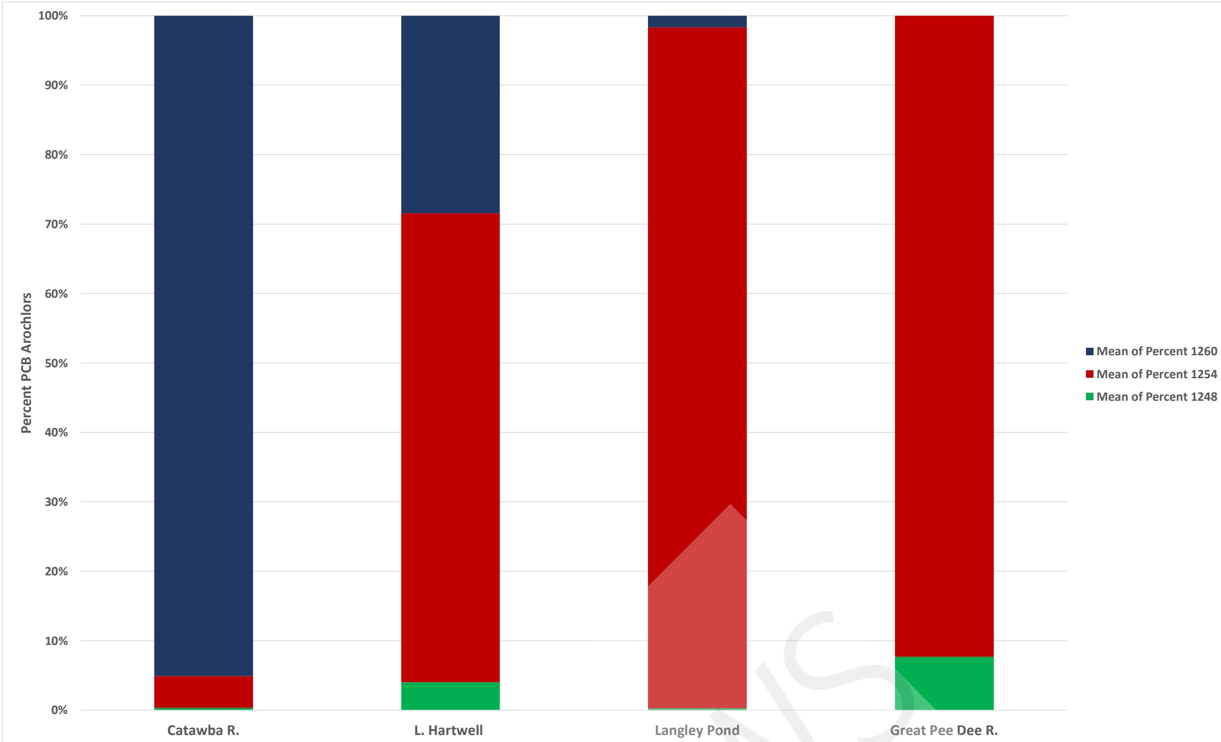

Figure 5. Percent Aroclors in largemouth bass from the Catawba River Basin, the Great Pee Dee River, Lake Hartwell, and Langley Pond in South Carolina.

DISCUSSION

THE USE OF OIL FOR MOSQUITO CONTROL—A BRIEF HISTORY

In the late nineteenth century, a campaign was initiated by the US military and public health officials to stop the spread of malaria, which included an aggressive effort to reduce the population of Anopheles mosquitoes (Ross 1900). In the first half of the twentieth century, tremendous volumes of waste oils were applied directly to US waterways, particularly on large reservoirs in the southeast, in an attempt to kill the larvae of the Anopheles mosquito (Carter 1913). While Dichlorodiphenyltrichloroethane (DDT), Paris Green, and other pesticides were applied to row crops in and around homes, great quantities were also applied directly to the water's surface, usually after being mixed with oils of various grades (Johnson 1922).

While malaria in the early part of the twentieth century had been on the decline in the US, there was great concern in the southeast about the many large reservoirs that were being built (Le Prince 1927). The still waters along the shores and in coves provided an ideal habitat for *Anopheles* mosquito larvae. Henry Carter, Senior Surgeon of the US Public Health Service, recognized early in the twentieth century the potential impact on public health of the many hydropower plants that were being planned and provided strong words of warning (Carter 1914). In addition to the concerns of public health officials, there were fiscal considerations, as lawsuits were common during this era (Kay 1915; Clark 1931). Williams (1958) reported that in the early twentieth century, "power companies were multiplying impoundments, malaria became epidemic around the new ponds, and resulting lawsuits threatened some companies with bankruptcy." Malaria was thus a major consideration in planning the construction of hydroelectric plants through the first half of the twentieth century and appears to have consumed much consideration and expense pre- and post-construction (Williams 1958). Duke Energy (2013) reported that their mosquito control program began in 1923 and was the "oldest continuous environmental program of any utility in the US, and one of the first in North America." Their program continued for 93 years before being terminated in 2016.

EVIDENCE OF TRANSFORMER OIL USED FOR MOSQUITO CONTROL IN THE CAROLINAS

It is not surprising that in May 1970 the participants at a workshop on mosquito control in North Carolina were well represented not only by public health officials and academia but also by utilities, the Tennessee Valley Authority (TVA), and the US Army Corps of Engineers. The proceedings were published in August 1970, which included the questions and answers for the speakers (Howells 1970). To the question of what chemicals hydroelectric plants use for mosquito

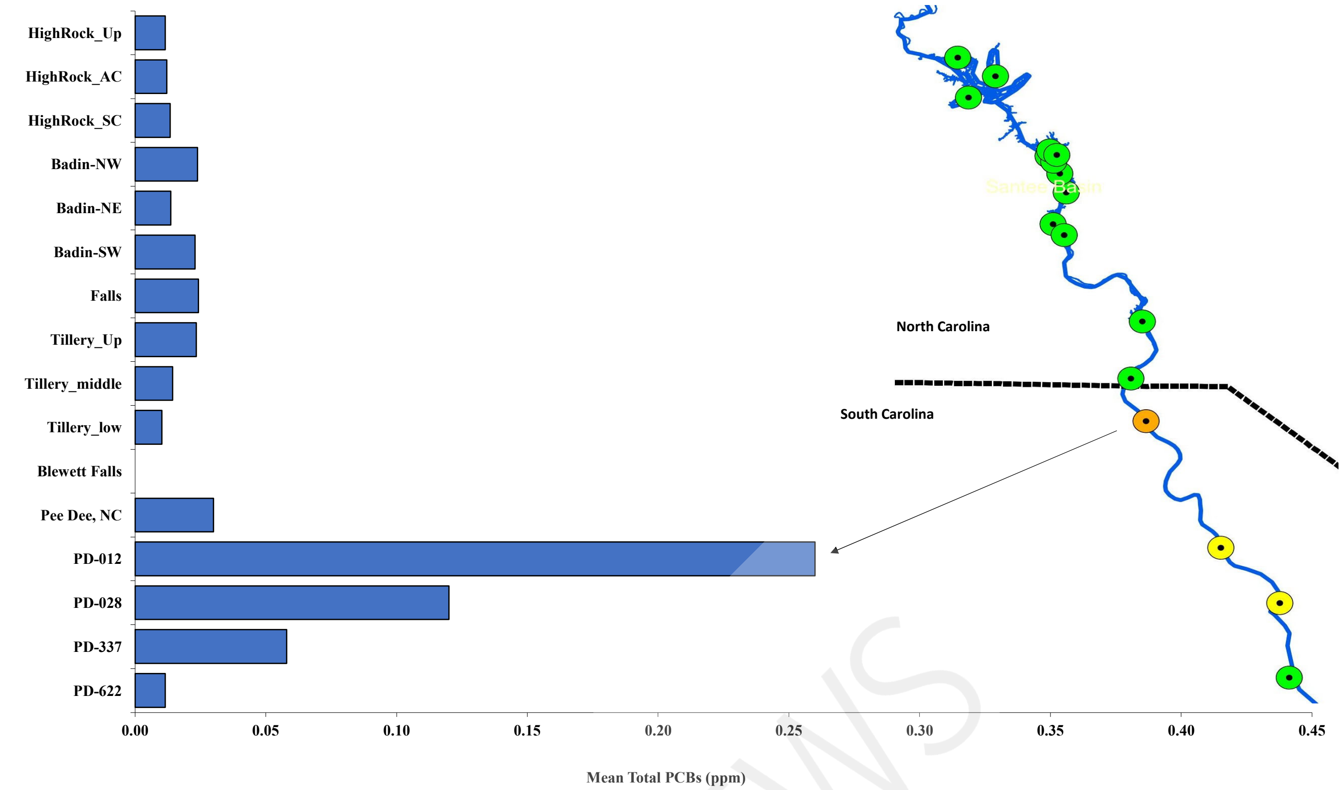

Figure 6. Mean total PCBs (ppm wet weight) in largemouth bass fillets from the Great Pee Dee River Basin. Mean total PCBs for green points <0.05 ppm, yellow points 0.05–0.19 ppm, and orange points 0.20–0.99 ppm.

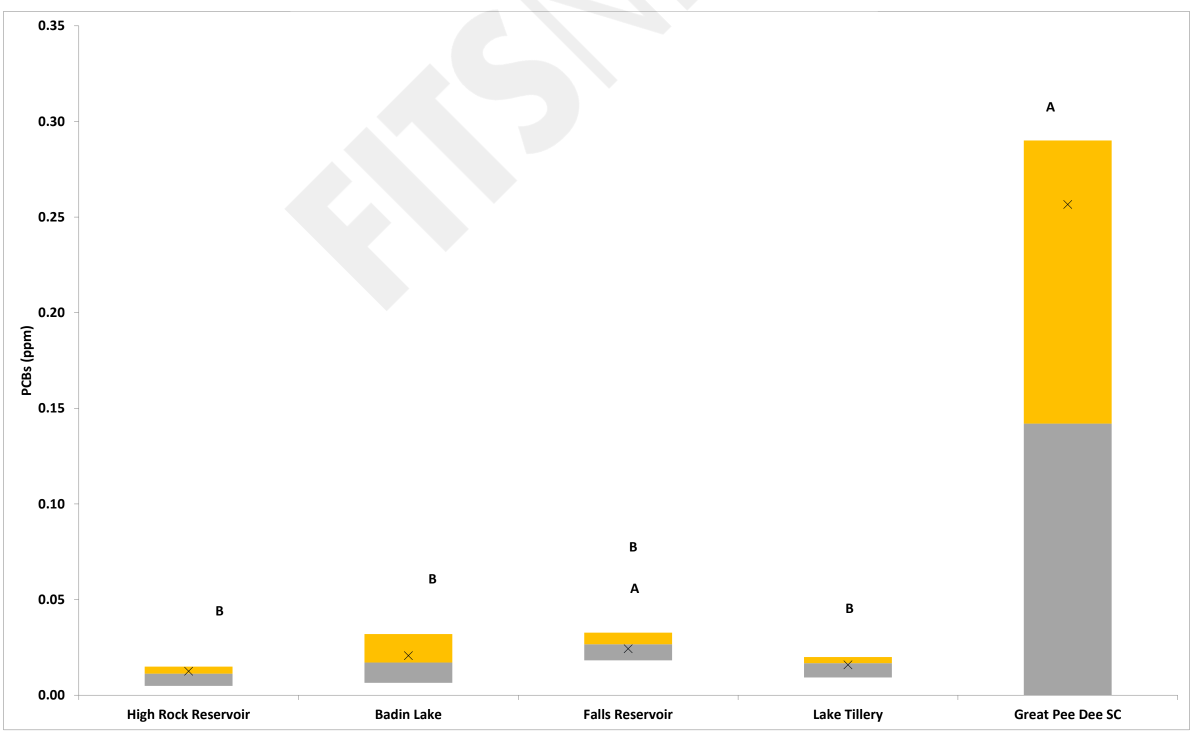

Figure 7. Box plot of PCBs in largemouth bass from waterbodies in the Great Pee Dee River Watershed showing quartile 1 (bottom of grey columns), median (top of grey columns, bottom of yellow columns), quartile 3 (top of yellow columns), and mean (X). Group means were significantly different (ANOVA $F=4.7$, $p < 0.01$). Groups with different letters were significantly different from each other ($p < 0.05$), while groups with the same letter were not significantly different (Tukey-Kramer post hoc test).

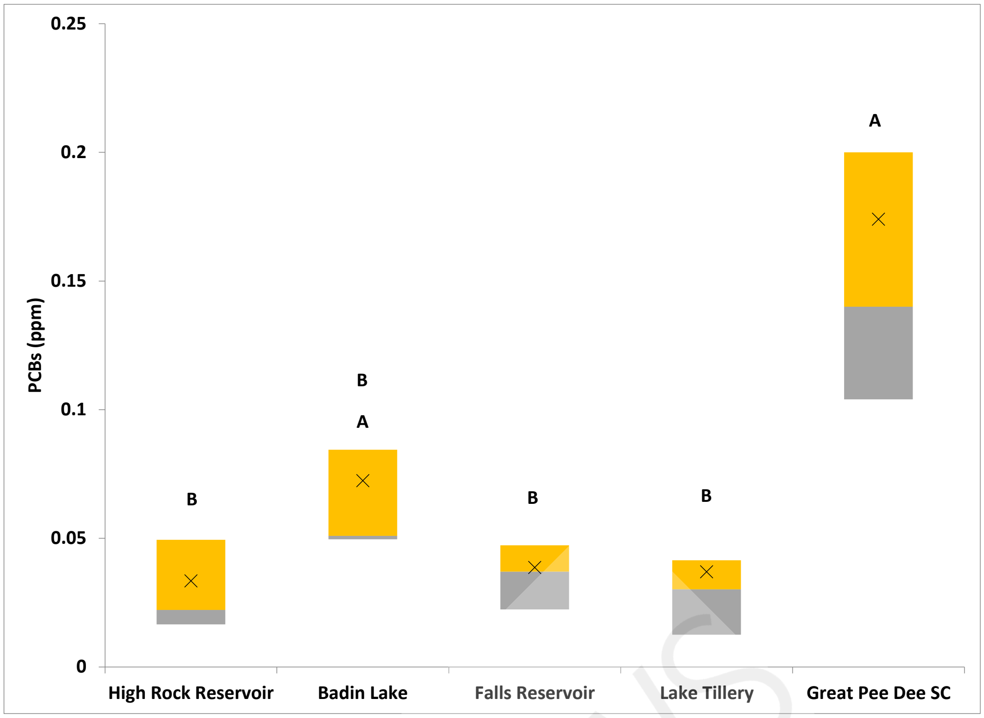

Figure 8. Box plot of PCBs in catfish (blue catfish and channel catfish combined) from water bodies in the Great Pee Dee River Watershed showing quartile 1 (bottom of grey columns), median (top of grey columns, bottom of yellow columns), quartile 3 (top of yellow columns), and mean (X). Group means were significantly different (ANOVA F=6.3, $p < 0.001$). Groups with different letters were significantly different from each other ($p < 0.05$), while groups with the same letter were not significantly different (Tukey-Kramer post hoc test).

control, Ashton (1970), with the North Carolina State Board of Health, indicated that a mixture of No. 2 fuel oil and used transformer oil was used by two electric companies. The oil was applied at the rate of 10 to 15 gallons per acre. Swearingen (1970), of Duke Power, reported that they used transformer oil and No. 2 diesel fuel in their mosquito control program. It was estimated that 2,500 miles of shoreline were being treated in North Carolina and South Carolina beginning in April and ending in October, with treatments occurring every 8 days. These are the reservoirs in the Catawba Basin shown in Figures 1 through 4. Harris (1970), with Carolina Power and Light (now Duke Energy) and superintendent of the Blewett Tillery Hydroelectric Project (see Figures 6 through 8) on the Pee Dee/Yadkin River, stated that when transformer oil was available it was used instead of motor oil because it was free. It was suggested that transformer oil had been used for many years, and it was preferred over motor oil because it left a better sheen on the water's surface, which helped the boat operators see where they had sprayed. Motor oil was reported at 20% by volume with the application rate of 150 gallons per day.

The spatial patterns of PCBs in fish shown in Figures 1 through 8 suggest that the past use of transformer oil on these reservoirs contributed, in part, to contemporary contamination. In general, PCBs are absent throughout most of the study area, including large reservoirs operated by other utilities in South Carolina. Where PCBs are present (Lake Hartwell, Langely Pond, and Great Pee Dee River in SC), there is a known point source. The fact that the PCBs found in fish from the Catawba River and Yadkin River reservoirs are Aroclor 1260 (Figure 5; Mort 2017) while those from other sites are mostly Aroclor 1254 further points to these compounds coming from a similar industrial source.

Taken together, these findings strongly suggest that the widespread contamination of PCBs within the Catawba River reservoirs (Figures 2 through 4) and the reservoirs on the Yadkin/Pee Dee (Figures 6 through 8) was caused, at least in part, by the direct application of used transformer oils as part of mosquito control efforts by the reservoir operators. We suggest that the culture of mosquito control and malarial eradication that coevolved with the creation of hydroelectric plants in the early part of the twentieth century likely made the use of waste oil from transformers likely, if not inevitable, at some locations in the Carolinas.

OTHER POTENTIAL SOURCES

Burlington Fibers Plant on Great Pee Dee River, South Carolina

In the fall of 2015, the SCDHEC discovered large quantities of PCBs in soil and sediment around a former Burlington Industrial Fibers plant in Cheraw, South Carolina (SCDHEC 2016). Further investigations found PCBs in the drainage that empties into the Great Pee Dee River near Cheraw, South Carolina (Figure 6). The US EPA, in cooperation with the SCDHEC, soon initiated an intensive investigation of the extent of the contamination. Remediation has occurred and is ongoing, with the site being listed on the National Priorities List (US EPA Superfund) in 2018 (US EPA 2021). The levels and extent of the PCBs in fish tissue of the Great Pee Dee River strongly points to this site as the source of contamination. The predominant Aroclor in the tissue of all fish species in the Great Pee Dee River in South Carolina was Aroclor 1254, but Aroclor 1248 was also present (Figure 5). This pattern was similar to Langley Pond, where the textile industry was also the suspected source. Aroclor 1248 and 1254 were also the primary mixture found in soil samples throughout the area of the former Burlington Fibers plant (SCDHEC 2016). The highest levels of PCBs in fish from the Great Pee Dee River were found at station PD-012, which is immediately downstream from the drainage of the former plant (Figure 6). The decreasing pattern of fish PCBs seen in the Great Pee Dee River moving downriver strongly points to this plant being the source of contamination. This is similar to patterns of PCB in fish from other waterways, with the highest concentrations being near the origin of contamination (e.g., Lake Hartwell in South Carolina; see Gaymon 1992a). The levels of PCBs were much lower in fishes upriver in the Great Pee Dee River in North Carolina, the Yadkin River, and the constructed reservoirs in North Carolina. The homolog pattern from largemouth bass from this basin in North Carolina resembled Aroclor 1260, which was similar to that of the Catawba River fishes (Figure 5). Similar findings were provided in the comprehensive evaluation of fish from the Yadkin Reservoirs conducted by Mort (2017), who showed that the patterns of individual congeners and homologs suggested Aroclor 1260 for most species of fish. It is unlikely that the oiling conducted by the utilities' mosquito control program in the Yadkin reservoirs or the contamination of Badin Lake sediments by the Alcoa plant (see below) contributed to the PCBs present in fish tissue of the Great Pee Dee River in South Carolina.

Alcoa Plant on Badin Lake, North Carolina

The Alcoa plant on Badin Lake also has been shown to have released PCBs into this reservoir and was implicated in the contamination of the fish there (NCDHHS 2012). Remediation, to include capping of lake sediment near the Alcoa plant, was initiated in 2012 as authorized through the North Carolina's Inactive sites and Hazardous Substance Response Act (NCDEQ 2012). However, High Rock Reservoir, where PCBs in fish have also been found, is upriver from Badin Lake (Figure 1 and Figures 6 through 8), and many of the other Yadkin reservoirs, where PCBS have been found in fish, appeared in the early literature related to malaria eradication and have been managed for mosquitoes for decades (Carter 1915; Gage 1925; Clark 1931). Further, there is strong evidence, including that reported here for the Burlington site, that where point source contamination exists, fish tissue PCBs are highest near the source and progressively lower further from the point of highest contamination (Gaymon 1992a). However, in the three Badin Lake sampling sites, no obvious trends were seen in largemouth bass PCB levels (Figure 6), with the two distant sites having similar levels of PCBs in fish as the site close to the Alcoa plant. The NCDHHS concluded that it was not feasible to link PCBs in fish and people who ate fish from Badin Lake to the Alcoa plant, for reasons that were enumerated by NCDHHS (2012). We support this view and suggest that application of transformer oils for mosquito control may have contributed to the contamination seen in Badin Lake.

Past Industrial Pollution

The discovery of large quantities of PCBs at the former Burlington Fibers site on the Great Pee Dee River in 2015 serves as a reminder that past industrial releases may remain undiscovered. A large portion of the city of Charlotte is in the Catawba River drainage, with much of the urban land area draining into the Catawba River through Sugar Creek (Figure 1). Data from the 1970s (Aldridge 1978) to date shows that levels of PCBs in fish tissue are consistently higher in fish below the confluence of Sugar Creek and the Catawba River in South Carolina (Catawba River, Fishing Creek Reservoir, Cedar Creek Reservoir, Lake Wateree, and Wateree River) than above in North Carolina (Lake Wylie, Mountain Island Reservoir, and Lake Norman) (Figures 1 through 4). The detection of PCBs in fish tissue of the Catawba River proper near the Catawba Indian Nation (US EPA 2016), where mosquito oiling likely did not occur, further points to additional sources (Figure 1). However, most of these reservoirs on the Catawba River were completed before 1930 while Lake Norman is a relatively newer reservoir (formed in1963) (Table 1). There would thus be a shorter history of mosquito control on Lake Norman than on other Catawba Reservoirs, which could explain the relatively lower levels of PCBs in that reservoir. However, this does not explain the relatively lower levels in Mountain Island Reservoir (constructed in 1924), suggesting that other sources of PCBs may have contributed to the relatively higher levels of contamination in the downriver reservoirs of South Carolina.

Regardless of potential other sources, the widespread contamination in the reservoirs managed by Duke Power and Carolina Power and Light (both now Duke Energy), and Alcoa Corporation (now Cube Energy, LLC), along with the acknowledged use of transformer oil in the operations mosquito control programs, strongly suggests that some of the contamination is from the direct application for mosquito control.

CONCLUSION

The global ramifications of these findings are numerous. It is unknown whether PCB waste oils were used by other mosquito control programs. The only documentation we found was from the proceedings of the Mosquito Control Workshop in North Carolina (Howells 1970). However, even here this could have escaped scrutiny if the post-presentation questions and answers had not been captured in the proceedings. Outside of this workshop and the published proceedings, it is also unclear how widely the idea of using spent transformer oils for mosquito control was shared with other regional operators. While the Tennessee Valley Authority (TVA) used oiling on their many reservoirs, water elevation management became an increasingly more important part of its mosquito control program than oiling (Gartrell and Ludvick 1954; Breeland et al. 1961).

In reservoirs where PCBs have been detected, but an obvious source has not been identified, a reevaluation of data and literature would seem appropriate. Many of these hydroelectric projects occur in the southeastern US, but oiling has been a staple for mosquito control throughout the world, even in regions where malaria does not exist. Early research found that PCBs were toxic to the larvae of Anopheles mosquitos (Deonier et al. 1947), but it appears that the primary application was to extend the effective life of other pesticides such as Lindane (Duda 1957). A prerequisite to suppose an association of contemporary PCB contamination with legacy mosquito control would require the same components that were seen in our study, such as a mature and well-funded mosquito control programs, presence of PCBs in the fish of the treated reservoirs but absent in others, and a ready supply of PCB oils that were expendable, likely in combination with other waste oils.

We believe consideration should also be given to remote locations where PCBs have been discovered. There is a large body of literature on this topic, with much of it focused on atmospheric transport and deposition of PCBs traveling great distances from their original source (Bright et al. 1995). For example, some of these projects have occurred in the artic, with PCBs being discovered far from any known source and in remote areas in this region (Freidman and Selin 2016). Mosquito control in the artic and subartic, as elsewhere, is well documented in the historic literature, with the same means and methods employed for eradication (Twinn 1950). For example, using aircraft, 2,777 gallons of a diesel oil mixed with Dichlorodiphenyltrichloroethane DDT was used to cover 3,500 acres of test plots near Churchill, Manitoba, Canada, in 1947 (Goldsmith et al. 1949). In a thorough review on control of biting flies in Northern Canada, Twinn (1950) reported on experimental control measures being carried out at Fort St. John and Fort Nelson, British Columbia; Watson Lake and Whitehorse, Yukon Territory; Goose Bay, Labrador; Rockcliff, Ontario; and Churchill, Manitoba. As was common during this period, DDT mixed with oil sprayed from aircraft was the primary form of treatment in these experiments, with up to 43 square miles being treated in some studies. The mixtures of these treatments were well documented by Twinn (1950) and appeared to consist solely of fuel oil, kerosene, and methylated naphthalenes as emulsifiers for DDT. However, Nordin et al. (1993) suggested that some petroleum oils in the Yukon Territories could have been contaminated with PCBs and that these could have been used in the region's mosquito control programs. It is unclear if PCB oils, when available, were used intentionally in subartic mosquito control programs, but our findings suggest that this should be considered in future investigations.

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