

Distribution and sources of PCBs (Aroclor 1268) in the Sapelo Island National Estuarine Research Reserve

E. F. Wirth · P. L. Pennington · C. Cooksey ·
L. Schwacke · L. Balthis · J. Hyland · M. H. Fulton

Received: 27 January 2014 / Accepted: 1 September 2014
© Springer International Publishing Switzerland (outside the USA) 2014

Abstract Aroclor 1268 is a highly chlorinated PCB mixture that was released into the aquatic environment near Brunswick, GA (BR), as a result of decades of local industrial activity. This extensive contamination has led to US EPA Superfund designation in estuarine areas in and around Purvis Creek, GA. Roughly 50 km to the northeast is the Sapelo Island National Estuarine Research Reserve (SI) where previous studies have documented unexpectedly high Aroclor 1268-like PCB levels in blubber and plasma samples of resident bottlenose dolphins. This result led to a collaborative effort to assess the PCB patterns and concentrations in SI sediment and fish (as potential vectors for PCB transfer to SI resident dolphins). Thirty SI randomly assigned stations were sampled for sediment PCB levels. Additionally, fish were collected and analyzed from SI ($n=31$) and BR ($n=33$). Results were pooled with regional assessments of PCB concentrations from South Carolina and North Carolina in an effort to determine the association of Aroclor 1268 levels in SI samples. Results indicated that PCB levels in sediment and fish are much lower in the SI estuary compared to BR sediment and fish concentrations. However, PCB congener profiles for both sediments and fish were similar between the two locations and consistent with the Aroclor

1268 signature, indicating possible transport from the Brunswick area. A likely source of Aroclor 1268 in dolphins from SI is contaminated fish prey.

Keywords Aroclor 1268 · PCB · Regional assessment · Sapelo Island NERRs

Introduction

Numerous environmental studies have described the magnitude and distribution of PCBs attributed to Aroclor 1268 contamination in the Brunswick, GA, area attributed to the LCP Superfund site (Kannan et al. 1997, 1998; Maruya and Lee 1998). This site was listed on the US EPA National Priorities List (Superfund) in 1994 after nearly 75 years of industrial activities. Contaminants of concern at this site include several metals (Hg, Cr, and Pb) as well as PAHs and PCBs (Aroclor 1268). Aroclor 1268 is an uncommon mixture of PCB congeners dominated by octa- through deca-chlorinated homolog groups (>90 % of total PCB content) (Ishikawa et al. 2007; Kannan et al. 1997). The composition of Aroclor 1268 is dominated by a suite of PCB congeners (IUPAC nomenclature) including 180, 187, 194, 196, 199, 200, 201 202, 206, 207, 208, and 209. Environmental sampling in and around Brunswick, GA, has revealed high levels of Aroclor 1268 contamination in sediments, invertebrates (blue crab), commercially important fishes, turtles, birds, (Kannan et al. 1998) as well as bottlenose dolphins (Balmer et al. 2011; Kucklick et al. 2011, Pulster et al. 2009).

E. F. Wirth (✉) · L. Schwacke
NCCOS, HML, NOAA, National Ocean Service,
Charleston, SC, USA
e-mail: ed.wirth@noaa.gov

E. F. Wirth · P. L. Pennington · C. Cooksey · L. Balthis ·
J. Hyland · M. H. Fulton
NCCOS, CCEHBR, NOAA, National Ocean Service,
Charleston, SC, USA

Sapelo Island (SI) is a barrier island located roughly 50 km northeast of the LCP site. SI is mostly undeveloped and is home to NOAA's Sapelo Island National Estuarine Research Reserve (SI NERR). This reserve was established in 1976, but this designation follows decades of agricultural use beginning in the 1800s as well as conservation and land-management efforts led by the State of Georgia. Historically, SI has been lightly populated and used mainly for agriculture and coastal environmental/ecological research (SI NERR Management Plan 2008). In 2007 and 2008, blubber samples from bottlenose dolphins resident to the SI NERR were reported to have elevated levels of PCBs (Kucklick et al. 2011) and these elevated levels of PCBs were suggested to be associated with Aroclor 1268 contamination from the LCP site (Balmer et al. 2011). To date, the levels of PCBs reported in dolphins from the SI estuary are some of the highest values reported for coastal dolphin studies (Kucklick et al. 2011) and in contrast to the undeveloped nature of SI.

The present study was undertaken to characterize the distribution and congener composition of PCBs in sediments and fish in the SI NERR in order to investigate possible linkages between the previously documented PCB concentrations in dolphins and Aroclor 1268 contamination at the LCP Superfund site in Brunswick, GA. The study was a component of a larger study that examined the overall ecological condition of the SI NERR (Balthis et al. 2012). A reserve-wide assessment of ecological conditions, including concentration of chemical contaminants, was lacking prior to this study.

Methods

Station assignment, sample collection, and preservation

Sampling stations ($n=30$) were randomly assigned within the SI NERR boundary using a generalized stratified methodology detailed in Balthis et al. (2012). Sediment samples were collected during June 2009 from each station (Fig. 1a.) using a 0.04-m² Young grab sampler. The top 2–3 cm from multiple grabs at each

station was removed, composited for analysis, and frozen. While on station, researchers attempted to collect

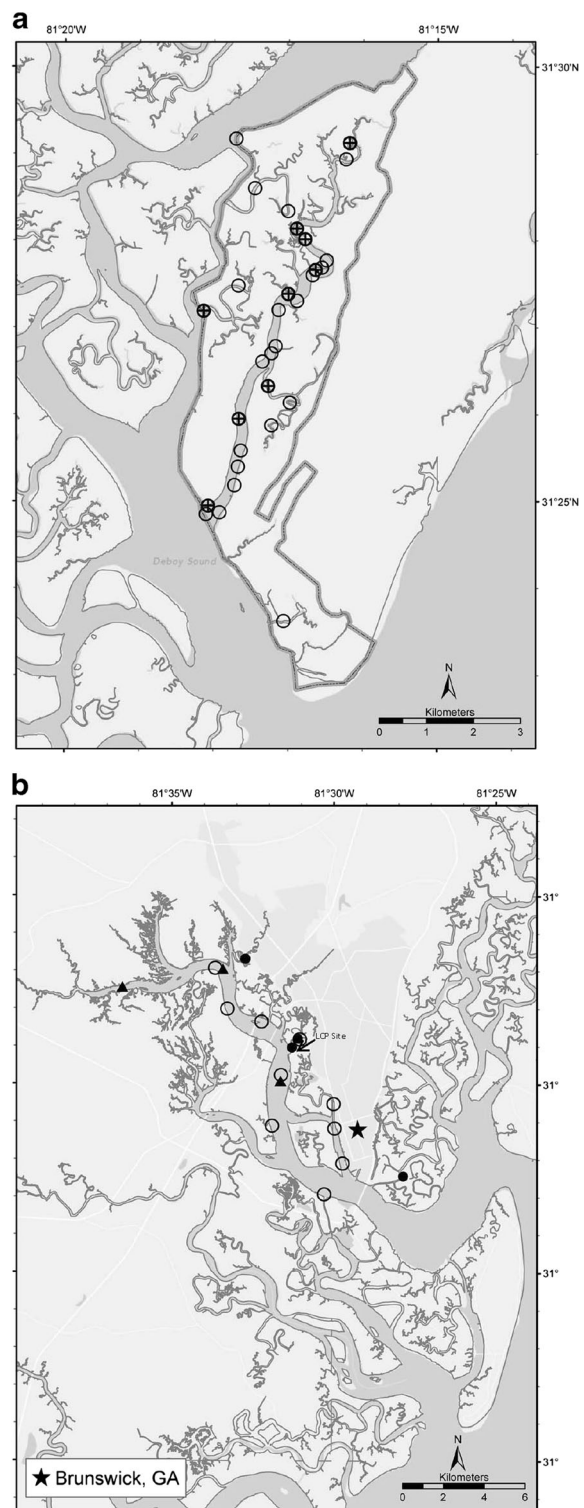


Fig. 1 **a** Sampling stations from within the Sapelo Island NERR; **▶** open circles are stations where sediment was collected; cross-hatched circles are stations where sediment and fish were collected. **b** Stations from the greater Brunswick, GA, estuarine system. Open circles are archival sediment samples, closed triangles are stations where fish were collected by GA DNR (7/2009), closed circles are stations where fish were collected by the authors for this study

specific species of fish by either hook and line or cast net. Targeted species included *Mugil cephalus* (striped mullet), *Micropogonias undulatus* (Atlantic croaker), *Cynoscion nebulosus* (spotted seatrout), *Paralichthys lethostigma* (southern flounder), *Sciaenops ocellatus* (red drum), *Menticirrhus sp.* (whiting), *Bairdiella chrysoura* (silver perch), and *Leiostomus sp.* (spot) based on human recreational consumption or predation by dolphin. Individual fish were wrapped in solvent-rinsed aluminum foil, frozen, and transported to the laboratory. Concurrently and in order to reduce collection costs during this field study, similar fish species were also captured by cast net or obtained from Georgia Department of Natural Resources Annual Shrimp Trawls from estuarine locations near the LCP Superfund site (Fig. 1b.). All samples were transported to National Centers for Coastal Ocean Science (NCCOS) laboratories in Charleston, SC, and stored frozen (-40°C) until analyzed. Sediment was not collected from Brunswick, GA, estuaries during this study, although sediment from the Turtle-Brunswick River Estuary (TBRE) near the LCP site was previously collected by NOAA staff and has been consistently maintained at the Charleston, SC, facility since collection in 1996 (Long et al. 1998). These archival samples were re-extracted and analyzed for this study to serve as a positive environmental Aroclor 1268 signature.

Sediment extraction and analysis

Sediment samples were thawed and approximately 10 g of wet sediment was extracted using pressurized fluid extraction (PFE) (ASE 200, Dionex Inc.). Prior to extraction, each sediment aliquot was combined with ~27 g of anhydrous sodium sulfate and homogenized using a mortar and pestle. The dried mixture was transferred into a 33 mL ASE cell and spiked with a suite of labeled ^{13}C -PCB congeners (Cambridge Isotope Laboratory, Inc.) and extracted with a mixture of dichloromethane and acetone (50:50 volume fraction). The volume of the resulting extract was reduced under nitrogen and passed through an SX-3 gel permeation column (GPC, J2 Scientific, Inc.) to remove lipids, pigments, and sulfur. Post-GPC extracts were again evaporated under nitrogen to ~1 mL and further cleaned via solid phase extraction (SPE) using ~2 g of 5 % water-deactivated alumina. The final extract was solvent exchanged into hexane and the final volume was adjusted to ~0.5 mL. A recovery standard (^{13}C - δ -hexachlorocyclohexane) was

added prior to instrumental analysis in order to evaluate extracted internal standard recoveries.

Tissue preparation and fillet analysis

Fish were prepared by partially thawing the fish, descaling, and removing each fillet with the skin included. At least 20 g of tissue was required for homogenization in TeflonTM containers using a ProScientific Tissue Homogenizer with a titanium or stainless-steel homogenization probe. Wet tissues (~5 g) were weighed into anhydrous sodium sulfate (~33 g) and ground to dryness. Dried samples were transferred into 33 mL ASE extraction cells, spiked with internal standard as above, and samples were extracted by PFE with 100 % dichloromethane. Samples were passed through GPC and SPE cleanup steps, and the final extract was solvent exchanged into hexane. The final volume was diluted to ~0.5 mL. Prior to instrumental analysis, ^{13}C - δ -HCH was also added as a recovery standard to these extractions.

Sample analysis and data quality assurance

Samples were analyzed using an Agilent 6890 GC equipped with a 5973 Mass Selective Detector (MSD; EI mode). GC parameters included a DB-5ms column (J&W; 30 m $\times$ 0.25 mm diameter $\times$ 0.25- μm film thickness) and a Programmable Temperature Vaporization (PTV) inlet. Concentrations of 88 PCB congeners were determined for both sediments and tissues. Blanks, fortified samples, and SRMs (NIST 1941b Organics in Marine Sediment and 1944 New York/New Jersey Water Way Sediment for sediment and NIST 1947 Lake Michigan Fish Tissue and 2977 Mussel Tissue (Organic Contaminants and Trace Elements) for tissues) were used to ensure data quality. Recoveries of method spikes and matrix spikes averaged (standard deviation) 103.8 (21.2)% and 98.6 (20.4)%, respectively. NIST SRMs 1941b for sediments and NIST SRMs 1947 and 2977 for tissues were also analyzed; PCB congener concentrations were within 10 % (SRM 1941b), 5 % (SRM 1947), and 20 % (SRM 2977) of certified values.

Data analysis

Congener patterns of PCBs in sediment and fish from both Brunswick and Sapelo NERR were compared to other regional datasets for sediment and fish tissues

from coastal South Carolina (Van Dolah et al. 2013) and North Carolina NERRs (Cooksey et al. 2008). Fish collections for the SC and NC projects were collected in a similar manner as samples for this study. These datasets were chosen because each represented approximately the same spatial scale, were analyzed using similar protocols, and the targeted fish species were similar in nature.

Descriptive statistics for sediment and fish were determined (on a per sample basis) for total PCB (PCB_T), defined as the sum of the 65 PCB congeners common to all four regional assessments. The congener-specific proportion of PCB_T was calculated as well. The geometric mean and the standard error of the mean are reported throughout the manuscript. The standard error of the mean was calculated by determining the standard deviation of the natural log-transformed data that was reportable, converting the transformed data ($\exp^{\text{stddev} \ln}$) and dividing by the square root of the count data.

The PCB data was also reduced to include 22 congener peaks that were measured across all projects and associated with Aroclor 1268 as detailed in previous publications (Ishikawa et al. 2007; Maruya and Lee 1998; Pulster and Maruya 2008). The PCB congener list (PCB_r) used to define regional differences included 29 congeners (PCBs 3, 8/5, 18, 20, 28/31, 44, 52, 77, 101/90/89, 105, 118/106, 126, 149, 153, 170/190, 180/193, 187, 194, 202, 206, 207, and 209) and is similar to the list used by Pulster et al. (2005) to evaluate regional differences in PCB composition in the southeastern USA. This list of 22 congeners includes 7 congeners (PCBs 180/193, 194, 206, 209, 187, 202, and 207) that are of importance to Aroclor 1268 and account for ~75 % of the Aroclor 1268 profile (Maruya and Lee 1998; Kucklick et al. 2011). The proportion of PCB_r to PCB_T was then calculated, and sites were compared using an ANOVA (SAS version 9.3) for both sediments and tissues. The congener pattern observed in archived Brunswick, GA, sediments collected in 1996 and reanalyzed for this study has previously been associated with Aroclor 1268 (Kannan et al. 1997; Maruya and Lee 1998).

Results

General description of sediments

The mean sediment concentration for PCB_T for the SI NERRS was 0.205 ng/g dry weight (dw) (standard error

(SE) of 0.637 ng/g dw). Station concentrations ranged from 0.015 to 3.84 ng/g dw (Table 1). No station exceeded published sediment toxicity guidelines such as the effects range low (ERL; 22.7 ng/g dw) or effects range median (ERM; 180 ng/g dw) (Long et al. 1995) or the probable effects level (PEL; 189 ng/g dw) (Canadian environmental quality guidelines 2002). For comparison, PCB_T concentrations from archived and reanalyzed Brunswick, GA, sediment samples averaged 79.3 ng/g dw (SE 2.47 ng/g dw) and ranged from 5.37 to 4200 ng/g dw. Five of the 10 archived BR stations had PCB levels that exceeded the ERM (180 ng/g dw), and three of the remaining five stations had PCB levels greater than the ERL (22.7 ng/g dw) (Long et al. 1995). Congener profiles were similar in both SI and BR sediment samples. Dominant congeners (in descending rank order; 1–3) for both locations (Table 2) were PCB 206, 209, and 202, accounting for nearly 70 % of PCB_T. The remaining congeners (rank order 4–10) included PCB 194, 187, 207, 195, 198, 180/193, and 52 for BR sediments and PCB 187, 153, 194, 99, 52, 28/31, and 183 for SI sediments.

General description of analyzed fish fillets

A total of 22 fish fillets from the SI NERR were analyzed for PCBs and included *M. cephalus* ($n=9$), *M. undulatus* ($n=3$), *C. nebulosus* ($n=3$), *Menticirrhus sp.* ($n=1$), *B. chrysoura* ($n=3$), *S. ocellatus* ($n=2$), and *P. lethostigma* ($n=1$). PCB_T concentrations averaged 3.90 (SE 0.577) ng/g wet weight (ww) (Table 3) and individual fillet concentrations ranged from 0.60 (silver perch) to 41.0 (whiting) ng/g ww. A total of 29 fillets were analyzed from the area around Brunswick, GA (BR), and species included *M. undulatus* ($n=8$), *Menticirrhus sp.* ($n=1$), *M. cephalus* ($n=9$), *B. chrysoura* ($n=4$), *C. nebulosus*

Table 1 PCB_T sediment concentrations (reported as the geometric mean) from BR, SI, SC (van Dolah et al. 2013), and NC (Cooksey et al. 2008)

Site	PCB _T concentration ± standard error (ng/g dry mass)	PCB _r /PCB _T proportion
BR	79.3 (2.47)	0.967
SI	0.205 (0.637)	0.904
SC	0.107 (0.3.06)	0.382
NC	0.005 (0.15.1)	0.212

Table 2 Rank order (by congener proportion) of the congeners from BR, SI, SC (van Dolah et al. 2013), and NC (Cooksey et al. 2008) sediments and the associated PCB_r congener proportion

Rank order	BR_sediment	SI_sediment	SC_sediment	NC_sediment
1	PCB 206 (0.611)	PCB 206 (0.665)	PCB 52 (0.079)	PCB 206 (0.589)
2	PCB 209 (0.121)	PCB 209 (0.104)	PCB 66 (0.071)	PCB 37 (0.226)
3	PCB 202 (0.094)	PCB 202 (0.049)	PCB 110 (0.067)	
4	PCB 194 (0.056)	PCB 187 (0.049)	PCB 153 (0.050)	
5	PCB 187 (0.043)	PCB 153 (0.019)	PCB 18 (0.041)	
6	PCB 207 (0.036)	PCB 194 (0.018)	PCB 15 (0.038)	
7	PCB 195 (0.004)	PCB 99 (0.015)	PCB 156 (0.036)	
8	PCB 198 (0.004)	PCB 52 (0.015)	PCB 99 (0.035)	
9	PCB 180/193 (0.003)	PCB 28/31 (0.009)	PCB 92 (0.035)	
10	PCB 52 (0.003)	PCB 183 (0.008)	PCB 101/90/89 (0.024)	

($n=1$), and *Leiostomus sp.* ($n=6$). Tissue fillet concentrations of PCB_r at BR averaged 141 ng/g ww (SE 0.478 ng/g ww) and ranged from 32.0 to 838 ng/g ww (Table 3). Congener profiles were again similar in both SI and BR tissue samples (Table 4). Dominant congeners for both BR and SI included PCB 206, 187, and 202, accounting for between ~45 % (SI) and 65 % (BR) of the PCB_r. The remaining congeners included PCB 194, 209, 207, 183, 180/193, 153, and 154 for BR tissues and PCB 153, 194, 180/193, 99, 209, 101/90/89, and 183 for SI tissues.

Species-specific descriptive statistics are found in Table 5. Comparisons were limited to those species for which there were matched data from the SI NERR and Brunswick, GA, and the sample size was greater than one from both regions (silver perch, mullet, and Atlantic croaker). Mean tissue concentrations were generally between 50 and 100 times greater in fish from the Brunswick, GA, sites relative to the fish from SI. Additionally, when PCB_r for fish was compared to US EPA consumption guidelines (four meals per month) (U.S. EPA 2000), all fish from Brunswick exceeded the lower

threshold for non-cancer risks (23 ng/g ww), and of these, 24 exceeded the upper threshold of 47 ng/g ww for non-cancer risks. Only two fish collected from the SI NERR exceeded the lower non-cancer threshold, and none exceeded the upper threshold. An additional 10 fish from the SI NERR were found to have PCB_r concentrations above the lower cancer-risk threshold of 5.9 ng/g ww; five had concentrations that also exceeded the upper cancer-risk endpoint of 12 ng/g ww (Fig. 2.).

The dominant congeners in both sediments (Table 2) and tissues (Table 4) from both SI and Brunswick, GA, were PCB 206, 202, and 187 and also included PCB 209, 194, 180/193, and 207. This profile was similar to the Aroclor 1268 profile described in Maruya and Lee (1998) for fish from Purvis Creek near Brunswick, GA.

Regional comparison

The mean PCB_r/PCB_t proportions for sediments ranged from 0.212 (NC) to 0.967 (BR). SI sediments matched closely (0.904) with the BR sediment PCB_r proportion. Results from the ANOVA (least squares means comparison) comparing the PCB_r/PCB_t proportion indicated significant differences between BR and both NC and SC datasets while BR and SI PCB_r proportions for sediments were not different ($p=0.936$). PCB_r proportions for SI sediments were different from both NC and SC as well ($p<0.0001$). NC and SC were not significantly different ($p=0.106$). A similar trend was observed in PCB_r/PCB_t tissue proportions. Mean proportions ranged from 0.881 (BR) to 0.464 (SC). BR and SI PCB_r proportion (tissue) results were not significantly different ($p=0.736$). PCB_r tissue proportions from SI

Table 3 PCB_r tissue (fillet) concentrations (reported as the geometric mean) from BR, SI, SC (van Dolah et al. 2013), and NC (Cooksey et al. 2008)

Site	PCB _r concentration±standard error (ng/g wet mass)	PCB _r /PCB _t proportion
BR	141 (0.478)	0.881
SI	3.90 (0.577)	0.824
SC	2.93 (0.451)	0.464
NC	0.087 (17.0)	0.470

Table 4 Rank order (by congener proportion) of the congeners from BR, SI, SC (van Dolah et al. 2013), and NC (Cooksey et al. 2008) fish fillets and the associated PCB_i congener proportion

Rank order	BR_tissues	SI_tissues	SC_tissues	NC_tissues
1	PCB 206 (0.311)	PCB 206 (0.170)	PCB 153 (0.159)	PCB 187 (0.401)
2	PCB 202 (0.181)	PCB 187 (0.143)	PCB 99 (0.093)	PCB 206 (0.388)
3	PCB 187 (0.148)	PCB 202 (0.130)	PCB 15 (0.047)	PCB 101/90/89 (0.057)
4	PCB 194 (0.063)	PCB 153 (0.093)	PCB 63 (0.043)	PCB 153 (0.049)
5	PCB 209 (0.045)	PCB 194 (0.032)	PCB 12 (0.037)	PCB 99 (0.042)
6	PCB 207 (0.034)	PCB 180/193 (0.029)	PCB 180/193 (0.035)	PCB 95 (0.037)
7	PCB 183 (0.034)	PCB 99 (0.028)	PCB 118/106 (0.028)	PCB 180/193 (0.029)
8	PCB 180/193 (0.028)	PCB 209 (0.023)	PCB 52 (0.019)	PCB 44 (0.016)
9	PCB 153 (0.020)	PCB 101/90/89 (0.023)	PCB 66 (0.013)	
10	PCB 154 (0.012)	PCB 183 (0.022)	PCB 202 (0.012)	

were different from both NC and SC as well ($p < 0.0001$). NC and SC were not significantly different ($p = 0.999$). Sediments from BR and SI shared 6 of the 10 most influential congeners, and there were no congeners among the 10 highest rank-ordered congeners that were shared among all four sediment datasets (Table 2). For tissues, 8 of 10 rank-ordered congeners were shared between BR and SI tissues and two congeners (PCB 153 and 180/193) were identified in each of the four regional datasets (although the ranks differed; Table 4).

Discussion

Sediment PCB_i concentrations in the SI NERR (0.015–3.84 ng/g dw) did not exceed the published ERL or ERM (Long et al. 1995) at any station, and concentrations were similar to other coastal assessments along the NC and SC coasts (Bergquist et al. 2011; Cooksey et al. 2008; Sanger et al. 2008; Van Dolah et al. 2013). Sediment PCB_i concentrations reported along the South Carolina coast over a 10-year (2000–2010) period ranged from not detectable (nd)–30.5 ng/g dw (annual

Table 5 Descriptive statistics for PCB_i (ng/g ww) in fish fillets collected in the SI NERR and the greater Brunswick, GA, estuary; no significant differences for lipid fraction were identified between locations and between species

Species	Count	Arithmetic mean lipid fraction (standard deviation)	Arithmetic mean (standard error) concentration (ng/g ww)	Concentration range (ng/g ww)
Sapelo Island				
Silver perch	3	0.0042 (0.0013)	1.34 (1.96)	0.604–18.8
Spotted seatrout	3	0.0115 (0.0036)	8.14 (0.38)	4.44–18.5
Striped mullet	9	0.0216 (0.0149)	3.32 (0.17)	1.61–7.08
Atlantic croaker	3	0.0249 (0.0165)	5.16 (0.47)	5.84–11.0
Whiting	1	0.0218	25.2	
Southern flounder	1	0.0013	0.873	
Red drum	2	0.0111 (0.0087)	2.17 (1.20)	0.877–13.3
Brunswick				
Silver perch	4	0.0320 (0.0252)	146 (0.51)	107–478
Spotted seatrout	1		42.3	
Spot	6	0.0253 (0.0327)	69.1 (0.56)	33.3–770
Striped mullet	9	0.0141 (0.0030)	207 (0.23)	32.0–838
Atlantic croaker	8	0.0482 (0.0425)	150 (0.27)	70.3–779
Whiting	1	0.0187	188	

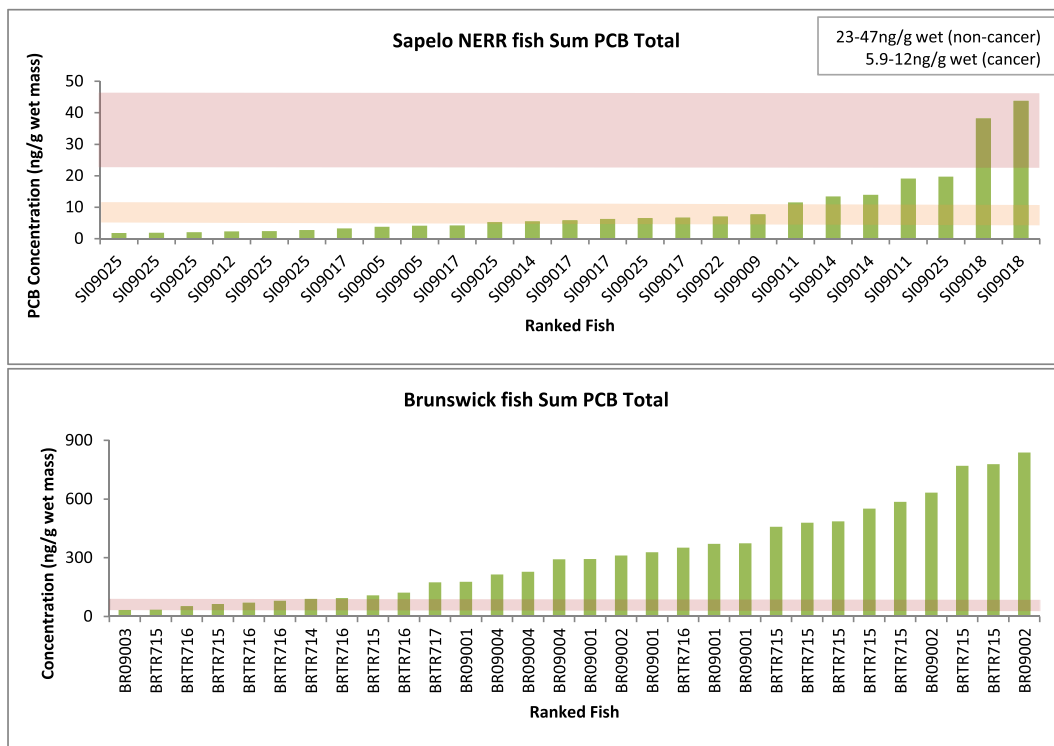


Fig. 2 Rank-ordered arithmetic mean fillet PCB_f concentrations from SI and BR fish plotted in relation to the US EPA cancer and non-cancer fish consumption limits estimating four meals per month (U.S. EPA 2000)

mean 0.750 ng/g dw) (Bergquist et al. 2009; Van Dolah et al. 2013; VanDolah et al. 2004, 2006). The dominant congeners reported for SC sediments were PCBs 138/163/164, 44, 52, and 153 (Bergquist et al. 2009; VanDolah et al. 2004, 2006). The mean PCB_f sediment concentration reported for NC NERR was 0.240 ng/g dw (range, nd–1.24 ng/g dw), dominated by PCB 206, but there were very few congener detections reported (Cooksey et al. 2008) (Table 2). PCBs were not detected in sediment from the Grays Reef National Marine Sanctuary, 32 km off the coast of Sapelo Island, GA (Balthis et al. 2007).

Several publications have highlighted the need for information on the trophic transfer of PCBs from sediments and prey species to apex and sentinel species such as coastal dolphins (Balmer et al. 2011; Kucklick et al. 2011; Pulster et al. 2005). Prey species from the Brunswick, GA, area had relatively high levels of PCBs (32.0 to 838, mean 141 ng/g ww), in agreement with other reports from the Brunswick area (Balmer et al. 2011; Pulster et al. 2005). Concentrations of PCB_f recently reported in SC fish range from 1.49 to 15.1 (mean 5.06) ng/g ww (Van Dolah et al. 2013), and the dominant

congeners expressed in these fish were PCBs 153, 99, and 15. Reported PCB_f concentrations in fish from NC NEER ranged from 0.29 to 4.03 (mean of 1.36) ng/g ww and were primarily driven by PCBs 187, 101/90/89, and 206 (Cooksey et al. 2008). Concentrations reported in fish from SI NERR ranged from 0.604 to 41.0 (mean 3.90) ng/g ww (current study). The congener patterns from SI NERR were more similar to the pattern found in samples from Brunswick estuaries and dominated by congeners PCBs 206, 187, and 202 (Table 4). The rank order of the Aroclor 1268 specific congeners for sediment and tissues follows the typical Aroclor 1268 pattern described by Kannan et al. (1998) and Maruya and Lee (1998).

The similar congener patterns in both sediment and tissues and the expected decrease in PCB concentration in SI relative to BR strongly indicate that PCB transport into SI NERR has occurred and could be attributed to several environmental routes, although the question of how Aroclor 1268 came to occur in SI cannot be defined in this experimental design. It is well documented that PCB transport occurs via downstream sediment transport (Feng et al. 1998). Feng et al. (1998) described a

10-fold decrease over an approximately 80-km stream transect along the Hudson River. In the case of transport from BR to SI, our data seem to indicate a 100-fold decrease over a similar spatial scale. Another possible route of PCB movement is through the migration of fish populations, realizing that fish migration is species and season dependent. There are subtle differences in PCB homolog patterns in fish from SI and BR (Table 4). Congener patterns appear to be relatively similar for the more highly chlorinated congeners (homolog groups 7, 8, 9, and 10), though congeners in homolog groups 4, 5, and 6 are enhanced in SI fish compared with BR. It is interesting to note that several congeners found in SI tissue were not often detected in SI sediment samples. For example, PCBs 105, 110, 177, 188, and 195 were detected in tissues but not detected in SI sediments. It seems that the PCB profiles between SI and BR are more similar for tissues than sediments and may indicate that tissues are a more likely source of transport, thus identifying fish as an influential vector for PCBs to be passed onto predators found in the SI NERR. Balmer et al. (2011) reported distinct dolphin populations that are resident in SI and distinct from dolphin populations found in the BR estuary; yet both populations exhibited clear Aroclor 1268-type congener patterns in blubber samples. Understanding the presence of an Aroclor 1268-type pattern in SI as observed in this study implies an influence from the likely source (BR) and helps define needed research into predator/prey associations and Aroclor 1268 movement mechanisms into this protected estuary.

US EPA consumption guidelines (U.S. EPA 2000) were used in an attempt to understand potential risks associated with the levels of PCB found in fish from Sapelo Island, NERR. Total PCB levels in some SI NERR fish exceeded US EPA lower and upper endpoints for cancer risks (48 % > 5.9 ng/g ww and 16 % > 12 ng/g ww, respectively) and the lower endpoint for non-cancer/systemic-health risks (6.5 % > 23 ng/g ww) based on predictions associated with consumption of four 8-oz meals per month (U.S. EPA 2000); none exceeded the upper non-cancer endpoint of 47 ng/g ww. Due to the restricted access to the SI NERR (boat only), the extent of recreational or subsistence fishing that occurs in these waters is unclear. All fish from BR exceeded both the lower and upper endpoints for cancer risks and the lower endpoint for non-cancer risks; most (73 %) also exceeded the upper non-cancer endpoint. Mullet and croaker from Purvis Creek (adjacent to the

LCP site in Brunswick, GA) are identified in Georgia EPD publications as “do not eat,” and red drum and flounder are listed as “1 meal per month” due to PCB contamination (Guidelines for Eating Fish from Georgia Waters 2010).

While SI PCB levels in both sediment and fish were considerably lower than the PCB levels found in Brunswick, GA, the congener patterns strongly suggest transport of Aroclor 1268 away from the LCP site and into the SI NERR (Tables 1, 2, 3, and 4). The rank order for the first four congeners (based on proportion) is identical in both SI and BR sediment samples from 1994 that were reanalyzed for this study (PCB 206 > 199 > 203/196 > 208), and 8 of the 10 most abundant congeners are similar between BR and SI sediments. Congeners 153 and 52 are among the 10 most abundant congeners from SI and are not associated with published Aroclor 1268 patterns (Maruya and Lee 1998; Kucklick et al. 2011), and this signal likely indicates ambient background PCBs (Hoekstra et al. 2003). Kucklick et al. (2011) showed a high correlation of PCB 153 with coastal human population supporting the assertion of PCB 153 as an indicator of general urban PCB contamination (non-Aroclor 1268). Congener profiles follow a similar pattern of agreement in tissues (Table 4). Congener ranks are identical for PCB 199 > 206 > 203/196 > 202 > 187 and 8 of the 10 most abundant congeners are again the same in both SI and BR tissues.

An earlier study examined dolphin prey species from three coastal regions including Brunswick, GA; Jacksonville, FL, to the south; and Savannah, GA, to the north of the LCP site (Pulster et al. 2005). Using discriminant analysis, these authors reported that the Aroclor 1268 patterns (using congeners 194, 138, 180, and 196) were recognizable to the south (in samples from Jacksonville, FL) but not in PCB profiles from Savannah. This apparent pattern movement is not specific to fish, as Aroclor 1268 specific signatures in turtles were reported as far south as Port Canaveral, FL (Ragland et al. 2011). Our data clearly indicated that the SC and NC datasets were distinct and different from SI and BR data for both sediments and tissues.

In closing, while the magnitude of PCB contamination at the SI NERR is much lower than that in the Brunswick, GA, area near the LCP site, the similarity of congener profiles suggests that much of the PCB contamination at the SI NERRS is likely due to transport from the LCP site. Sediment advection may account for the PCB transport into SI as only a small proportion of

highly contaminated BR sediment could explain the PCB levels found in SI. Additionally, PCB transport could also be a result of fish migration from BR into SI. The more similar PCB patterns were found between tissue PCB profiles. Fish tissue concentrations occasionally exceed US EPA human-health guidelines for cancer and non-cancer health endpoints (based on consumption of four 8-oz meals per month). Without better understanding of the amount of prey dolphins consume, it may be difficult to gauge if consumption of SI resident fish is enough to explain the high levels of PCB reported in SI resident dolphin populations. Generally, the Aroclor 1268 signature around BR and SI appears to be closely bounded to the north. Future research includes similar estuary wide assessments in northern Florida and southern South Carolina to help better understand the extent of the movement of Aroclor 1268 along the southeastern coast and especially among coastal-protected research areas (NERRs).

Acknowledgments This work was funded in part by the NOAA Oceans and Human Health Initiative. The authors would also like to acknowledge the contributions of Dorset Hurley, Research Coordinator at the Sapelo Island NERR for project management and insight, Georgia Department of Natural Resources for fish collections from the Brunswick, GA estuaries, Dan Liebert, JHT contractor to NOAA for chemical analysis, and the many manuscript reviewers both internal to NOAA and the journal who helped complete this manuscript.

Disclaimer The National Ocean Service (NOS) does not approve, recommend, or endorse any proprietary product or material mentioned in this publication. Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

References

- Balmer, B. C., Schwacke, L. H., Wells, R. S., George, R. C., Hoguet, J., Kucklick, J. R., Lane, S. M., Martinez, A., McLellan, W. A., Rosel, P. E., Rowles, T. K., Sparks, K., Speakman, T., Zolman, E. S., & Pabst, D. A. (2011). Relationship between persistent organic pollutants (POPs) and ranging patterns in common bottlenose dolphins (*Tursiops truncatus*) from coastal Georgia, USA. *Science of the Total Environment*, 409(11), 2094–2101. doi:10.1016/j.scitotenv.2011.01.052.
- Balthis, L., Hyland, J., Cooksey, C., Fulton, M., & McFall, G. (2007). Long-term monitoring of ecological conditions in Gray's Reef National Marine Sanctuary: comparison of soft-bottom benthic assemblages and contaminant levels in sediments and biota in spring 2000 and 2005 (Vol. NOAA Technical Memorandum NOS NCCOS 68, pp. 36). Charleston, SC: NOAA/NOS/NCCOS.
- Balthis, L., Hyland, J., Cooksey, C., Wirth, E., Fulton, M., Moore, J. C., & Hurley, D. (2012). *Support for integrated ecosystem assessment of NOAA's National Estuarine Research Reserve system (NERRS): assessment of ecological condition and stressor impacts in subtidal waters of the Sapelo Island National Estuarine Research Reserve* (p. 79). Charleston: NOAA Center for Coastal Environmental Health and Biomolecular Research.
- Bergquist, D. C., VanDolah, R. F., Riekerk, G. H. M., Levinsen, M. V., Crowe, S. E., Brock, L., Greenfield, D. I., Chestnut, D. E., McDermott, W., Fulton, M. H., Wirth, E., & Harvey, J. (2009). *The condition of South Carolina's estuarine and coastal habitats during 2005–2006* (p. 74). Charleston: South Carolina Department of Natural Resources.
- Bergquist, D. C., VanDolah, R. F., Riekerk, G. H. M., Levinsen, M. V., Crowe, S. E., Chestnut, D. E., McDermott, W., Fulton, M. H., Wirth, E., & Harvey, J. (2011). *The condition of South Carolina's estuarine and coastal habitats during 2007–2008. South Carolina Marine Resources Division Technical Report 106* (p. 64). Charleston: SC Department of Natural Resources.
- Canadian environmental quality guidelines*. (2002). Canadian sediment quality guidelines for the protection of aquatic life: summary tables, updated. In: Canadian environmental quality guidelines, 1999, Canadian Council of Ministers of the Environment. Winnipeg, Canada.
- Cooksey, C., Hyland, J., Wirth, E., Balthis, W. L., Fulton, M., & Whittall, D. (2008). *Support for integrated ecosystem assessments of NOAA's national estuarine research reserve system (NERRS). Volume II: assessment of ecological condition and stressor impacts in subtidal waters of the North Carolina NERRS*. Charleston, SC: NOAA Technical Memorandum NOS NCCOS 83.
- EPA, U. S. (2000). *Guidance for assessing chemical contaminant data for use in fish advisories, volume 2: risk assessment and fish consumption limits*. (EPA 823-B-00-008). Washington: United States Environmental Protection Agency.
- Feng, H., Cochran, J. K., Lwiza, H., Brownawell, B. J., & Hirschberg, D. J. (1998). Distribution of heavy metals and PCB contaminants in the sediments of an urban estuary: the Hudson River. *Marine Environmental Research*, 45(1), 69–88.
- Georgia Department of Natural Resources. (2010). Guidelines for eating fish from Georgia waters, 2010 update. Atlanta, GA. 59pp.
- Hoekstra, P. F., O'Hara, T. M., Fisk, A. T., Borgå, K., Solomon, K. R., & Muir, D. C. G. (2003). Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort–Chukchi Seas. *Environmental Pollution*, 124(3), 509–522. doi:10.1016/S0269-7491(02)00482-7.
- Ishikawa, Y., Noma, Y., Mori, Y., & Sakai, S.-i. (2007). Congener profiles of PCB and a proposed new set of indicator congeners. *Chemosphere*, 67(9), 1838–1851. doi:10.1016/j.chemosphere.2006.05.075.

- Kannan, K., Maruya, K. A., & Tanabe, S. (1997). Distribution and characterization of polychlorinated biphenyl congeners in soil and sediments from a superfund site contaminated with Aroclor 1268. *Environmental Science & Technology*, 31(5), 1483–1488. doi:10.1021/es960721r.
- Kannan, K., Nakata, H., Stafford, R., Masson, G. R., Tanabe, S., & Giesy, J. P. (1998). Bioaccumulation and toxic potential of extremely hydrophobic polychlorinated biphenyl congeners in biota collected at a superfund site contaminated with Aroclor 1268. *Environmental Science & Technology*, 32(9), 1214–1221. doi:10.1021/es9709435.
- Kucklick, J., Schwacke, L., Wells, R., Hohn, A., Guichard, A., Yordy, J., Hansen, L., Zolman, E., Wilson, R., Litz, J., Nowacek, D., Rowles, T., Pugh, R., Balmer, B., Sinclair, C., & Rosel, P. (2011). Bottlenose dolphins as indicators of persistent organic pollutants in the Western North Atlantic Ocean and Northern gulf of Mexico. *Environmental Science & Technology*, 45(10), 4270–4277. doi:10.1021/es1042244.
- Long, E., MacDonald, D., Smith, S., & Calder, F. (1995). Incidences of adverse biological effects within ranges of chemical concentrations in marine and estuarine sediments. *Environmental Management*, 19, 81–97.
- Long, E., Scott, G.I., Kucklick, J., Fulton, M., Thompson, B., Carr, R.S., Biedenbach, J., Scott, K.J., Thursby, G.B., Chandler, G.T., Anderson, J.W., & Sloane, G.M. (1998). Magnitude and extent of sediment toxicity in selected estuaries of South Carolina and Georgia. Washington, DC: NOAA Technical Memorandum NOS ORCA128.
- Maruya, K. A., & Lee, R. F. (1998). Aroclor 1268 and toxaphene in fish from a southeastern U.S. estuary. *Environmental Science & Technology*, 32(8), 1069–1075. doi:10.1021/es970809k.
- Pulster, E.L., Smalling, K.L., Zolman, E., Schwacke, L., & Maruya, K. (2009). Persistent organochlorine pollutants and toxaphene congener profiles in bottlenose dolphins (*Tursiops truncatus*) frequenting the Turtle/Brunswick River estuary, Georgia, USA. *Environmental. Toxicological and Chemistry*. 28(7):1390–1399.
- Pulster, E. L., & Maruya, K. A. (2008). Geographic specificity of Aroclor 1268 in bottlenose dolphins (*Tursiops truncatus*) frequenting the Turtle/Brunswick River Estuary, Georgia (USA). *Science of the Total Environment*, 393(2–3), 367–375. doi:10.1016/j.scitotenv.2007.12.031.
- Pulster, E. L., Smalling, K. L., & Maruya, K. A. (2005). Polychlorinated biphenyls and toxaphene in preferred prey fish of coastal southeastern U.S. bottlenose dolphins (*Tursiops truncatus*). *Environmental Toxicology and Chemistry*, 24(12), 3128–3136.
- Ragland, J. M., Arendt, M. D., Kucklick, J. R., & Keller, J. M. (2011). Persistent organic pollutants in blood plasma of satellite-tracked adult male loggerhead sea turtles (*Caretta caretta*). *Environmental Toxicology and Chemistry*, 30(7), 1549–4556. doi:10.1002/etc.540.
- Sanger, D., Blair, A., DiDonato, G., Washburn, T., Jones, S., Chapman, R., Bergquist, D., Riekerk, G., Wirth, E., Stewart, J., White, D., Vandiver, L., White, S., & Whittall, D. (2008). Support for integrated ecosystem assessment of NOAA's national estuarine research reserve system (NERRS, Volume I: The impacts of coastal development on the ecology and human well-being of tidal creek ecosystems of the US Southeast (Vol. NOAA Technical Memorandum NOS NCCOS 82). Charleston, SC.
- Sapelo Island National Estuarine Reserve. (2008). Sapelo Island National Estuarine Research Reserve Management Plan, 2008–2013: *Sapelo Island National Estuarine Research Reserve*. http://nerrs.noaa.gov/Doc/PDF/Reserve/SAP_MgmtPlan.pdf.
- Van Dolah, R. F., Sanger, D. M., Riekerk, G. H. M., Crowe, S. E., Levinsen, M. V., Bergquist, D. C., Chestnut, D. E., McDermott, W., Fulton, M. H., & Wirth, E. (2013). *The condition of South Carolina's estuarine and coastal habitats during 2009–2010* (p. 64). Charleston: South Carolina Marine Resources Division.
- VanDolah, R. F., Jutte, P. C., Riekerk, G. H. M., Levinsen, M. V., Crowe, S. E., Lewitus, A., Chestnut, D. E., McDermott, W., Bearden, D., Fulton, M. H., & Harvey, J. (2004). *The condition of South Carolina's estuarine and coastal habitats during 2001–2002* (p. 70). Charleston: South Carolina Department of Natural Resources.
- VanDolah, R. F., Berquist, D. C., Riekerk, G. H. M., Levinsen, M. V., Crowe, S. E., Wilde, S. B., Chestnut, D. E., McDermott, W., Fulton, M. H., Wirth, E., & Harvey, J. (2006). *The condition of South Carolina's estuarine and coastal habitats during 2003–2004* (p. 70). Charleston: South Carolina Department of Natural Resources.