

## **Supporting Information**

### **Temporal Trends in Per- and Polyfluoroalkyl Substances in Bottlenose Dolphins (*Tursiops truncatus*) of Indian River Lagoon, Florida and Charleston, South Carolina**

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**Table S1.** Per- and polyfluoroalkyl substances (PFAS) group, abbreviation, full name, and limit of detection (LOD) for sample years 2012-2013 and 2015.

PFAS group	PFAS	Full name	LOD (ng/g wet wt.)	
			2012-2013	2015
PFSA	PFBS	Perfluorobutane sulfonate	0.005	
PFSA	PFHxS	Perfluorohexane sulfonate	0.030	0.293
PFSA	PFHpS	Perfluoroheptane sulfonate	0.005	
PFSA	PFOS	Perfluorooctane sulfonate	1.046	0.600
PFSA	PFDS	Perfluorodecane sulfonate	0.005	0.275
PFSA	PFOSA	Perfluorooctane sulfonamide	0.005	0.287
PFSA	PFECHS	Perfluoroethylcyclohexane sulfonate	0.047	
	ΣPFSA <sub>s</sub>	Sum perfluorosulfonates		
PFCA	PFHxA	Perfluorohexanoate	0.046	0.275
PFCA	PFHpA	Perfluoroheptanoate	0.042	0.276
PFCA	PFOA	Perfluorooctanoate	0.135	0.420
PFCA	PFNA	Perfluorononanoate	0.044	0.279
PFCA	PFDA	Perfluorodecanoate	0.039	0.228
PFCA	PFUnDA	Perfluoroundecanoate	0.028	0.235
PFCA	PFDoDA	Perfluorododecanoate	0.005	0.433
PFCA	PFTriA	Perfluorotridecanoate	0.005	
PFCA	PFTA	Perfluorotetradecanoate	0.005	
PFCA	PPeA	Perfluoropentanoate		4.370
	ΣPFCAs	Sum perfluorocarboxylates		

**Table S2.** Summary of median, geometric mean (GM), and range of per- and polyfluoroalkyl substances (PFAS) plasma concentrations (ng/g wet weight), by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC.

PFAS	N	Median	GM	Min	Max	N	Median	GM	Min	Max
2003					2004					
PFHpA <sup>a</sup>	2	0.6	0.6	0.6	0.6	0	-	-	-	-
PFOA	46	35.8	33.0	4.7	163	19	35.7	34.6	11.2	123
PFNA	46	54.6	51.5	11.2	214	19	65.3	66.7	29.2	278
PFDA	46	121	132	40.7	542	19	106	132	59.3	483
PFUnDA	46	53.9	50.4	10.2	320	19	76.6	95.0	23.9	583
PFDoDA	46	8.7	8.1	0.4	62.4	19	7.7	10.0	1.5	135
PFHxS	46	41.6	33.0	4.6	165	19	36.4	31.7	5.9	322
PFOS	46	1260	1190	492	3070	19	708	813	317	2310
PFDS <sup>a</sup>	2	15.4	15.3	14.9	15.8	0	-	-	-	-
2005					2013					
PFHpA	17	0.6	3.0	0.6	95.2	19	6.7	6.1	0.9	38.6
PFOA	17	36.5	32.9	0.2	561	19	23.3	23.8	6.3	55.4
PFNA	17	108	131	28.0	561	19	59.9	54.3	18.6	113
PFDA	17	225	180	54.9	667	19	70.8	69.3	41.4	110
PFUnDA	17	121	104	25.3	496	19	25.6	27.0	14.8	69.2
PFDoDA	17	10.1	4.7	0.2	30.1	19	7.2	7.4	3.0	36.4
PFHxS	17	126	83.3	8.5	471	19	11.8	13.3	4.1	45.4
PFOS	17	1890	1710	491	6260	19	571	652	288	1830
PFDS	17	28.7	36.5	10.0	267	19	3.9	4.7	2.0	24.0

<sup>a</sup>PFAS not measured in 2004

**Table S3.** Summary of median, geometric mean (GM), and range of per- and polyfluoroalkyl substances (PFAS) plasma concentrations (ng/g wet weight), by year, in bottlenose dolphins (*Tursiops truncatus*) from Indian River Lagoon, FL.

PFAS	N	Median	GM	Min	Max	N	Median	GM	Min	Max
	2003					2004				
PFHpA <sup>a</sup>	0	-	-	-	-	0	-	-	-	-
PFOA	38	6.8	6.7	0.4	70.3	30	7.3	7.2	0.2	38.1
PFNA	38	10.6	11.1	3.2	50.9	30	8.1	7.1	0.3	34.1
PFDA	38	15.3	14.8	4.4	53.2	30	12.0	11.5	2.5	78.6
PFUnDA	38	10.1	10.9	1.8	64.1	30	11.2	10.5	2.6	73.8
PFDoDA	38	1.9	1.4	0.4	6.1	30	1.5	1.1	0.3	12.9
PFHxS	38	21.1	30.8	2.3	332	30	40.7	34.4	2.6	227
PFOS	38	411	473	69.2	2010	30	395	520	92.8	1870
PFDS <sup>a</sup>	0	-	-	-	-	0	-	-	-	-
	2005					2006				
PFHpA	12	0.6	0.8	0.6	26.6	6	1.0	1.0	0.6	1.4
PFOA	12	12.3	11.2	2.1	65.6	6	6.0	6.0	4.0	9.6
PFNA	12	17.4	18.7	8.8	65.6	6	7.0	7.1	4.9	11.2
PFDA	12	14.9	18.6	8.1	138	6	5.2	4.8	3.5	6.2
PFUnDA	12	11.5	13.5	7.3	68.4	6	6.6	6.7	4.8	9.9
PFDoDA	12	0.2	0.3	0.2	14.0	6	1.5	1.4	0.8	2.0
PFHxS	12	230	147	16.9	757	6	10.4	11.9	6.8	26.2
PFOS	12	2330	1450	303	3620	6	399	455	275	890
PFDS	12	17.3	14.6	3.2	51.0	6	1.8	1.8	1.3	3.0
	2010					2011				
PFHpA	14	1.0	0.9	0.5	2.7	13	0.8	0.7	0.2	1.1
PFOA	14	6.8	7.1	2.9	29.6	13	5.2	5.1	2.6	9.8
PFNA	14	6.9	8.4	3.7	29.8	13	5.7	6.0	2.8	10.5
PFDA	14	4.7	4.9	3.3	11.6	13	3.5	3.5	2.1	5.4
PFUnDA	14	7.1	6.8	4.5	12.7	13	5.8	5.6	2.8	8.6
PFDoDA	14	1.3	1.3	0.8	2.5	13	1.1	1.1	0.7	1.6
PFHxS	14	19.2	17.3	3.3	111	13	14.2	12.8	2.8	42.2
PFOS	14	250	368	137	1990	13	237	237	81.5	990
PFDS	14	1.3	1.4	1.0	2.7	13	1.2	1.2	0.9	1.5
	2012					2015				
PFHpA	17	0.9	0.7	0.1	1.7	16	0.6	0.6	0.2	1.2
PFOA	17	4.9	3.9	0.7	11.0	16	5.1	5.6	3.2	13.8
PFNA	17	9.0	8.5	3.3	19.8	16	7.2	7.6	4.1	17.6
PFDA	17	12.1	11.8	4.6	22.2	16	3.8	4.2	2.6	9.5
PFUnDA	17	6.3	6.1	3.2	11.0	16	5.9	6.3	4.4	9.2
PFDoDA	17	3.1	3.1	1.1	6.4	16	1.0	1.1	0.5	2.4
PFHxS	17	11.4	8.7	2.4	22.8	16	20.7	22.2	6.8	72.0
PFOS	17	344	317	116	1010	16	450	471	131	3490
PFDS	17	1.4	1.9	0.9	6.2	16	1.2	1.5	1.0	4.9

<sup>a</sup> PFAS not measured in 2003 or 2004

**Table S4.** Temporal trend (% change per year) and 95% confidence intervals (CI) of concentrations of per- and polyfluoroalkyl substances (PFAS) among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, stratified by location.

PFAS	Indian River Lagoon, FL			Charleston, SC		
	Trend	95% CI	N	Trend	95% CI	N
<b>PFCAs</b>						
PFHpA	3% decrease	-8, +2	78	12% increase	+1, +25	38
PFOA	4% decrease	-7, 0	146	3% decrease	-6, 0	101
PFNA	3% decrease	-5, -1	146	0%	-3, +2	101
PFDA	9% decrease	-11, -7	146	7% decrease	-8, -5	101
PFUnDA	6% decrease	-8, -4	146	8% decrease	-10, -5	101
PFDoDA	3% increase	0, +6	146	1% decrease	-5, +3	101
<b>PFSAs</b>						
PFHxS	9% decrease	-13, -5	146	9% decrease	-13, -6	101
PFOS	4% decrease	-7, -1	146	6% decrease	-8, -3	101
PFDS	16% decrease	-22, -11	78	21% decrease	-26, -15	38

**Table S5.** Temporal trend (% change per year) and 95% confidence intervals (CI) of concentrations of plasma per- and polyfluoroalkyl substances (PFAS) among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, combined.

PFAS	Trend	95% CI	N
<b>PFCAs</b>			
PFHpA	2% decrease	-8, +6	116
PFOA	7% decrease	-10, -4	247
PFNA	7% decrease	-10, -4	247
PFDA	14% decrease	-17, -11	247
PFUnDA	11% decrease	-13, -8	247
PFDoDA	3% decrease	-6, 0	247
<b>PFSAs</b>			
PFHxS	9% decrease	-12, -7	247
PFOS	6% decrease	-9, -4	247
PFDS	21% decrease	-25, -17	116

**Table S6.** Temporal trend (% change per year) and 95% confidence intervals (CI) of concentrations of plasma per- and polyfluoroalkyl substances (PFAS) among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, stratified by location, using piecewise linear splines with a knot at 2005.

	Trend	95% CI	Trend	95% CI
Indian River Lagoon, FL	2003-2005		2005-2015	
<b>PFCAs</b>				
PFHpA	-- <sup>a</sup>	-- <sup>a</sup>	3% decrease	-8, +2
PFOA	13% increase	-14, +50	6% decrease	-11, -1
PFNA	1% increase	-15, +20	4% decrease	-7, 0
PFDA	14% decrease	-29, +4	8% decrease	-12, -4
PFUnDA	4% decrease	-20, +15	6% decrease	-9, -3
PFDoDA	31% decrease	-49, -5	11% increase	+3, +18
<b>PFSAs</b>				
PFHxS	23% increase	-18, +85	14% decrease	-21, -6
PFOS	25% increase	-2, +60	9% decrease	-14, -3
PFDS	-- <sup>a</sup>	-- <sup>a</sup>	16% decrease	-22, -11
Charleston, SC	2003-2005		2005-2013	
<b>PFCAs</b>				
PFHpA	126% increase	+44, +256	9% increase	-3, +23
PFOA	0%	-33, +51	4% decrease	-13, +5
PFNA	55% increase	+27, +90	9% decrease	-14, -5
PFDA	14% increase	-5, +38	11% decrease	-14, -7
PFUnDA	49% increase	+20, +84	17% decrease	-21, -13
PFDoDA	19% decrease	-46, +23	3% increase	-6, +13
<b>PFSAs</b>				
PFHxS	49% increase	+12, +99	18% decrease	-24, -13
PFOS	11% increase	-7, +33	9% decrease	-13, -4
PFDS	54% increase	+23, +93	23% decrease	-28, -17

<sup>a</sup> PFAS not measured in 2003 and 2004

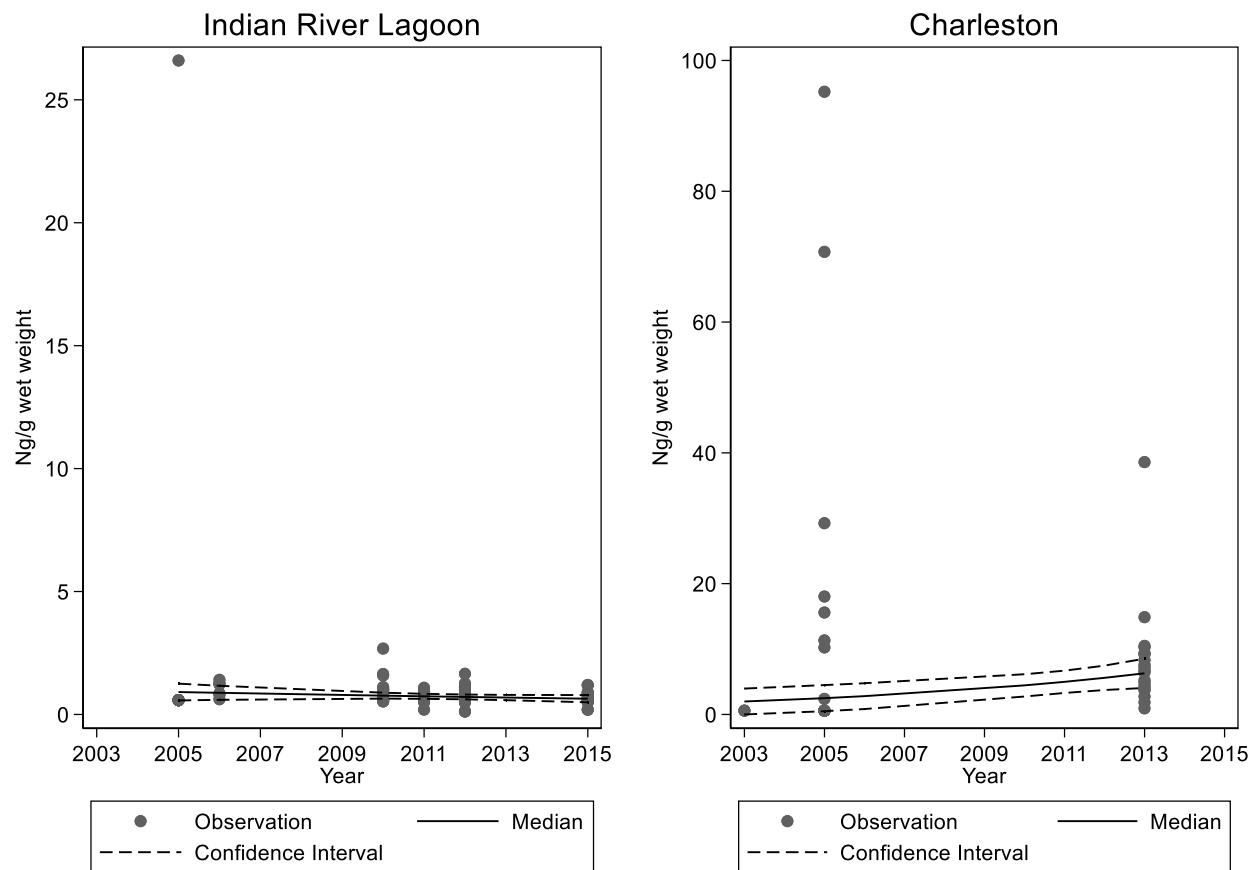
**Table S7.** Temporal trend (% change per year) and 95% confidence intervals (CI) of concentrations of plasma per- and polyfluoroalkyl substances (PFAS) among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, stratified by location and sex.

PFAS	Males			Females		
	Trend	95% CI	N	Trend	95% CI	N
<b>Indian River Lagoon, FL</b>						
<b>PFCAs</b>						
PFHpA	4% decrease	-9, +2	65	3% increase	-7, +13	10
PFOA	4% decrease	-7, -1	106	2% decrease	-11, +9	37
PFNA	4% decrease	-6, -2	106	2% increase	-4, +9	37
PFDA	10% decrease	-12, -7	106	5% decrease	-9, 0	37
PFUnDA	5% decrease	-7, -2	106	7% decrease	-11, -3	37
PFDoDA	2% increase	-1, +6	106	9% increase	+4, +16	37
<b>PFSAs</b>						
PFHxS	8% decrease	-12, -4	106	11% decrease	-21, +1	37
PFOS	4% decrease	-7, 0	106	3% decrease	-10, +5	37
PFDS	17% decrease	-23, -11	65	9% decrease	-21, +4	10
<b>Charleston, SC</b>						
<b>PFCAs</b>						
PFHpA	16% increase	-1, +36	20	4% increase	-11, +21	16
PFOA	4% decrease	-7, -1	60	5% decrease	-11, +2	39
PFNA	0%	-3, +3	60	2% decrease	-8, +3	39
PFDA	6% decrease	-9, -4	60	8% decrease	-11, -4	39
PFUnDA	8% decrease	-11, -4	60	8% decrease	-13, -3	39
PFDoDA	2% decrease	-6, +4	60	2% decrease	-7, +3	39
<b>PFSAs</b>						
PFHxS	11% decrease	-14, -8	60	8% decrease	-15, -1	39
PFOS	6% decrease	-9, -3	60	6% decrease	-10, -2	39
PFDS	17% decrease	-24, -11	20	26% decrease	-32, -19	16

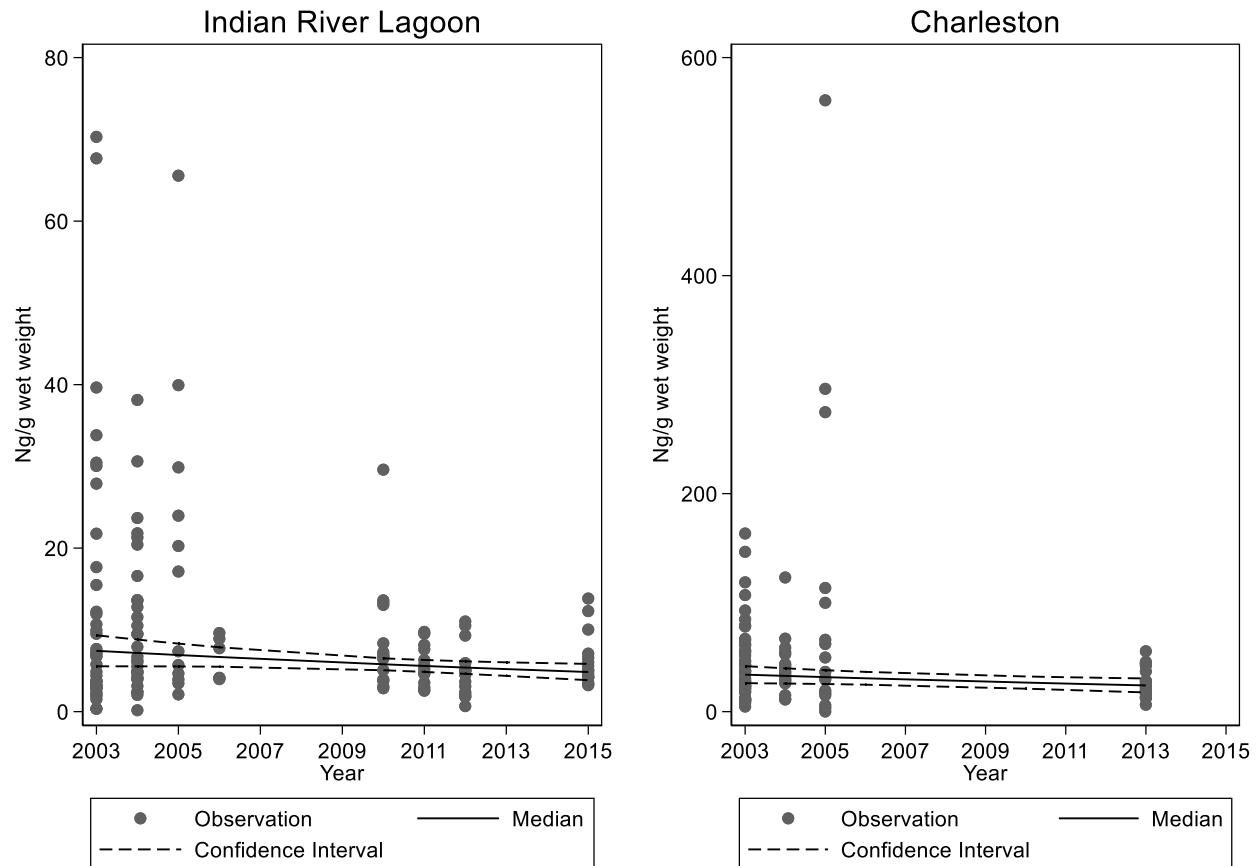
**Table S8.** Temporal trend (% change per year) and 95% confidence intervals (CI) of concentrations of plasma per- and polyfluoroalkyl substances (PFAS) among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, stratified by location for adult dolphins (classified as  $\geq 10$  years old for males and  $\geq 6$  years old for females).

PFAS	Indian River Lagoon, FL			Charleston, SC		
	Trend	95% CI	N	Trend	95% CI	N
<b>PFCAs</b>						
PFHpA	1% decrease	-5, +2	46	21% increase	+7, +37	23
PFOA	1% decrease	-5, +3	85	3% decrease	-6, +1	63
PFNA	2% decrease	-4, +1	85	1% increase	-2, +4	63
PFDA	9% decrease	-11, -7	85	5% decrease	-7, -3	63
PFUnDA	5% decrease	-7, -3	85	6% decrease	-9, -3	63
PFDoDA	1% increase	-2, +5	85	1% increase	-3, +5	63
<b>PFSAs</b>						
PFHxS	6% decrease	-10, -1	85	9% decrease	-12, -6	63
PFOS	2% decrease	-6, +1	85	6% decrease	-9, -4	63
PFDS	16% decrease	-24, -8	46	19% decrease	-24, -14	23

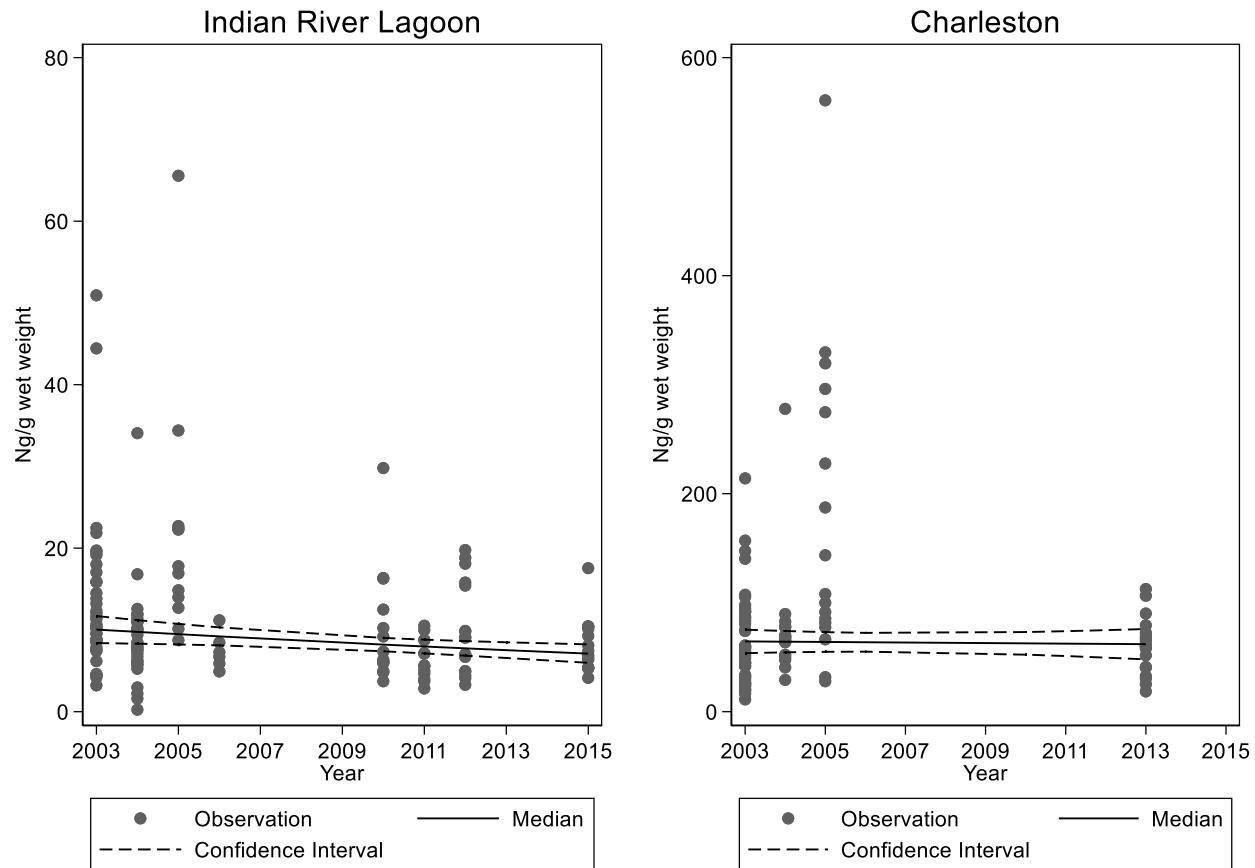
**Figure S1.** Concentrations of PFHpA in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



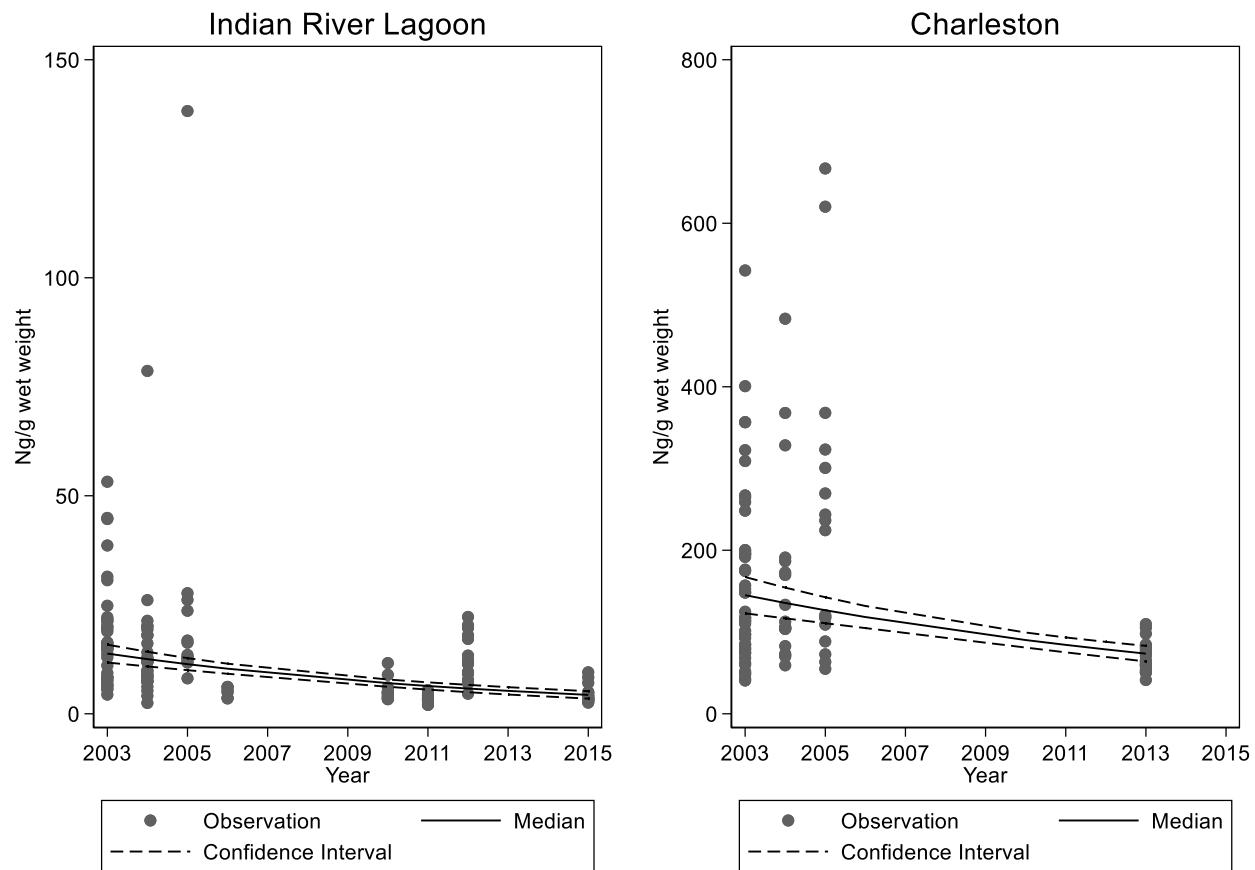
**Figure S2.** Concentrations of PFOA in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



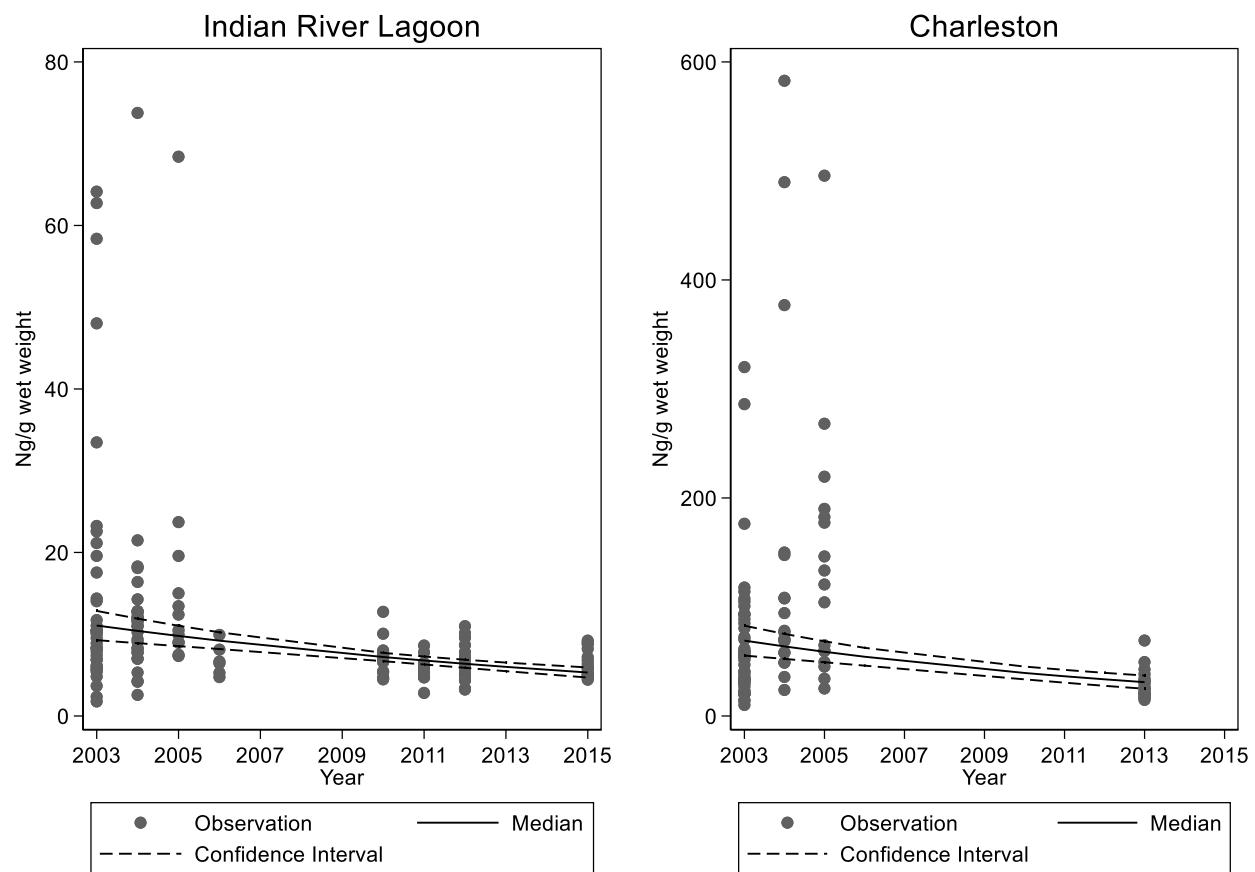
**Figure S3.** Concentrations of PFNA in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



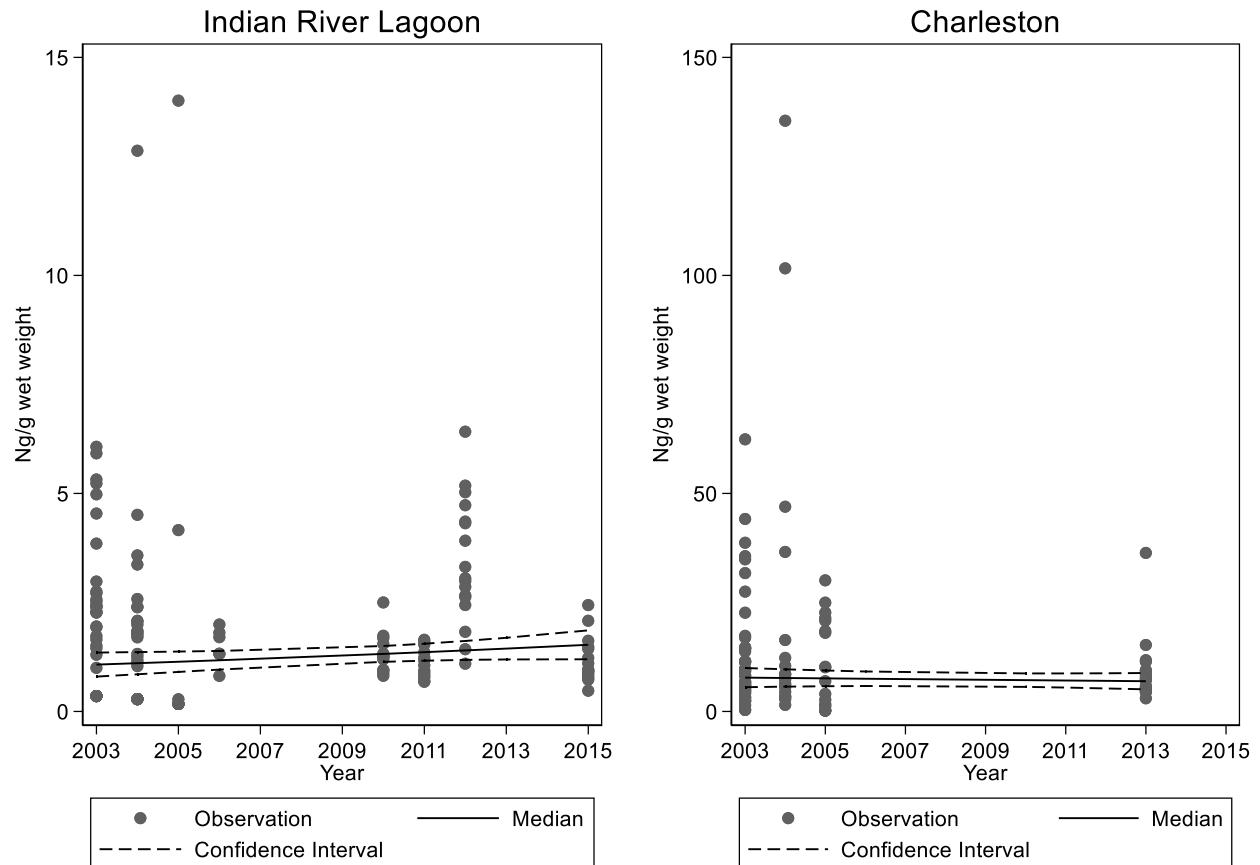
**Figure S4.** Concentrations of PFDA in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



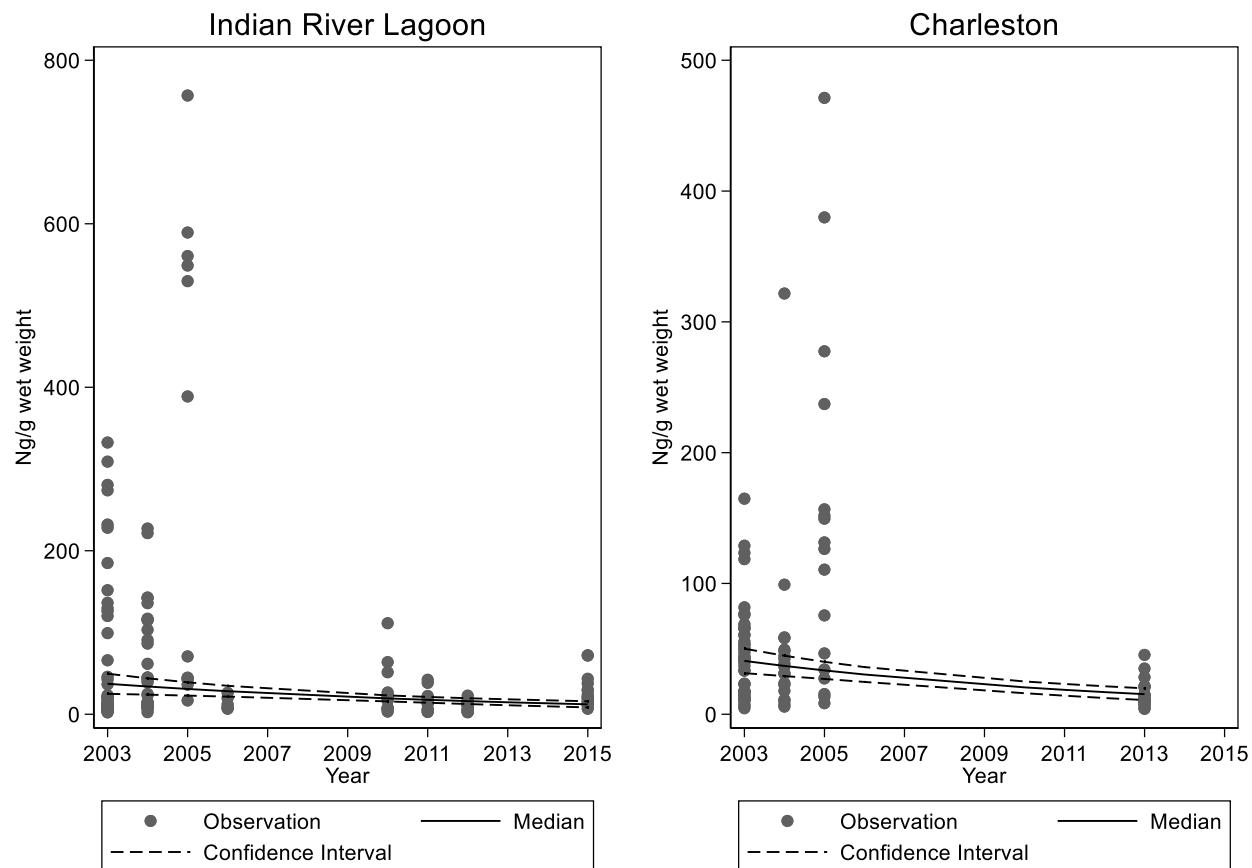
**Figure S5.** Concentrations of PFUnDA in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



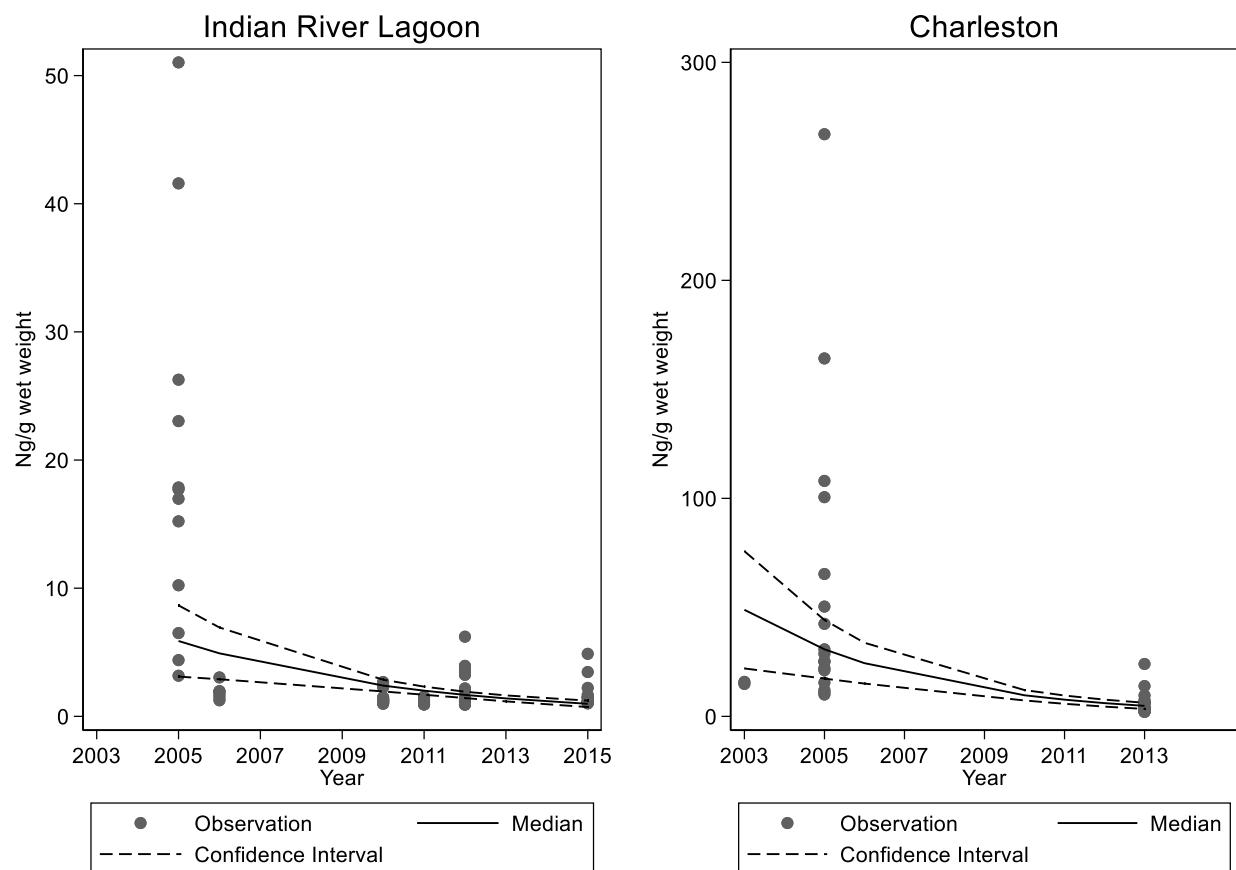
**Figure S6.** Concentrations of PFDoDA in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



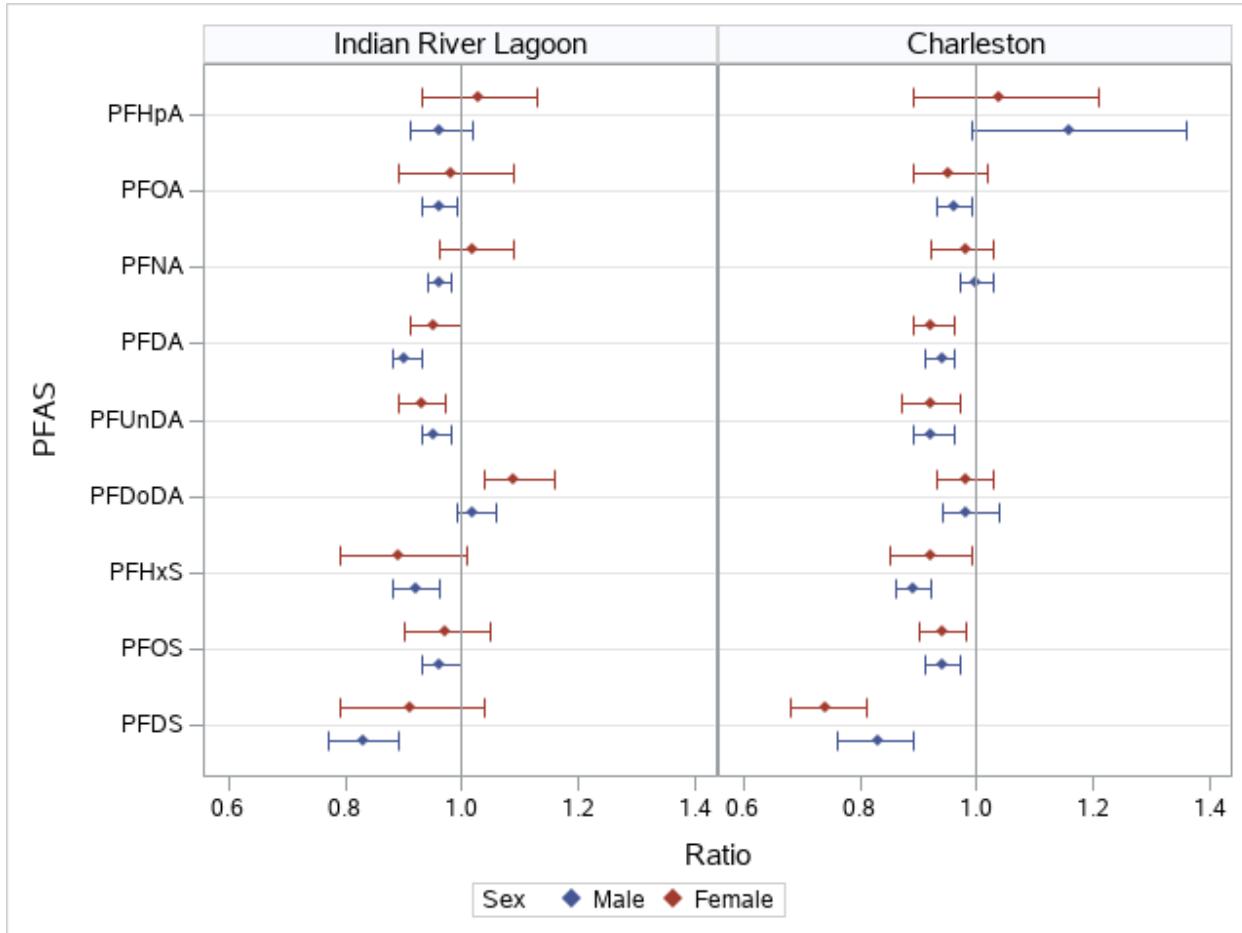
**Figure S7.** Concentrations of PFHxS in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



**Figure S8.** Concentrations of PFDS in plasma, with estimated population medians and 95% confidence intervals, by year, in bottlenose dolphins (*Tursiops truncatus*) from Charleston, SC and Indian River Lagoon, FL.



**Figure S9.** Relative concentrations of plasma per- and polyfluoroalkyl substances (PFAS) per increasing year, with 95% confidence intervals, among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, stratified by location and sex.



**Figure S10.** Relative concentrations of plasma per- and polyfluoroalkyl substances (PFAS) per increasing year with 95% confidence intervals, among bottlenose dolphins (*Tursiops truncatus*) sampled from Charleston, SC and Indian River Lagoon, FL, stratified by location, for adult dolphins (classified as  $\geq 10$  years old for males and  $\geq 6$  years old for females).

