Electronic Supplementary Information for

Spatial and temporal variability of perfluoroalkyl substances in the Laurentian Great Lakes

Christina K. Remucal[1,2]*

[1]Environmental Chemistry and Technology Program

University of Wisconsin – Madison

Madison, Wisconsin

[2]Department of Civil and Environmental Engineering

University of Wisconsin – Madison

Madison, Wisconsin

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Contents (18 pages): Figures: S1 – S16, Tables: S1 – S2

* Corresponding author address: 660 N. Park St., Madison, WI 53706; e-mail: remucal@wisc.edu; telephone: (608) 262-1820; fax: (608) 262-0454; Twitter: @remucal.

Aqueous Concentrations

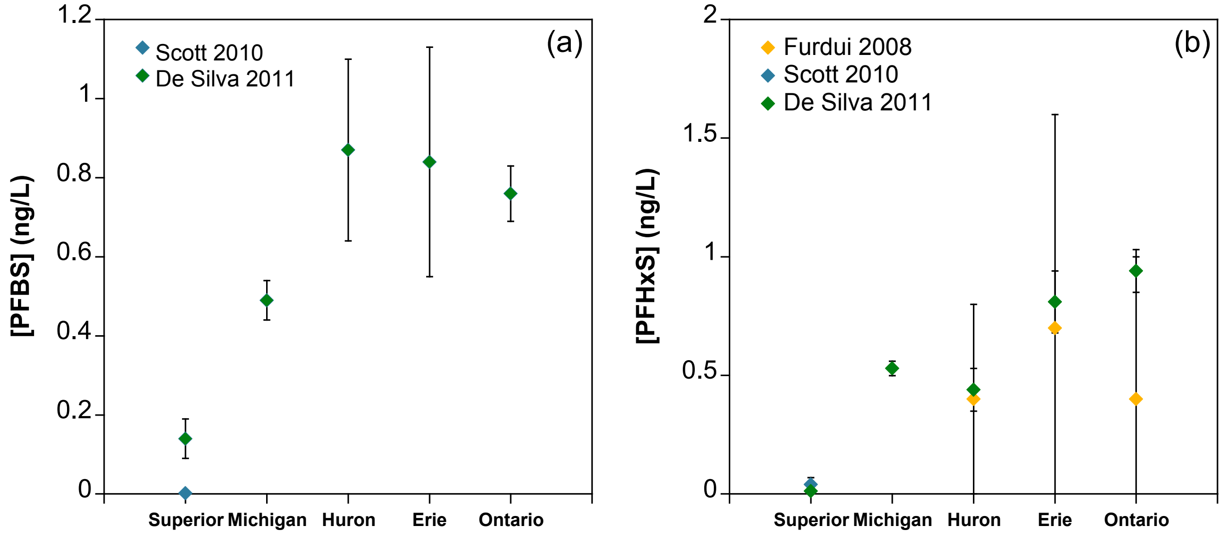

Figure S1. Mean surface (<10 m depth) concentrations of (a) PFBS and (b) PFHxS in the Great Lakes. Points represent the average value and error bars represent the standard deviation of concentrations within each study. Data is compiled from Furdui et al. 2008,[1] Scott et al. 2010,[2] and De Silva et al. 2011.[3]

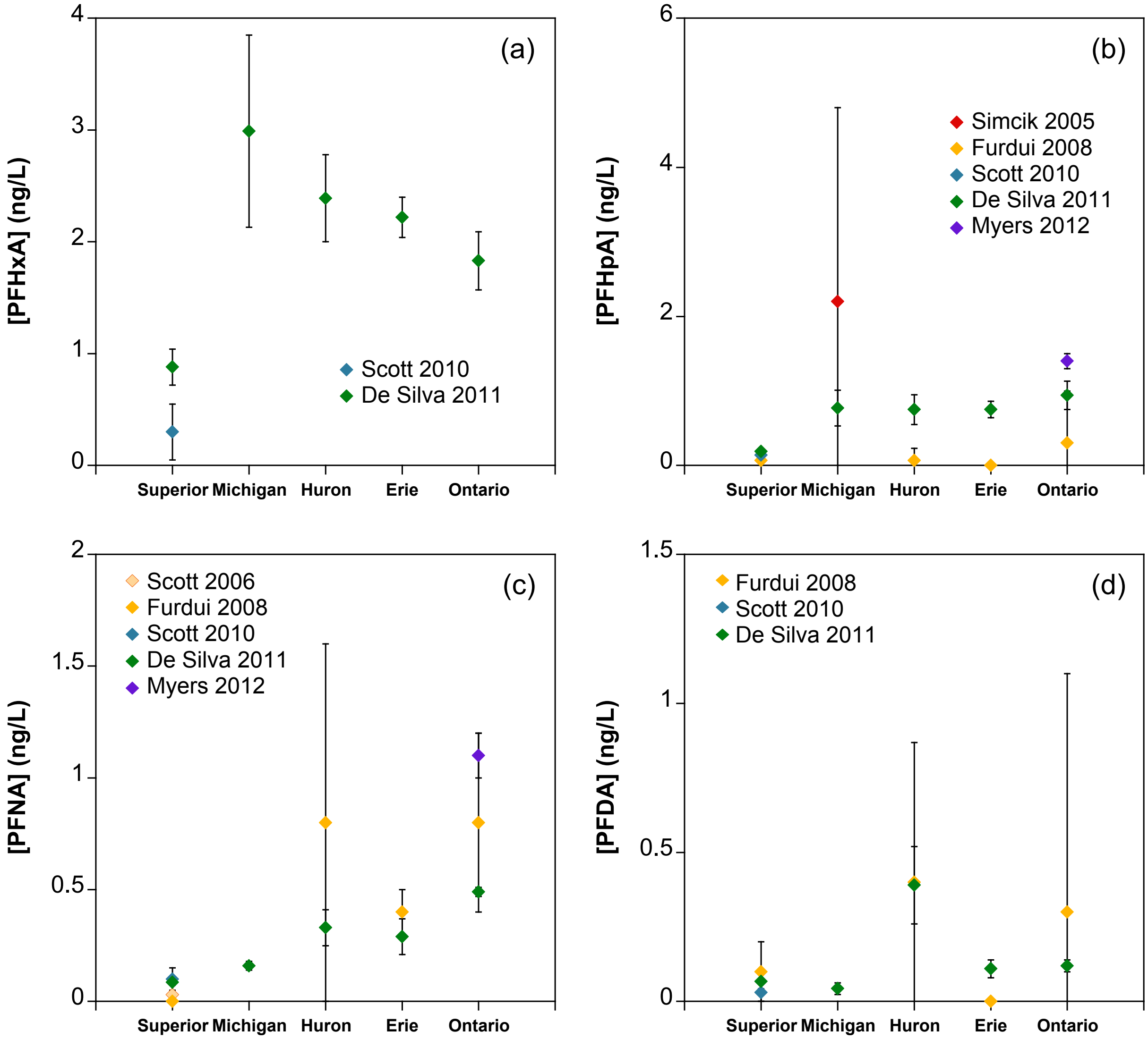

Figure S2. Mean surface (<10 m depth) concentrations of (a) PFHxA, (b) PFHpA, (c) PFNA, and (d) PFDA in the Great Lakes. Points represent the average value and error bars represent the standard deviation of concentrations within each study. Data is compiled from Simcik et al. 2005,[4] Scott et al. 2006,[5] Furdui et al. 2008,[1] Scott et al. 2010,[2] De Silva et al. 2011,[3] and Myers et al. 2012.[6]

Surface Sediment Concentrations

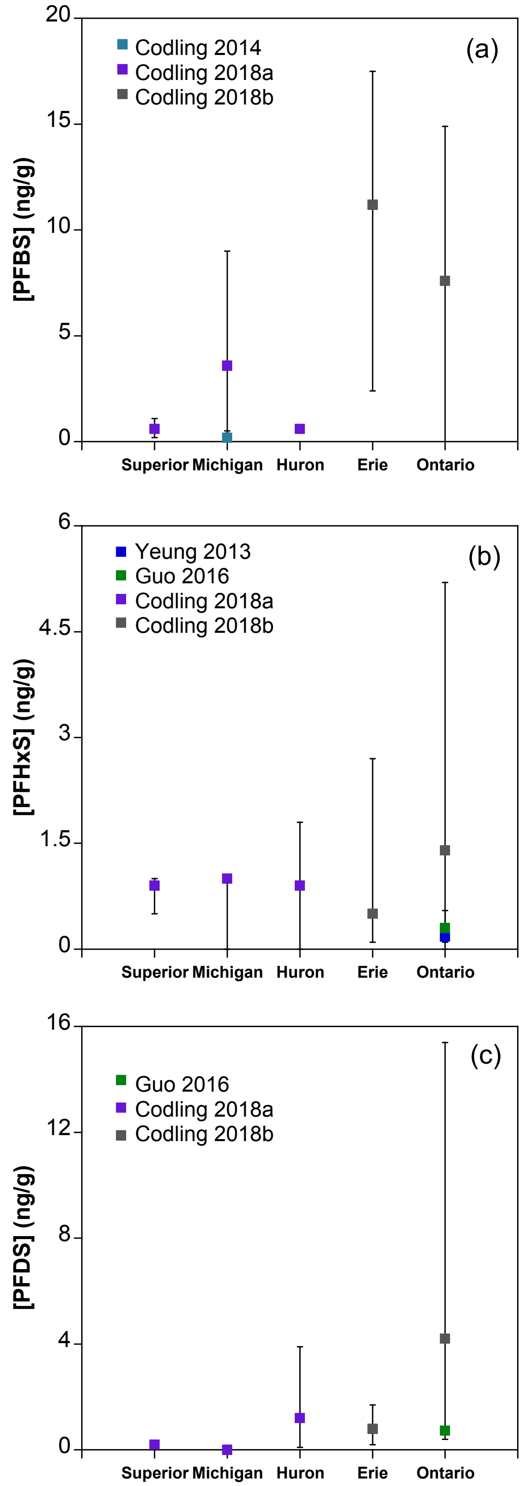

Figure S3. Mean surface sediment concentrations of (a) PFBS, (b) PFHxS, and (c) PFDS in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations within each study. Data is compiled from Yeung et al. 2013,[7] Codling et al. 2014,[8] Guo et al. 2016,[9] Codling et al. 2018a,[10] and Codling et al. 2018b.[11]

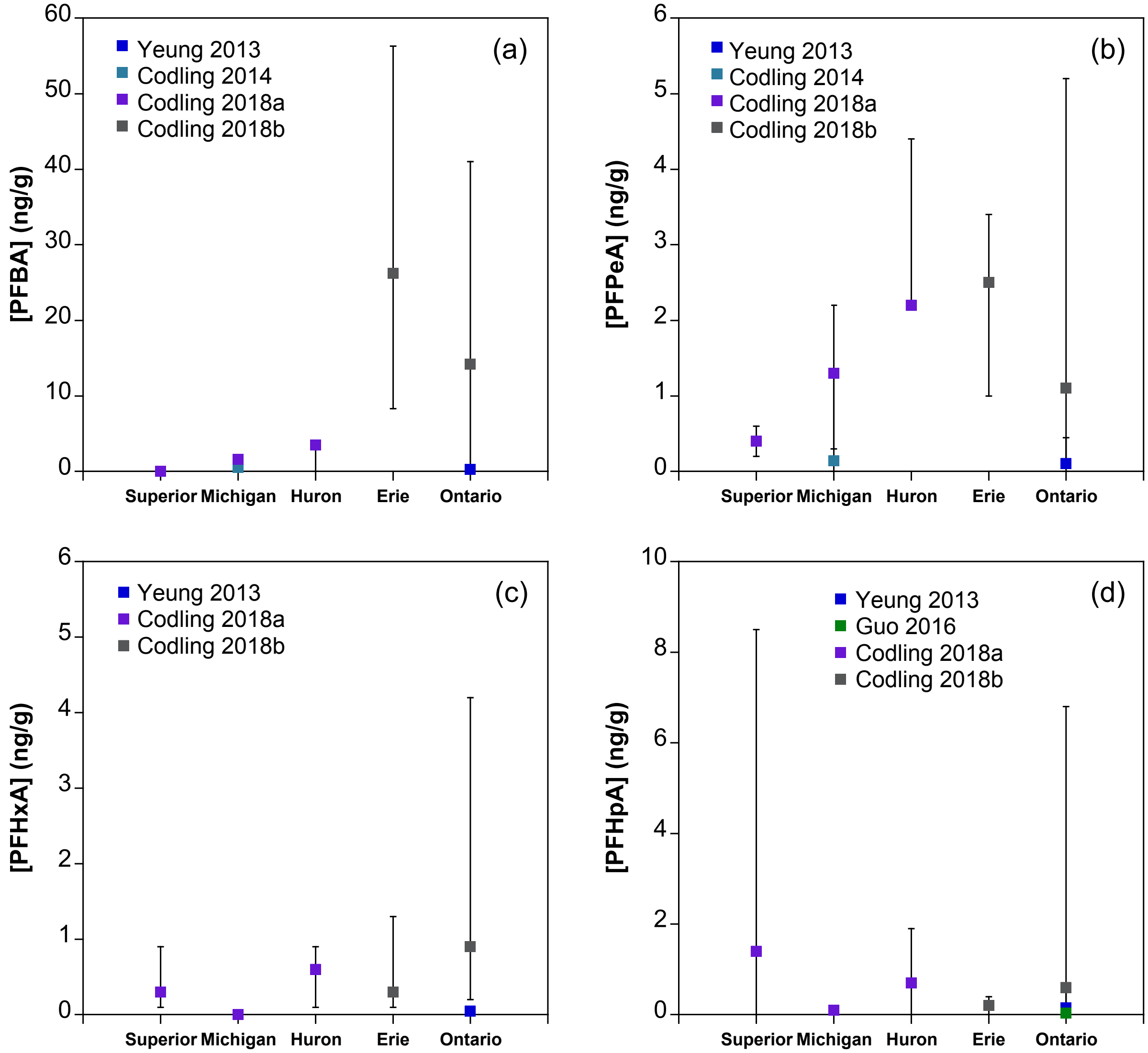

Figure S4. Mean surface sediment concentrations of (a) PFBA, (b) PFPeA, (c) PFHxA, and (d) PFHpA in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations within each study. Data is compiled from Yeung et al. 2013,[7] Codling et al. 2014,[8] Guo et al. 2016,[9] Codling et al. 2018a,[10] and Codling et al. 2018b.[11]

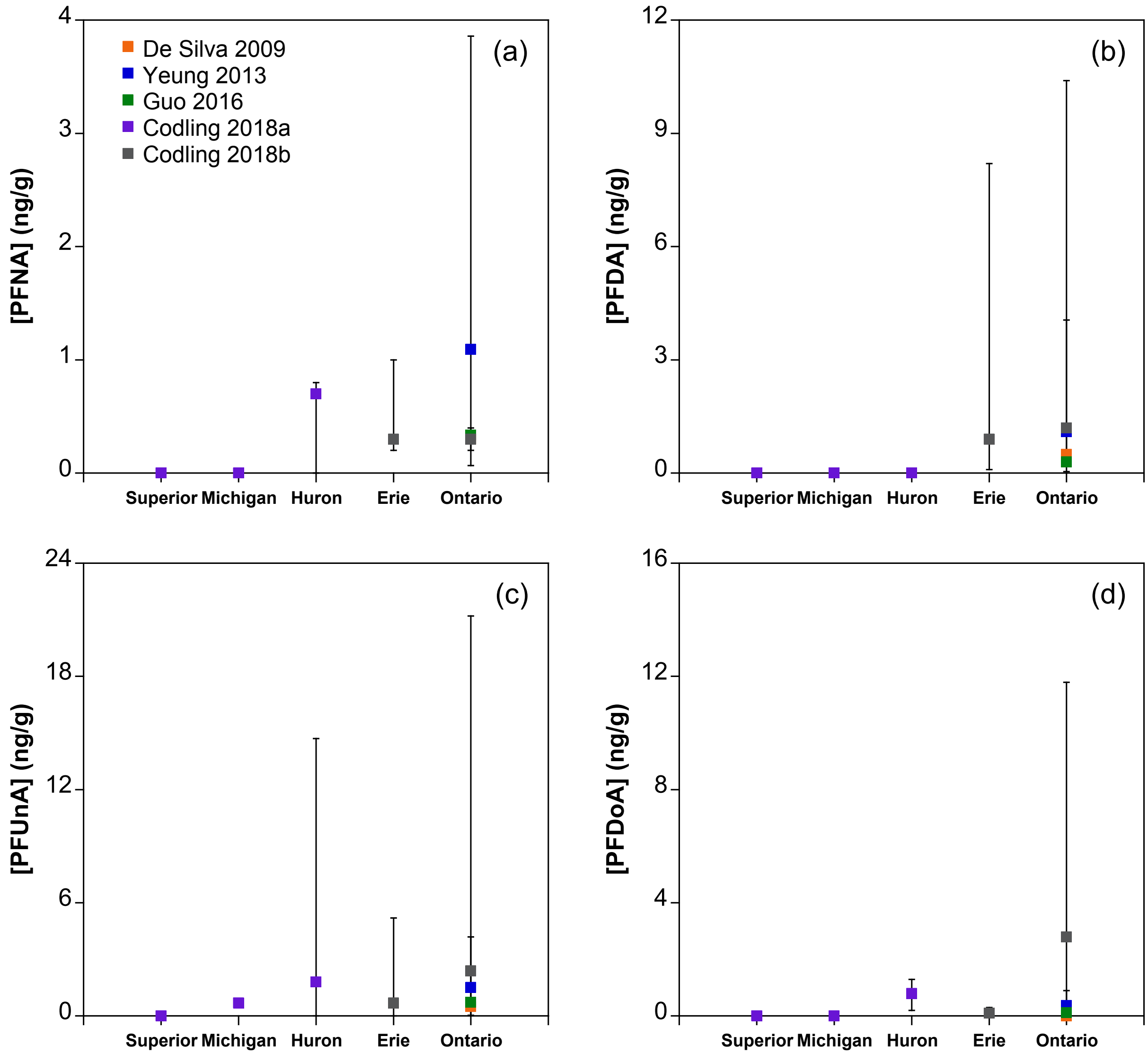

Figure S5. Mean surface sediment concentrations of (a) PFNA, (b) PFDA, (c) PFUnA, and (d) PFDoA in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations within each study. Data is compiled from De Silva et al. 2009,[12] Yeung et al. 2013,[7] Guo et al. 2016,[9] Codling et al. 2018a,[10] and Codling et al. 2018b.[11]

PFAS in Great Lakes Fish

Table S1. Reported K_{BAF} (L/kg) for fish in the Great Lakes and nearby waters (mean ± standard deviation when reported). All values are based on the PFAS concentration in the specified fish tissue divided by the PFAS concentration in the water.

Lake	Organism	PFOA	PFNA	PFDA	PFUnA	PFHxS	PFOS
Superior	Lake trout, whole	41 ± 41,[3] 1,995[13]	8,170 ± 1,870,[3] 5,000[13]	5,760 ± 2,410,[3] 7,940 ± 2,000[13]	N/A	320[13]	9,040 ± 2,210,[3] 19,950[13]
Michigan	Lake trout, whole	2,510[13]	N/A	N/A	N/A	N/A	6,310[13]
Huron	Lake trout, whole	3,980[3]	4,400 ± 1,540,[3] 3,980[13]	3,310 ± 1,593,[3] 5,010[13]	120,900 ± 48,600[3]	2,180 ± 1,200,[3] 2,000[13]	7,690 ± 2,550,[3] 10,000[13]
Erie	Lake trout, whole	10 ± 7,[3] 790[13]	8,900 ± 2,530,[3] 6,310[13]	56,600 ± 17,500,[3] 19,950[13]	N/A	1,710 ± 820,[3] 60[13]	33,700 ± 4,730,[3] 25,100[13]
Ontario	Lake trout, whole	200 ± 80,[3] 400[13]	1,840 ± 240,[3] 1,260[13]	11,750 ± 2,860,[3] 5,000[13]	156,400 ± 42,100[3]	750 ± 126,[3] 1,000[13]	9,400 ± 1,600,[3] 7,900[13]
Erie	Walleye, whole	90 ± 20[3]	4,160 ± 1,210[3]	33,580 ± 9,560[3]	N/A	N/A	18,970 ± 2,650[3]

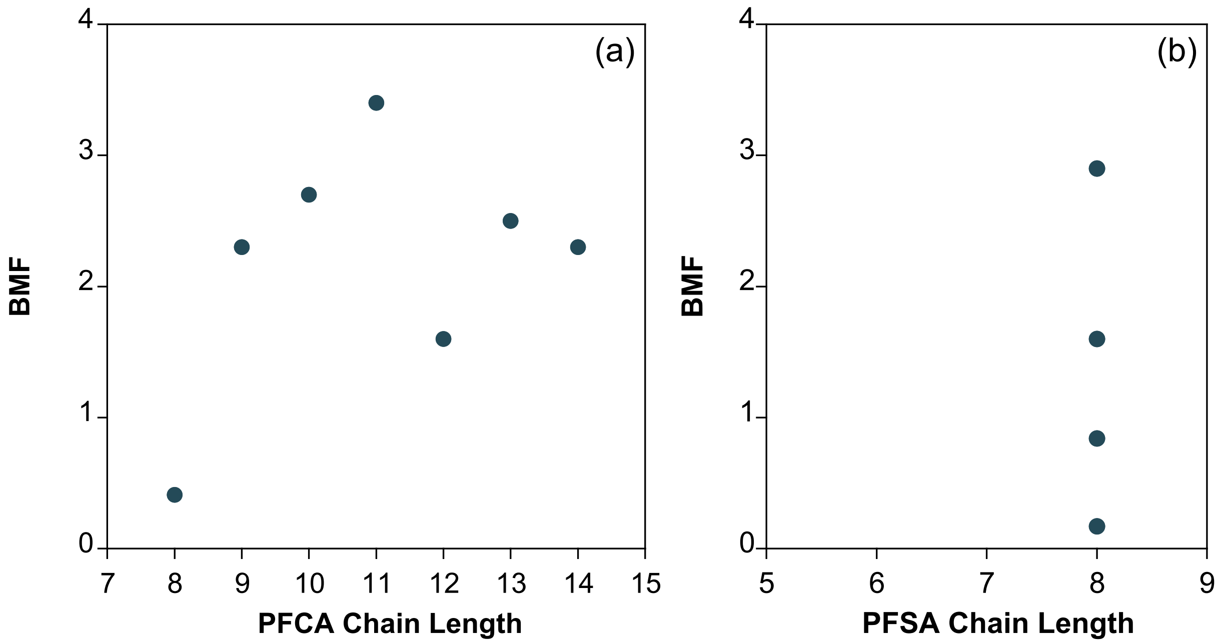

Figure S6. Biomagnification factors calculated for lake trout in Lake Ontario versus chain length for (a) PFCAs[14] and (b) PFSAs.[14, 15] BMF was calculated based on the fish concentration measured in whole fish divided by the concentration measured in their prey. The PFCA values and the highest PFOS value (2.9) are weighted according to different fish species consumed by lake trout,[14] whereas the other PFOS values are lake trout concentrations compared directly to alewife (1.6), smelt (0.8), and sculpin (0.2).[15]

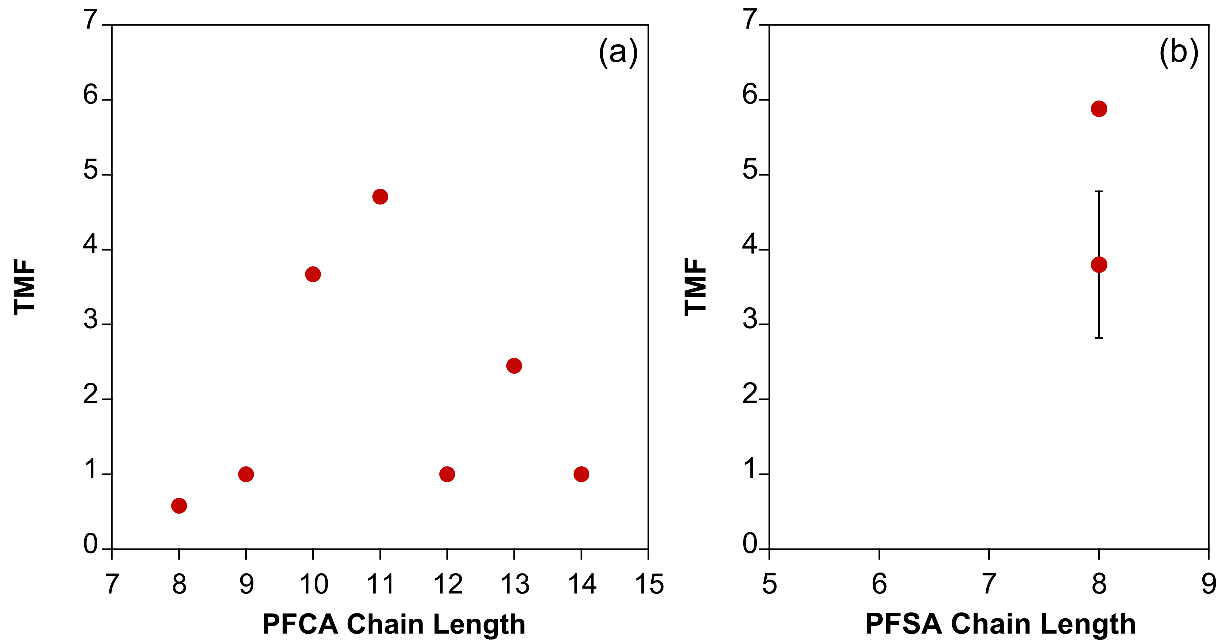

Figure S7. Trophic magnification factors calculated for four species (i.e., *Mysis*, alewife, smelt, and lake trout) in Lake Ontario versus chain length for (a) PFCAs and (b) PFSAs.[14, 15]

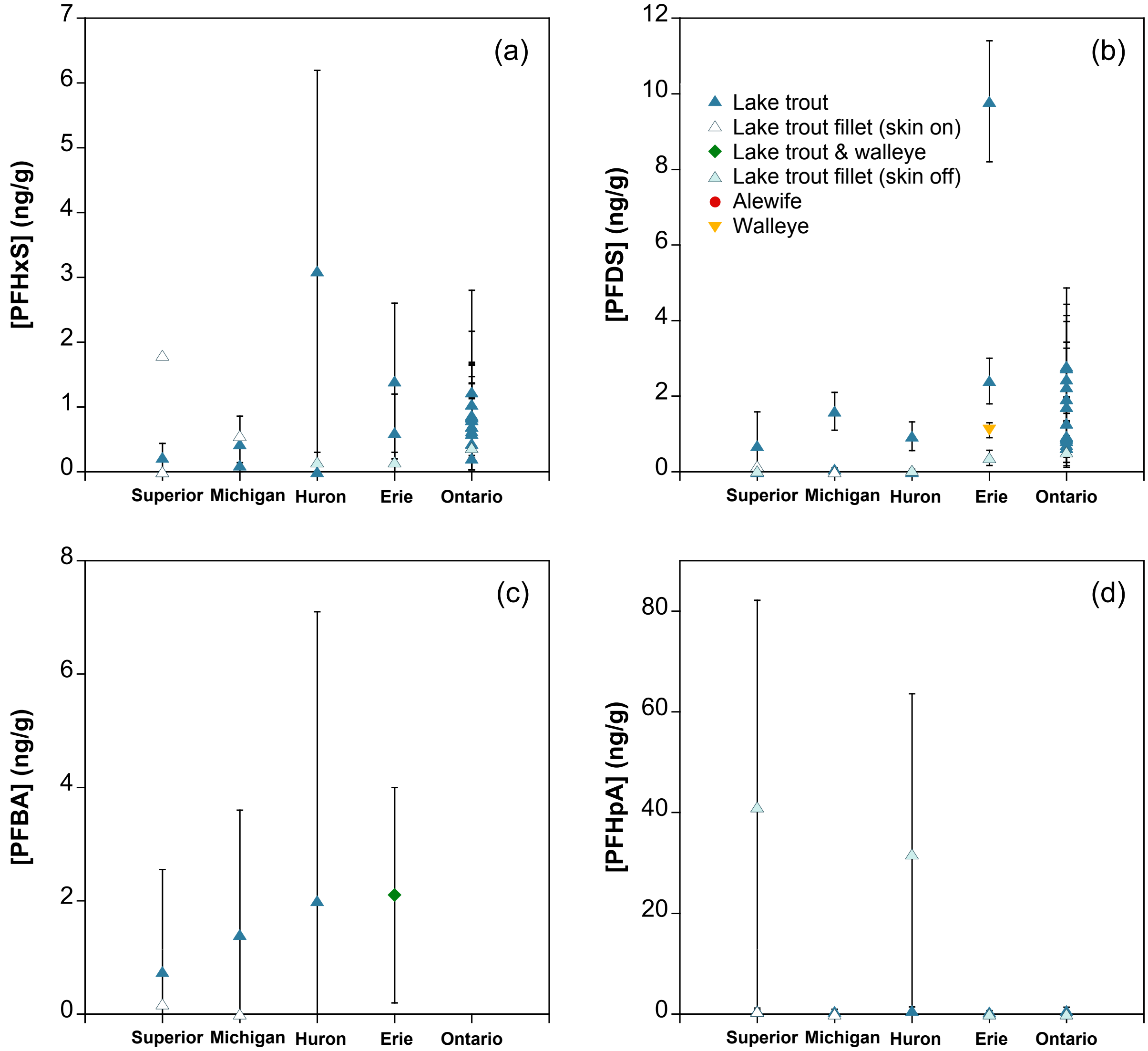

Figure S8. Mean concentrations of (a) PFHxS, (b) PFDS, (c) PFBA, and (d) PFHpA in fish in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations within each study. Data is presented for whole lake trout,[3, 13, 16, 17] skin on lake trout fillets,[18] skin off lake trout fillets,[19] and whole walleye.[3]

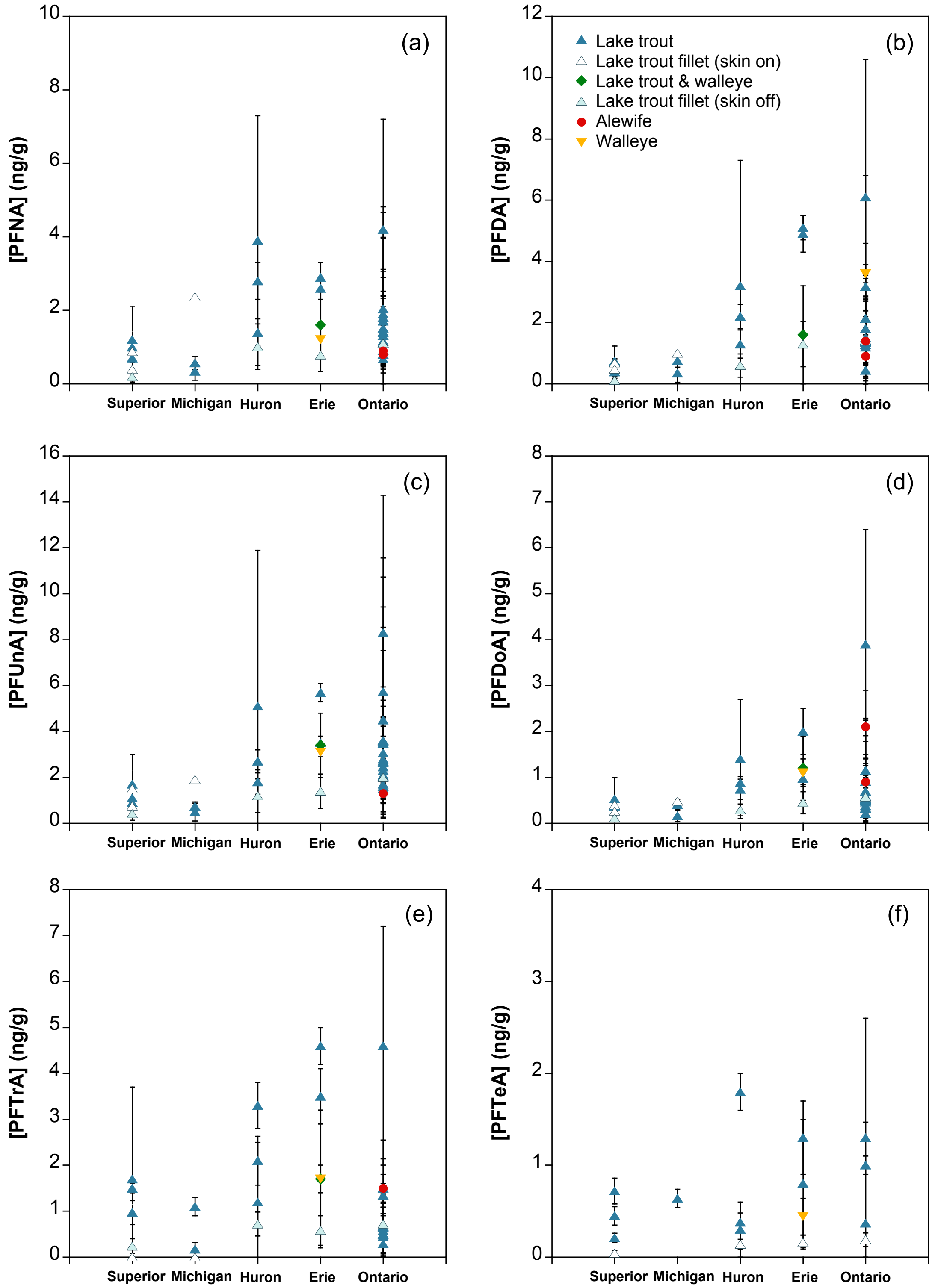

Figure S9. Mean concentrations of (a) PFNA, (b) PFDA, (c) PFUnA, (d) PFDoA, (e) PFTrA, and (f) PFTeA in fish in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations within each study. Data is presented for whole lake trout,[3, 13-17] skin on lake trout fillets,[18] combined concentrations for whole lake trout and walleye,[16] skin off lake trout fillets,[19] whole alewife,[14, 15] and whole walleye.[3]

PFAS in Herring Gulls and Other Terrestrial Biota

Table S2. Reported concentrations of PFAS in less frequently studied biota in or near Lakes Superior (SU), Michigan (MI), Huron (HU), Erie (ER), or Ontario (ON). Values in parentheses are the range of concentrations. Values outside of parentheses correspond to study means ± the standard deviation when available.

Compound	Invertebrates	Amphibians	Reptiles	Birds	Mammals
PFSAs					
PFHxS	N/A	Frog liver (HU): <LOD[20]	Snapping turtle plasma (HU): <LOD[20]	Bald eagle plasma (SU): 2.0 (<LOD – 8.6) ng/mL;[21] tree swallow plasma (SU): <LOD[22]	Mink liver (HU): (6.3 – 40) ng/g[20]
PFOS	*Mysis* (ON): (1.8 – 13) ng/g;[12, 14 23] *Diporeia* (ON): (21 – 280) ng/g;[14, 15, 23] zooplankton (ON): (1.4 – 6.5) ng/g[15, 23]	Frog liver (HU): (<LOD – 285) ng/g[20]	Snapping turtle plasma (HU): (<LOD – 169) ng/mL[20]	Bald eagle liver (MI): 394 (27 – 1,740) ng/g;[20] bald eagle muscle (MI): 32 (<LOD – 96) ng/g;[20] bald eagle plasma (SU): 345 (71 – 830) ng/mL;[21] cormorant plasma (HU): 215 (1 – 430) ng/mL;[24] tree swallow plasma (SU): 13.1 (5.6 – 30) ng/g[22]	Mink liver (HU): (64 – 4,870) ng/g[20]
PFDS	N/A	N/A	N/A	Bald eagle plasma (SU): 13.7 (<LOD – 100) ng/mL[21]	N/A
PFCAs					
PFBA	N/A	N/A	N/A	Bald eagle plasma (SU): 0.4 (<LOD – 22) ng/mL[21]	N/A
PFOA	*Mysis* (ON): (2.5 – 3.9) ng/g;[12, 14] *Diporeia* (ON): (67 – 90) ng/g;[14 12] zooplankton (ON): 1.3 ng/g[12]	Frog liver (HU): <LOD[20]	Snapping turtle plasma (HU): <LOD[20]	Bald eagle plasma (SU): 0.8 (<LOD – 14) ng/mL;[21] tree swallow plasma (SU): 11.9 (1.9 – 31.7) ng/g[22]	Mink liver (HU): (<LOD – 12) ng/g[20]
PFNA	*Mysis* (ON): (2.7 – 2.8) ng/g;[12, 14] *Diporeia* (ON): (37 – 57) ng/g;[12, 14] zooplankton (ON): 0.4 ng/g[12]	N/A	N/A	Bald eagle plasma (SU): 6.5 (4.9 – 160) ng/mL;[21] tree swallow plasma (SU): 9.1 (2.2 – 20.8) ng/g[22]	N/A
PFDA	*Mysis* (ON): (1.2 – 1.3) ng/g;[12, 14] *Diporeia* (ON): (16 – 32) ng/g;[12, 14] zooplankton (ON): 0.4 ng/g[12]	N/A	N/A	Bald eagle plasma (SU): 12 (0.1 – 77) ng/mL;[21] tree swallow plasma (SU): 1.7 (0.5 – 2.9) ng/g[22]	N/A

PFUnA	*Mysis* (ON): (0.8 – 1.3) ng/g;[12, 14] *Diporeia* (ON): (18 – 41) ng/g;[12, 14] zooplankton (ON): (0.2 – 0.8) ng/g[12]	N/A	N/A	Bald eagle plasma (SU): 14.7 (10.9 – 110) ng/mL;[21] tree swallow plasma (SU): 3.4 (1.3 – 5.5) ng/g[22]	N/A
PFDoA	*Mysis* (ON): (0.4 – 1.8) ng/g;[12, 14] *Diporeia* (ON): (5 – 14) ng/g;[12, 14] zooplankton (ON): 0.3 ng/g[12]	N/A	N/A	Bald eagle plasma (SU): 6.4 (2.0 – 27) ng/mL; [21] tree swallow plasma (SU): (<LOD – 0.9) ng/g[22]	N/A
PFTrDA	*Mysis* (ON): 1.5 ± 0.2 ng/g;[14] *Diporeia* (ON): 15 ± 0.8 ng/g[14]	N/A	N/A	Bald eagle plasma (SU): 3.5 (3.0 – 63) ng/mL[21]	N/A
PFTeDA	*Mysis* (ON): <LOD;[14] *Diporeia* (ON): 7.3 ± 0.6 ng/g[14]	N/A	N/A	Bald eagle plasma (SU): 1.4 (0.4 – 19) ng/mL[21]	N/A

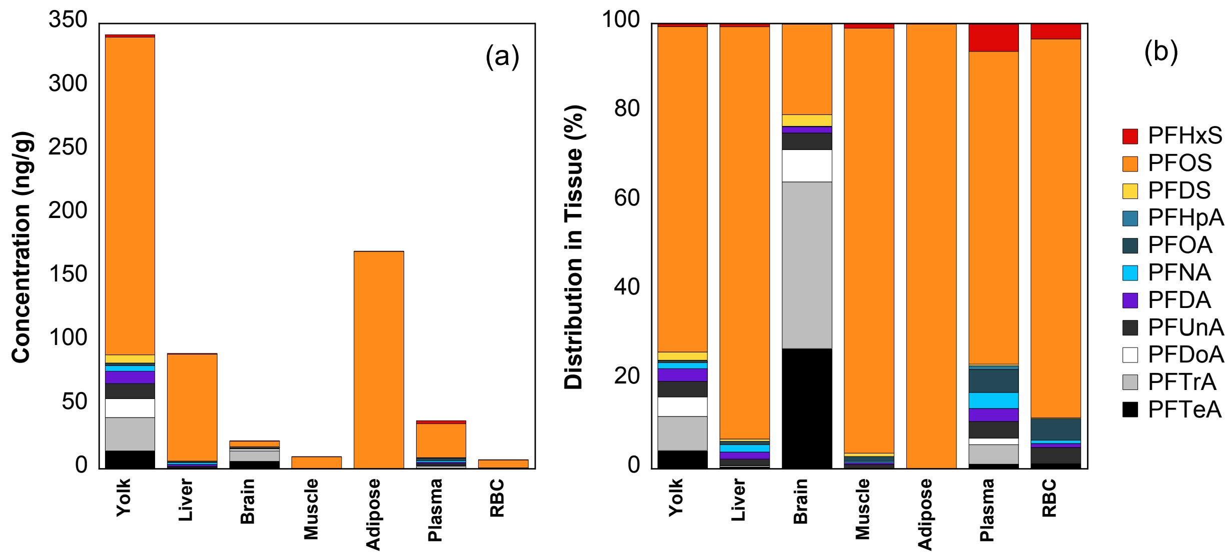

Figure S10. (a) Concentration and (b) percent distribution of commonly studied PFAS in female herring gull eggs and tissues collected in 2010 from Chantry Island, Lake Huron.[25]

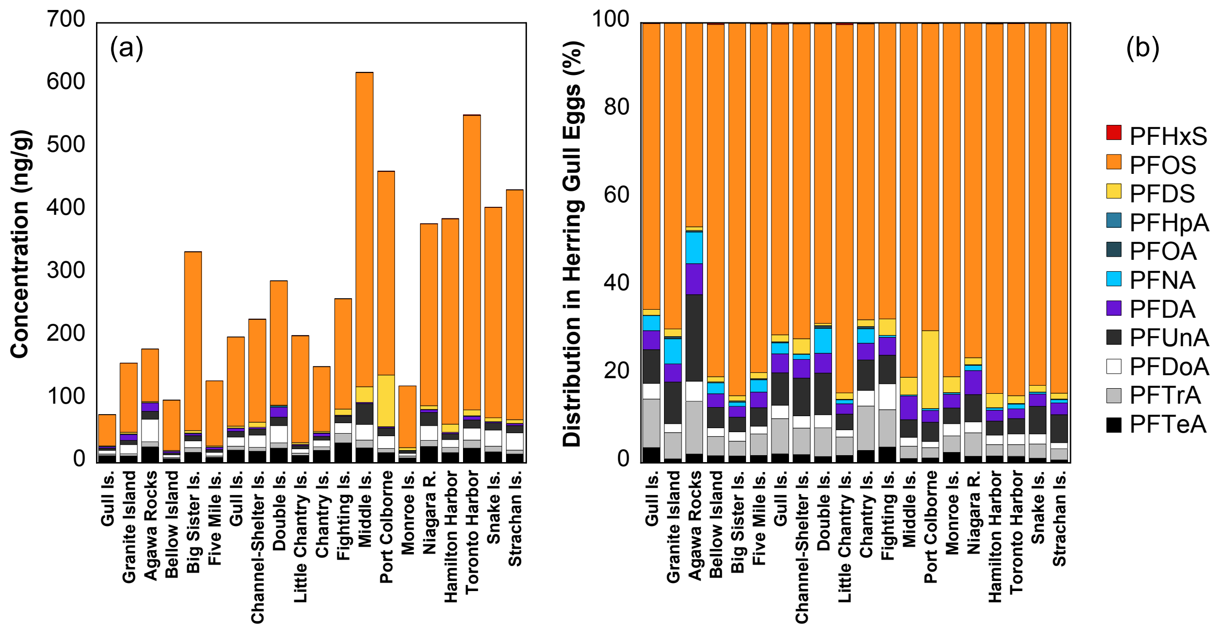

Figure S11. (a) Concentration and (b) percent distribution of commonly studied PFAS in herring gull eggs collected from colonies in the Great Lakes determined based on mean concentrations calculated across all studies shown in Figures 9, S12, and S13.[26-29] Colonies are arranged from west to east and include three riverine colonies (i.e., Detroit River, Niagara River, and Strachan Island).

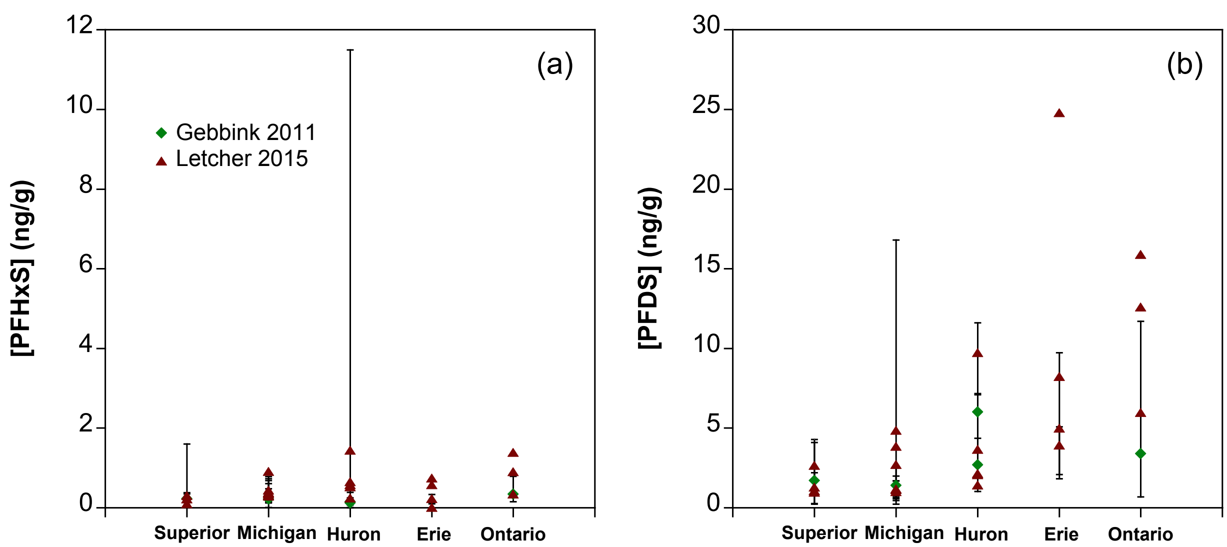

Figure S12. Mean concentrations of (a) PFHxS and (b) PFDS in herring gull eggs collected from 17 colonies in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations within each study.[27, 28] Data is compiled from Gebbink et al. 2011[27] and Letcher et al. 2015.[28]

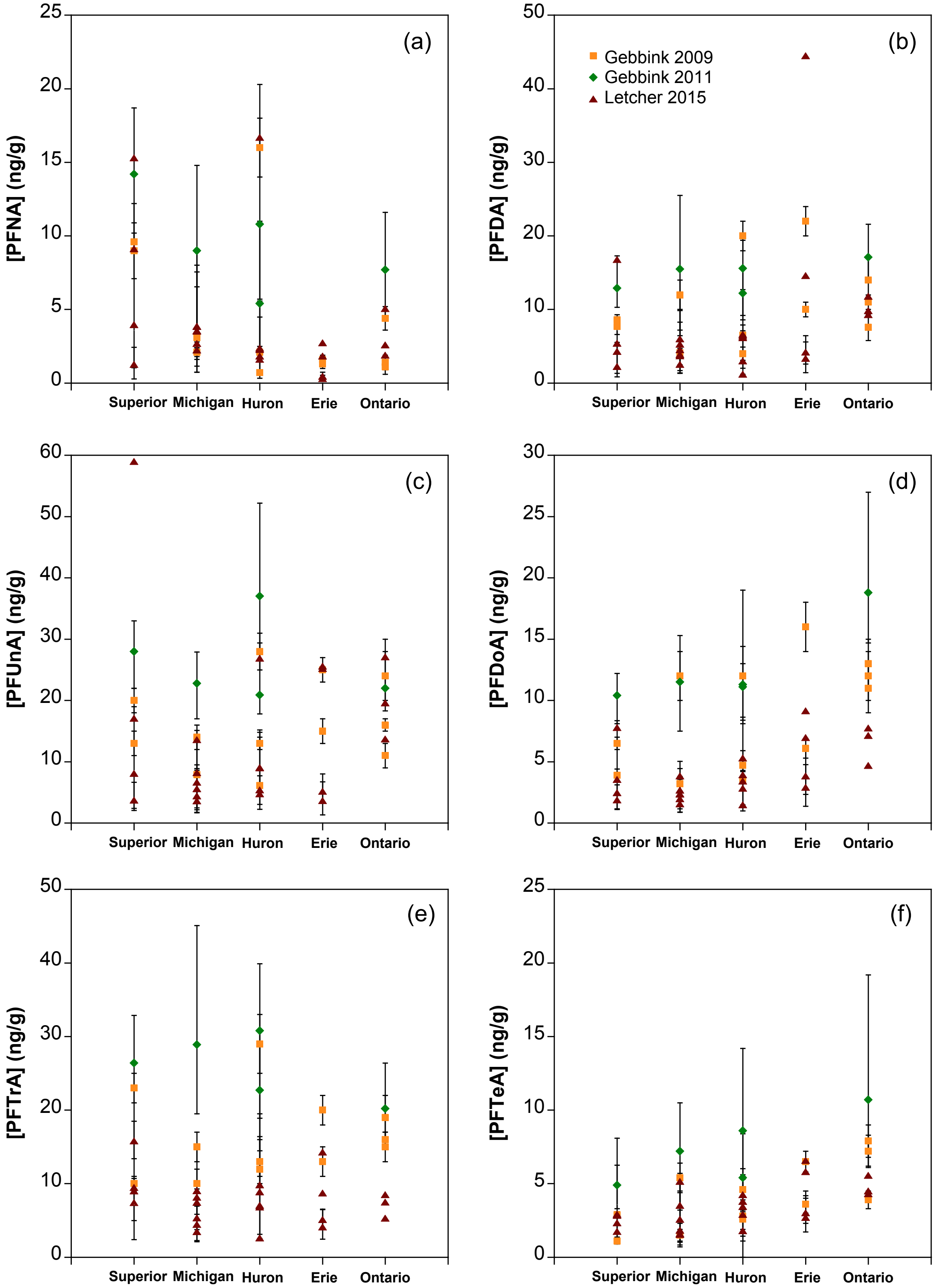

Figure S13. Mean concentrations of (a) PFNA, (b) PFDA, (c) PFUnA, (d) PFDoA, (e), PFTrA, and (f) PFTeA in herring gull eggs collected from 17 colonies in the Great Lakes. Points represent the average value and error bars represent the reported range of concentrations[27, 28] or standard deviation[29] within each study. Data is compiled from Gebbink et al. 2009,[29] Gebbink et al. 2011,[27] and Letcher et al. 2015.[28]

Temporal Trends in PFAS

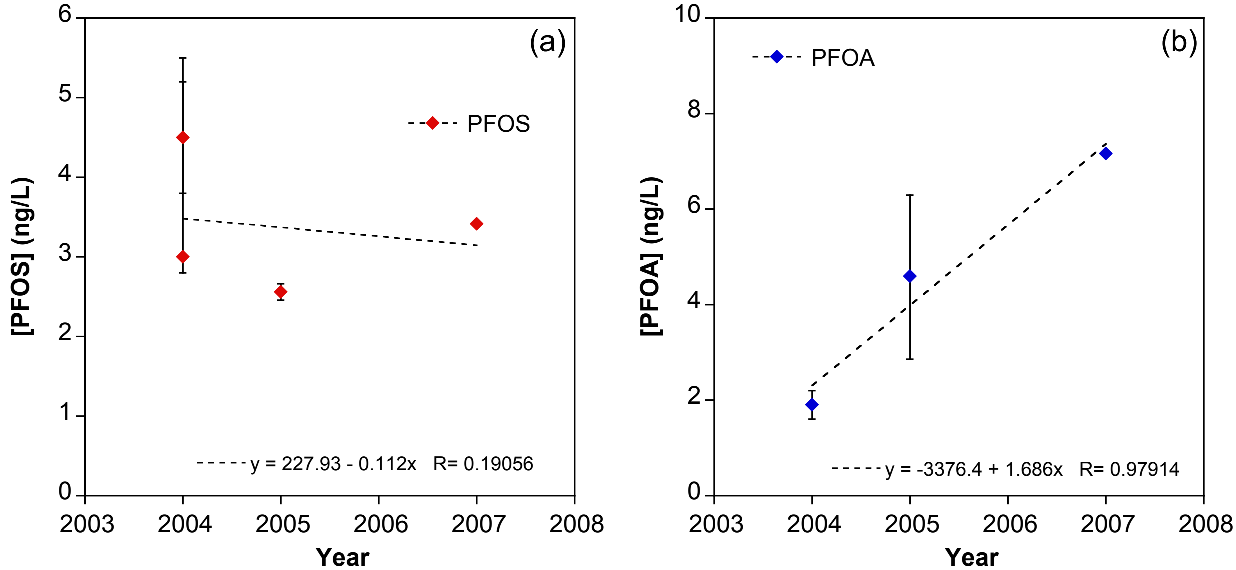

Figure S14. Temporal variability in the average surface concentrations of (a) PFOS and (b) PFOA in Lake Erie. Error bars correspond to reported standard deviations and are not available for all data points.[1, 3, 30]

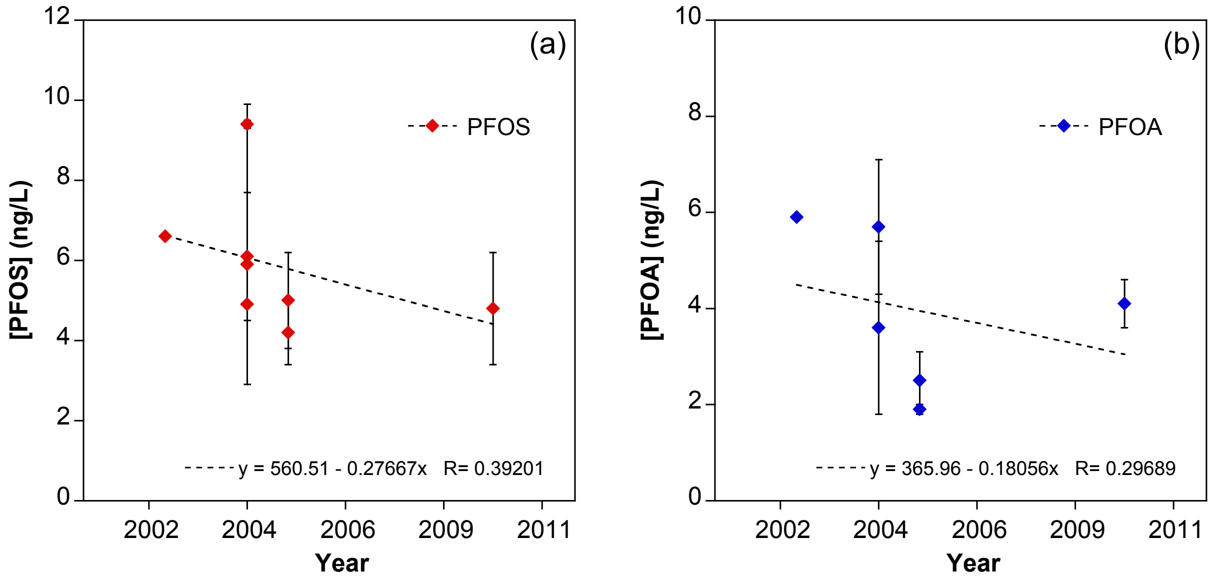

Figure S15. Temporal variability in the average surface concentrations of (a) PFOS and (b) PFOA in Lake Ontario. Error bars correspond to reported standard deviations and are not available for all data points.[1, 3, 6, 15, 30]

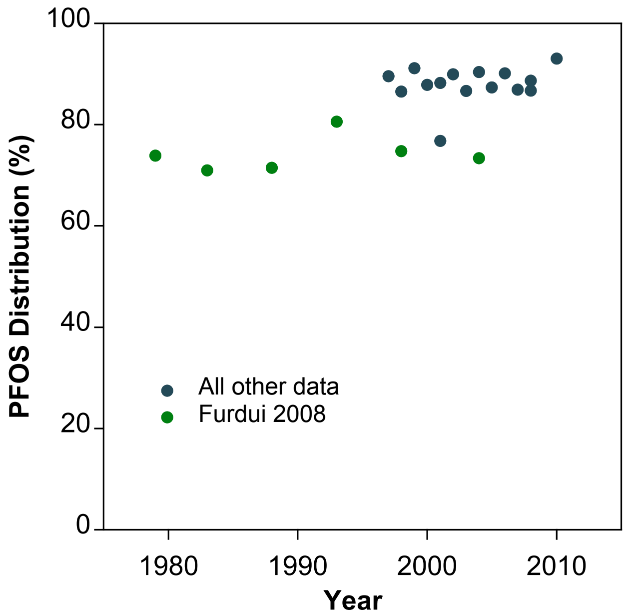

Figure S16. Percentage of total PFAS present at PFOS in whole lake trout collected in Lake Ontario. Data from Furdui (hollow)[31] was estimated from figures. All other data was summarized from tabulated data.[3, 13, 16, 17] Note that the 2010 data point was averaged for samples collected in 2008 – 2012.[16]

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