



# **Chemical Mixtures in Freshwater Fish in the US and the Risks They Pose for Fish Consumption**

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School of Public Health

**Celia Chen**, Department of Biological Sciences, Dartmouth College



## **The Problem:**

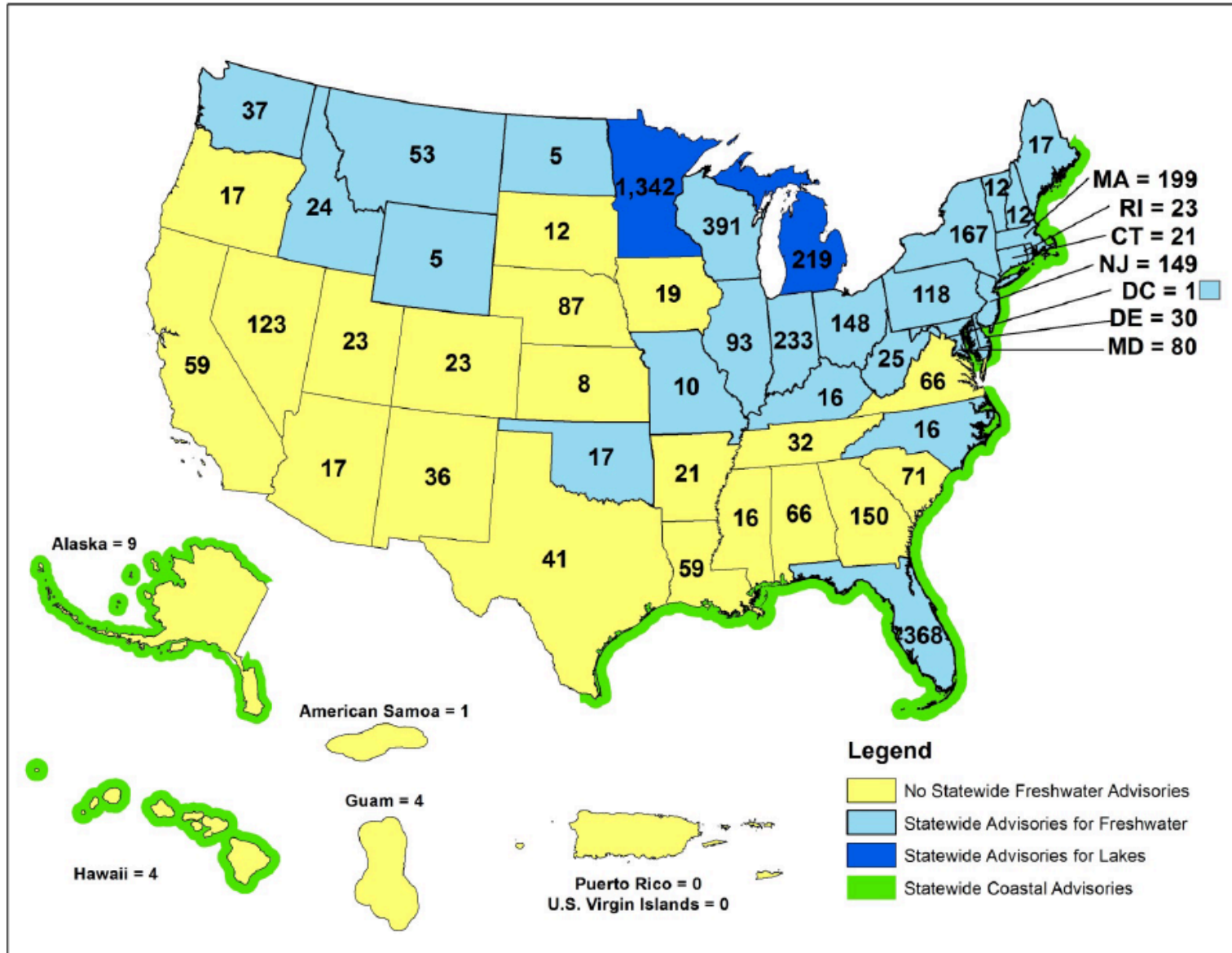
**Eating fish results in exposure to multiple contaminants, not just mercury!**

## **The Overarching Questions:**

**Which fish contaminants co-occur and should we be most concerned about mercury?**

**Is it safe to consume wild-caught freshwater fish?**

# US Fish Consumption Advisories (2011)

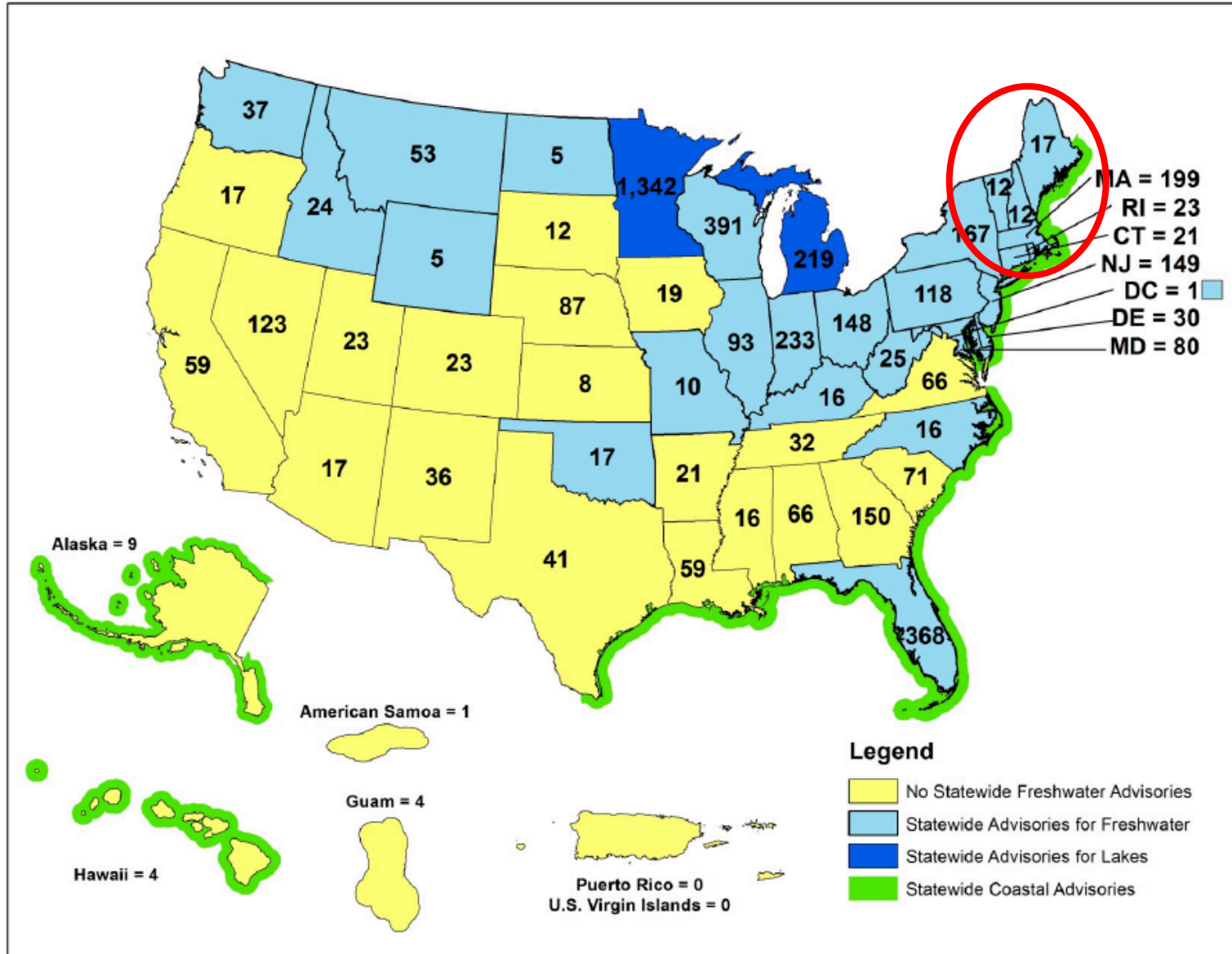


**Table 2. Summary of Statewide Advisories by Waterbody Type and Year Issued**

State	Lake	Issued	River	Issued	Coastal Waters	Issued
Alabama					Mercury	1996
Connecticut	Mercury	1996	Mercury	1996	PCBs	1993
Dist. of Columbia	PCBs	1993	PCBs	1993		
Florida	Mercury	2002	Mercury	2002	Mercury	1993
Georgia					Mercury	2000
Hawaii					Mercury*	2003
Illinois	Mercury	2002	Mercury	2002		
Indiana			Mercury PCBs	1996		
Kentucky	Mercury	2000	Mercury	2000		
Louisiana					Mercury	1997
Maine	Mercury	1994	Mercury	1994	Dioxins Mercury PCBs	1994
Maryland	Mercury PCBs Organo-chlorine pesticides	2002	Mercury PCBs Organo-chlorine pesticides	2002		
Massachusetts	Mercury	1996	Mercury	1996	PCBs Mercury	1994
Michigan	Mercury	1993				
Minnesota	Mercury	1999				
Mississippi					Mercury	1998
Missouri	Mercury	2001	Mercury	2001		
Montana	Mercury	2003	Mercury	2003		
New Hampshire	Mercury	1995	Mercury	1995	PCBs	1994
New Jersey	Mercury	1995	Mercury	1995	PCBs Cadmium Dioxins	1993
New York	PCBs Chlordane Mirex DDT	1994	PCBs Chlordane Mirex DDT	1994	Cadmium Dioxins PCBs	1995
North Carolina					Mercury	2000
North Dakota	Mercury	2001	Mercury	2001		
Ohio	Mercury	1997	Mercury	1997		
Pennsylvania	Mercury	2001	Mercury	2001		
Rhode Island	Mercury	2002	Mercury	2002	PCBs Mercury	1993
South Carolina					Mercury	2001
Texas					Mercury	1997
Vermont	Mercury	1995	Mercury	1995		
Washington	Mercury	2003	Mercury	2003		
Wisconsin	Mercury	2000	Mercury	2003		

\*Hawaii has a statewide advisory for mercury in marine fish.

# US Fish Consumption Advisories (2011)



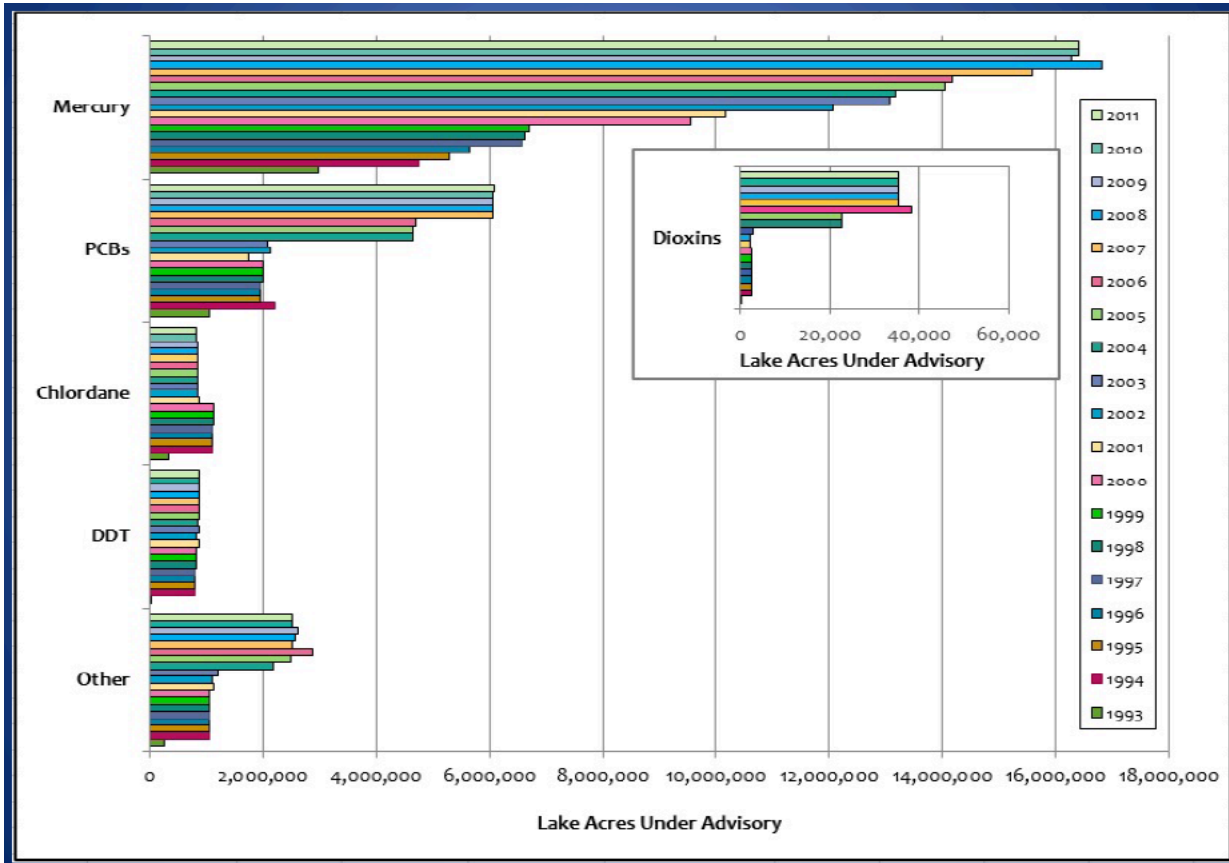
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Maine	Mercury	1994	Mercury	1994	Dioxins Mercury PCBs	1994
Maryland	Mercury PCBs Organo-chlorine pesticides	2002	Mercury PCBs Organo-chlorine pesticides	2002		
Massachusetts	Mercury	1996	Mercury	1996	PCBs Mercury	1994
Michigan	Mercury	1993				
Minnesota	Mercury	1999				
Mississippi					Mercury	1998
Missouri	Mercury	2001	Mercury	2001		
Montana	Mercury	2003	Mercury	2003		
New Hampshire	Mercury	1995	Mercury	1995	PCBs	1994
New Jersey	Mercury	1995	Mercury	1995	PCBs Cadmium Dioxins	1993
New York	PCBs Chlordane Mirex DDT	1994	PCBs Chlordane Mirex DDT	1994	Cadmium Dioxins PCBs	1995
North Carolina					Mercury	2000
North Dakota	Mercury	2001	Mercury	2001		
Ohio	Mercury	1997	Mercury	1997		
Pennsylvania	Mercury	2001	Mercury	2001		
Rhode Island	Mercury	2002	Mercury	2002	PCBs Mercury	1993
South Carolina					Mercury	2001
Texas					Mercury	1997
Vermont	Mercury	1995	Mercury	1995		
Washington	Mercury	2003	Mercury	2003		
Wisconsin	Mercury	2000	Mercury	2003		

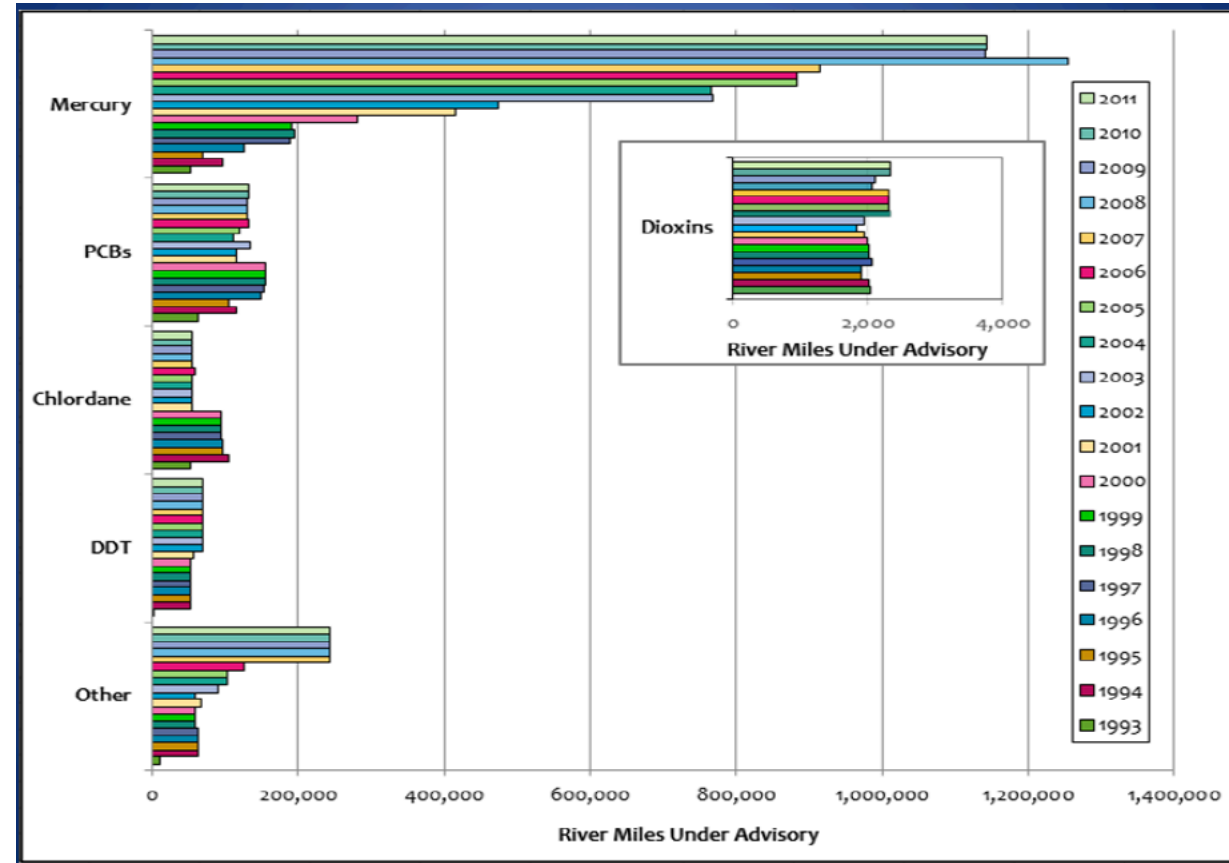
\*Hawaii has a statewide advisory for mercury in marine fish.

# Fish Consumption Advisories for Various Pollutants, 1993-2011

## Lake Acres



## River Miles



# July 2024 USEPA Recommendations

## EPA Releases New Science-Based Recommendations to Help More States, Tribes, and Territories Reduce Exposure to PFAS in Fish

July 11, 2024

**Contact Information**  
EPA Press Office ([press@epa.gov](mailto:press@epa.gov))

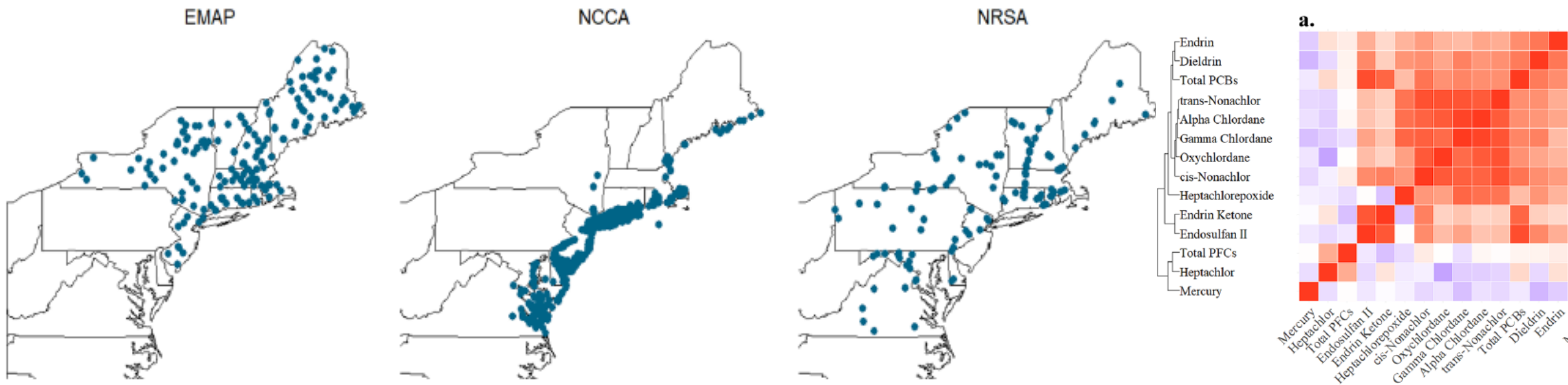
Group	Analyte
PFAS	Perfluorodecanoic acid (PFDA) Perfluorohexane sulfonic acid (PFHxS) Perfluorononanoic acid (PFNA) Perfluorooctanoic acid (PFOA) Perfluorooctane sulfonic acid (PFOS)
Cyanotoxins	Microcystins
Flame retardants	BDE-47
Pharmaceuticals	Amphetamine
Metals	Arsenic (inorganic) Mercury (methylmercury) Cadmium Selenium <b>Lead</b> Tributyltin
Organochlorine pesticides	Chlordane, total ( <i>cis-</i> and <i>trans-chlordane</i> , <i>cis-</i> and <i>trans-nonachlor</i> , <i>oxychlordane</i> ) Endrin DDT, total ( <i>2,4'-DDD</i> , <i>4,4'-DDD</i> , <i>2,4'-DDE</i> , <i>4,4'-DDE</i> , <i>2,4'-DDT</i> , <i>4,4'-DDT</i> ) Heptachlor epoxide Dicofol Lindane ( $\gamma$ -hexachlorocyclohexane; $\gamma$ -HCH) Hexachlorobenzene Dieldrin Mirex Endosulfan (I and II) Toxaphene
Organophosphate pesticides	Chlorpyrifos Diazinon Disulfoton Ethion Terbufos
Chlorophenoxy herbicides	Oxyfluorfen
Polycyclic aromatic hydrocarbons (PAHs)	Dibenz[ <i>a,h</i> ]anthracene Chrysene Benzo[ <i>a</i> ]pyrene Acenaphthene Benz[ <i>a</i> ]anthracene Acenaphthylene Benzo[ <i>b</i> ]fluoranthene Fluoranthene Benzo[ <i>k</i> ]fluoranthene Fluorene Indeno[1,2,3- <i>cd</i> ]pyrene Phenanthrene Anthracene Pyrene Benzo[ <i>g,h,i</i> ]perylene

Polychlorinated biphenyls (PCBs)	2,4' diCB	2,2',4,5,5' pentaCB	2,3',4,4',5,5' hexaCB
	2,2',5 triCB	2,3,3',4,4' pentaCB	2,3',4,4',5',6 hexaCB
	2,4,4' triCB	2,3,4,4',5 pentaCB	3,3',4,4',5,5' hexaCB
	3,4,4' triCB	2,3',4,4',5 pentaCB	2,2',3,3',4,4',5 heptaCB
	2,2'3,5' tetraCB	2,3',4,4',6 pentaCB	2,2',3,4,4',5,5' heptaCB
	2,2'4,5' tetraCB	2',3,4,4',5 pentaCB	2,2',3,4,4',5',6 heptaCB
	2,2',5,5' tetraCB	3,3',4,4',5 pentaCB	2,2',3,4,4',6,6' heptaCB
		2,3',4,4' tetraCB	2,2',3,3',4,4' hexaCB
	2,3',4',5 tetraCB	2,2',3,4,4',5' hexaCB	2,3,3',4,4',5,5' heptaCB
	2,4,4',5 tetraCB	2,2',3,5,5',6 hexaCB	2,2',3,3',4,4',5,6 octaCB
	3,3',4,4' tetraCB	2,2',4,4',5,5' hexaCB	2,2',3,3',4,4',5,6,6' octaCB
	3,4,4',5 tetraCB	2,3,3',4,4',5 hexaCB	2,2',3,3',4,4',5,5',6 nonaCB
	2,2',3,4,5' pentaCB	2,3,3',4,4',5 hexaCB	2,2',3,3',4,4',5,5',6,6' decaCB
	2,2',3,4',5 pentaCB	2,3,3',4,4',6 hexaCB	
Dioxins	2,3,7,8-TCDD	1,2,3,6,7,8-HxCDD	1,2,3,4,6,7,8-HpCDD
	1,2,3,7,8-PeCDD	1,2,3,7,8,9-HxCDD	OCDD
	1,2,3,4,7,8-HxCDD		
Furans	2,3,7,8-TCDF	1,2,3,6,7,8-HxCDF	1,2,3,4,6,7,8-HpCDF
	1,2,3,7,8-PeCDF	1,2,3,7,8,9-HxCDF	1,2,3,4,7,8,9-HpCDF
	2,3,4,7,8-PeCDF	2,3,4,6,7,8-HxCDF	OCDF
	1,2,3,4,7,8-HxCDF		

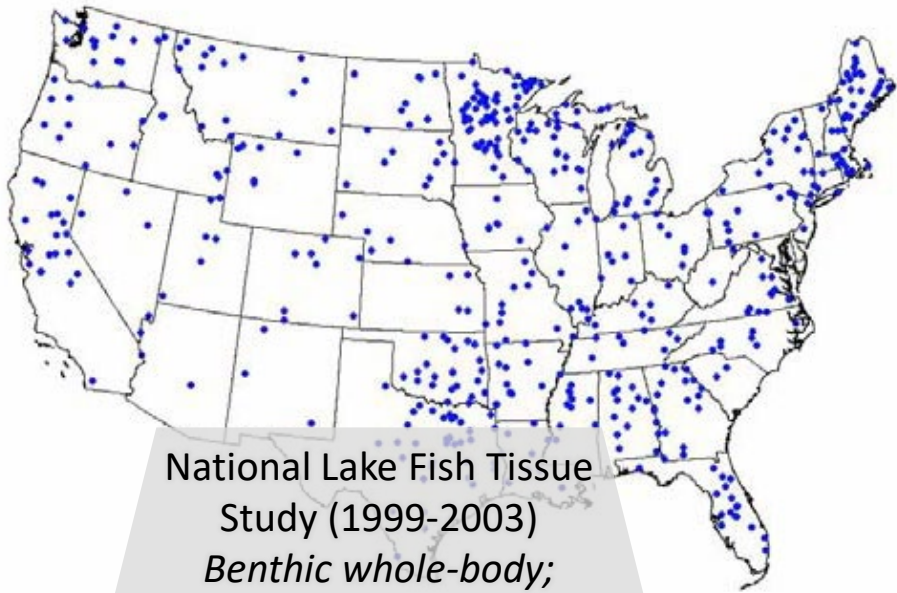


# Patterns of Co-contamination in Freshwater and Marine Fish of the Northeastern USA

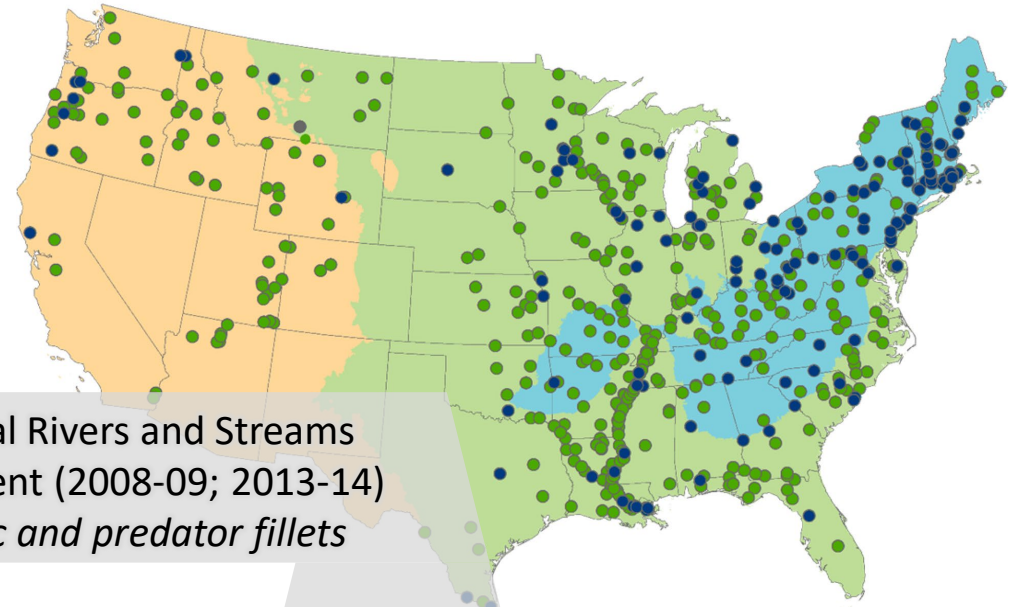
Kimberly Bourne<sup>1</sup> · Amanda N. Curtis<sup>2</sup> · Jonathan Chipman<sup>3</sup> · Cella Y. Chen<sup>4</sup> · Mark E. Borsuk<sup>1</sup>



**Fig. 1** Spatial representation of the northeastern US sample locations included in this study



National Lake Fish Tissue Study (1999-2003)  
*Benthic whole-body; predator fillets*

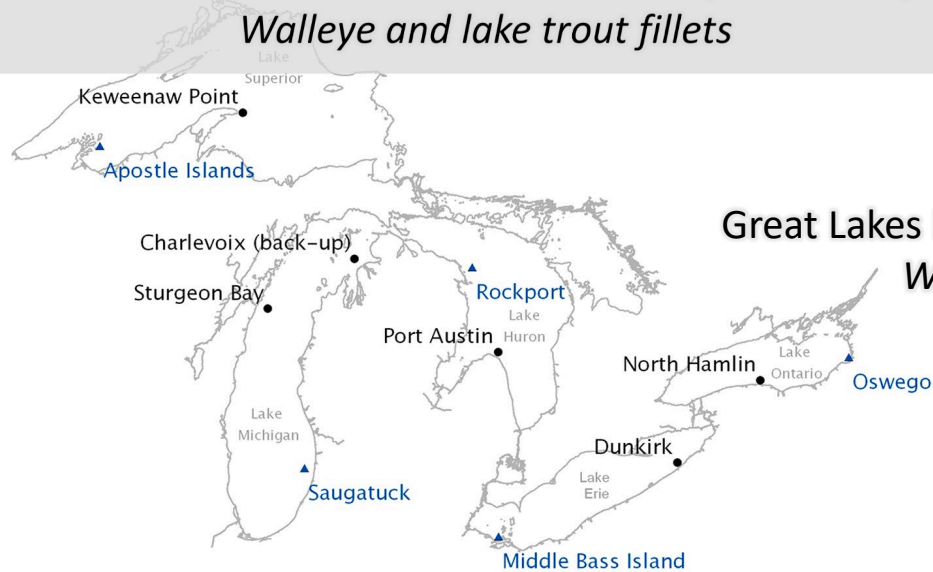


National Rivers and Streams Assessment (2008-09; 2013-14)  
*Benthic and predator fillets*

1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
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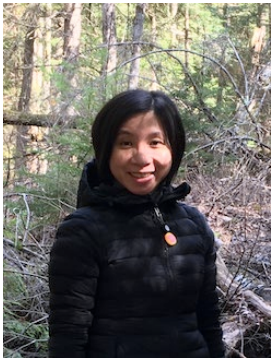
Great Lakes Environmental Database (1999-2018)

*Walleye and lake trout fillets*



Great Lakes Environmental Database (1999-2018)

*Walleye and lake trout fillets*



Pianpian Wu

# Datasets

	National Rivers and Streams Assessment (NRSA)		National Lake Fish Tissue Study (NLFTS)	Great Lakes Environmental Database (GLEND A)		
Years	2008-2009	2013-2014	1999-2003	1999-2003	2004-2009	2010-2018
Covariates studied	Species, feeding behavior, location		Species, feeding behavior, location	Species		
Elements	Hg, Se	Hg	Hg	Hg	Hg	Hg
PCBs	21 PCB congeners from PCB-8 to PCB-209	All 209 PCB congeners	All 209 PCB congeners	125 PCB congeners from PCB-1 to PCB-209	All 209 PCB congeners	All 209 PCB congeners
Pesticides	21 pesticides including dieldrin, mirex, and DDT		46 pesticides including dieldrin, kepone, DDE and DDT	25 pesticides including dieldrin, chlordane, mirex, toxaphene, and DDT	31 pesticides including dieldrin, chlordane, mirex, toxaphene, and DDT	31 pesticides including dieldrin, chlordane, mirex, toxaphene, and DDT
Dioxins			17 dioxins and furans including TCDD			
Flame retardants	5 PBDE congeners from PBDE-46 to PBDE-183			PBB-153 and 6 PBDE congeners from PBDE-47 to PBDE-154	5 PBDE congeners from PBDE-47 to PBDE-154	PBB-153 and 26 PBDE congeners from PBDE-007 to PBDE-209
PFAS	13 PFAS compounds including PFOS, PFDA, PFUnA	13 PFAS compounds including PFOS, PFDA, PFUnA, PFOSA				31 PFAS compounds including PFOS, PFOA, PFDA

# Research Questions

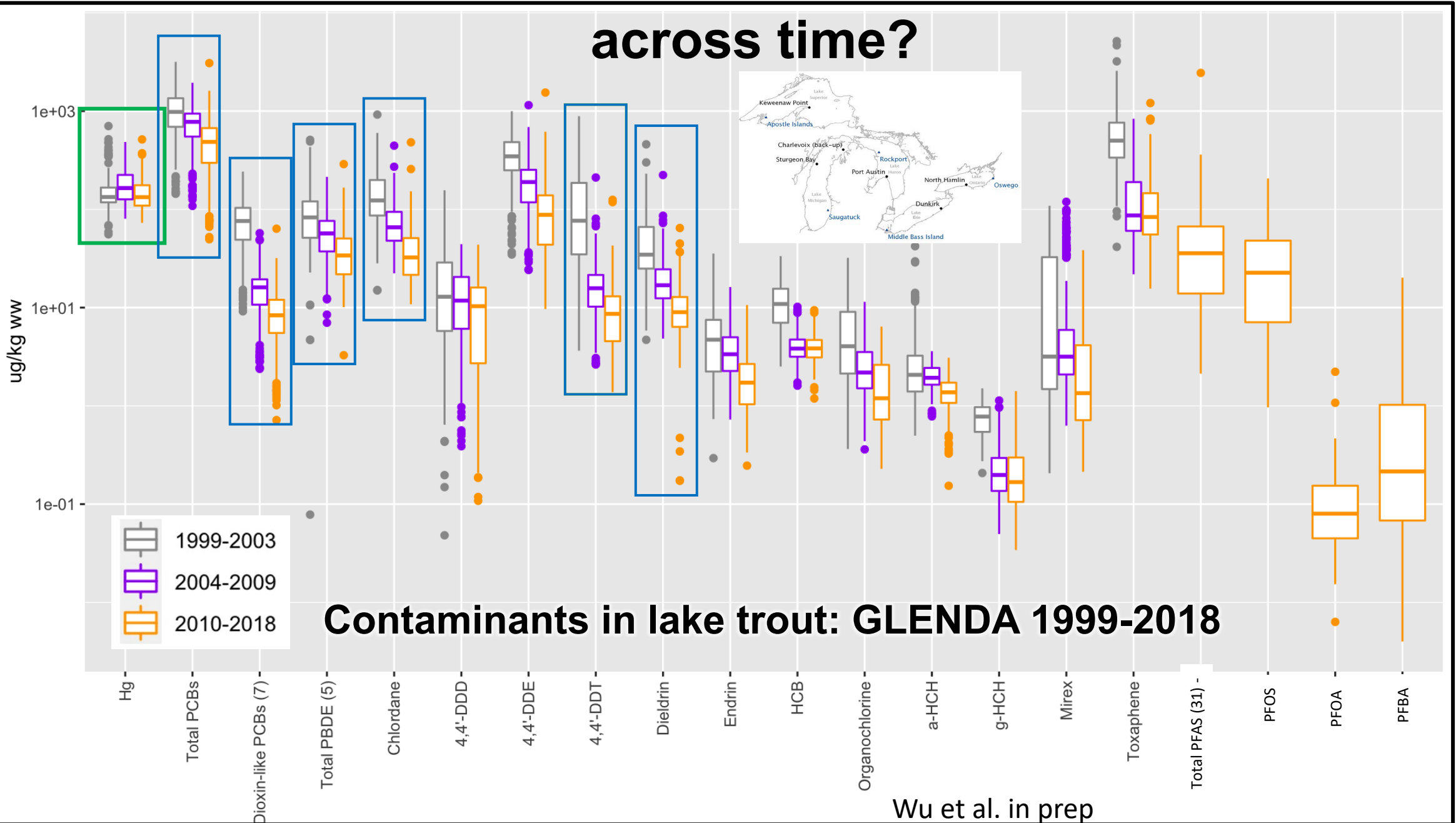
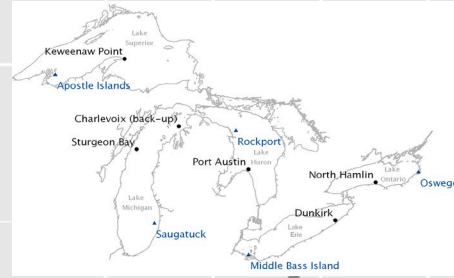
- How have contaminant levels varied in fish tissue across time?
- Do contaminants correlate with one another in fish tissue, and if so, how?
- How do observed contaminant levels compare with risk-based screening levels (RSLs)?



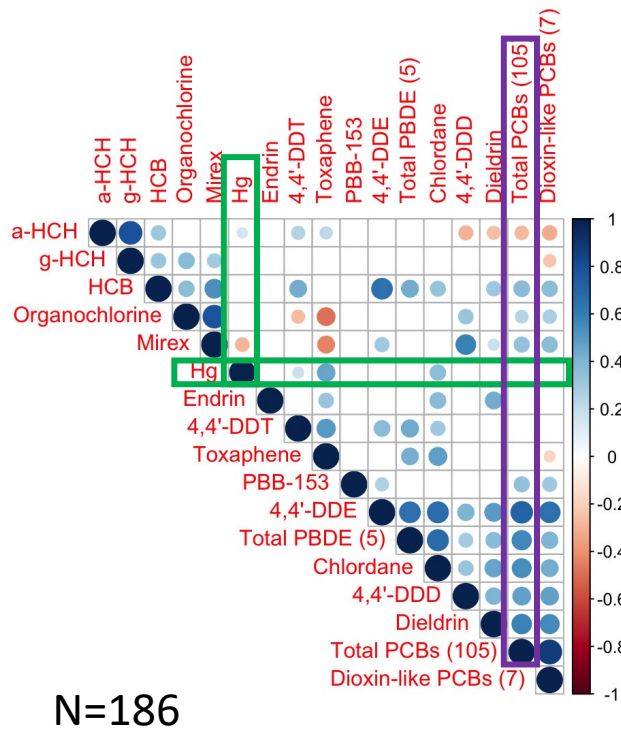
<https://www.epa.gov/choose-fish-and-shellfish-wisely/fish-and-shellfish-advisories-and-safe-eating-guidelines>

# How have contaminant levels varied in fish tissue

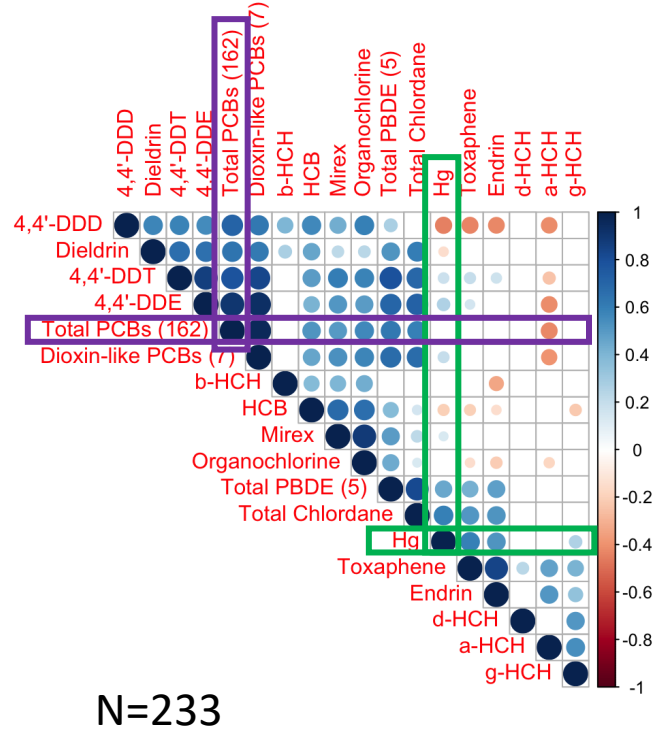
## across time?



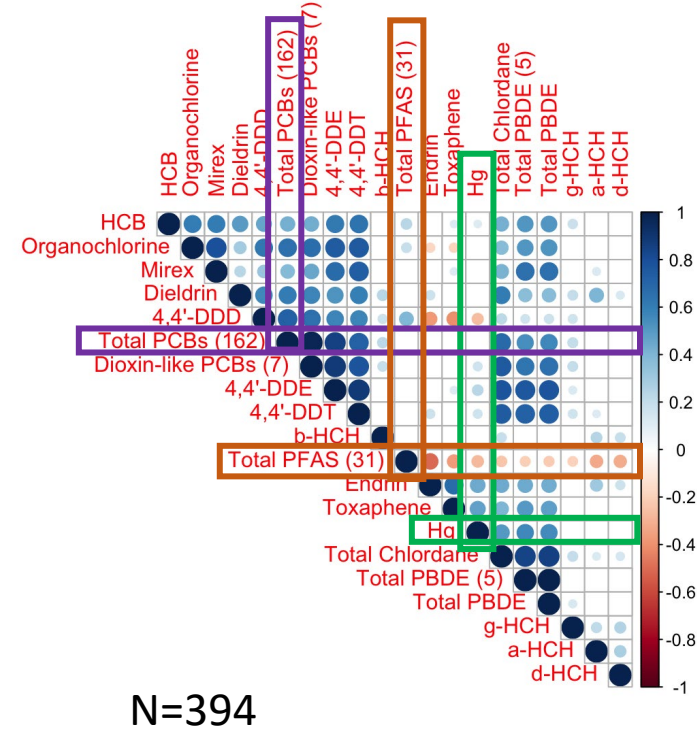
# Do contaminants correlate with one another in lake trout, and if so, how?



GLEND A 1999-2003



GLEND A 2004-2009



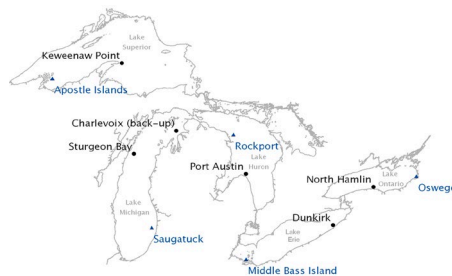
GLEND A 2010-2018

Correlates:

Green Hg

Purple PCBs

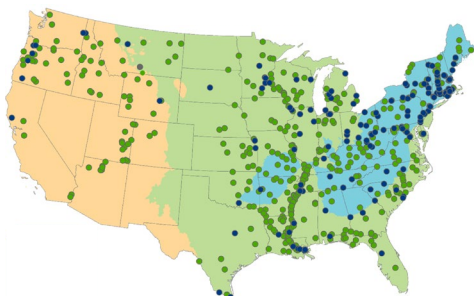
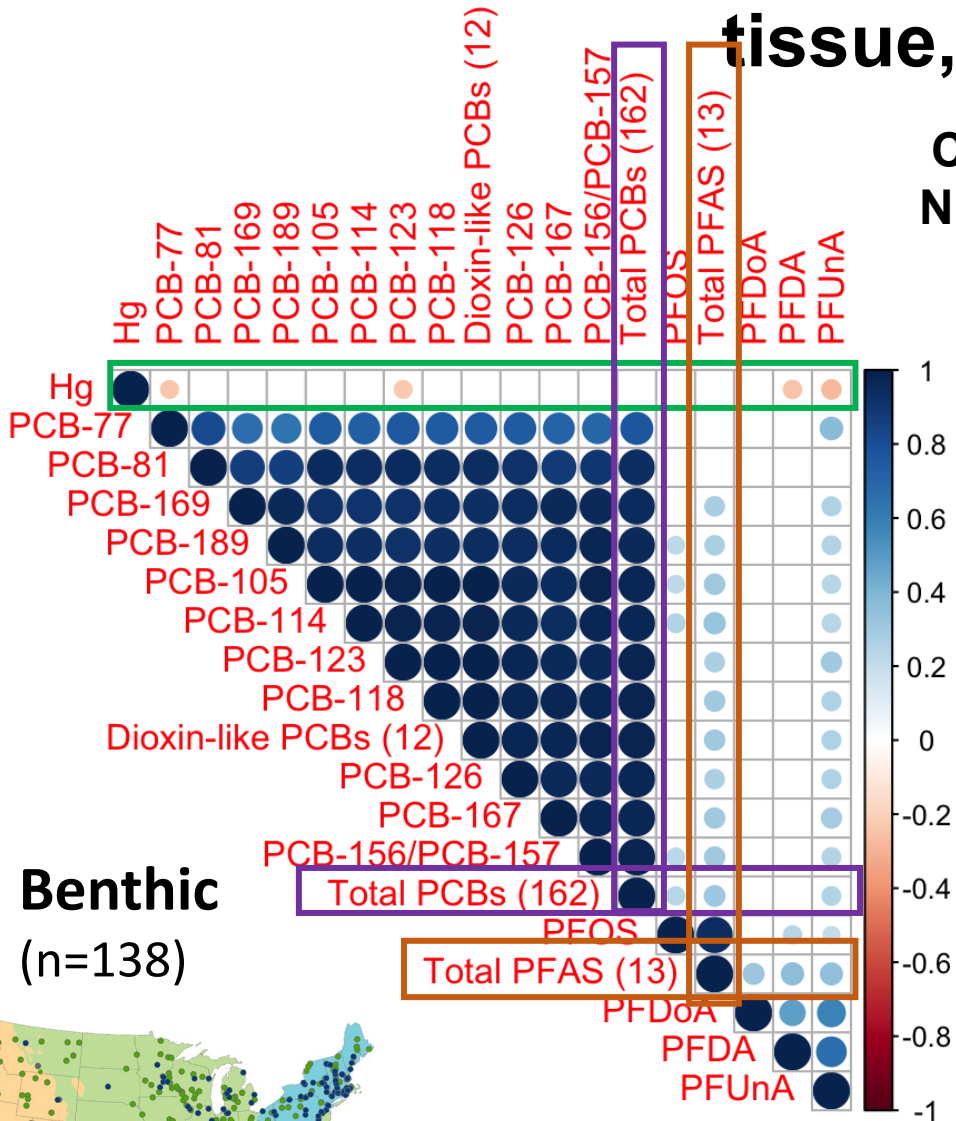
Orange PFAS



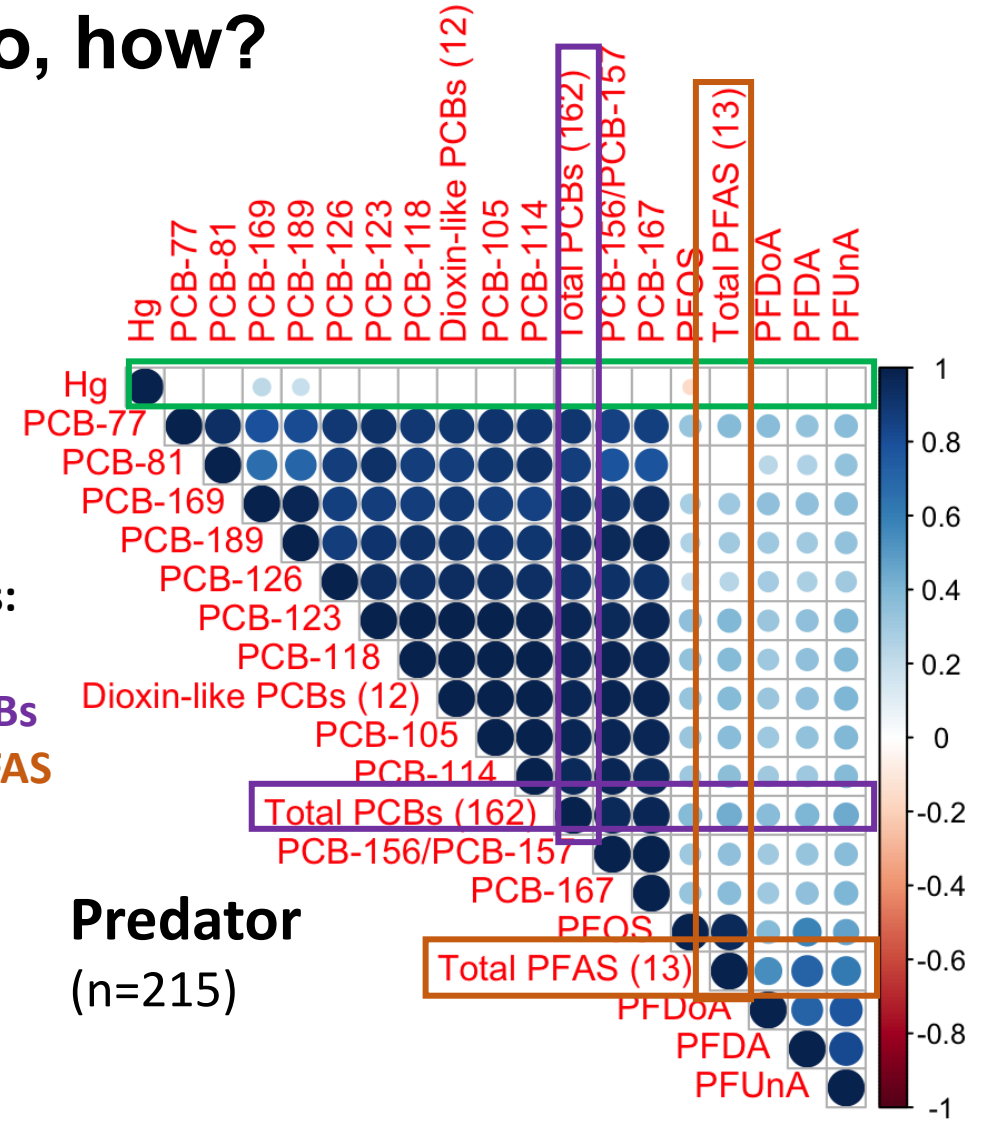
Contaminants in lake trout: GLEND A 1999-2018

# Do contaminants correlate with one another in fish tissue, and if so, how?

Correlations:  
NRSA 2013-14



Correlates:  
Green Hg  
Purple PCBs  
Orange PFAS



Mercury in fish do not track other organic chemicals significantly, while PCB congeners co-occur closely

# Regional Risk-Based Screening Levels (RSLs)

EPA tool to estimate concentrations of contaminants in fish tissue that are considered “safe”

## Target Risk Level

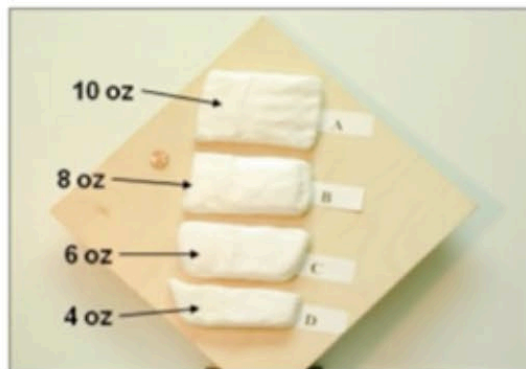
0.1 (Non-cancer)  
1E-6 (Cancer)

## Toxicity Values

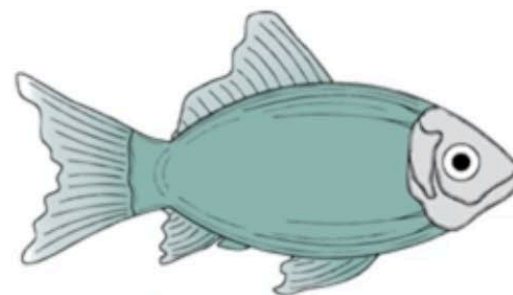
Reference Dose (RfD)

Cancer Slope Factors (CSF)

## Fish consumption rate



RSL  
Concentration in Fish Tissue



Hg  
PCBs  
Dieldrin  
Mirex  
PFAS - PFOS

# How do observed contaminant levels compare with risk-based screening levels (RSLs)?



Lake trout	Daily 100g intake			Weekly 100g intake			Monthly 100g intake		
	1999-2003	2004-2009	2010-2018	1999-2003	2004-2009	2010-2018	1999-2003	2004-2009	2010-2018
	Hg	●	●	●	●	●	●		
Dioxin-like PCBs	●	●	●	●	●	●	●		
Chlordane	●	●	●	●	●	●	●		
4,4'-DDD	●	●	●	●	●				
4,4'-DDE	●	●	●	●	●	●	●	●	
4,4'-DDT	●	●	●	●					
Dieldrin	●	●	●	●	●	●	●	●	●
Endrin									
Mirex	●	●	●	●	●	●	●	●	●
Toxaphene	●	●	●	●	●	●	●	●	●
a-HCH	●	●	●	●					
b-HCH	n/a			n/a					
PFOS	n/a	n/a	●	n/a	n/a	●			
PFOA	n/a	n/a	●	n/a	n/a				

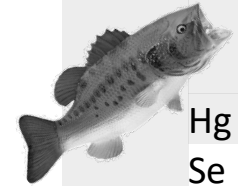
**Dieldrin, mirex, and toxaphene in lake trout still exceed RSL even at only monthly consumption.**

Red dot represents risk exceedance, calculated as 95% UCL/fish intake RSL.



**RSL exceedances in lake trout: GLEND A 1999-2018**

# Risk Screening Level exceedances: NRSA 2008-14



Predator fish

	Daily 100g intake		Weekly 100g intake		Monthly 100g intake	
	2008-09	2013-14	2008-09	2013-14	2008-09	2013-14
Hg	●	●	●	●	●	●
Se	●	n/a		n/a		n/a
Chlordane		n/a		n/a		n/a
4,4'-DDD		n/a		n/a		n/a
4,4'-DDE	●	n/a		n/a		n/a
4,4'-DDT		n/a		n/a		n/a
Dieldrin	●	n/a	●	n/a		n/a
PCB-105	●	●				
PCB-118	●	●	●	●		
Dioxin-like PCBs	●	●		●		
PFOS	●	●	●	●		

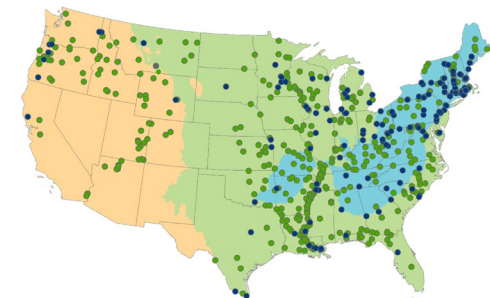
Benthic fish

Hg	●	●	●	●		
Se	●	n/a		n/a		n/a
Chlordane	●	n/a		n/a		n/a
4,4'-DDD	●	n/a		n/a		n/a
4,4'-DDE	●	n/a		n/a		n/a
4,4'-DDT		n/a		n/a		n/a
Dieldrin	●	n/a	●	n/a	●	n/a
PCB-105	●	●				
PCB-118	●	●	●	●		
Dioxin-like PCBs	●	●	●	●		
PFOS	●	●	●	●		



**Predator Hg and benthic dweller dieldrin still exceed RSL even at only monthly consumption.**

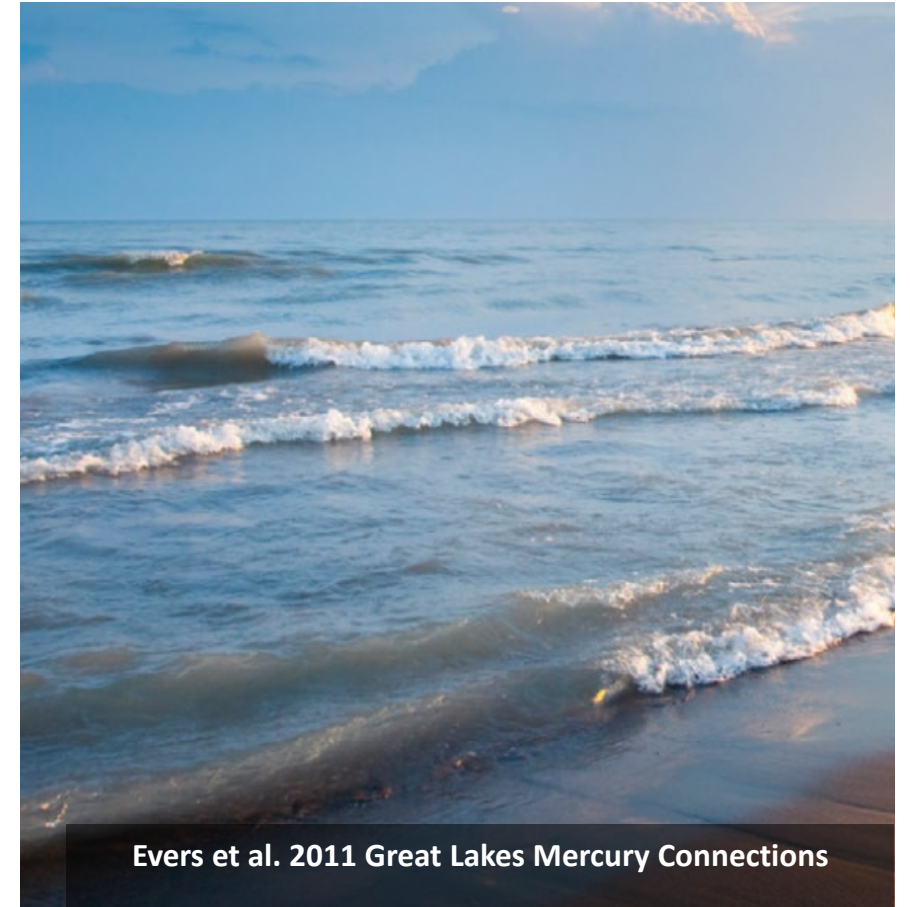
Red dot represents risk exceedance, calculated as 95% UCL/fish intake RSL.



# Chemical Mixtures Findings

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- **Temporal change:**
  - Organic contaminants in lake trout appear to be declining but Hg does not
  - Chemical substances banned 50 years ago were still detected and exceed RSLs after 2010
- **Chemical Correlations:**
  - Hg remains prevalent in all species
  - Hg doesn't strongly correlate with other organic chemicals,
- **Risk Screening Level Exceedances:**
  - Higher exposure and risks for all chemicals in high fish consumption frequency
  - At lowest consumption, pesticides still exceed RSLs



Evers et al. 2011 Great Lakes Mercury Connections

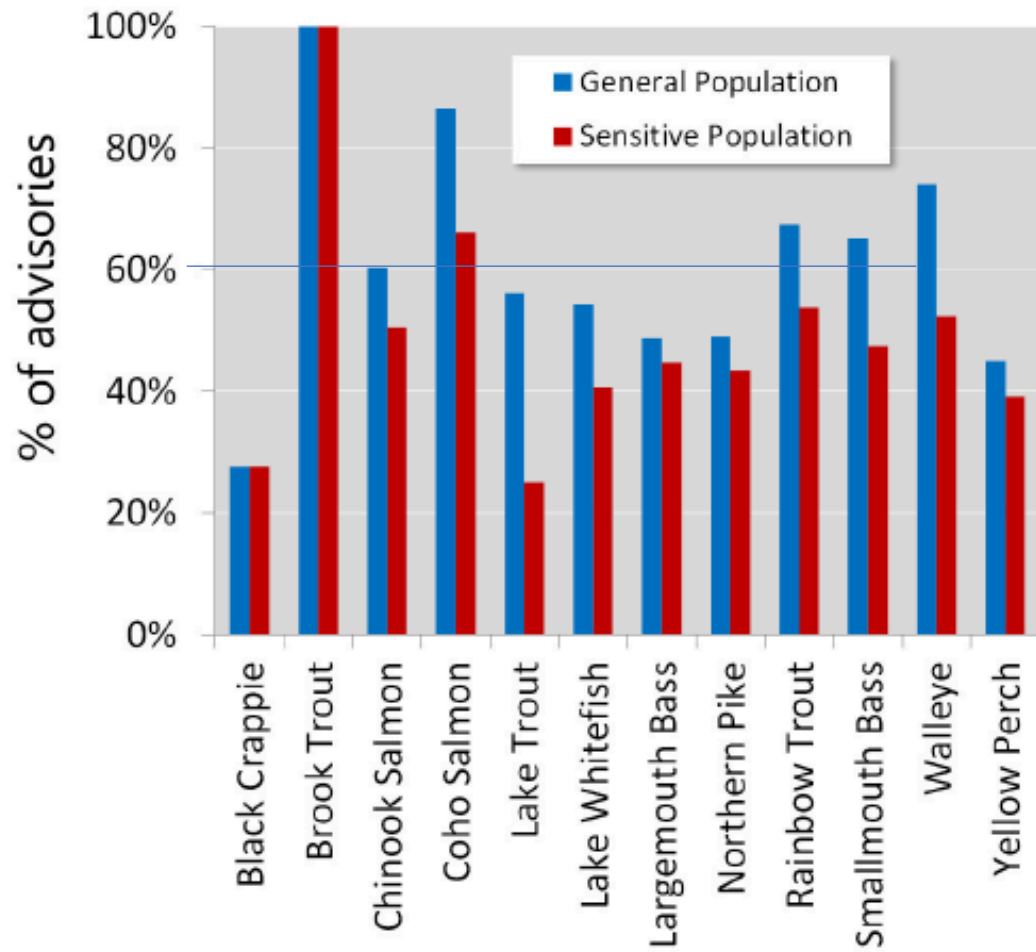
## Acknowledgments

Kim Bourne, Duke University  
Mark Borsuk, Duke University  
Kate Crawford, Middlebury College  
Amanda Curtis, US Geological Survey  
Jonathan Chipman, Dartmouth College

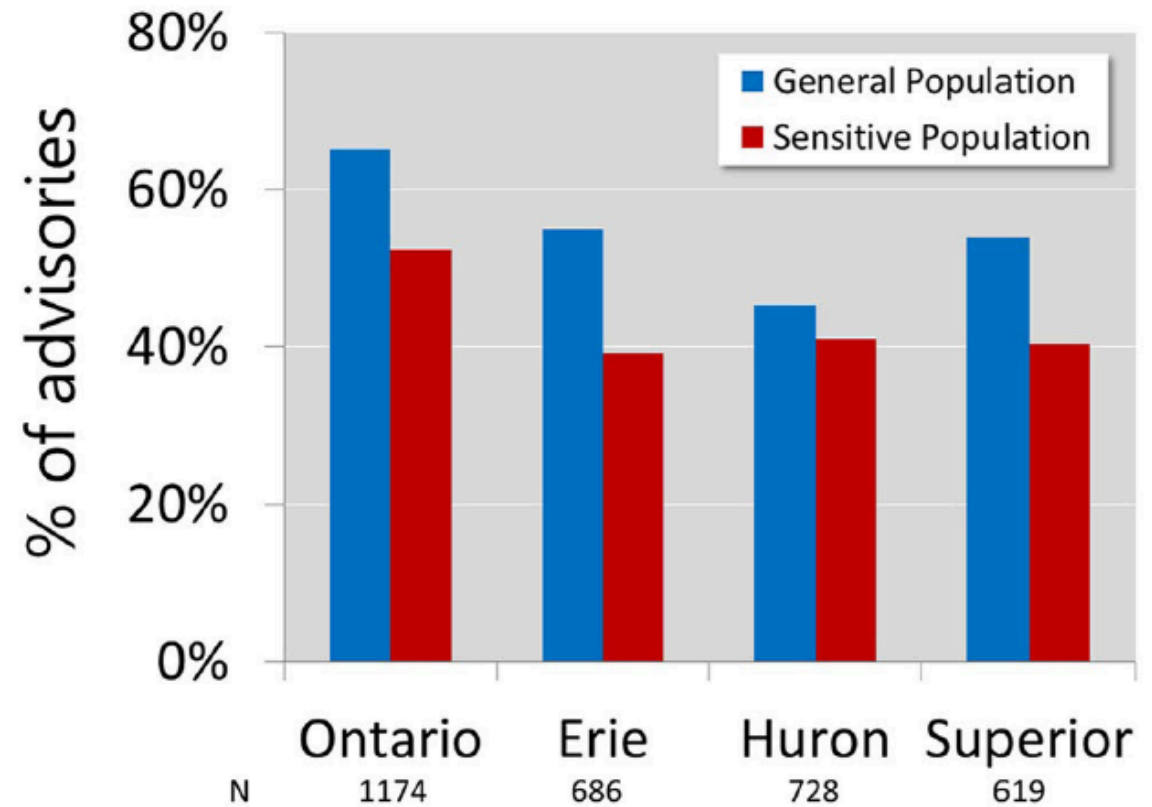




# Multi-chem vs. One chem Approach to FCA (Gandhi et al. 2017)




Percentage of the multi-chem approach–based advisories that were more stringent than the one-chem approach for fish favored by anglers in the region.



Percentage of the multi-chem approach–based advisories that were more stringent than the one-chem approach. *N* represents the total number of advisories for each population.

## Comparison of Recreational Fish Consumption Advisories Across the USA

Brittany M. Cleary<sup>1</sup> · Megan E. Romano<sup>2</sup> · Celia Y. Chen<sup>3</sup> · Wendy Heiger-Bernays<sup>4</sup> · Kathryn A. Crawford<sup>2,5</sup> 

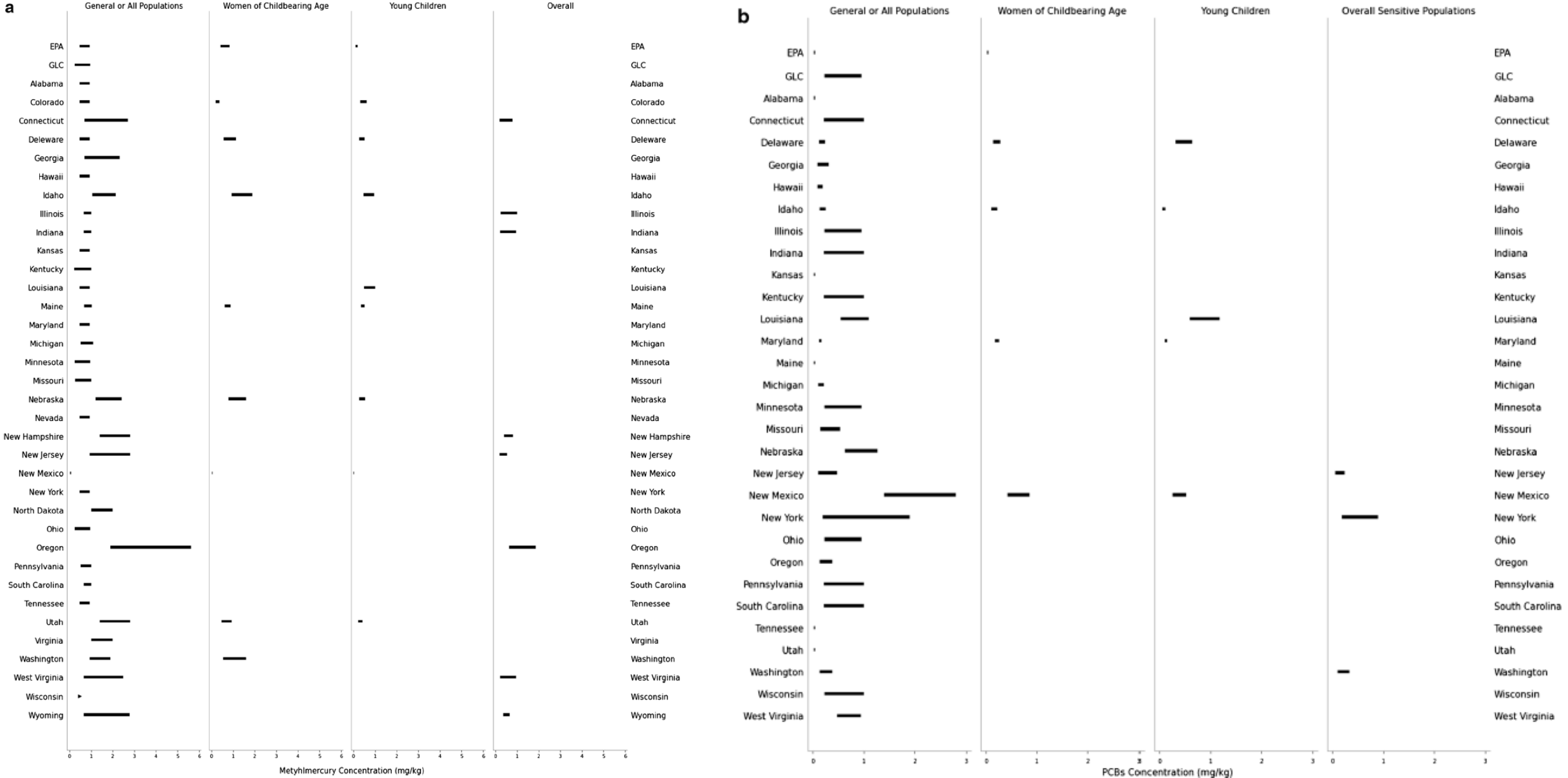
**Fish Consumption Advisories differ by state on the same waterbodies for the same species.**

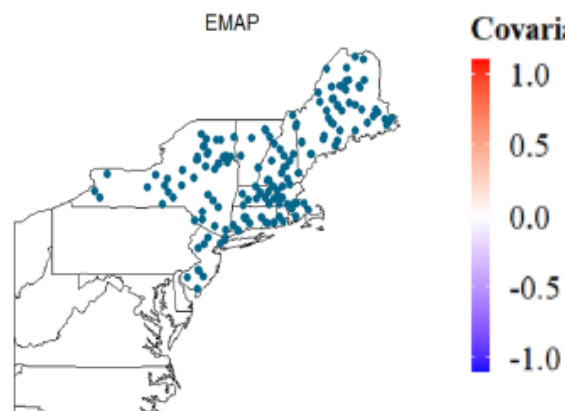
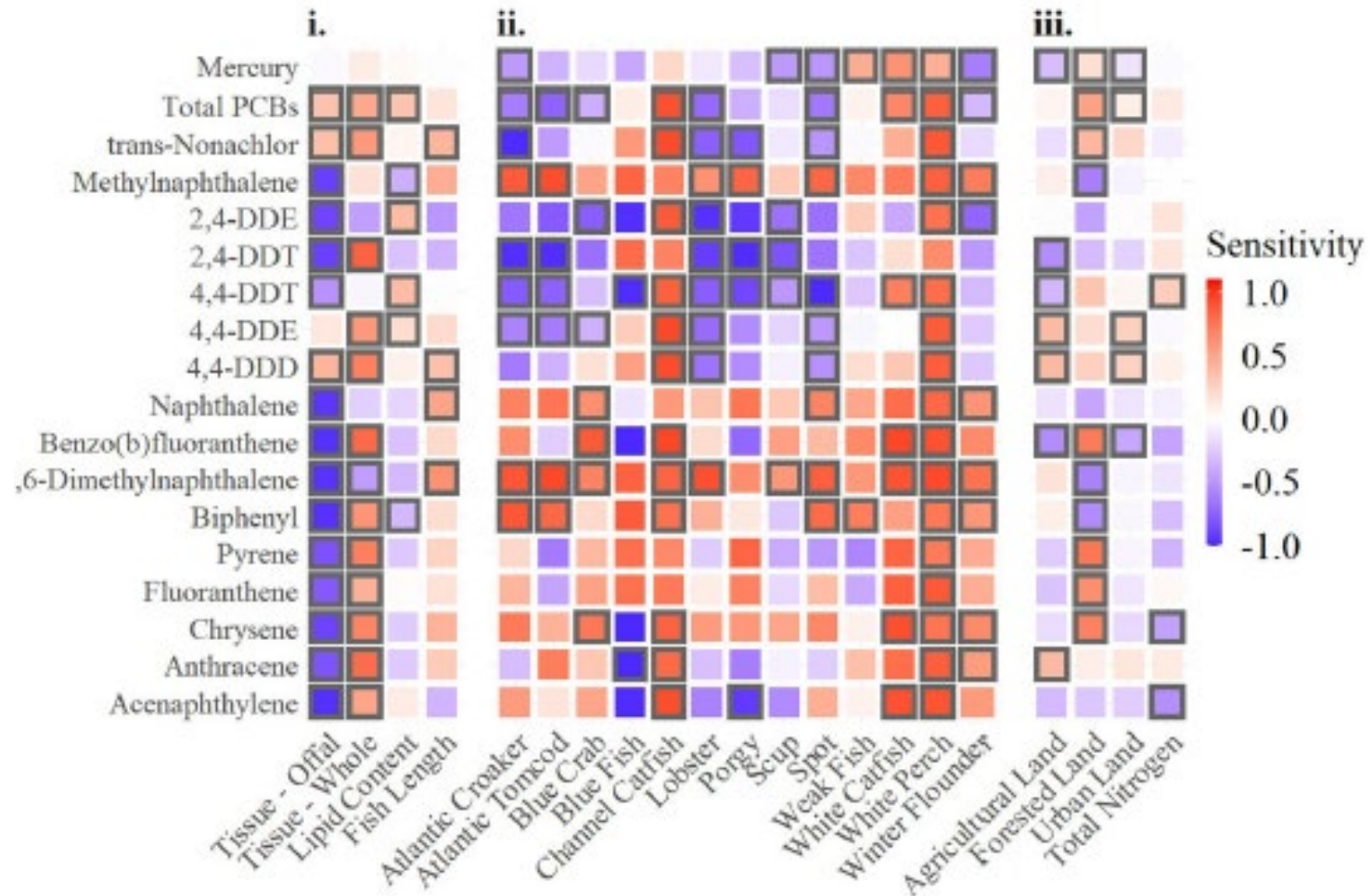
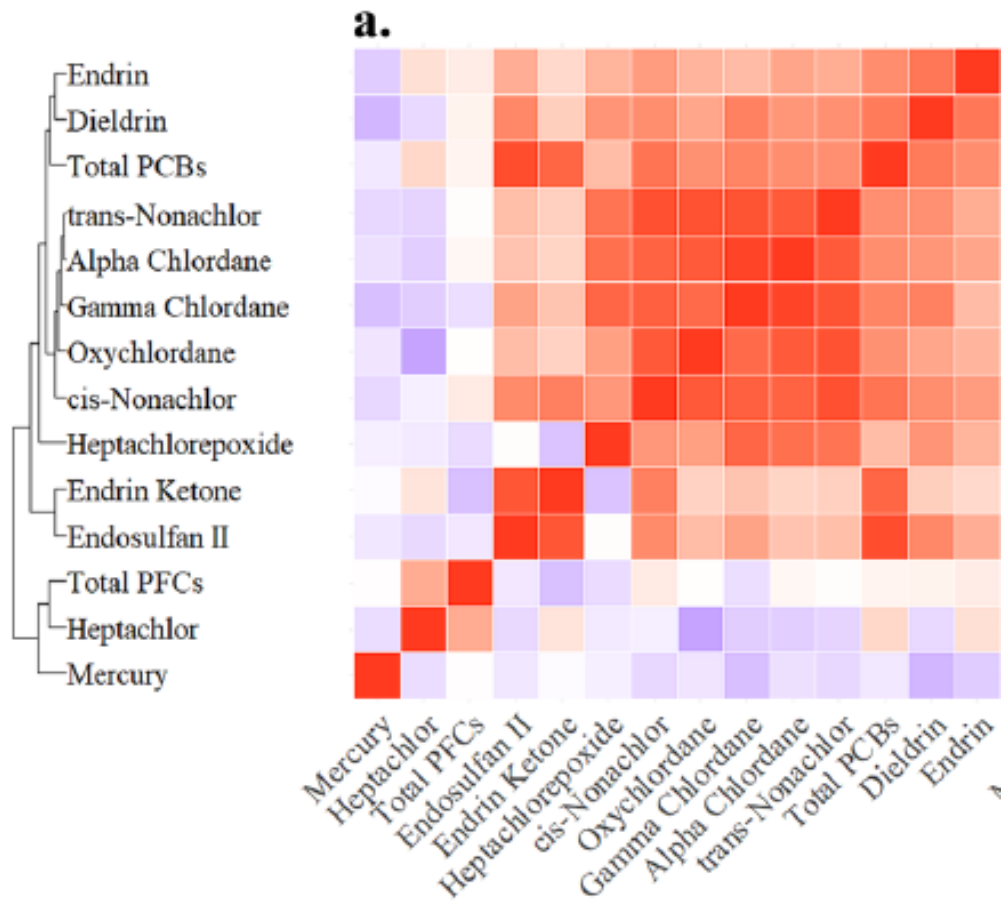
	Illinois	Indiana	Michigan	Wisconsin
Brown trout	1 meal/month	1 meal/month	Limited*	1 meal/month
Chinook salmon	1 meal/month	1 meal/month	6 meals/year	1 meal/month
Coho salmon	1 meal/week (< 24")	1 meal/week (≤ 24")	1 meal/month	1 meal/week (< 24")
	1 meal/month (≥ 24")	1 meal/month (> 24")		1 meal/month (> 24")
Lake trout	1 meal/month (< 30")	1 meal/week (≤ 22")	6 meals/year (< 24")	1 meal/month (< 30")
	Do not eat (≥ 30")	1 meal/month (22"-30")	Limited (> 24")*	Do not eat (> 30")
		Do not eat (> 30")		
Rainbow trout	1 meal/week (< 28")	1 meal/week	2 meals/month (< 20")	1 meal/week (< 28")
	1 meal/month (≥ 28")		6 meals/year (> 20")	1 meal/month (> 28")
Yellow perch	1 meal/week (< 11")	1 meal/week (≤ 11")	4 meals/month	1 meal/week (< 11")
	1 meal/week (≥ 11", GP)	1 meal/month (> 11")		1 meal/month (> 11")
	1 meal/month (≥ 11", SP)			

If not otherwise indicated, advisories are specific to Lake Michigan (rather than general statewide advisory). If both statewide and Lake Michigan advisories were available, the Lake Michigan advisory was recorded

\*= Michigan employs a "limited" advisory category, which indicates that for the type of fish in question, SP should avoid consumption, and the GP should consume a maximum of one to two meals/year

# Target tissue concentration range corresponding to a one meal/month FCA (Cleary et al. 2021)





- Mercury doesn't correlate with other compounds in lakes
- On the coast, channel catfish and white perch have most chemicals

