

Pharmaceuticals in Surface Water: Extent and Sources of Contamination

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EPA National Aquatic Resource Sampling State-wide contaminant studies

Five-year rotating studies of lakes and rivers

293 total chemicals tested

184 pharmaceuticals

2012, 2017, 2022 Lakes

2015, 2020, 2025 Rivers and Streams

50 randomly selected locations for each study



Pharmaceuticals tested in surface water

- Antibiotics
- Antidepressants
- Hormones
- Cardiovascular medications
- Lipid regulators
- Blood pressure
- Sedatives
- Antiseptics
- Anxiolytics
- NSAIDs
- Cytostatics

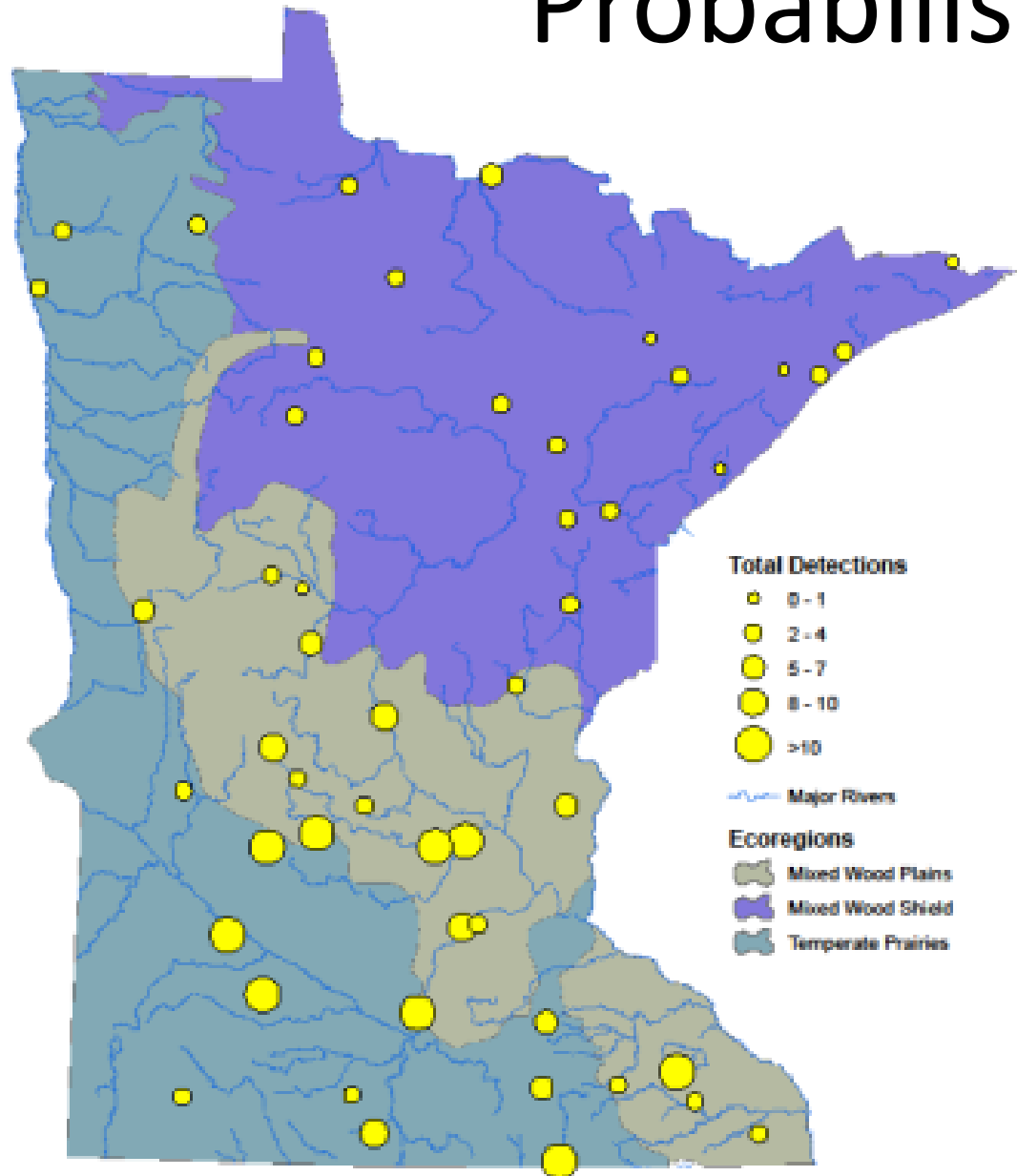
Pharmaceutical Categories Detected

2012 Lake Study

Classification	Pharmaceuticals	Detections
Neurological	6	46
Anti-infectives	8	18
Cardiovascular	3	7
Dermatological	4	5

(Ferrey et al., 2015. Environ. Toxicol. Chem. 34:2475-88)

Probabilistic studies



47 of 50 lakes

49 of 50 river locations

With at least one
chemical detection

Northern Light Lake

DEET

Bisphenol A

Diltiazem

Ciprofloxacin

Erythromycin

Trimethoprim

Clinafloxacin



Antibiotics Tested

Azithromycin

Carbadox

Ciprofloxacin

Clarithromycin

Clinafloxacin

Cloxacillin

Enrofloxacin

Erythromycin

Lincomycin

Lomefloxacin

Moxifloxacin

Norfloxacin

Ofloxacin

Oxacillin

Oxolinic Acid

Penicillins

Roxithromycin

Sarafloxacin

Sulfachloropyridazine

Sulfadiazine

Sulfadimethoxine

Sulfamerazine

Sulfamethazine

Sulfamethizole

Sulfamethoxazole

Sulfanilamide

Sulfathiazole

Trimethoprim

Tylosin

Virginiamycin M1

Tetracycline

Doxycycline

Isochlortetracycline

Chlortetracycline

Anhydro-chlortetracycline

Demeclocycline

4-Epianhydro-chlortetracycline

4-Epianhydro-tetracycline

4-Epichlortetracycline

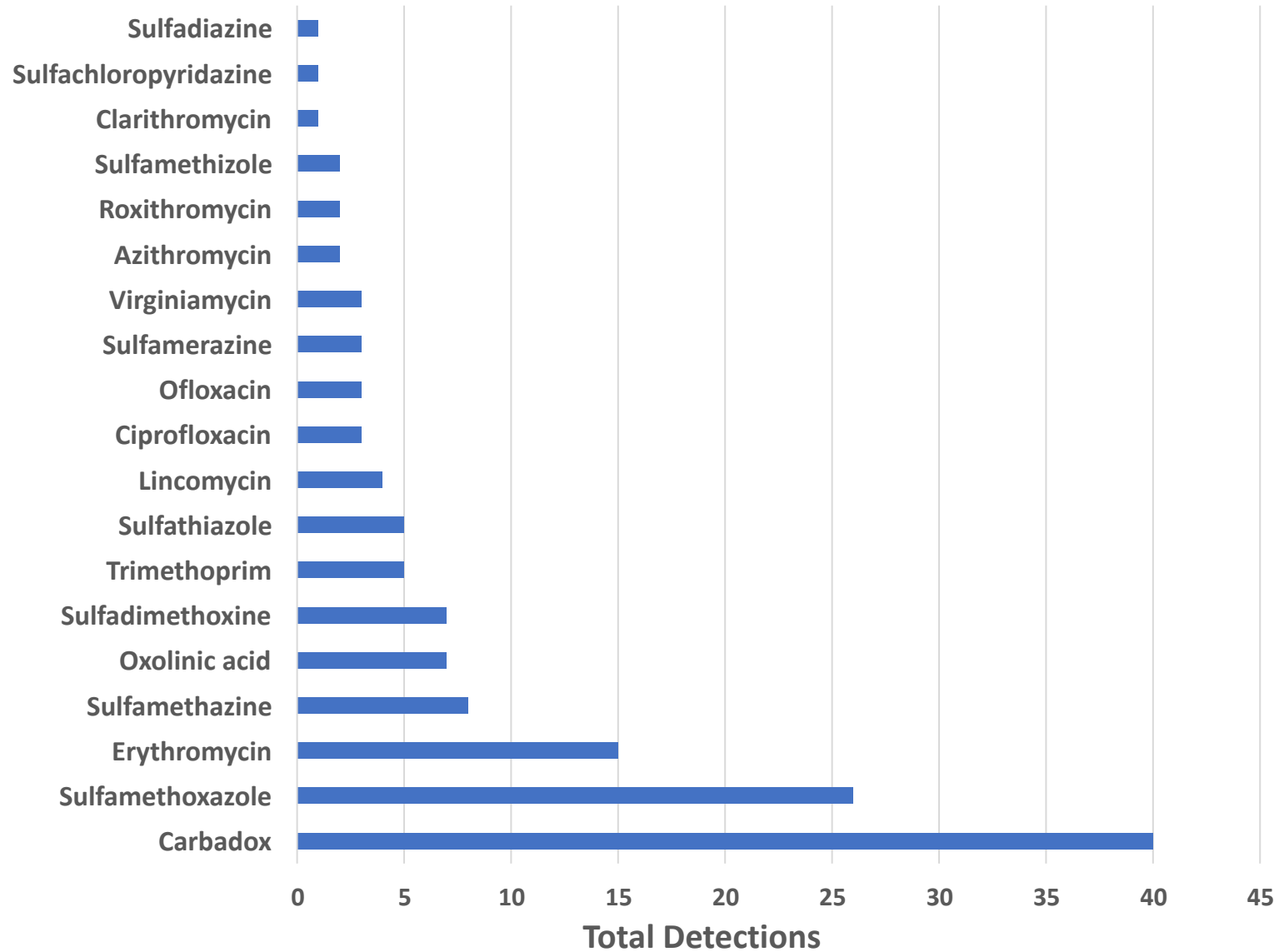
4-Epioxytetracycline

4-Epitetracycline

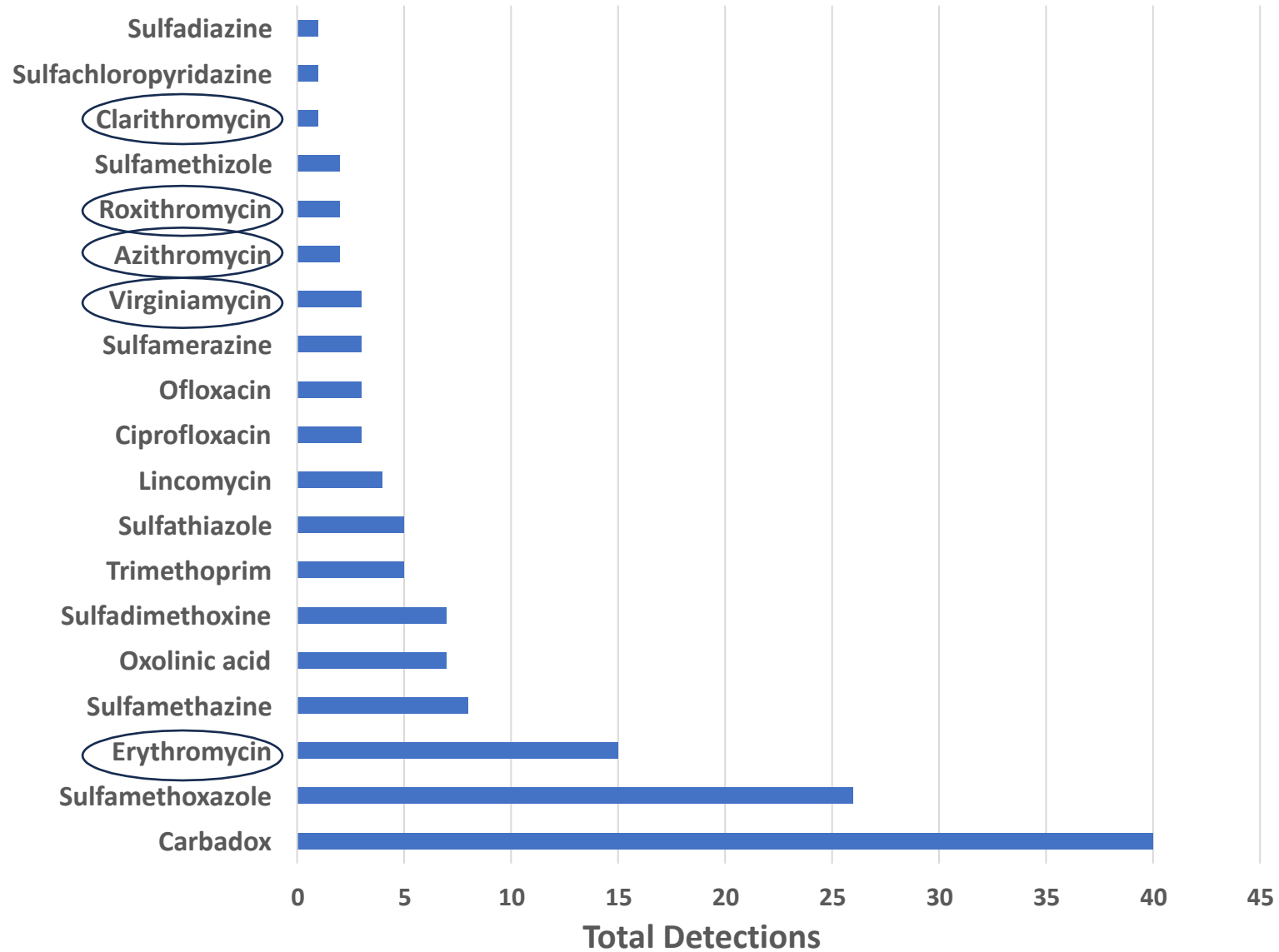
Minocycline

Oxytetracycline

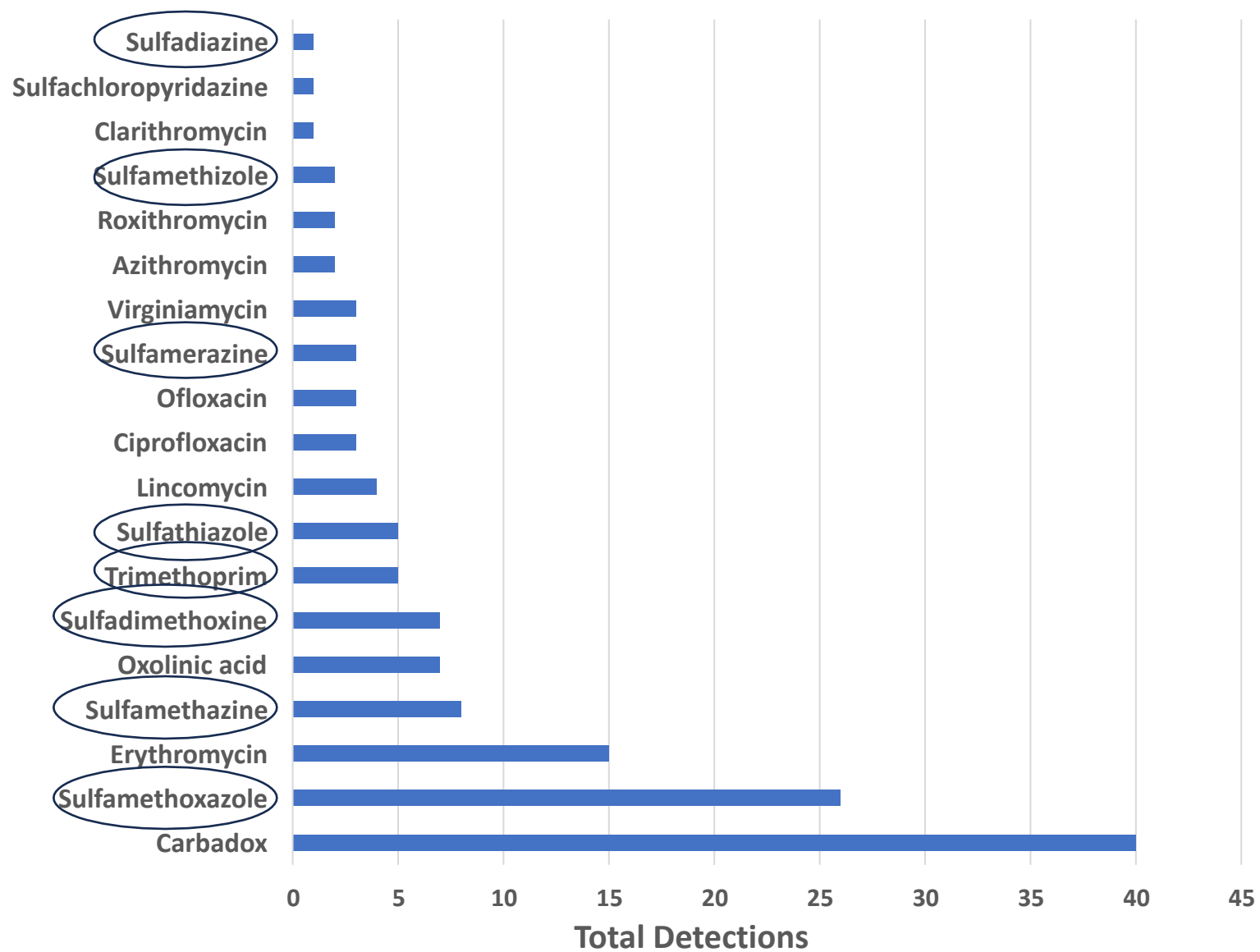
Antibiotics detected in surface water



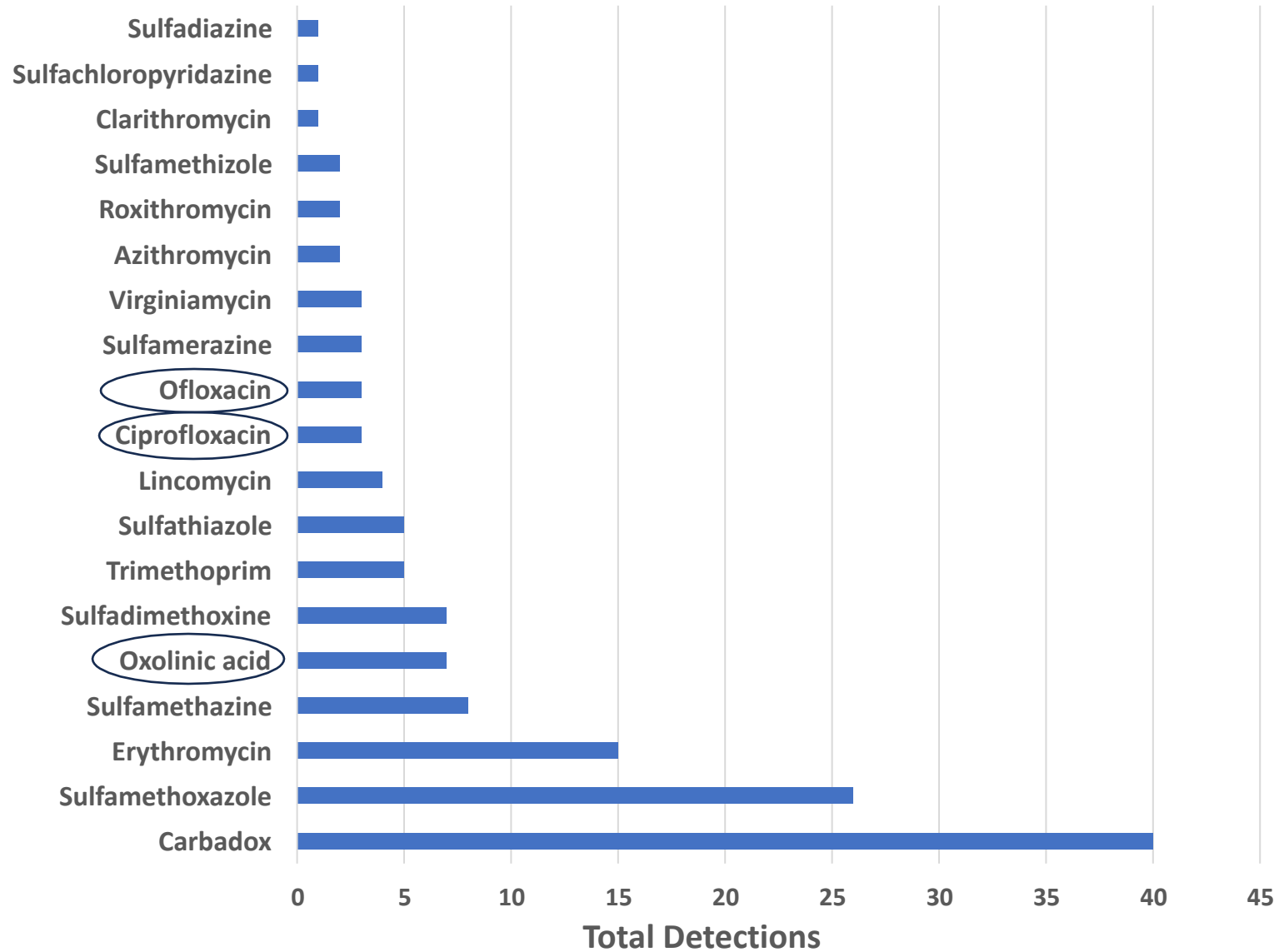
Antibiotics detected in surface water



Antibiotics detected in surface water



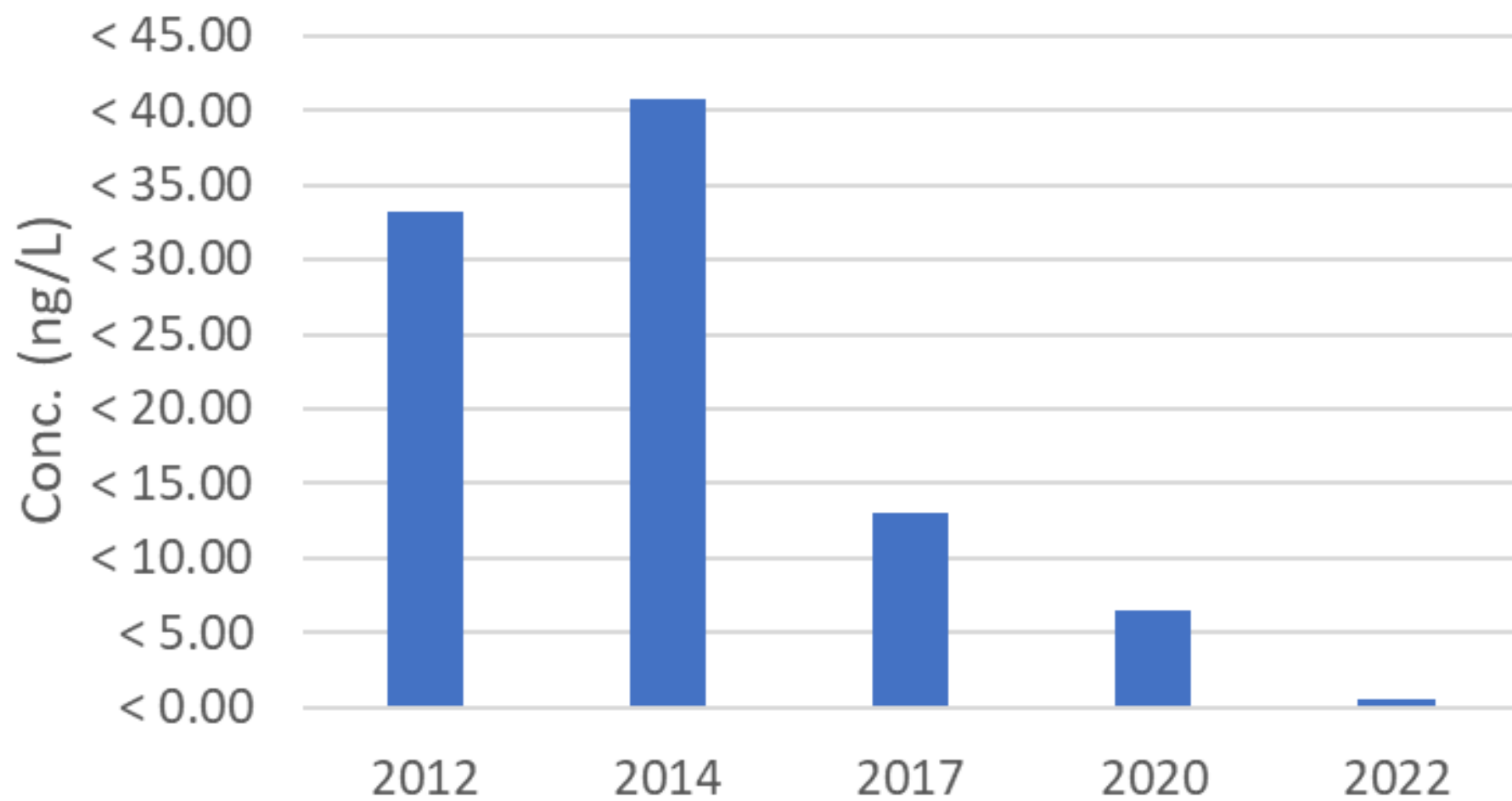
Antibiotics detected in surface water



2017 Lake Study

Antibiotic	% Samples	Mean Conc. (ng/L)
Carbadox	18	2.7
Erythromycin-H2O	16	2.1
Azithromycin	4	1.5
Virginiamycin M1	4	2.8
Ofloxacin	2	1.5
Oxolinic Acid	2	0.6
Roxithromycin	2	0.3
Sulfadimethoxine	2	0.4
Sulfamethizole	2	0.7
Sulfamethoxazole	2	1

Sulfamethoxazole 95th percentile



Pharmaceutical	Max Conc. (ng/L)	
Amitriptyline	9.8	Tricyclic antidepressant
Sertraline	2.7	SSRI antidepressants
Fluoxetine	25.3	
Venlafaxine	11	
Citalopram	3.19	
Carbamazepine	54.5	Anticonvulsive
Topiramate	145	
Lamotrigine	226	
Diazepam	0.39	Sedative
Meprobamate	29.4	
Amphetamine	4.7	Stimulant
Methamphetamine	1.0	

Pharmaceutical	Max. Conc. (ng/L)	
Metformin	667	Anti-diabetic
Theophylline	90.6	Bronchodilator
Diphenhydramine	1.8	Antihistamine
Iopamidol	1650	X-ray contrast agent
Metoprolol	16.3	Beta blocker

Contaminant	River miles affected (%)
Iopamidol	81.8
Sertraline	40.9
Amitriptyline	50.7
Cotinine	35.8
DEET	39.8
Sulfamethoxazole	19.7
Metformin	13.1
Bisphenol A	7.6
4-Methyl Benzotriazole	26.4
5,6-Dimethyl Benzotriazole	36.5
5-Methyl Benzotriazole	40.4
Benzotriazole	22.9

Effects?

- *Oxycodone human therapeutic dose: 10 mg*
- *WWTP effluents: frequently detected at >40ug/L*
→ *250L effluent = one therapeutic dose*
- *WWTP effluent contributes 20% flow*
→ *1,250L river water = one therapeutic dose*

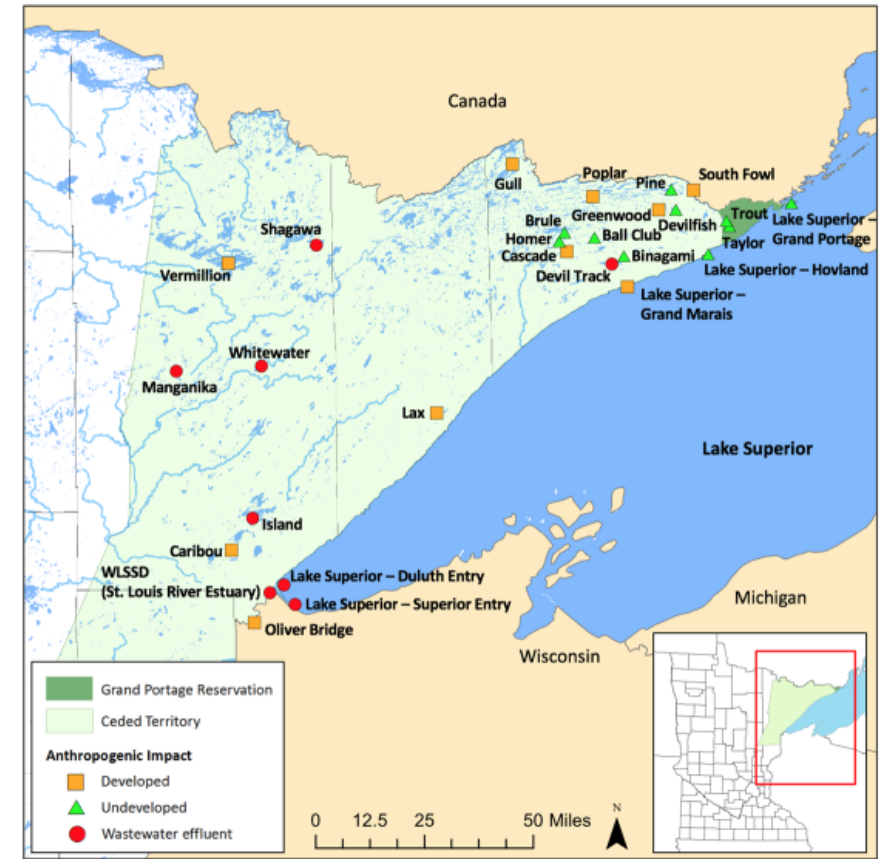
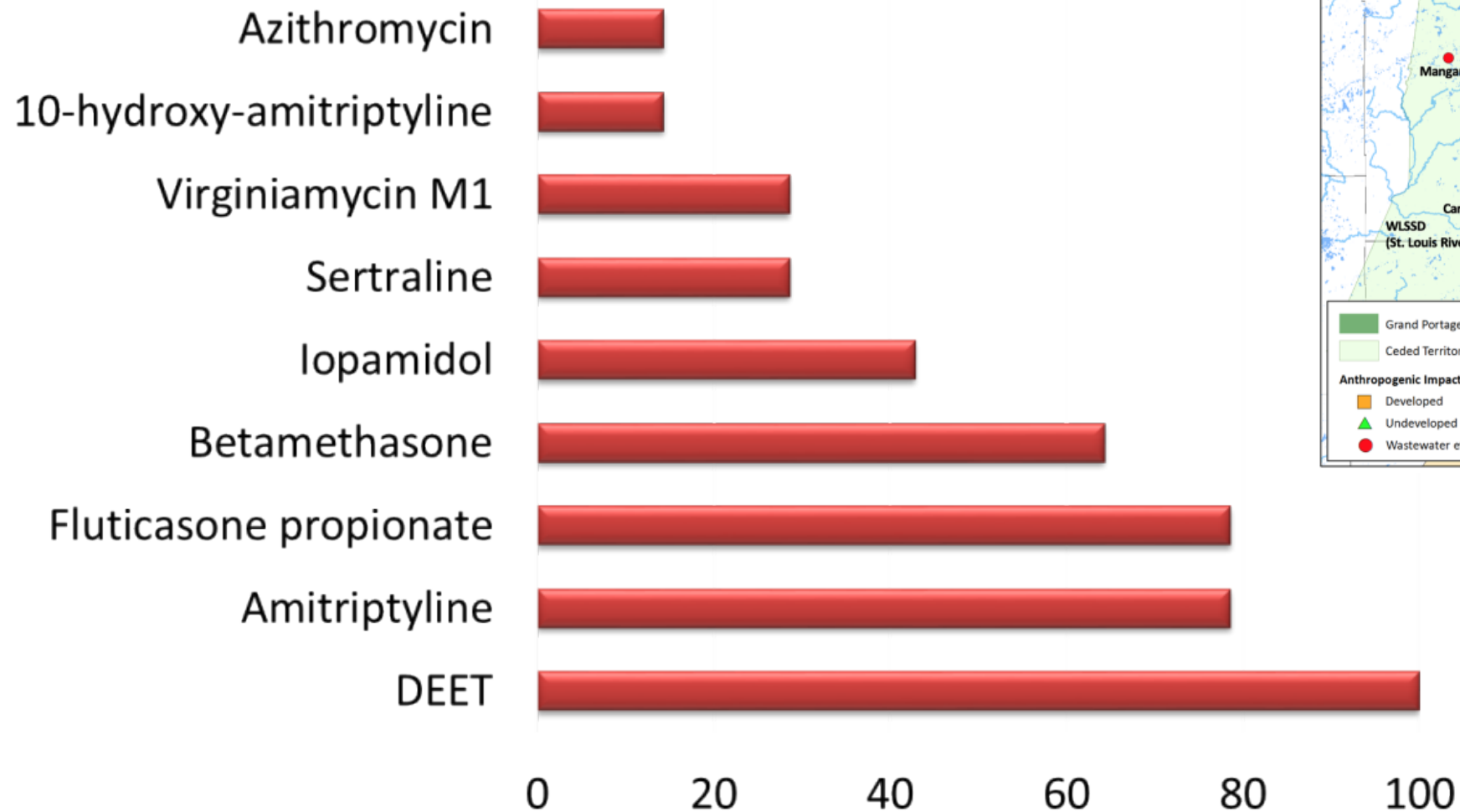
Water flow over fish gill:

5g fathead minnow: 795 Liter/day

830 g smallmouth bass: 6,134 Liter/day

5 therapeutic doses per day!

Percent Sites with Chemical Detections in Fish



Biological Effects

- Dosing an experimental lake with 5 ng/L of the synthetic estrogen ethinylestradiol caused the minnow population to collapse. (*Kidd et al., 2007. PNAS*)
- 30 ng/L fluvoxamine (an SSRI) in water triggers immediate spawning in mussels. (*Fong, 1998. Biol. Bull.*)
- Fish are affected after exposure to 3 ng/L estradiol. (*Elliott et al., 2014. J. Amer. Water Res. Assoc.*)
- Iopamidol + Chloramine = iodinated disinfection byproducts (highly toxic compounds).

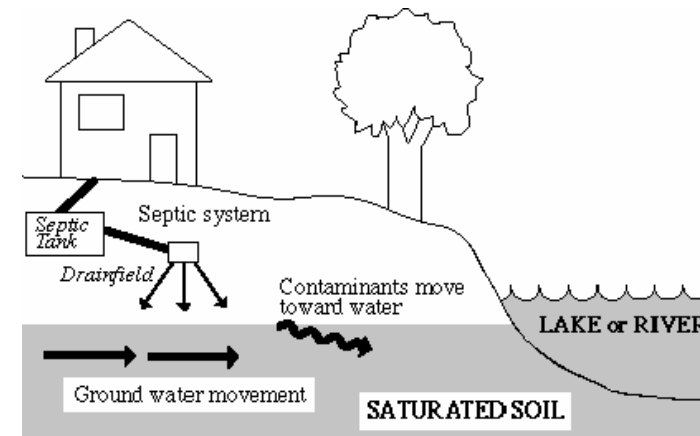
2023 Iowa Streams Study

- Total coliform resistance to extended spectrum beta-lactams (ESBL) in 94% of samples
- *E. coli* growth resistant to ESBLs (44%), cephalosporin (42%), and carbapenems (6%)
- Enterococci and staphylococci: “near ubiquitous” resistance with 94% vancomycin resistance (VRE) and 100% penicillinase-resistant *S. aureus* (MRSA)
- Antibiotic resistance gene signatures were more prevalent than antibiotics in streams.

Givens et al., 2023. Sci. Tot. Environ. 904: 166753

15 pharmaceuticals and other chemicals detected in the littoral zone of selected Minnesota lakes

(Guyader et al., 2018. *Sci. Tot. Environ.*)



(University of Minnesota Extension, 2008)

123 chemicals of 384 tested found in storm water, including human and veterinary pharmaceuticals

(Fairbairn et al., 2018. *Water Res.*)



	Rain (ng/L)	Snow (ng/L)	Air (ng/m ³)	Lake Water (ng/L)
Naproxen		3.74		
Iopamidol		228		685
Ciprofloxacin	10.3			19.4
Enrofloxacin	2.97			
Sulfamethoxazole		2.84		57.1
Ofloxacin			0.013	8.9
Caffeine			0.111	42.9
Cocaine	0.806	0.171	0.033	5.3
Benzoyllecgonine		0.477		9.5

Other sources?

- 7 million tons of biosolids produced by WWTPs annually
- Pharmaceuticals are commonly found in biosolids, including antibiotics ciprofloxacin, sulfamethoxazole, enrofloxacin, and ofloxacin from 3 – 6800 ppm (*McLellan and Halden, 2010*)
- 60% of biosolids are applied to agricultural fields in the U.S.
- 7 mg of PM₁₀ produced for every 1 kg of land-spread biosolids (*Paez-Rubio, 2007*)

Contaminants detected in airborne particulates and precipitation, including polar chemicals with low vapor pressure such as antibiotics and iopamidol, could originate from the land spreading of biosolids.



MPCA

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Summer Streets

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Mille Lacs Band of Ojibwe

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The Minnesota *Clean Water, Land and Legacy Amendment* was approved in 2008. The amendment increased the sales and use tax rate by three-eighths of one percent on taxable sales from 2009 through 2034.

Approximately 33% of the revenue goes into the *Clean Water Fund* to protect, enhance, and restore water quality in lakes, rivers, streams, and groundwater in Minnesota.