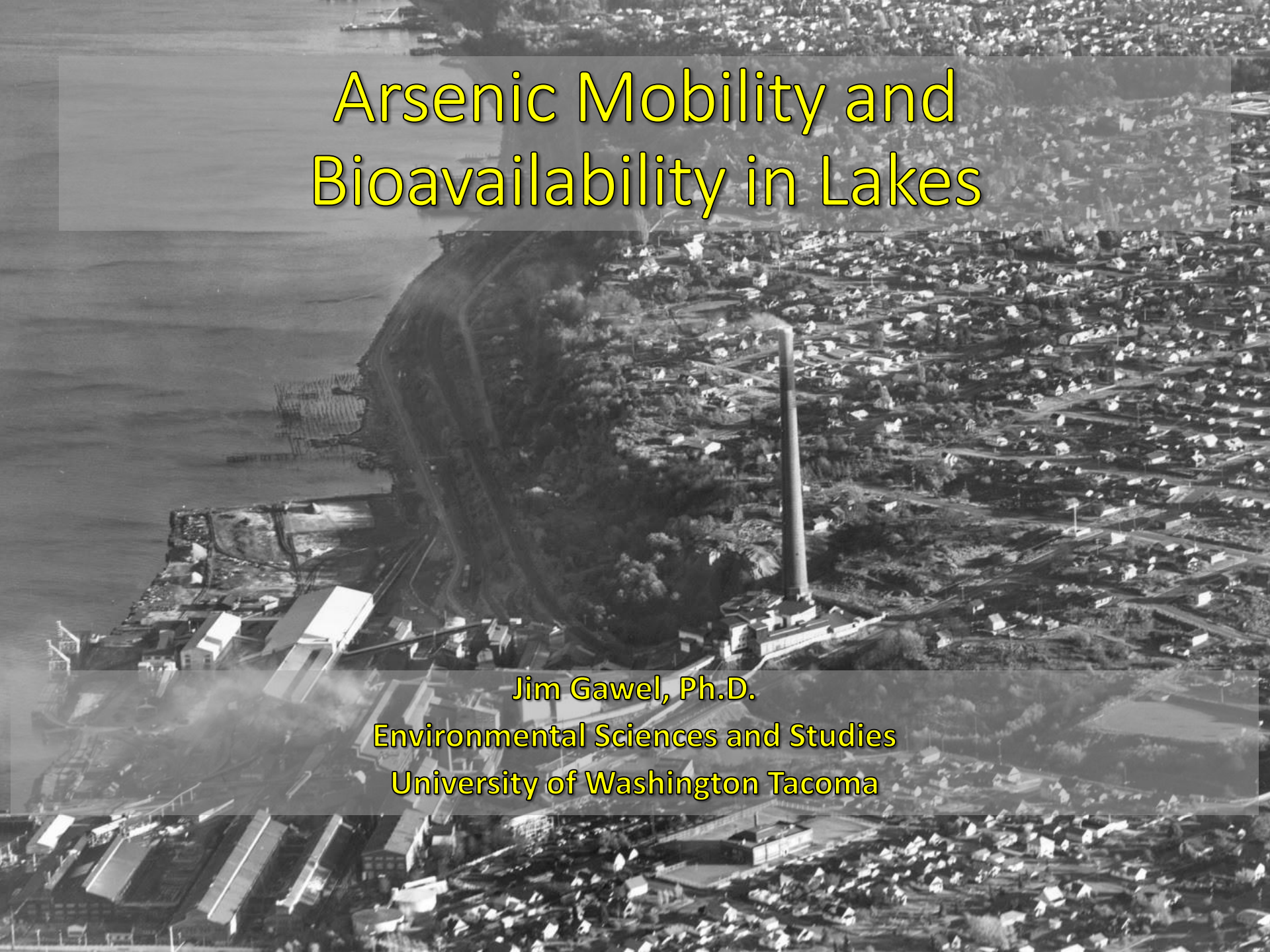
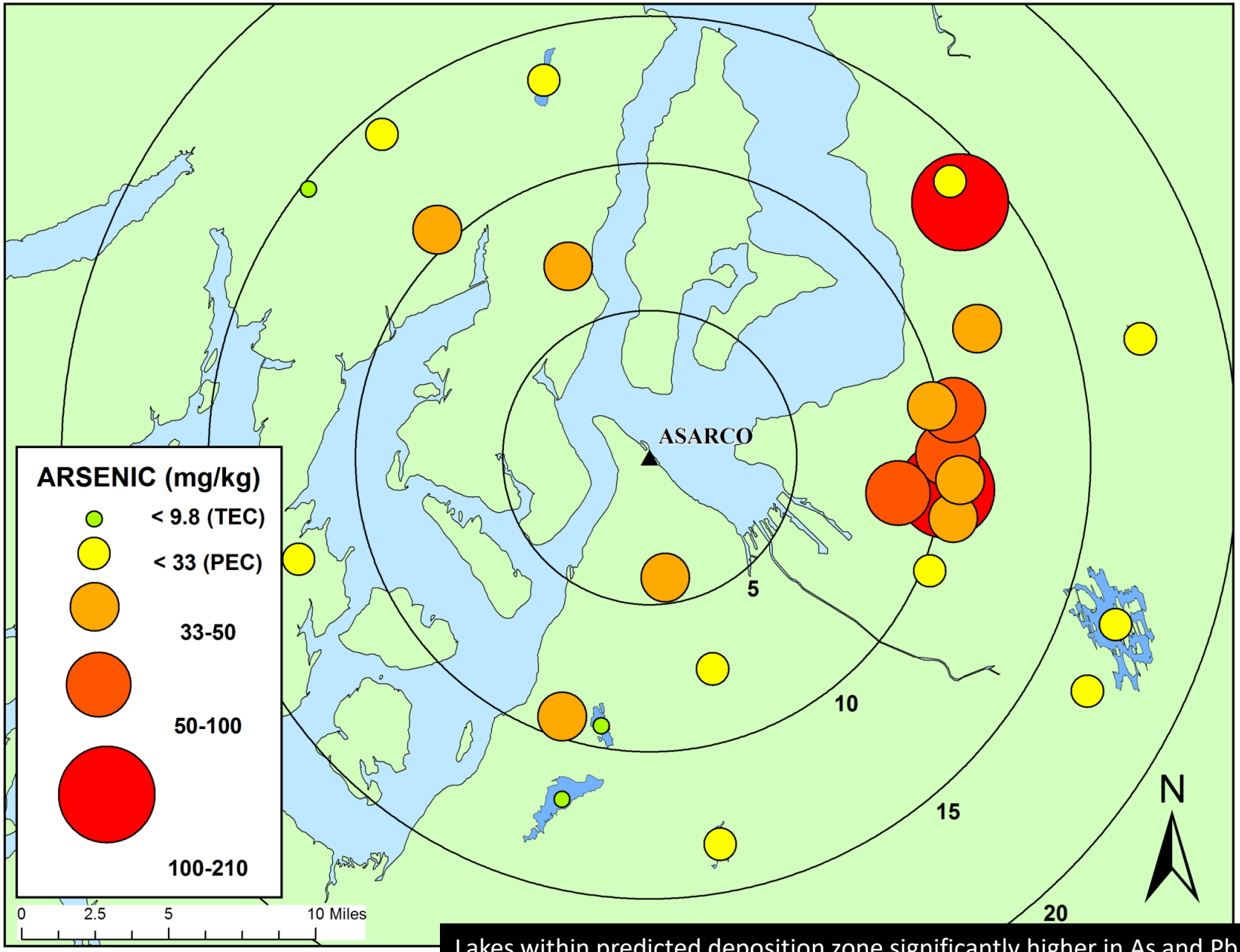
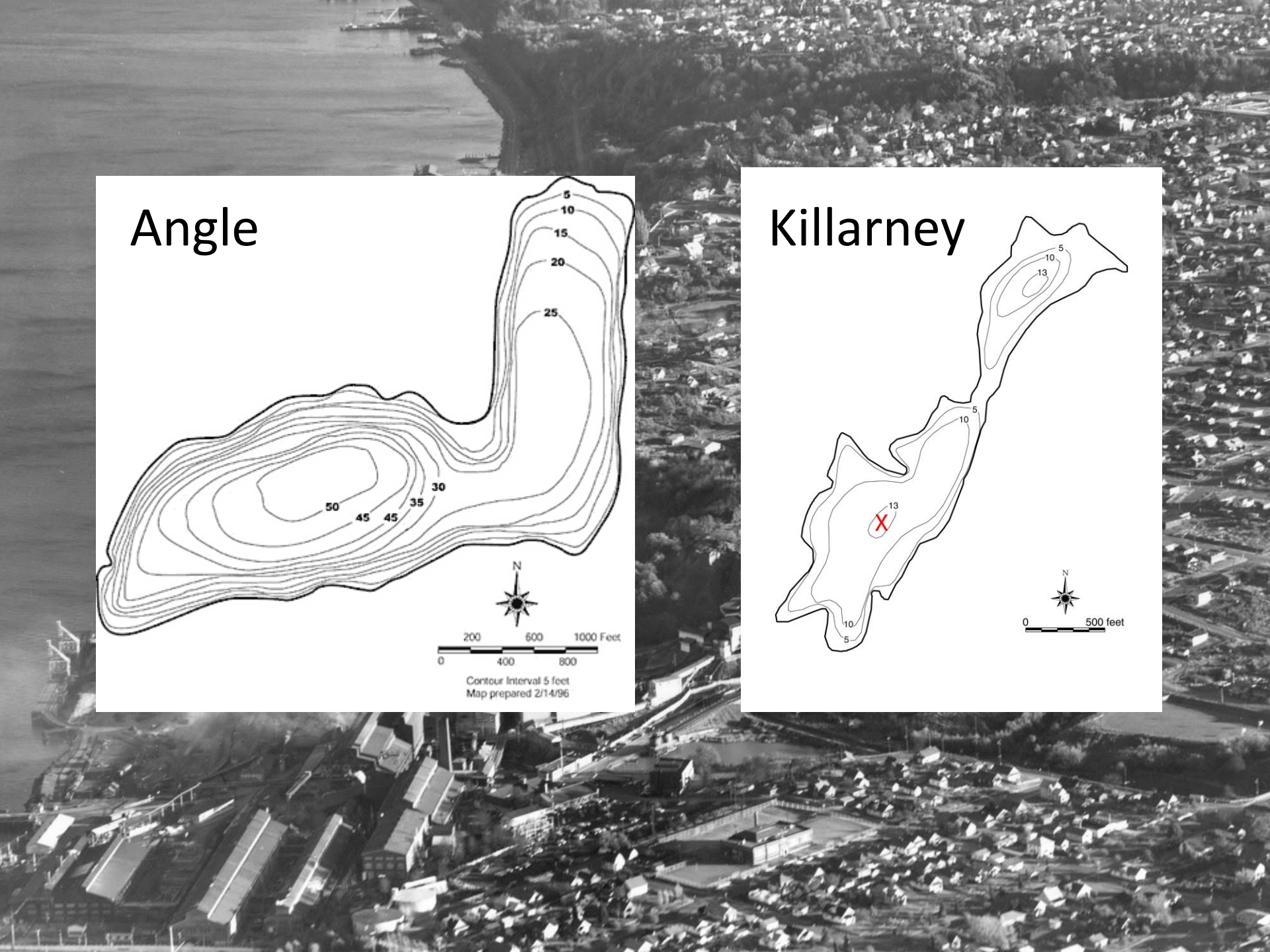


Arsenic Mobility and Bioavailability in Lakes

Jim Gawel, Ph.D.
Environmental Sciences and Studies
University of Washington Tacoma

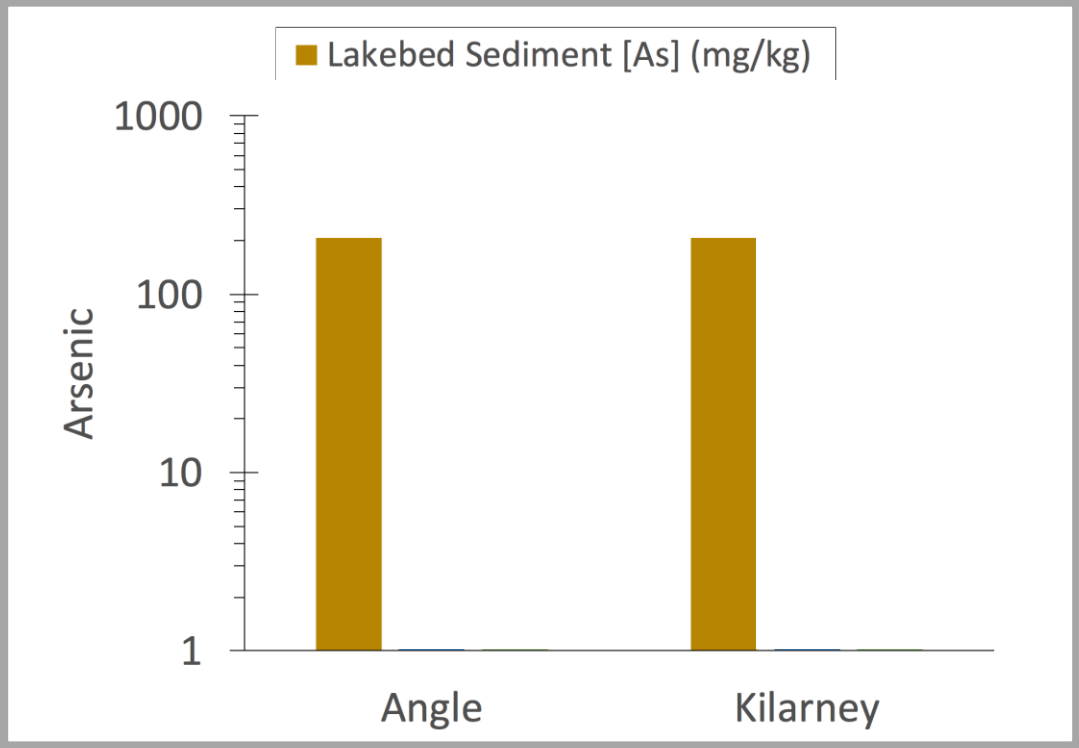


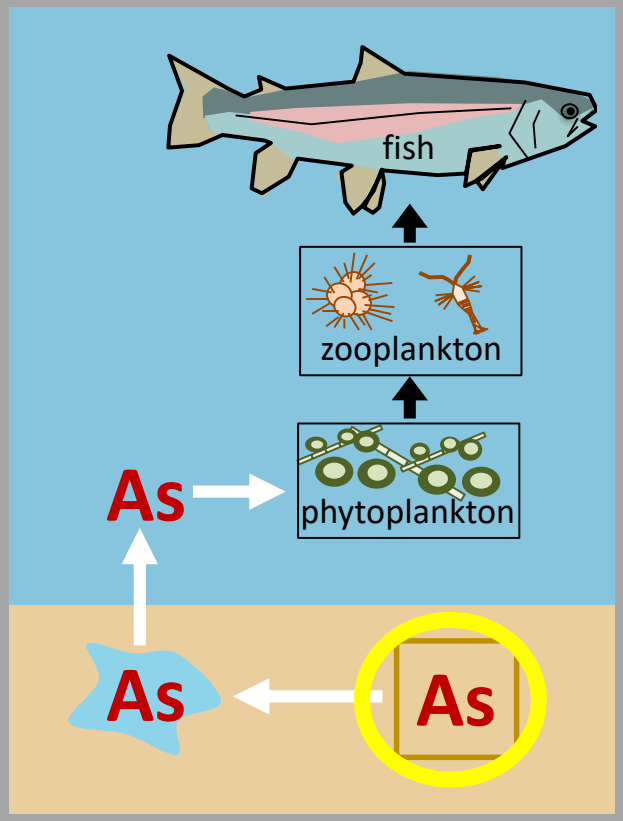
Lakes within predicted deposition zone significantly higher in As and Pb


Angle
5
10
15
20
25
30
35
45
45
50
N
200
600
1000 Feet
0
400
800
Contour Interval 5 feet
Map prepared 2/14/96
Killarney
5
10
13
5
10
13
X
10
5
N
0
500 feet

Arsenic uptake into food web

Note: log scale!

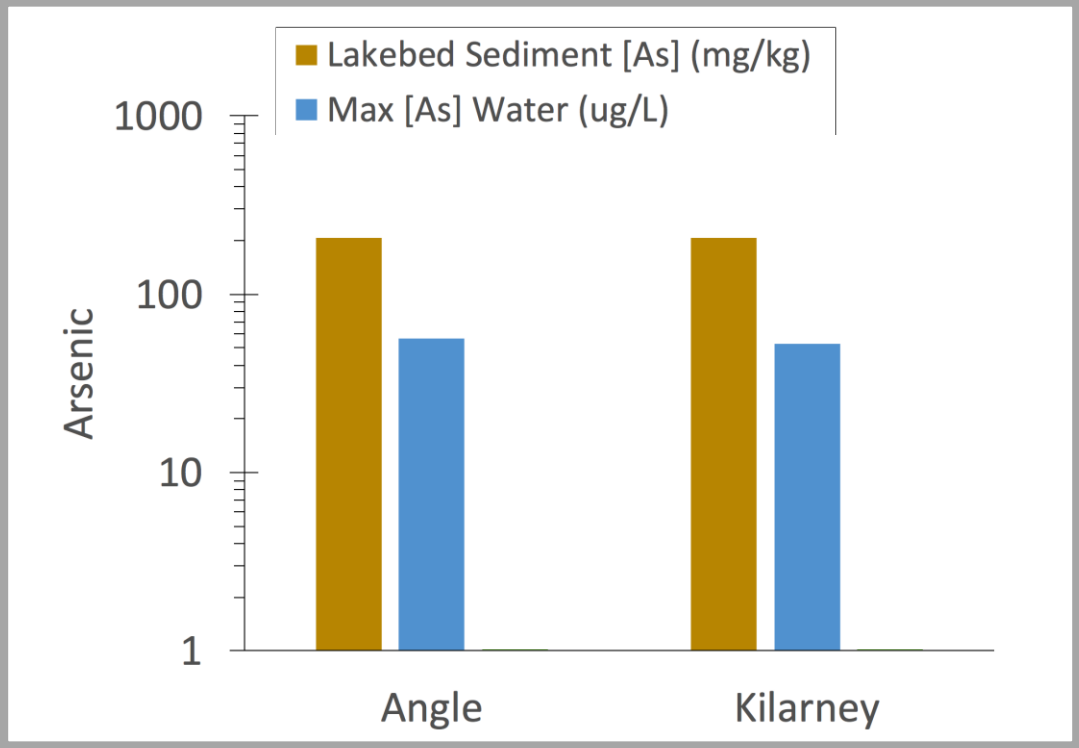


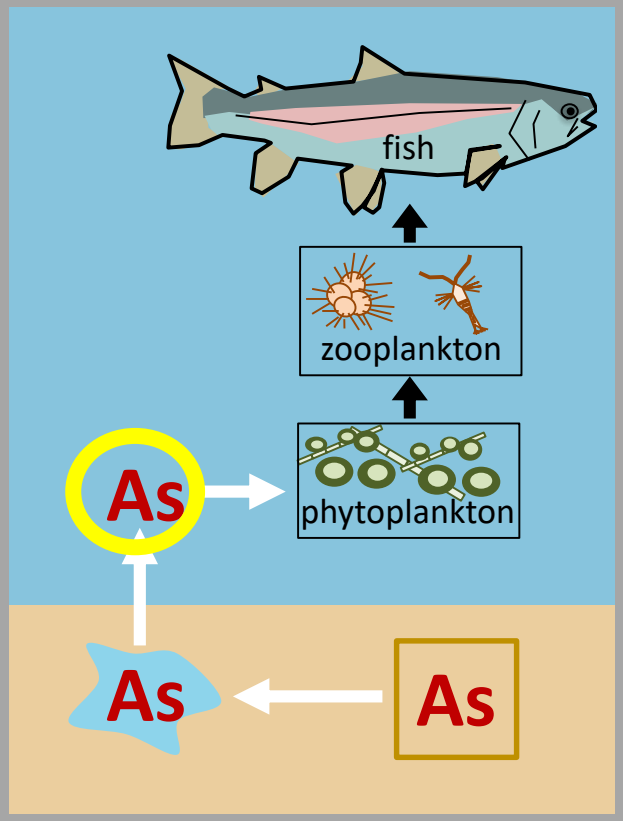


Barrett, P. M., Hull, E. A., King, C. E., Burkart, K., Ott, K. A., Ryan, J. N., Gawel, J. E., Neumann, R. B. (2018). Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. *Science of The Total Environment*, *625*, 1606–1614.

Arsenic uptake into food web

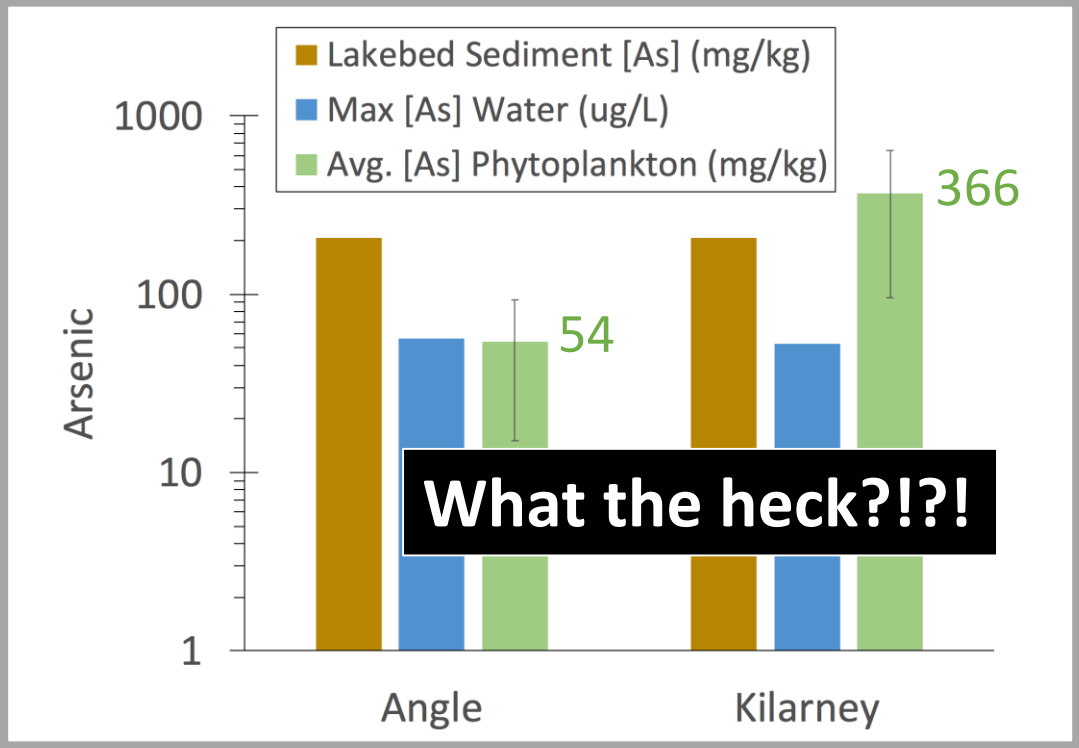
Note: log scale!



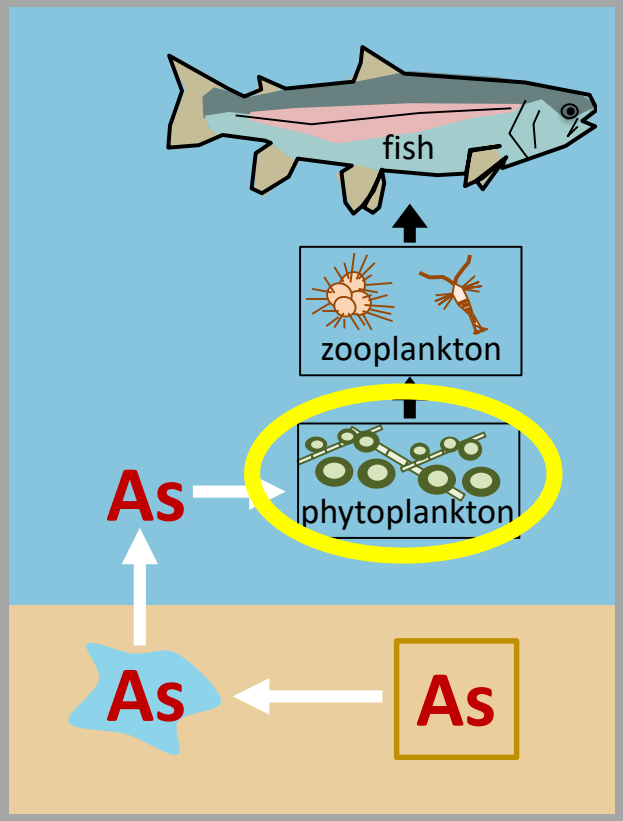


Barrett, P. M., Hull, E. A., King, C. E., Burkart, K., Ott, K. A., Ryan, J. N., Gawel, J. E., Neumann, R. B. (2018). Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. *Science of The Total Environment*, *625*, 1606–1614.

Arsenic uptake into food web

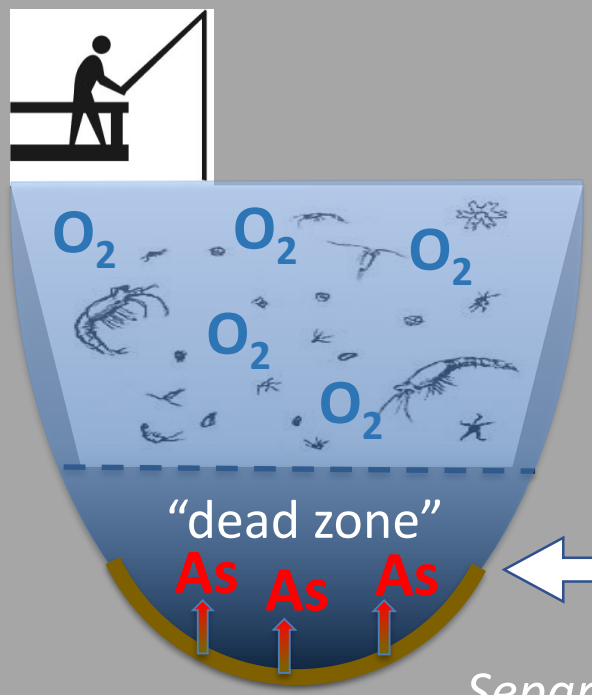


Note: log scale!

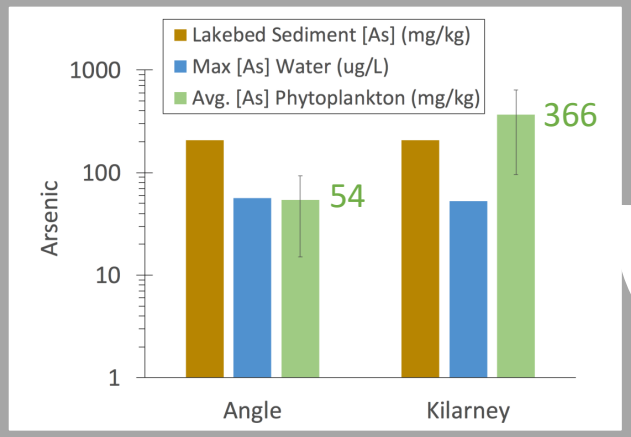


Barrett, P. M., Hull, E. A., King, C. E., Burkart, K., Ott, K. A., Ryan, J. N., Gawel, J. E., Neumann, R. B. (2018). Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. *Science of The Total Environment*, *625*, 1606–1614.

Arsenic uptake into food web



Arsenic in waters **WITHOUT** oxygen

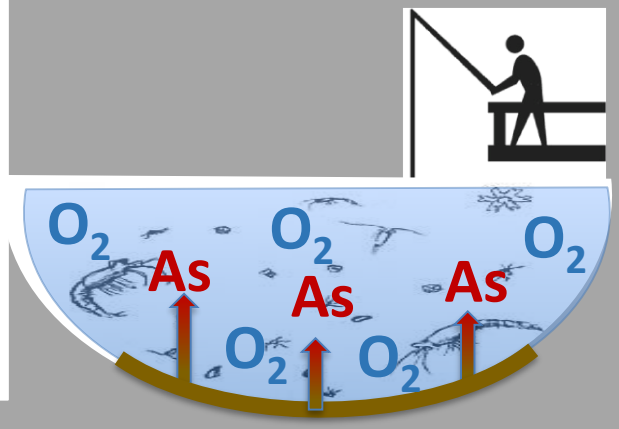


Deep Lake

Separation between arsenic & aquatic life

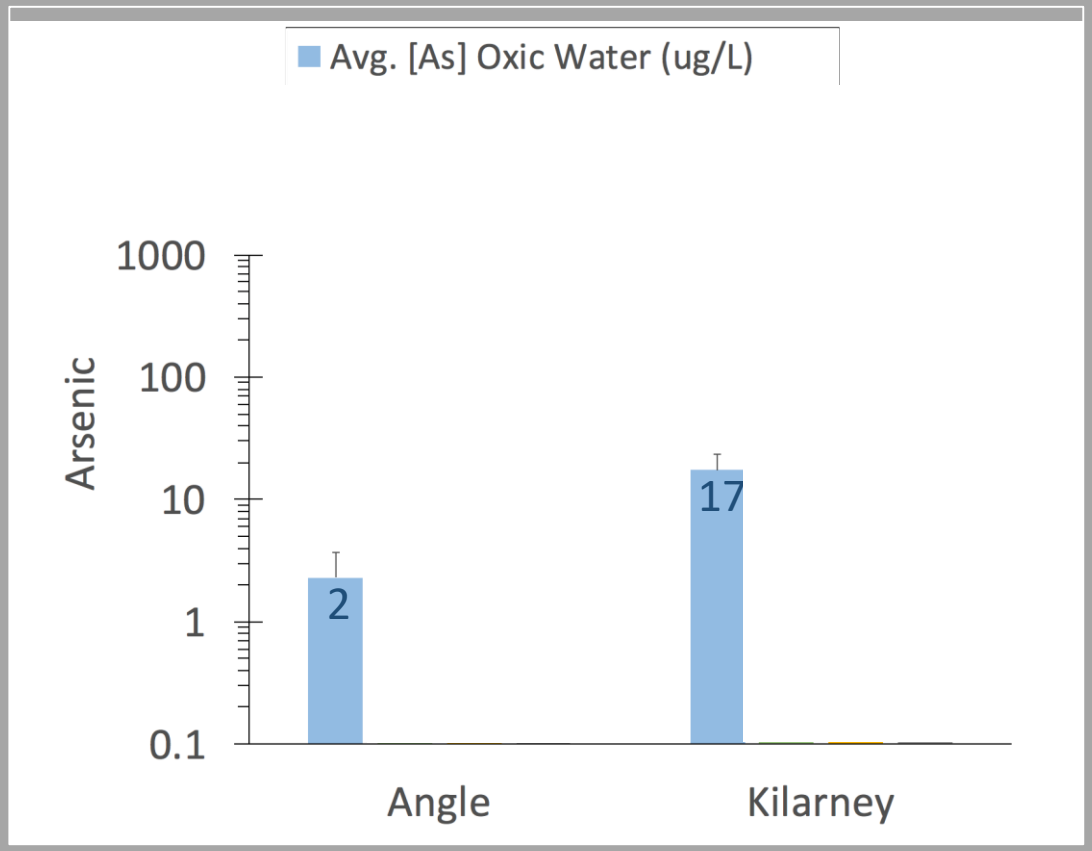
Shallow Lake

Overlap between arsenic & aquatic life

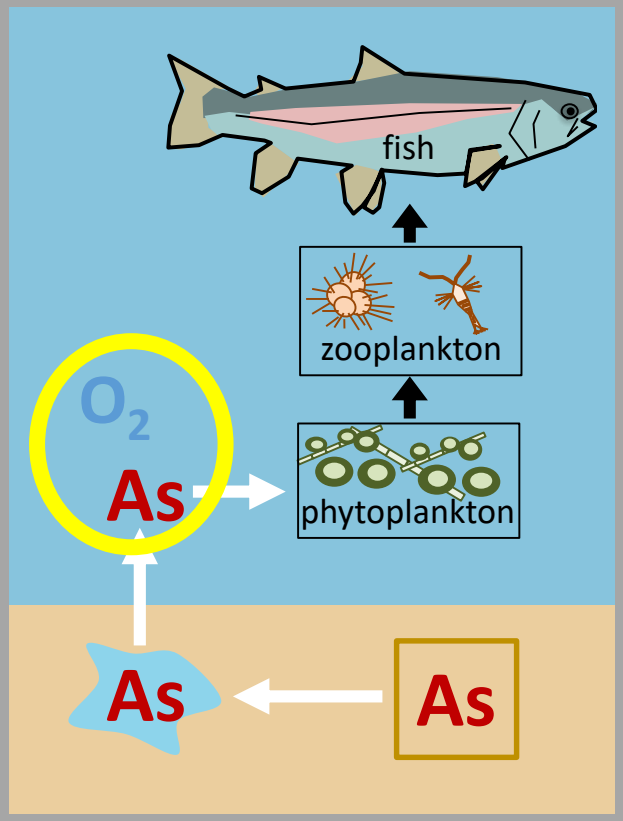


Arsenic in waters **WITH** oxygen

Arsenic uptake into food web

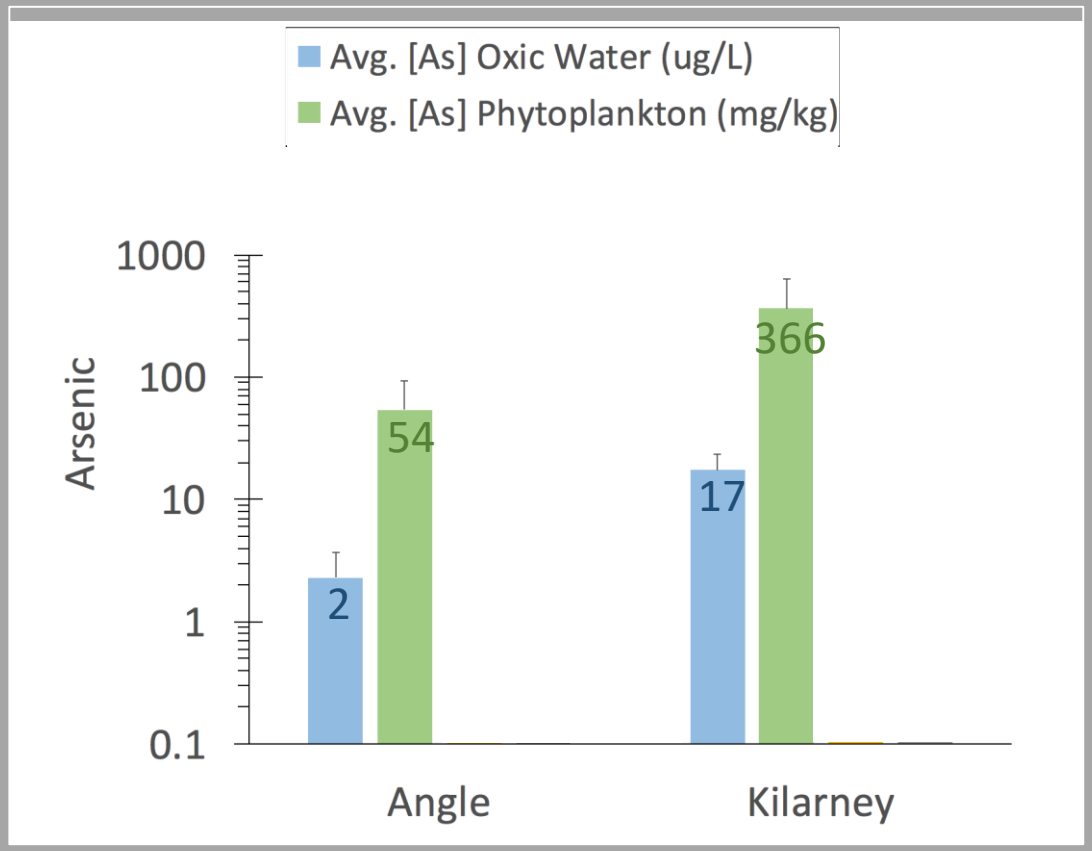


Note: log scale!

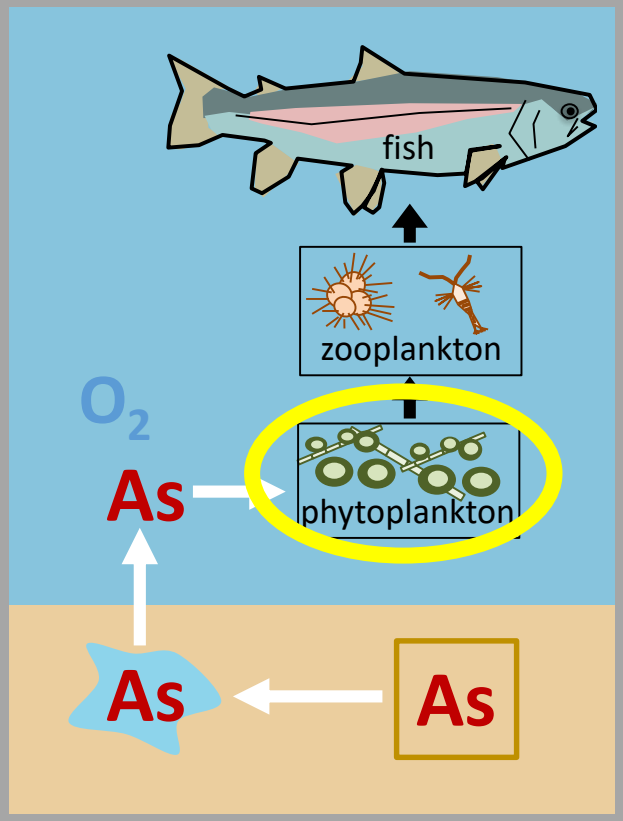


Barrett, P. M., Hull, E. A., King, C. E., Burkart, K., Ott, K. A., Ryan, J. N., Gawel, J. E., Neumann, R. B. (2018). Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. *Science of The Total Environment*, *625*, 1606–1614.

Arsenic uptake into food web

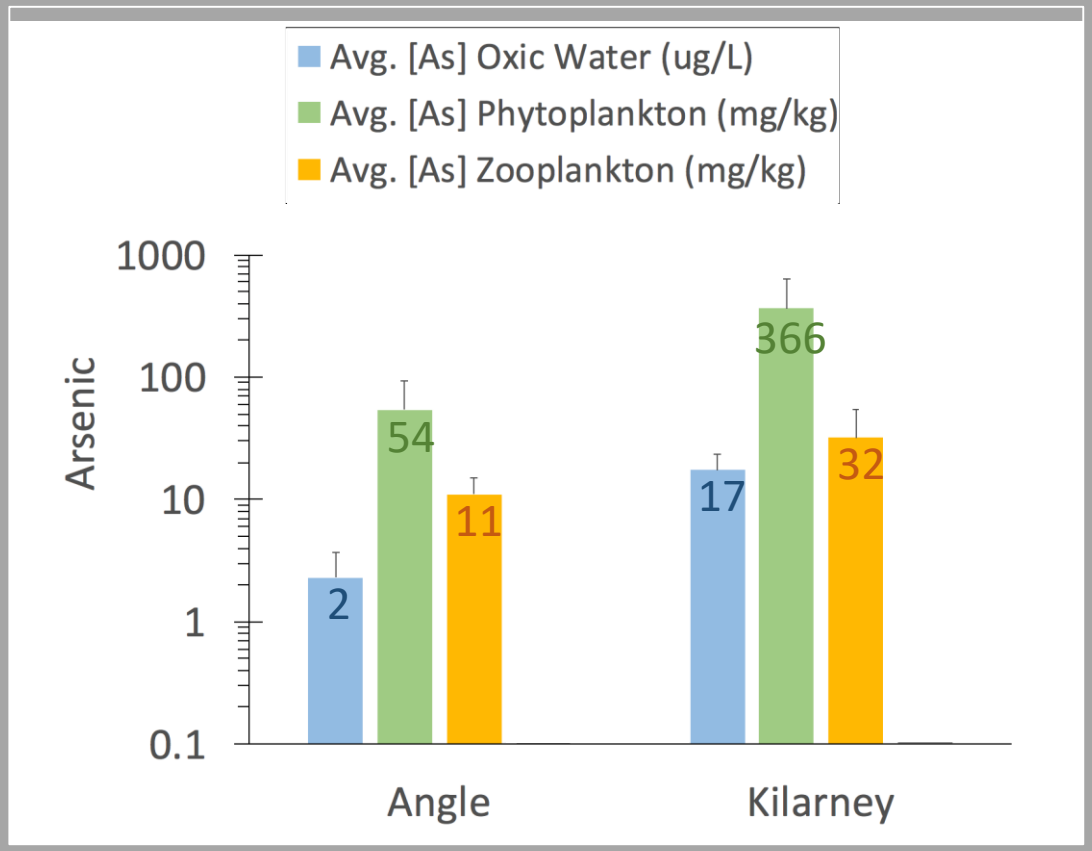


Note: log scale!

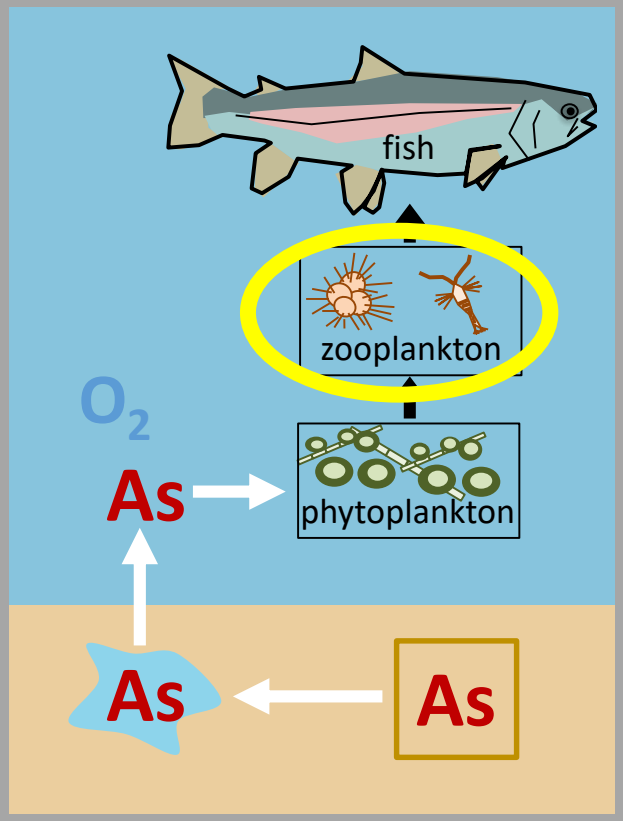


Barrett, P. M., Hull, E. A., King, C. E., Burkart, K., Ott, K. A., Ryan, J. N., Gawel, J. E., Neumann, R. B. (2018). Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. *Science of The Total Environment*, *625*, 1606–1614.

Arsenic uptake into food web



Note: log scale!



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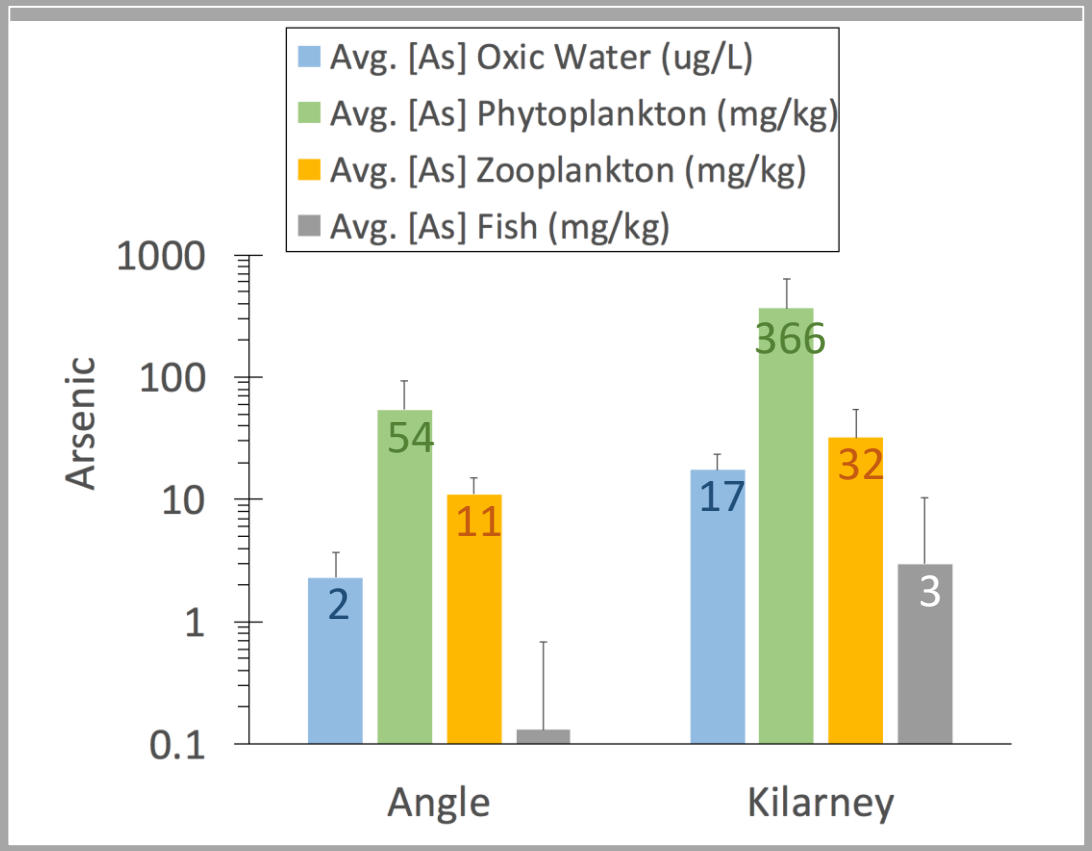
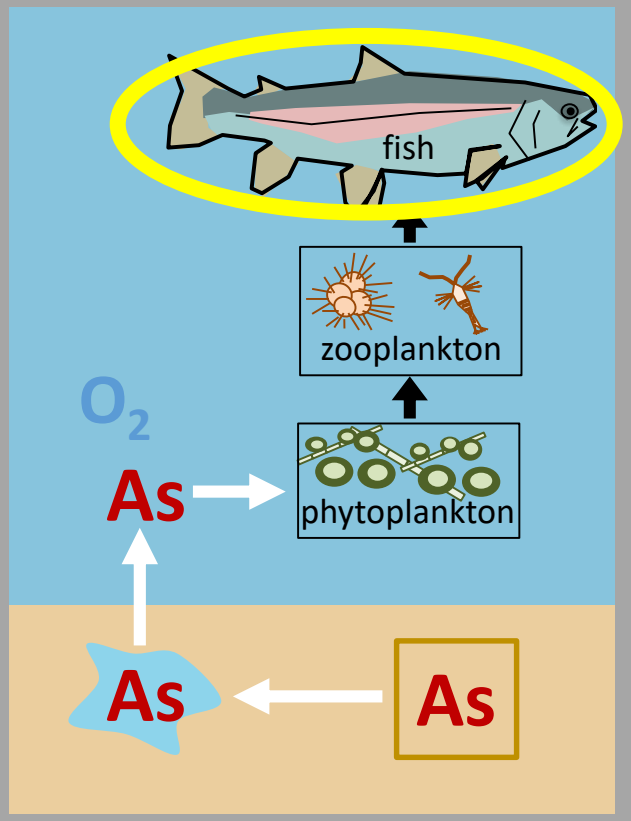
Arsenic uptake into food web

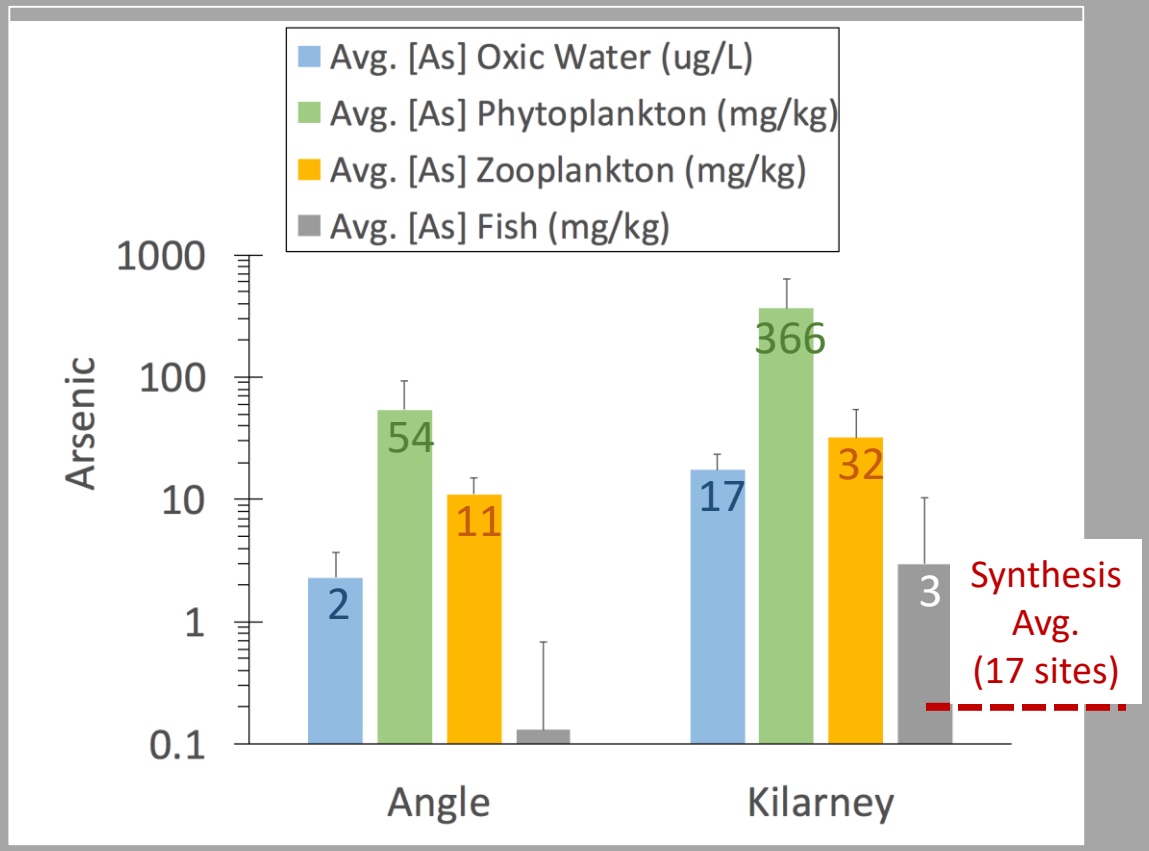


Note: log scale!

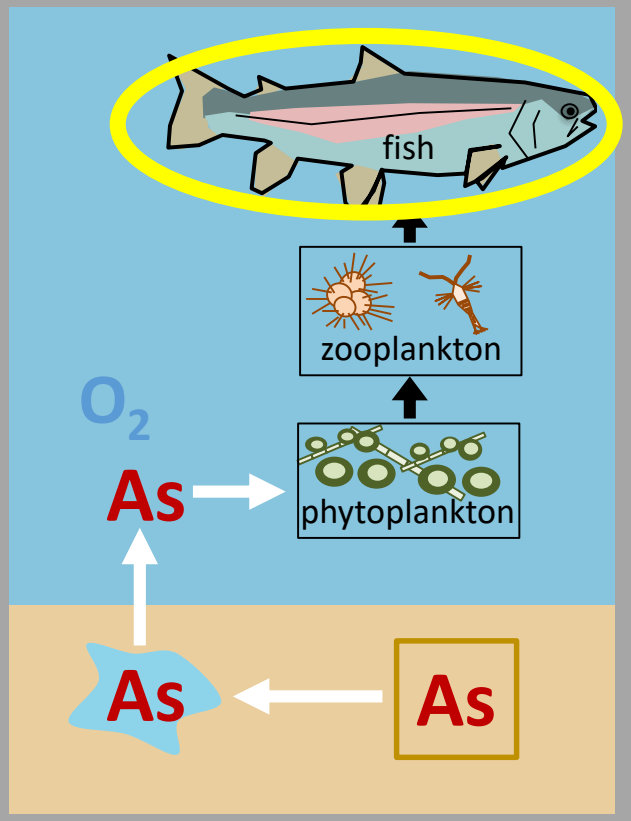


Barrett, P. M., Hull, E. A., King, C. E., Burkart, K., Ott, K. A., Ryan, J. N., Gawel, J. E., Neumann, R. B. (2018). Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. *Science of The Total Environment*, *625*, 1606–1614.

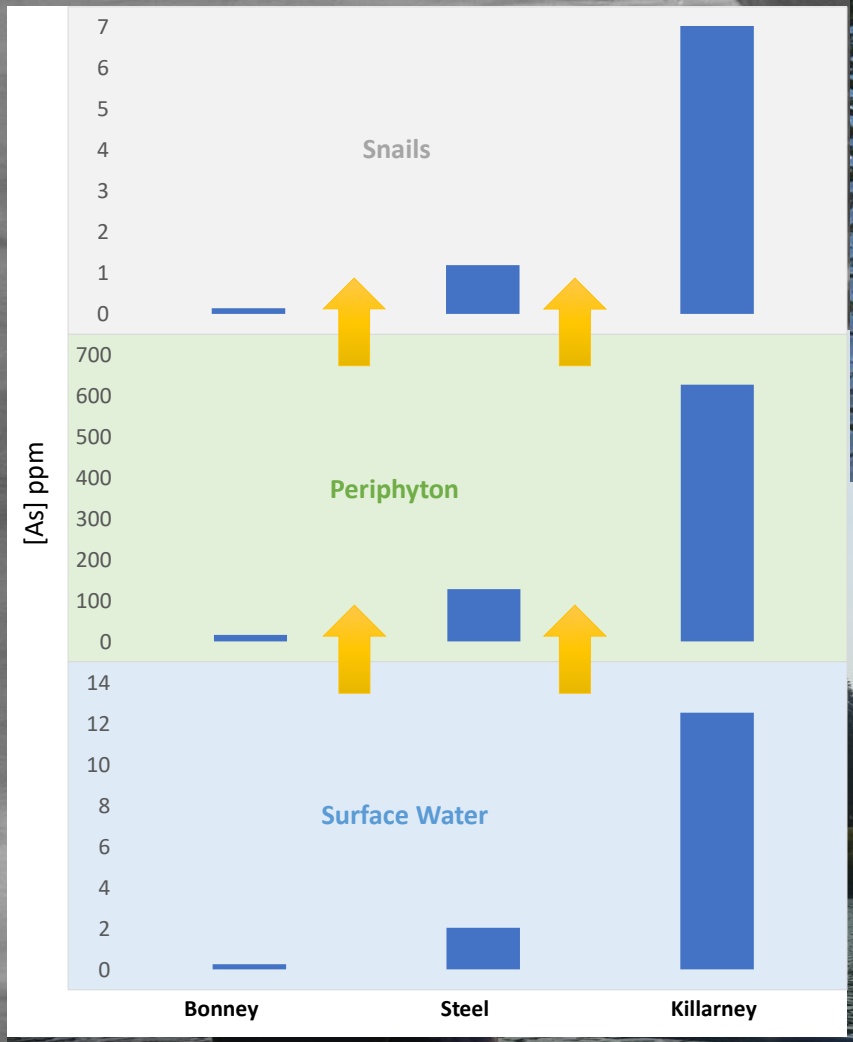
Arsenic uptake into food web

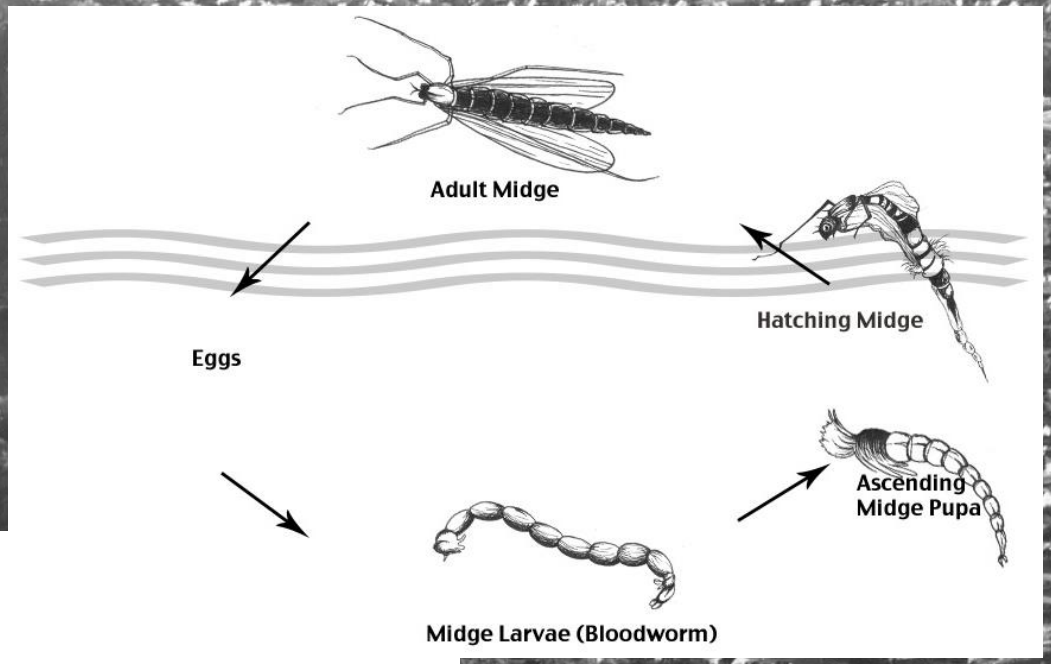


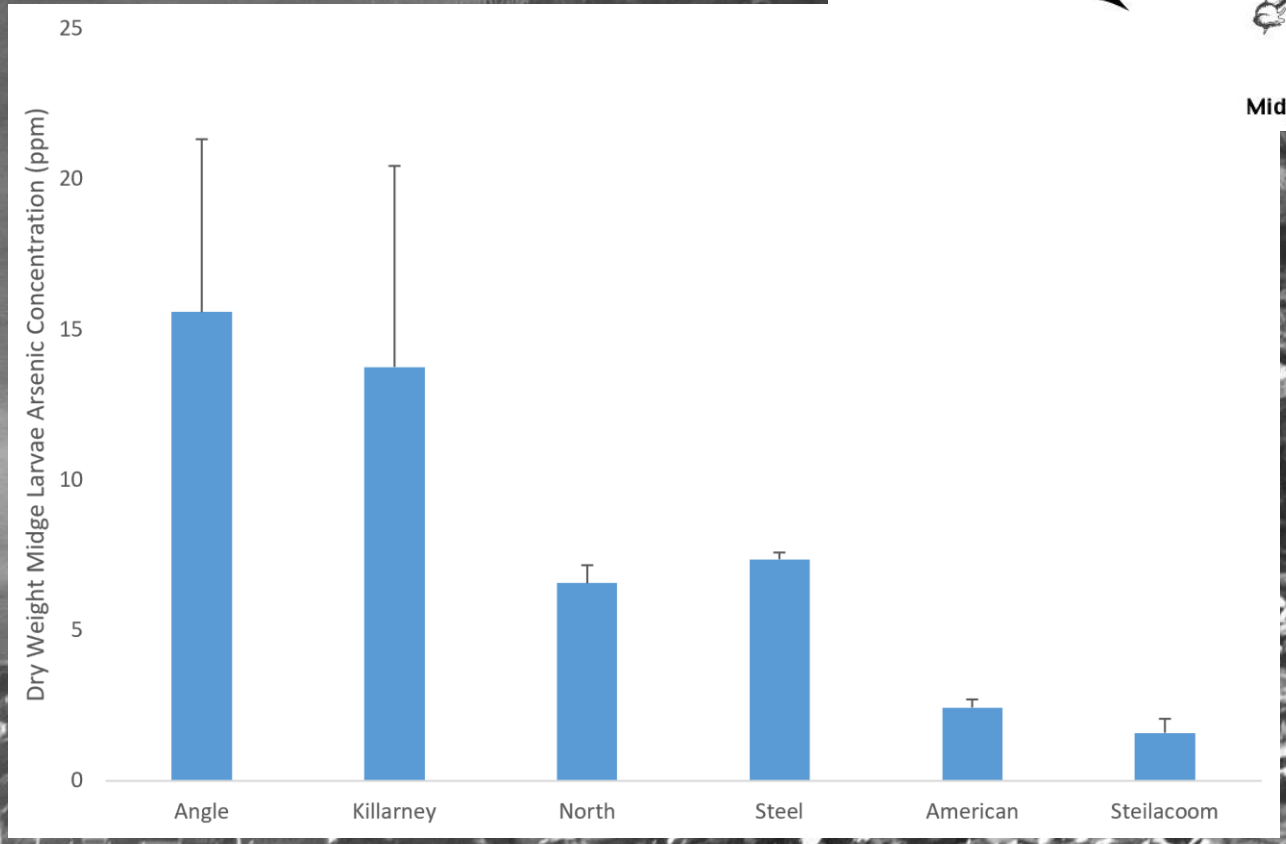
Note: log scale!

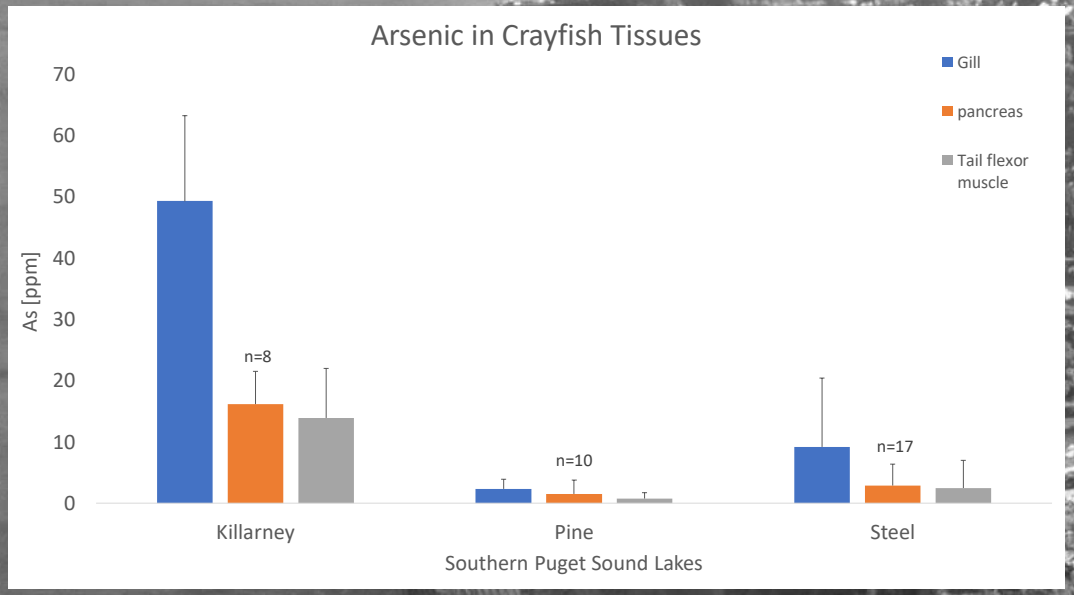


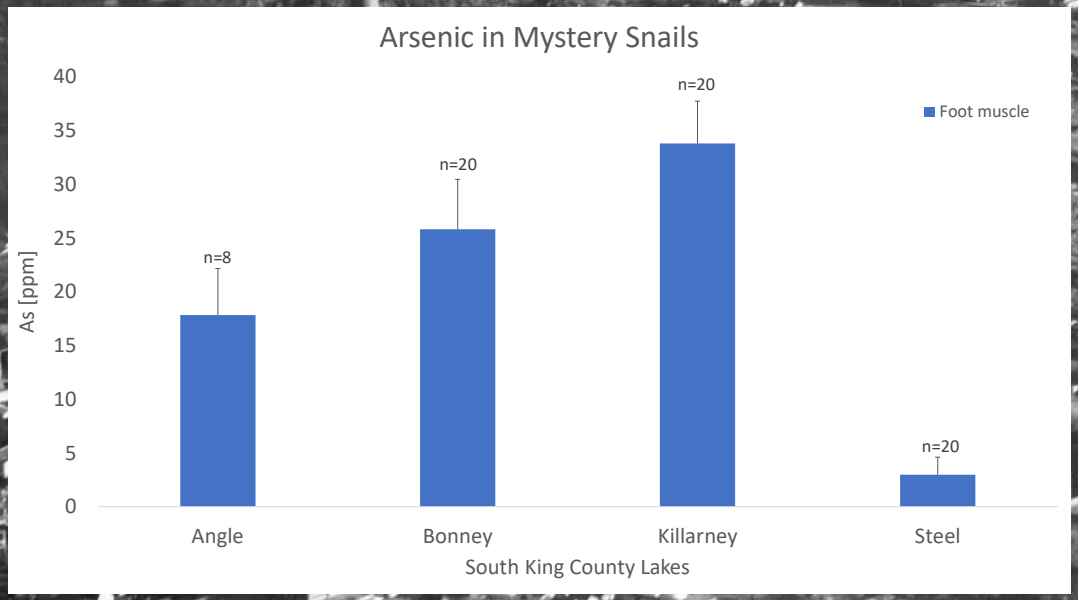
Williams, L., Schoof, R. A., Yager, J. W., & Goodrich-Mahoney, J. W. (2006). Arsenic Bioaccumulation in Freshwater Fishes. *Human and Ecological Risk Assessment: An International Journal*, *12*(5), 904–923.


[As] ppm
Snails
Periphyton
Surface Water
7
6
5
4
3
2
1
0
700
600
500
400
300
200
100
0
14
12
10
8
6
4
2
0
Bonney
Steel
Killarney


Adult Midge
Hatching Midge
Eggs
Ascending Midge Pupa
Midge Larvae (Bloodworm)


Dry Weight Midge Larvae Arsenic Concentration (ppm)
25
20
15
10
5
0
Angle
Killarney
North
Steel
American
Steilacoom


Arsenic in Crayfish Tissues
Gill
pancreas
Tail flexor muscle
As [ppm]
70
60
50
40
30
20
10
0
n=8
n=10
n=17
Killarney
Pine
Steel
Southern Puget Sound Lakes


Arsenic in Mystery Snails
Foot muscle
As [ppm]
40
35
30
25
20
15
10
5
0
n=8
n=20
n=20
n=20
Angle
Bonney
Killarney
Steel
South King County Lakes

Acknowledgements:

- Volunteer divers Chuck Neudorf, Dave DeGroot
- This work was sponsored (in whole or in part) by the University of Washington Superfund Research Program, Grant #: NIEHS P42ES004696

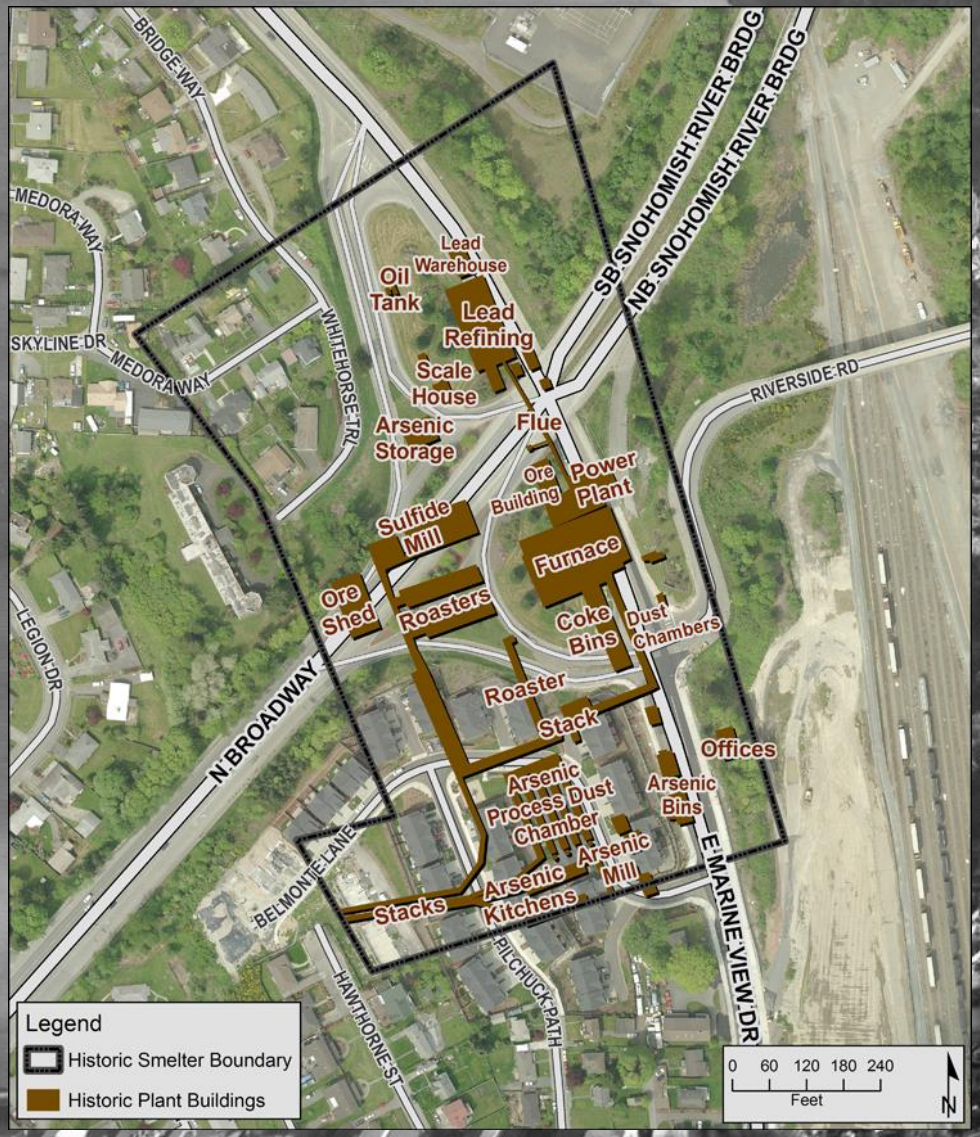
Research Team

Current Team:

- (UW Tacoma) Erin Hull, Ken Burkart, Noelle Hogan, Marco Barajas, Adam Matulich, Dr. Jack Vincent
- (UW Seattle) Dr. Becca Neumann, Dr. Alex Horner-Devine, Dr. Julian Olden, Dr. Evan Gallagher, Dr. Theo Bammler, and Samantha Fung

Past Team Members

- (UW Tacoma) Phil Hite, Ricky Pendergrass, Suji Kim, Corey King, Sarah Burdick, Michelle Miller, Jessica Asplund, Shawna Peterson, Kara Ziegler, Lindsay Tuttle, Patti Sandvik, Joe Chynoweth, Dena Reaugh, Ali Ehle, Anthony Endresen
- (UW Seattle) Dr. Pam Barrett
- (UC Boulder) Dr. Joe Ryan and Kimberly Ott
- (Bellarmine High School) Dave DeGroot, Tyler Vu, Lance Joseph, Amanda Tollefson, Brian Rurik



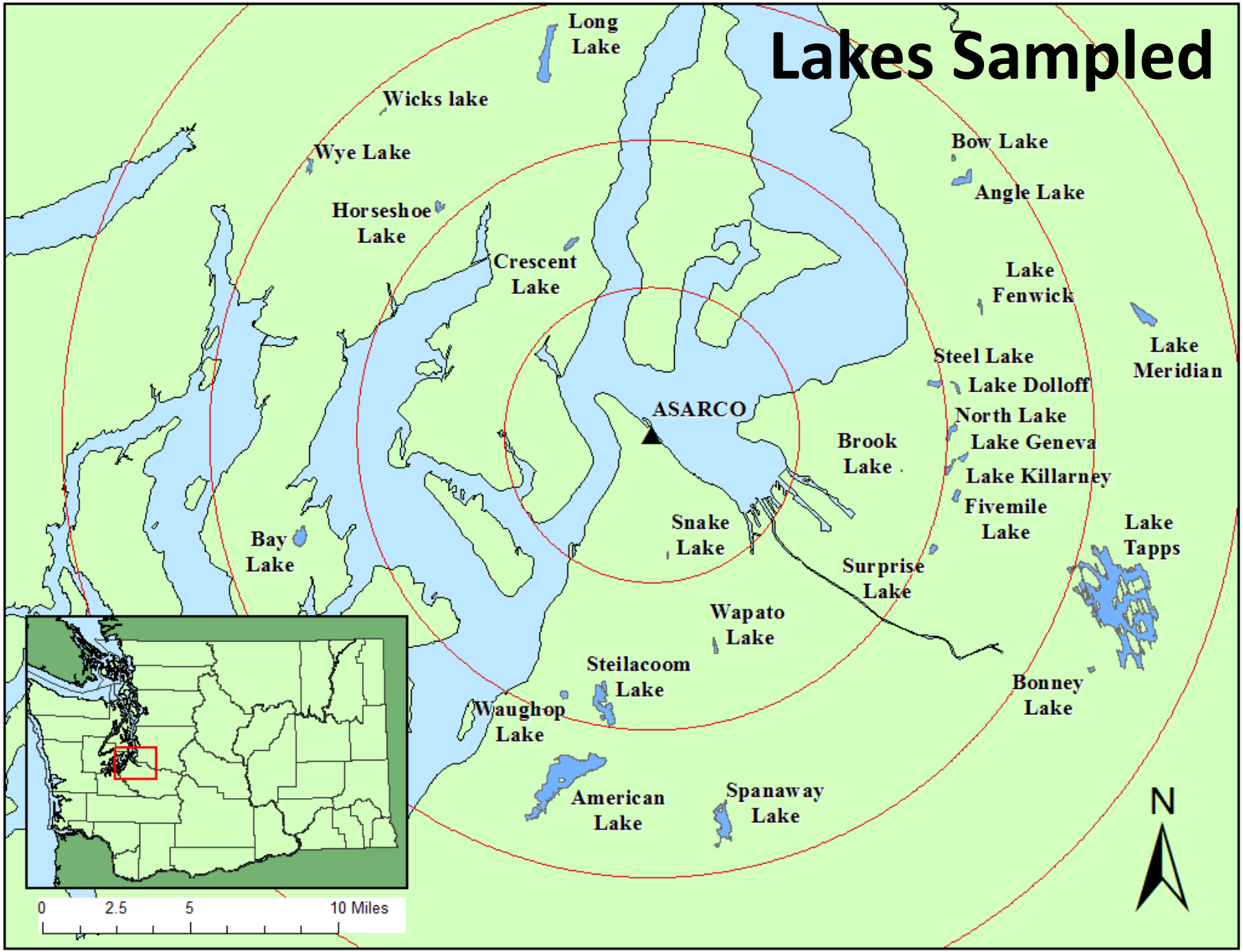
ASARCO History in Ruston, WA

- 1890 Lead smelting begins
- 1905 Conversion to copper smelter
- 1912 Arsenic recovery begun in Tacoma
- 1917 Tall stack constructed (700 ft asl) + electrostatic precipitators
- 1970 Meteorological Curtailment Program
- 1986 All smelting operations cease

Urbanization & Arsenic Pollution

- Urban poor rely on local, inexpensive recreational water resources; some rely on water sources for cultural ties and diet augmentation
- If remobilized periodically As may migrate to surface sediments
- Excess nutrients (eutrophication) can increase As release from sediments

Lakes Sampled

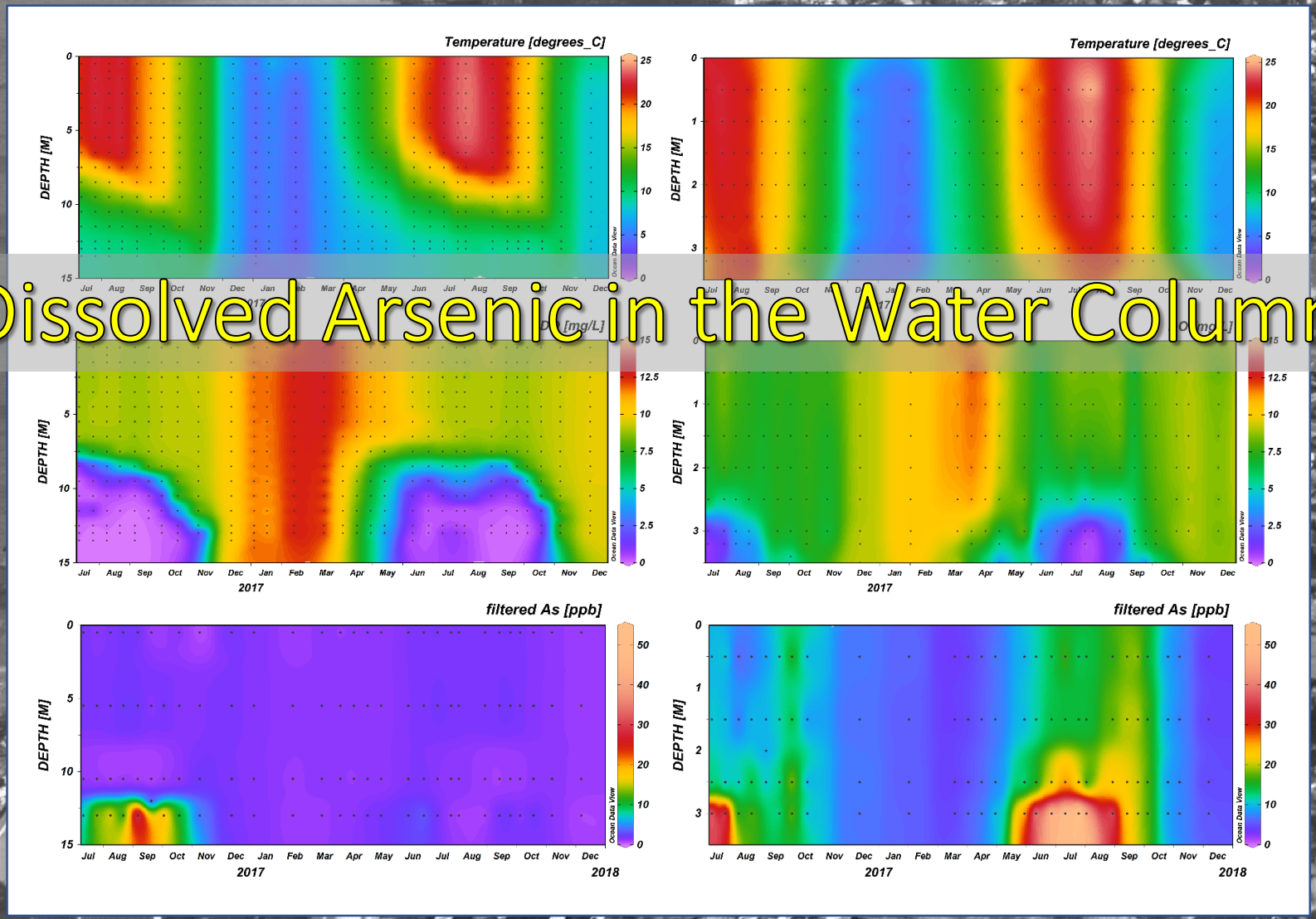


Wind Patterns

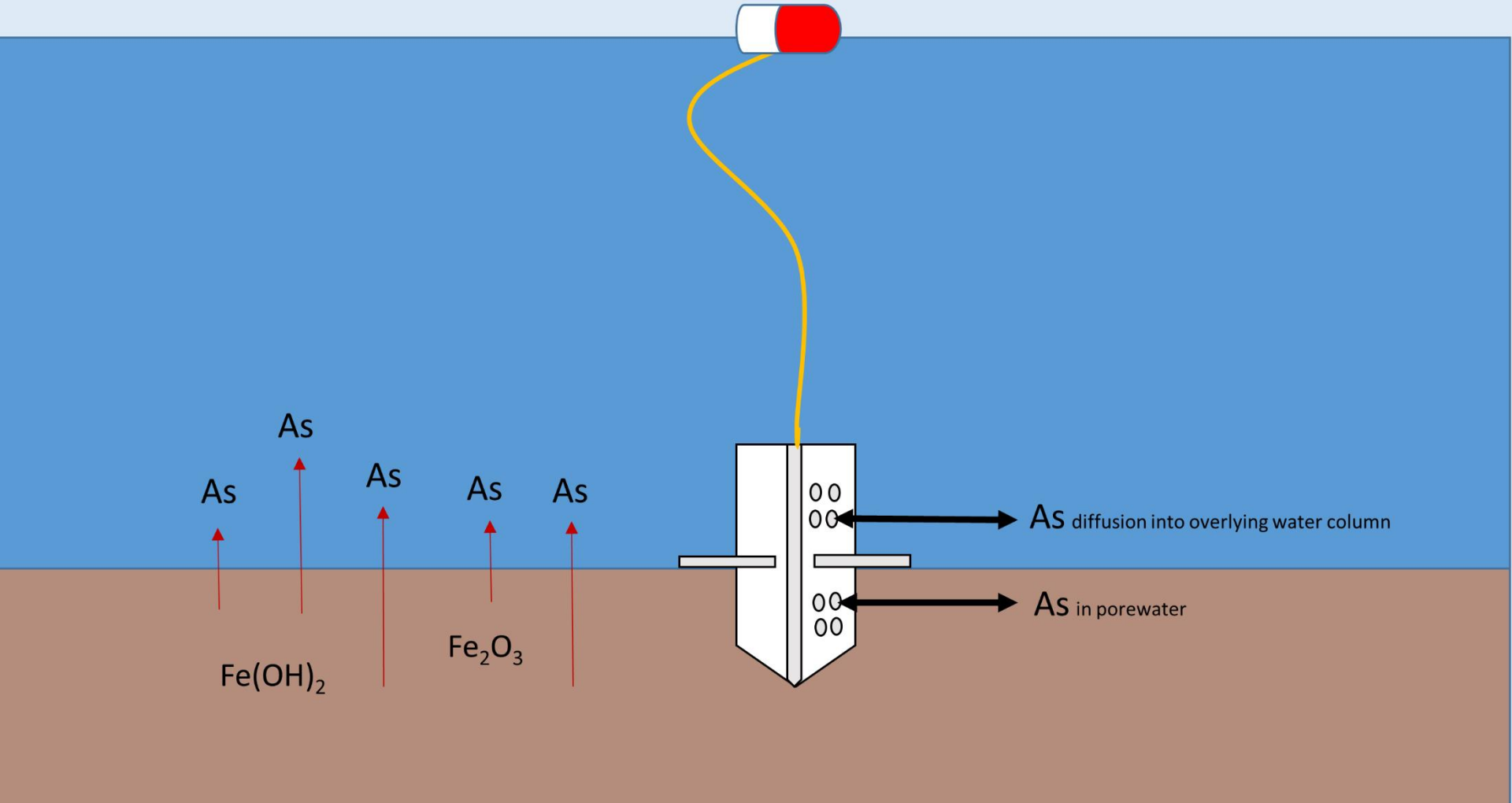
Angle Lake

Lake Killarney

Dissolved Arsenic in the Water Column

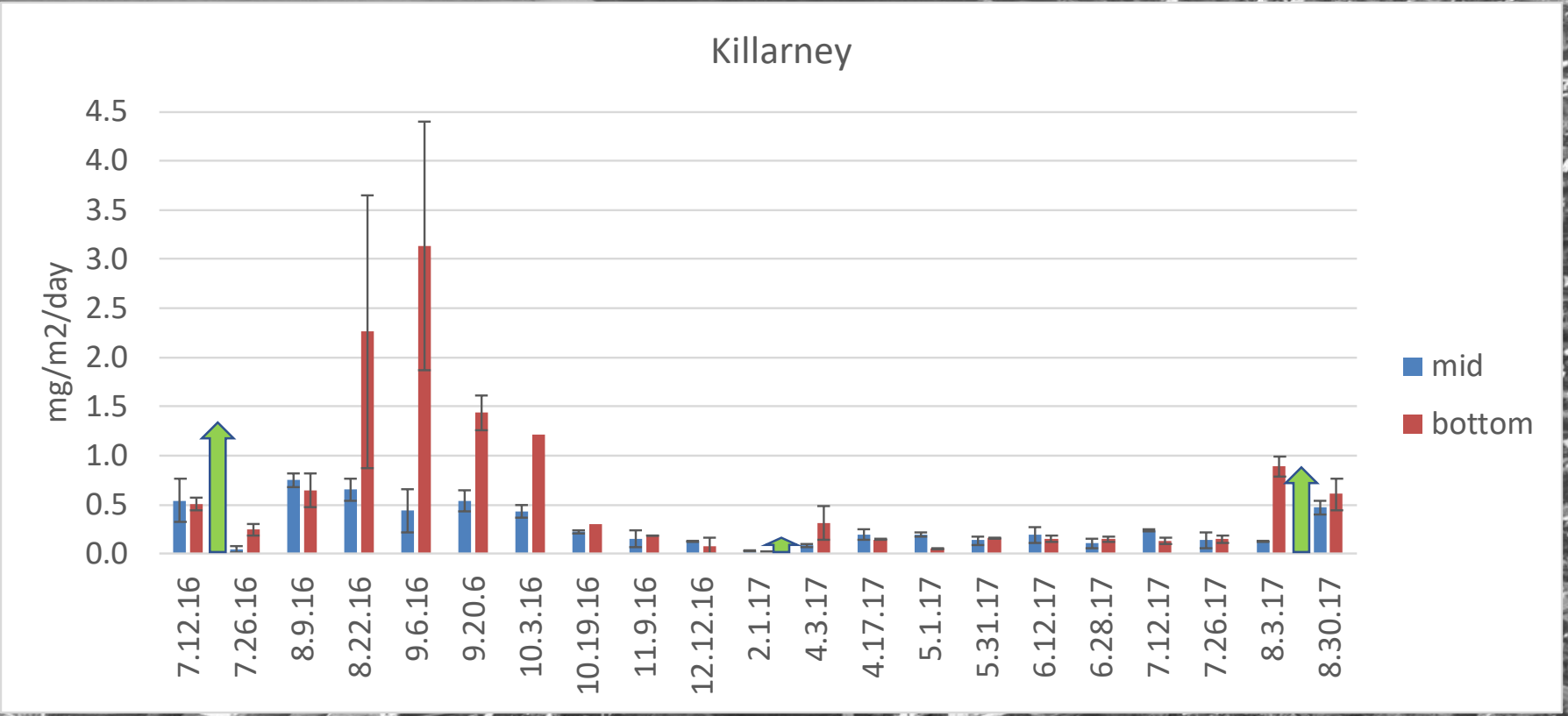


pore water sampling using peepers



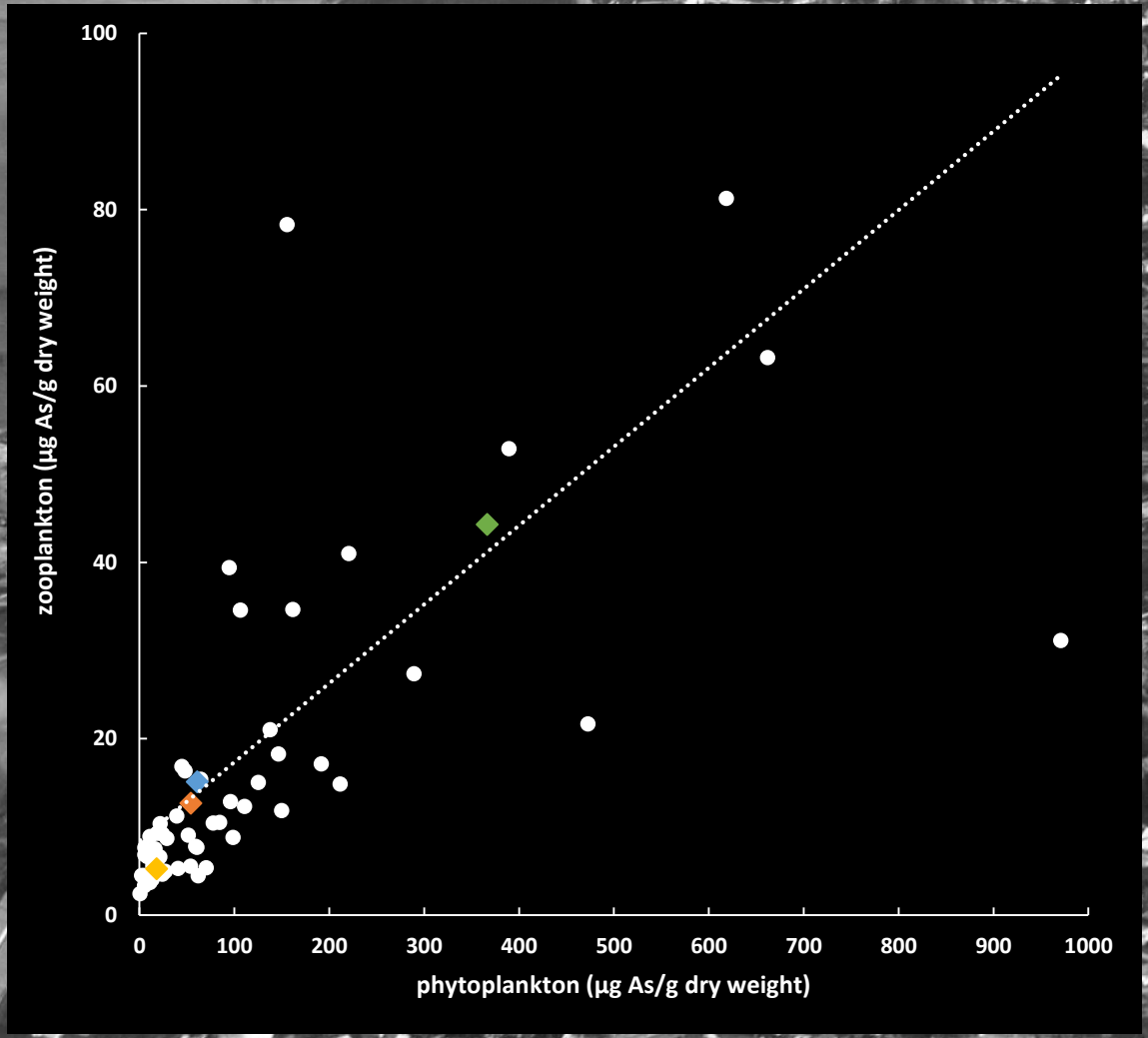
Arsenic in Sediment Traps

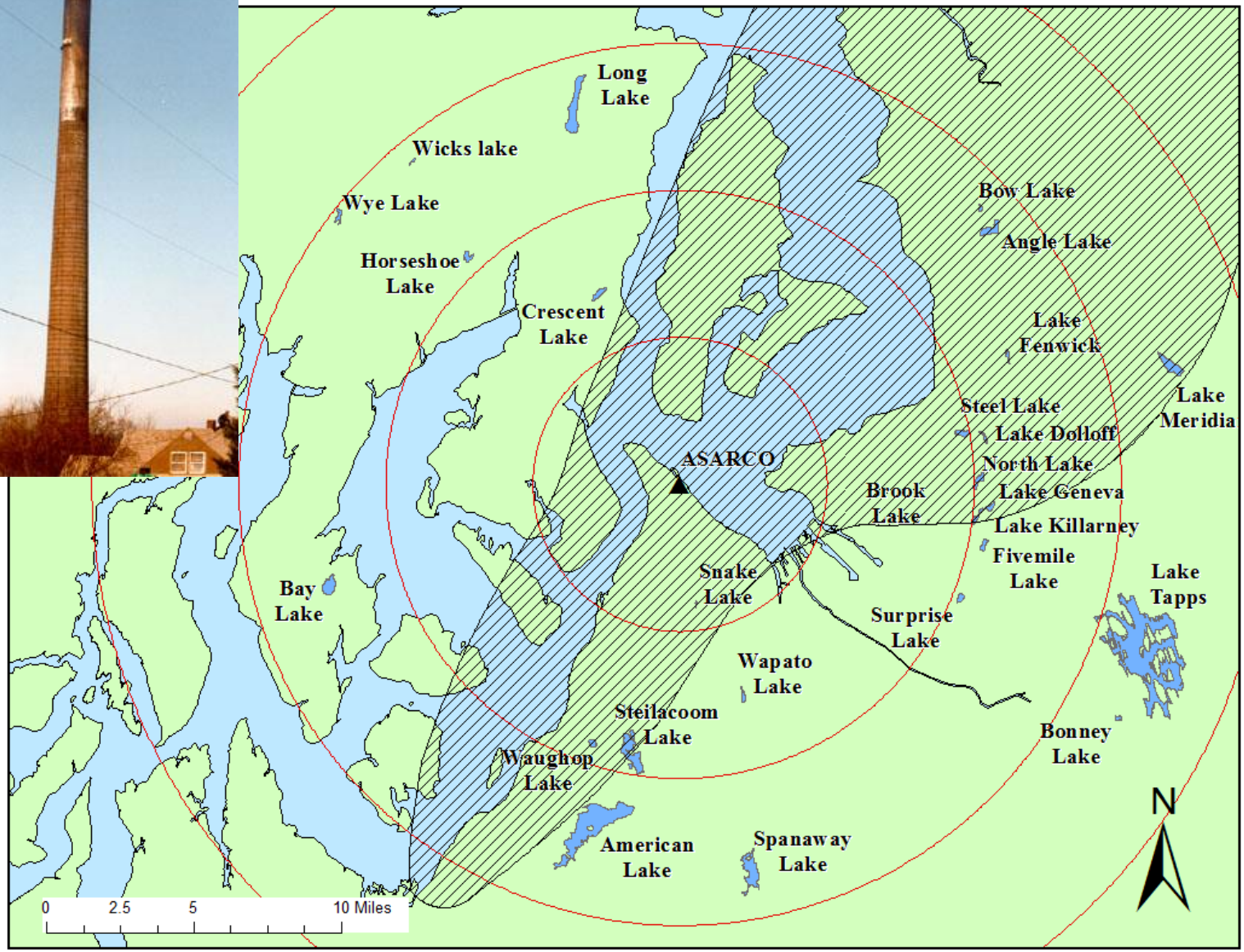
Diffusion v. Sedimentation Rates



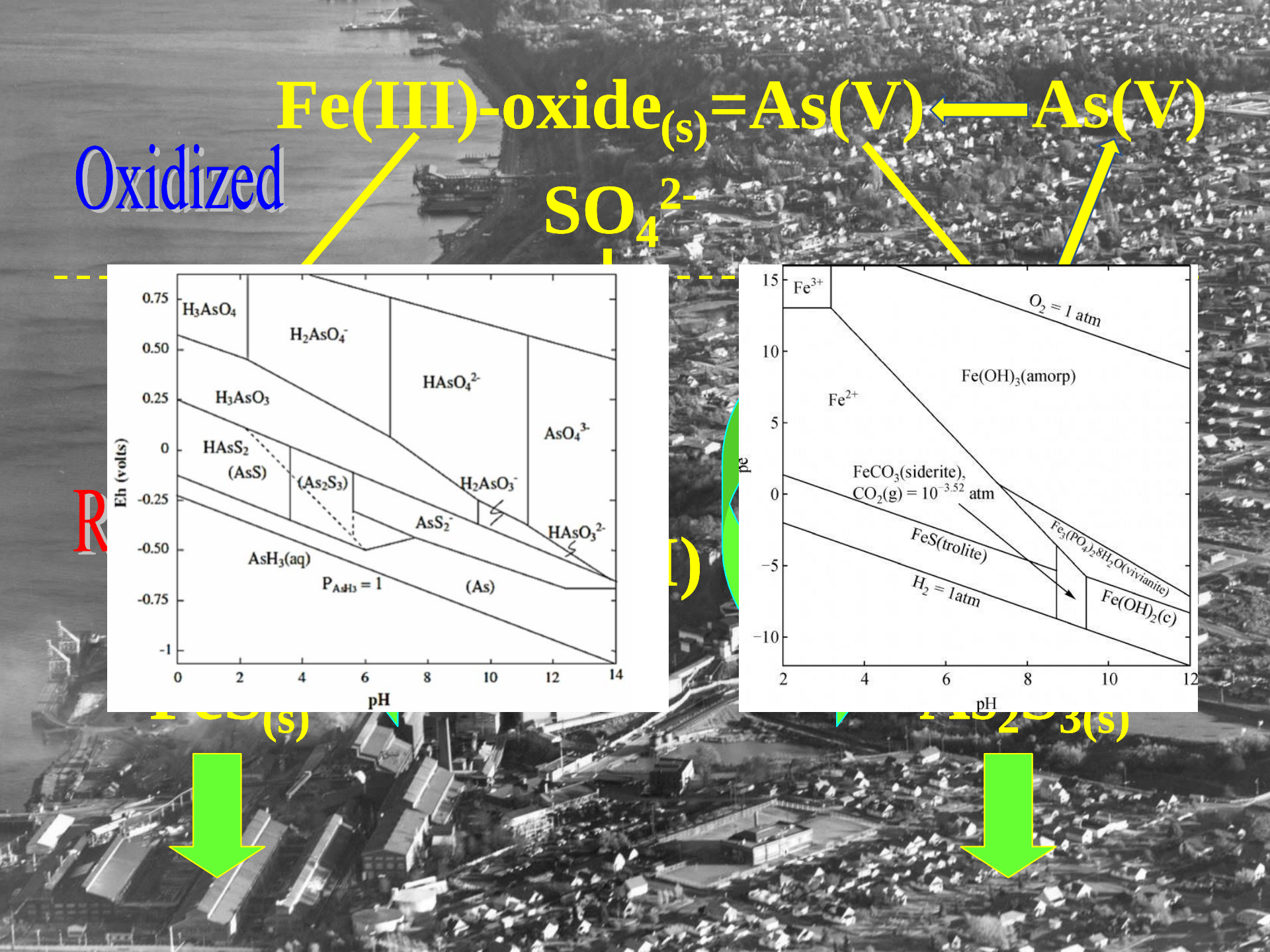
site	date	As flux from sediments[a] ($\mu g\ m^{-2}\ d^{-1}$)	As flux to sediments[b] ($\mu g\ m^{-2}\ d^{-1}$)
Angle Lake	Sept 2016	11	15.7
	Aug 2017	14	5.1
	Dec 2017	127	51
Lake Killarney	July 2016	1302	647
	Aug 2017	873	964
	Dec 2017	15	174

Arsenic Moving Up the Food Chain

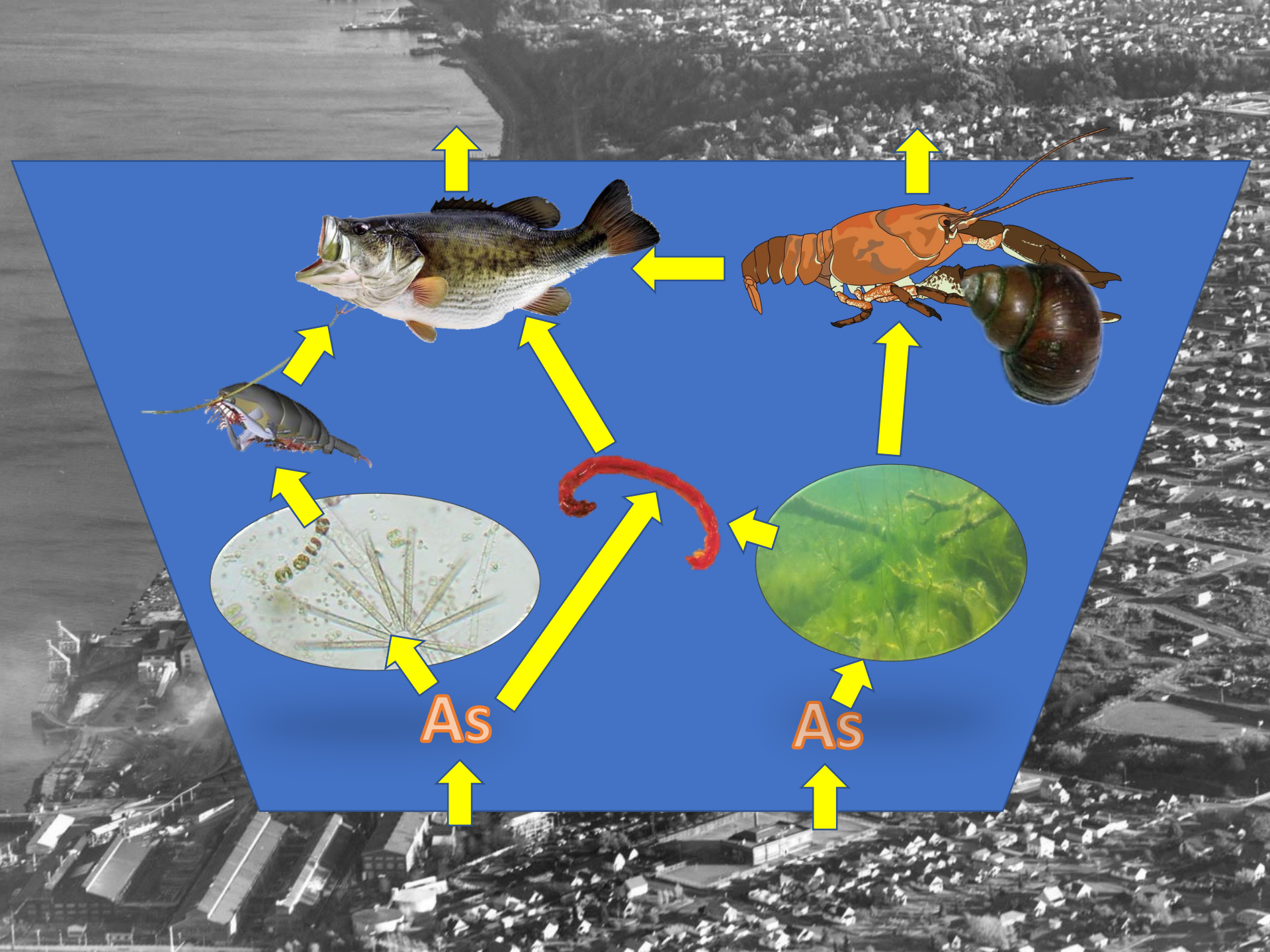




http://www.ecy.wa.gov/PROGRAMS/tcp/area_wide/AWToolbox_chap2_figures/Tier2Maps.pdf



Fe(III)-oxide(s)=As(V)
As(V)
Oxidized
SO4 2-
H3AsO4
H2AsO4-
HAsO4 2-
H3AsO3
AsO4 3-
HAsS2
(AsS)
(As2S3)
H2AsO3-
AsS2-
HAsO3 2-
AsH3(aq)
PAsH3 = 1
(As)
Eh (volts)
pH
Fe3+
O2 = 1 atm
Fe(OH)3(amorp)
Fe2+
FeCO3(siderite),
CO2(g) = 10-3.52 atm
FeS(trolite)
Fe3(PO4)2 8H2O(vivianite)
H2 = 1atm
Fe(OH)2(c)
pe
pH


As
As

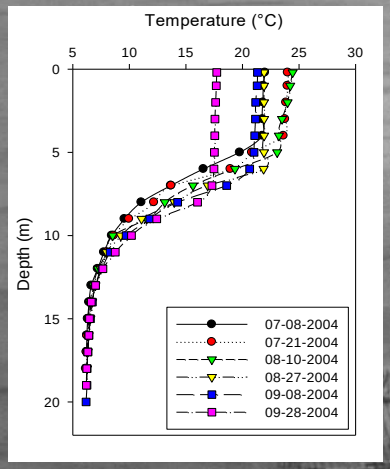
Lake Meridian

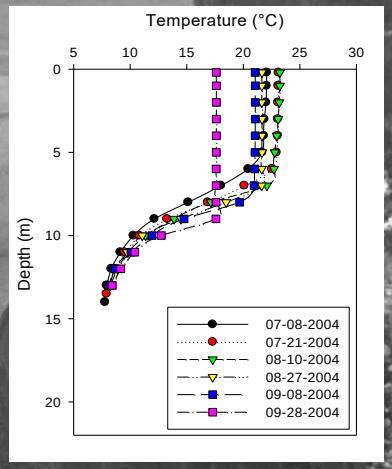
Angle Lake

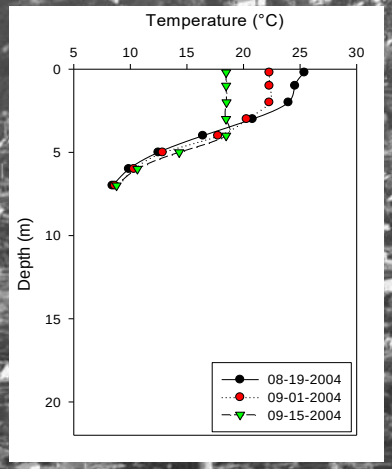
North Lake

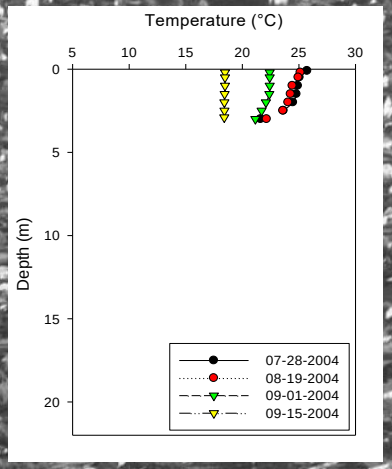
Lake Killarney

Temperature

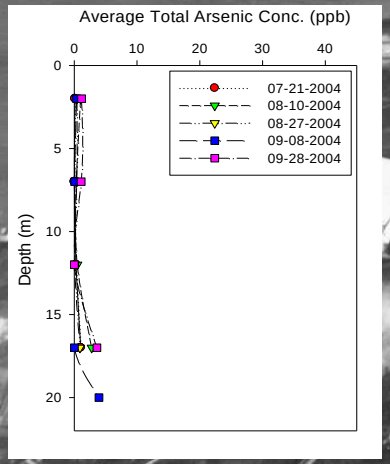


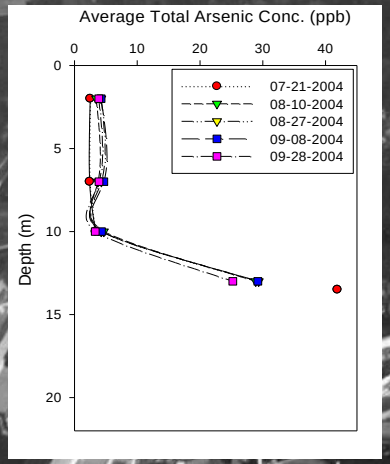


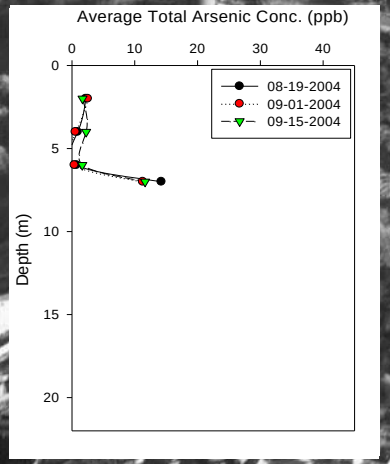


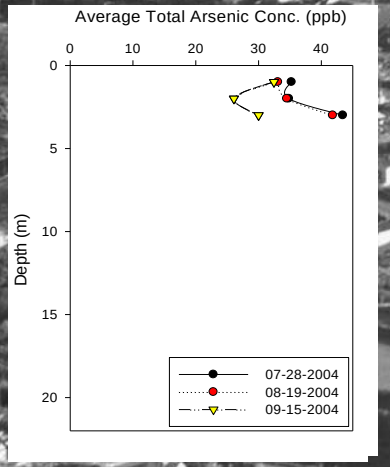


Dissolved Oxygen / Total Arsenic

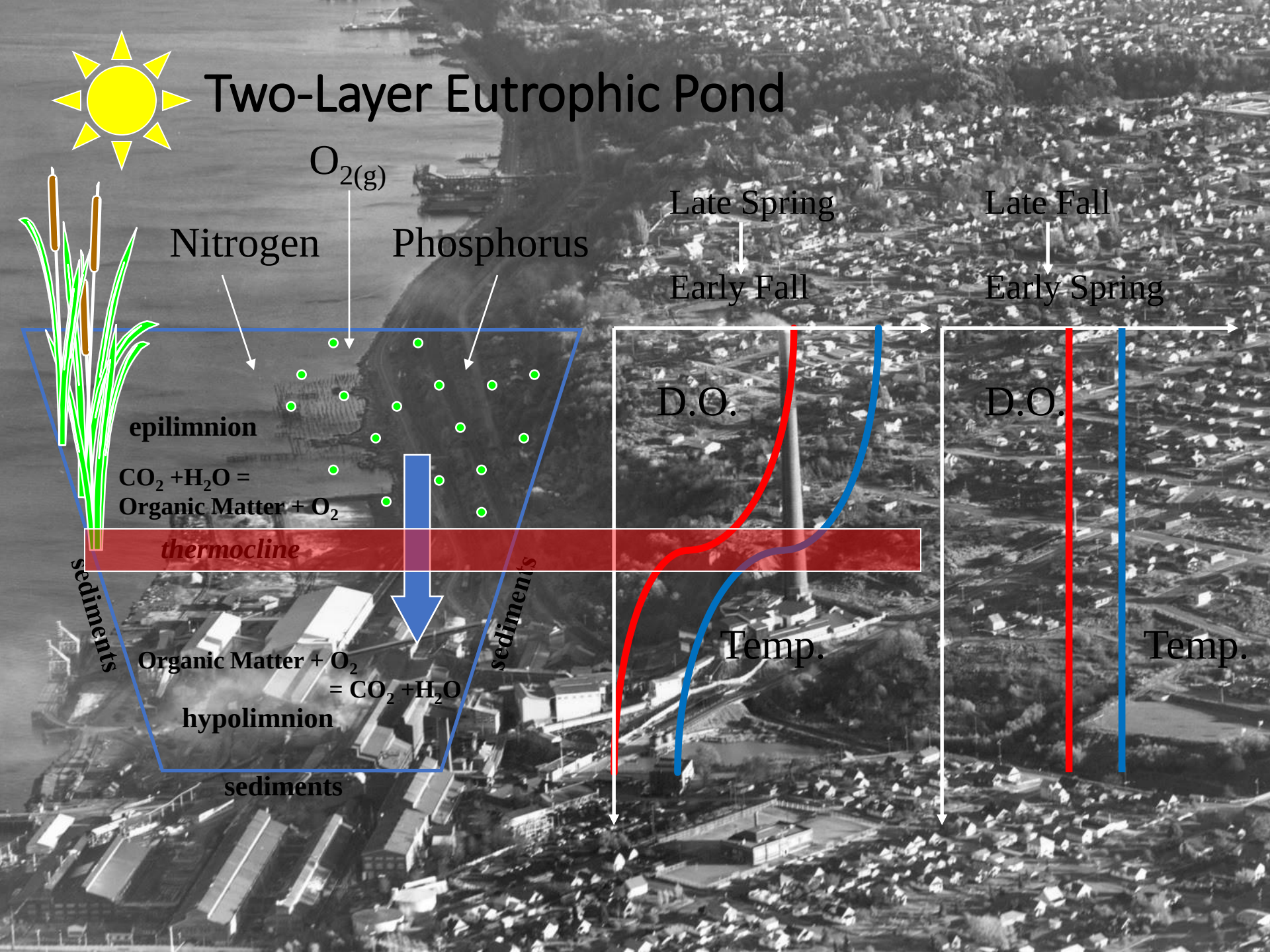


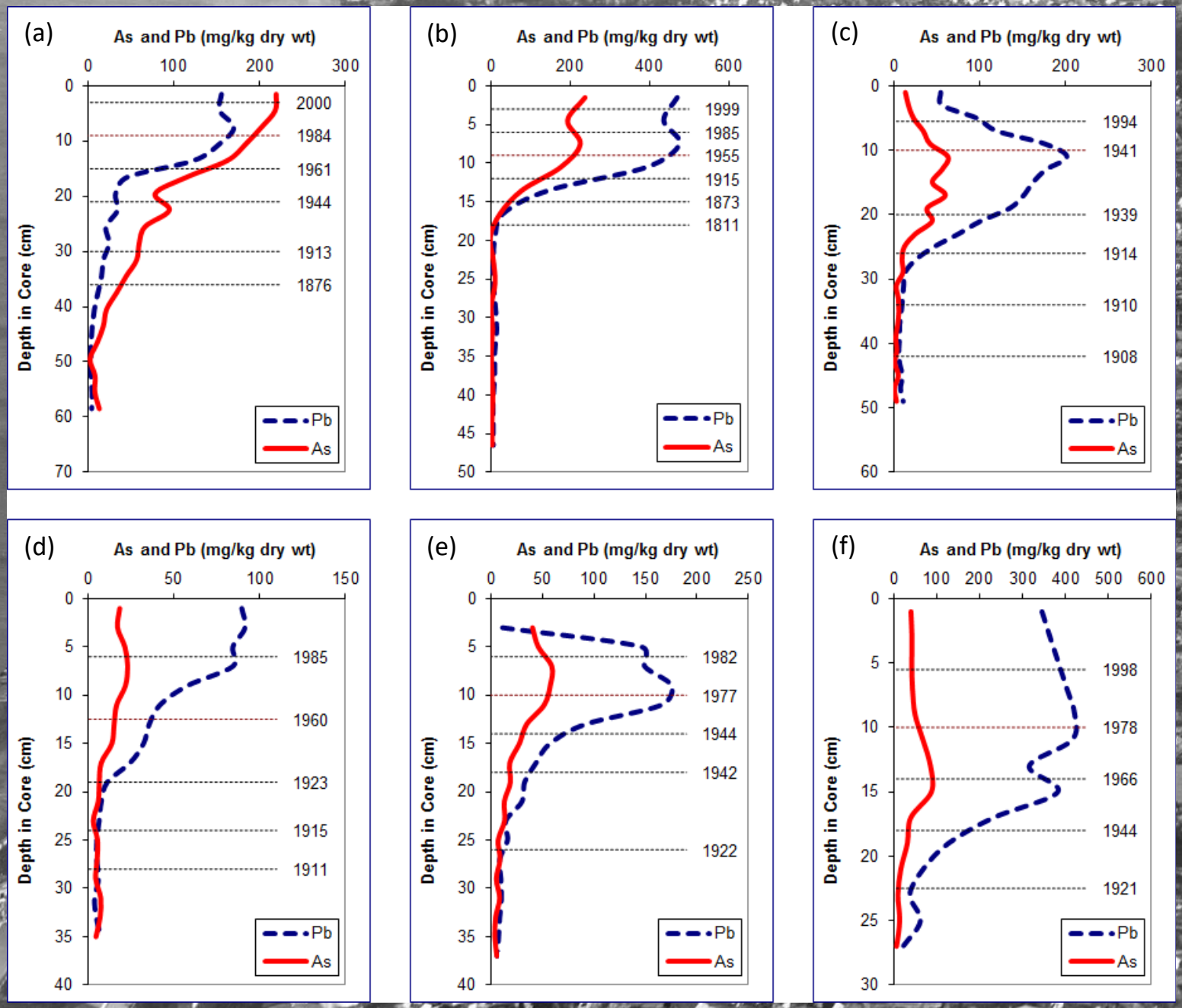




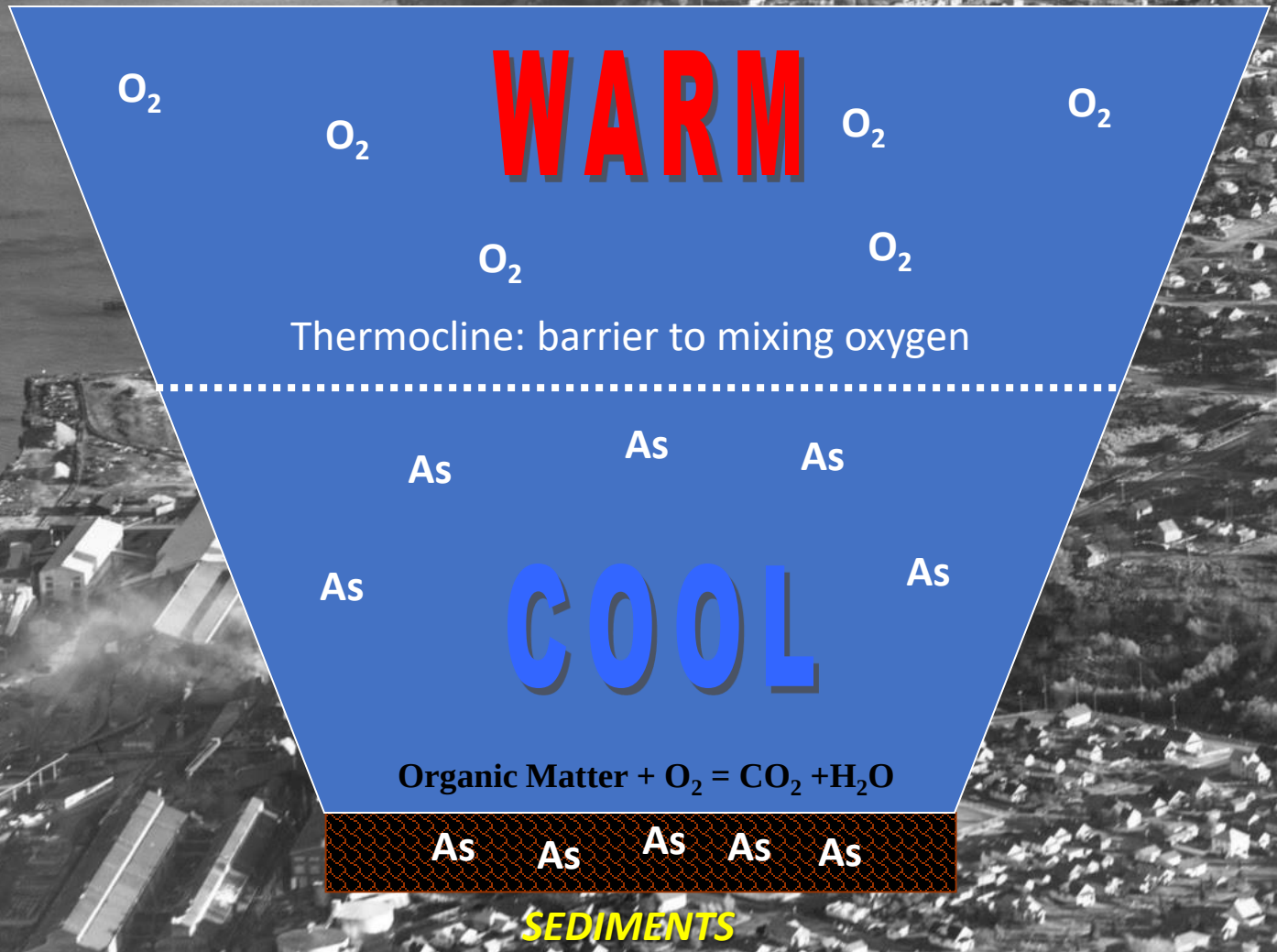


Two-Layer Eutrophic Pond




(a)
As and Pb (mg/kg dry wt)
Depth in Core (cm)
2000
1984
1961
1944
1913
1876
Pb
As
(b)
As and Pb (mg/kg dry wt)
Depth in Core (cm)
1999
1985
1955
1915
1873
1811
Pb
As
(c)
As and Pb (mg/kg dry wt)
Depth in Core (cm)
1994
1941
1939
1914
1910
1908
Pb
As
(d)
As and Pb (mg/kg dry wt)
Depth in Core (cm)
1985
1960
1923
1915
1911
Pb
As
(e)
As and Pb (mg/kg dry wt)
Depth in Core (cm)
1982
1977
1944
1942
1922
Pb
As
(f)
As and Pb (mg/kg dry wt)
Depth in Core (cm)
1998
1978
1966
1944
1921
Pb
As

Arsenic Remobilization

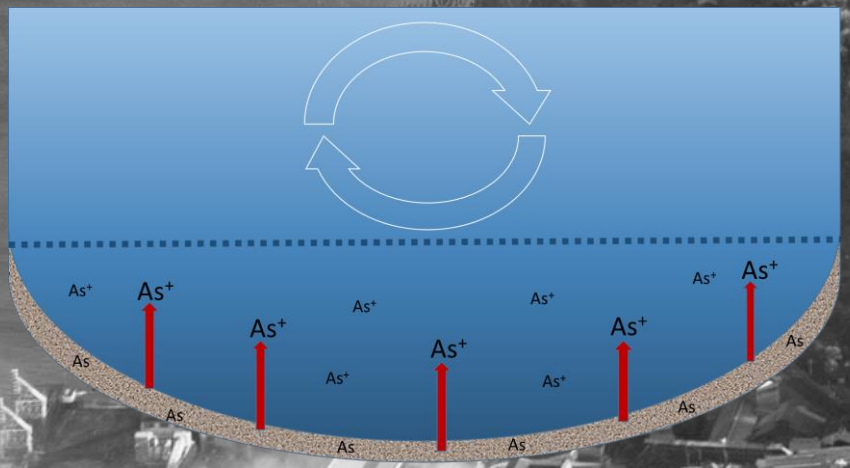


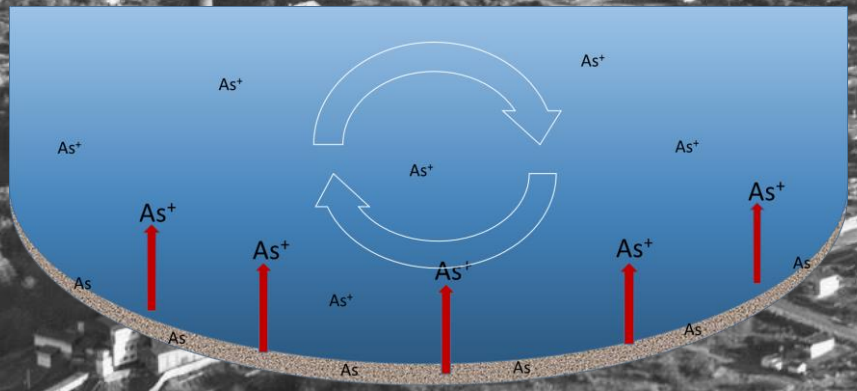
Deep Lakes:

- Seasonal thermocline, strongly stratified
- Anoxic hypolimnion

Shallow Lakes:

- Polymictic
- Short-term anoxia at sediment interface, followed by mixing





Angle Lake (50 ft) High sed As

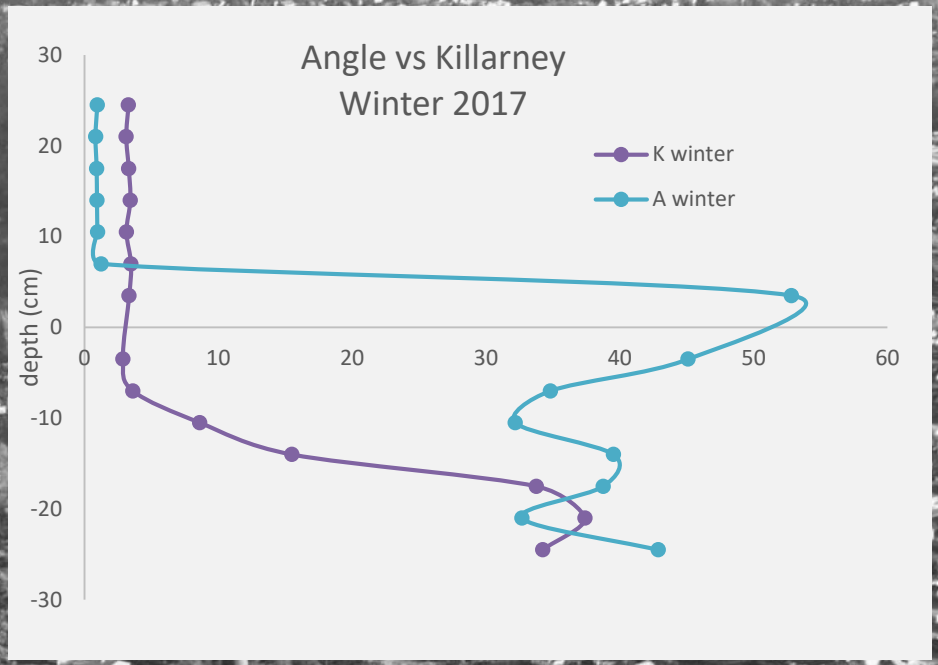
North Lake (25 ft) Med sed As

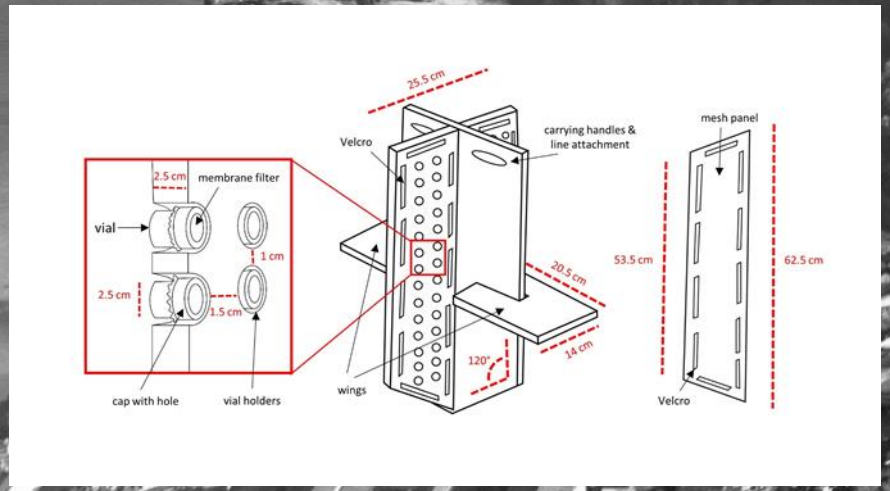
Lake Killarney (13 ft) Hi sed As

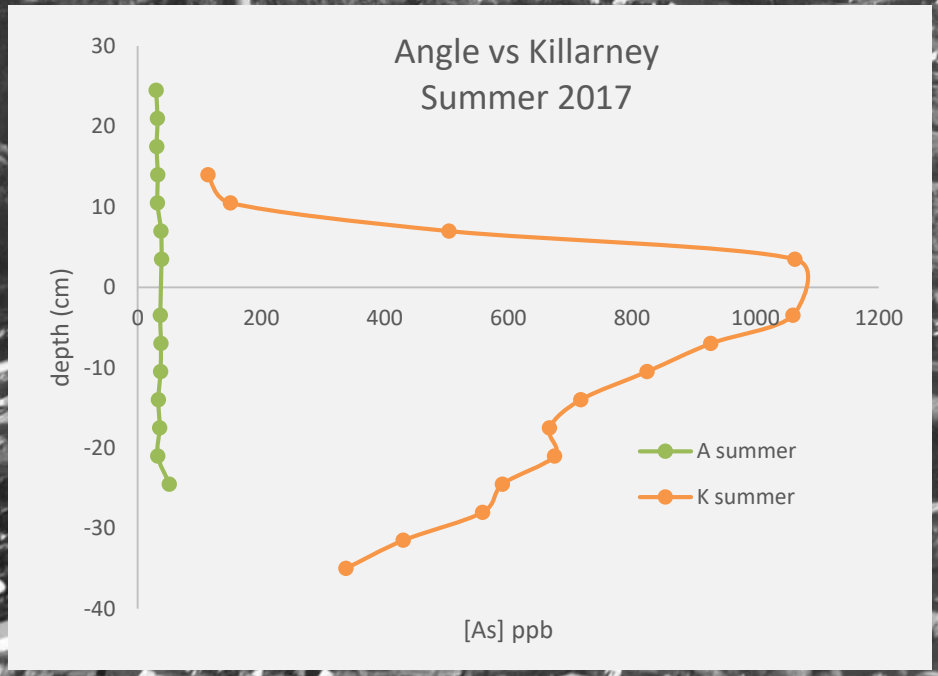
Steel Lake (20 ft) Med sed As

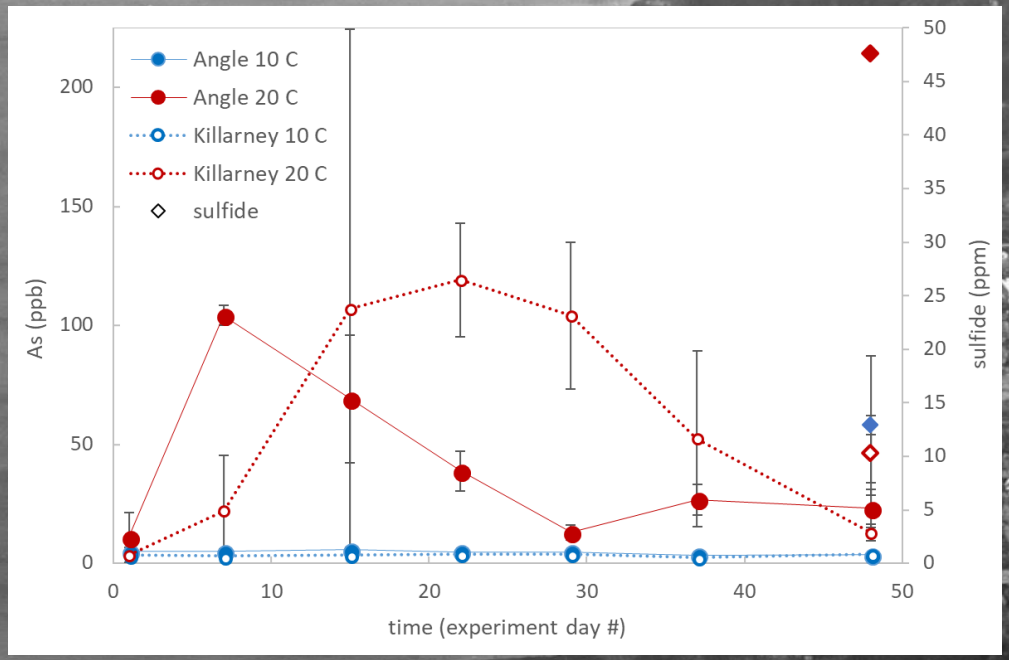
As(V)
As(V)
As(III)

Porewater Peepers Diffusive Flux

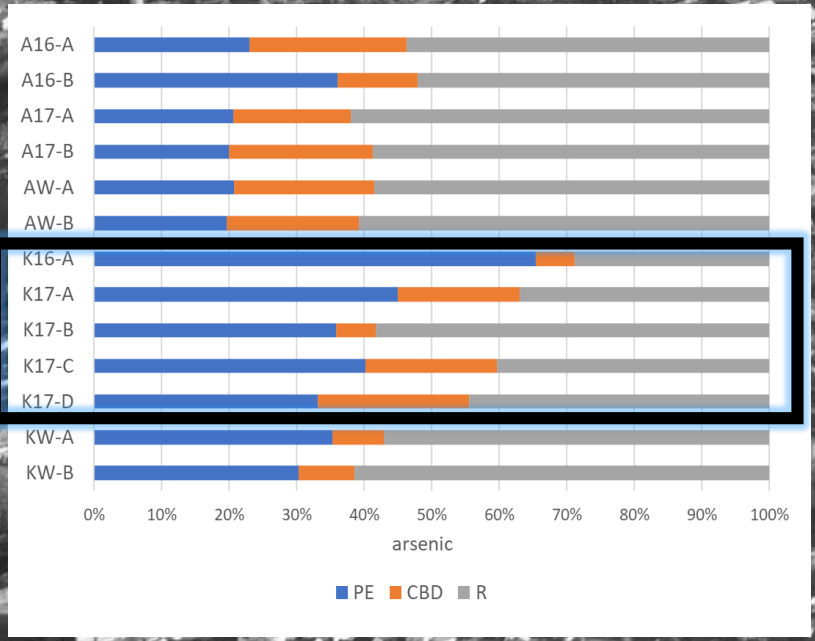






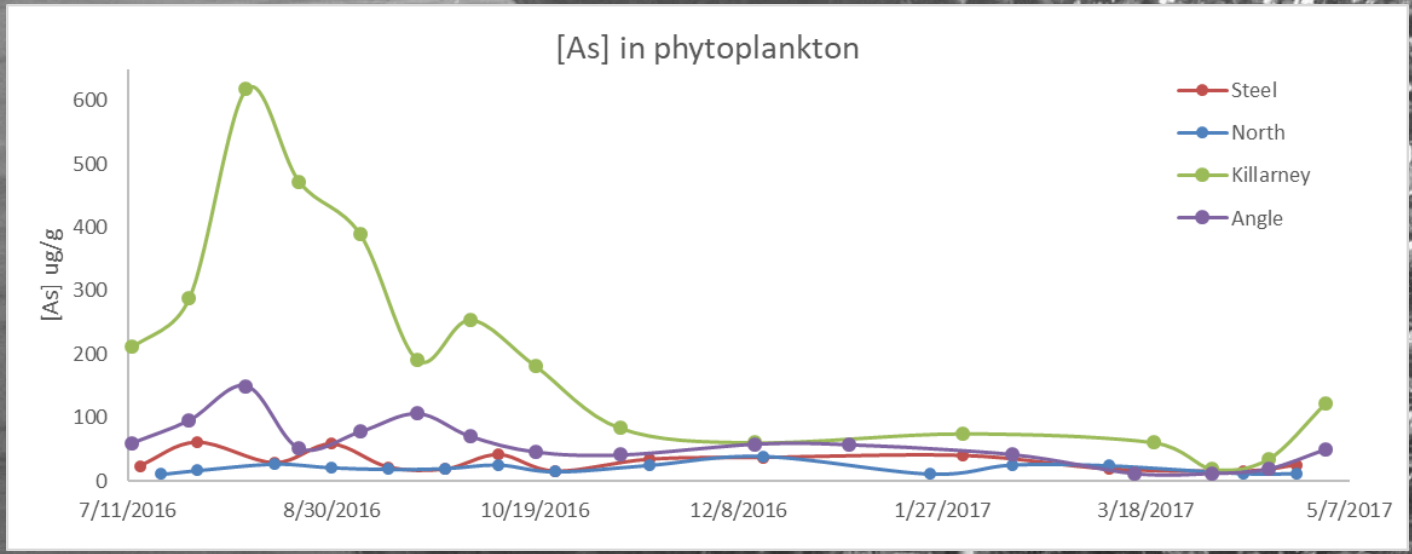


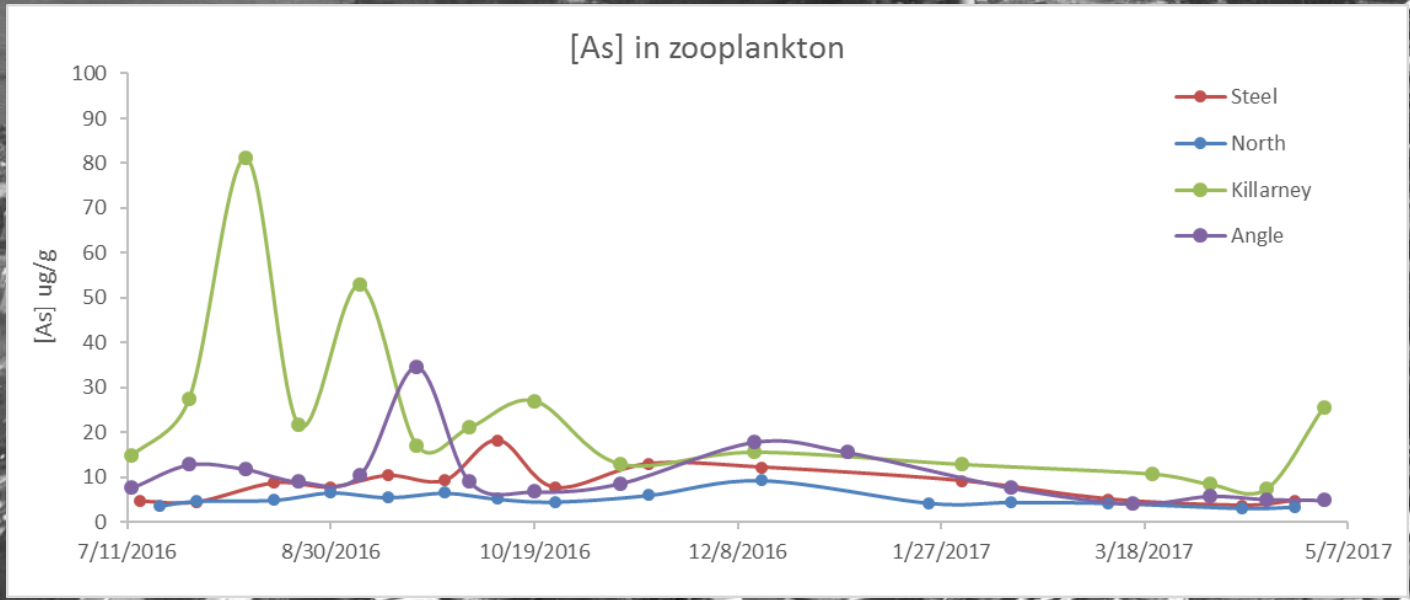
- Temperature has a large effect on arsenic release
- Similar As concentrations reached in water column at same temperatures, same sediment concentrations (≈200 ppm)
- In the field we do not see these levels of sulfide

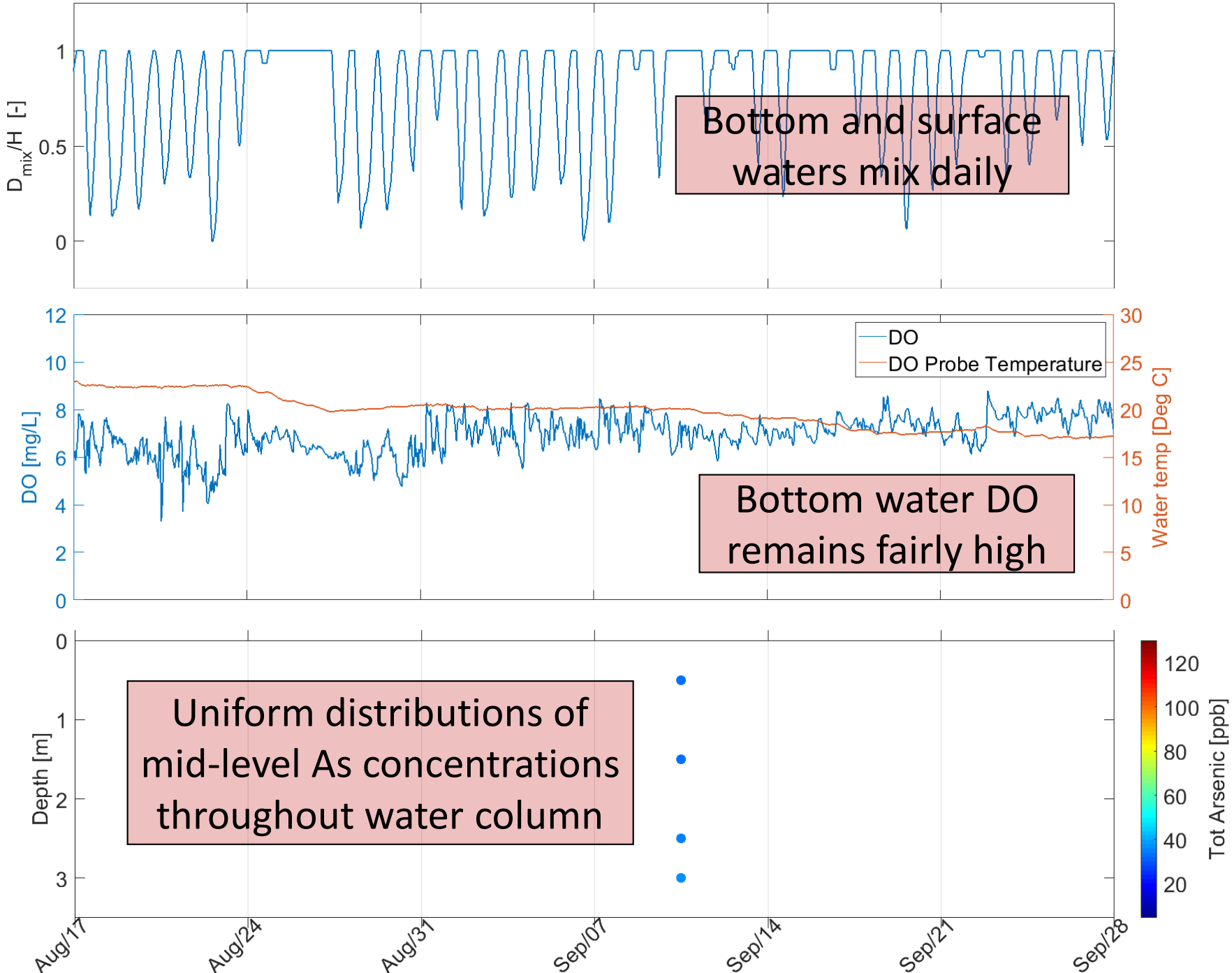


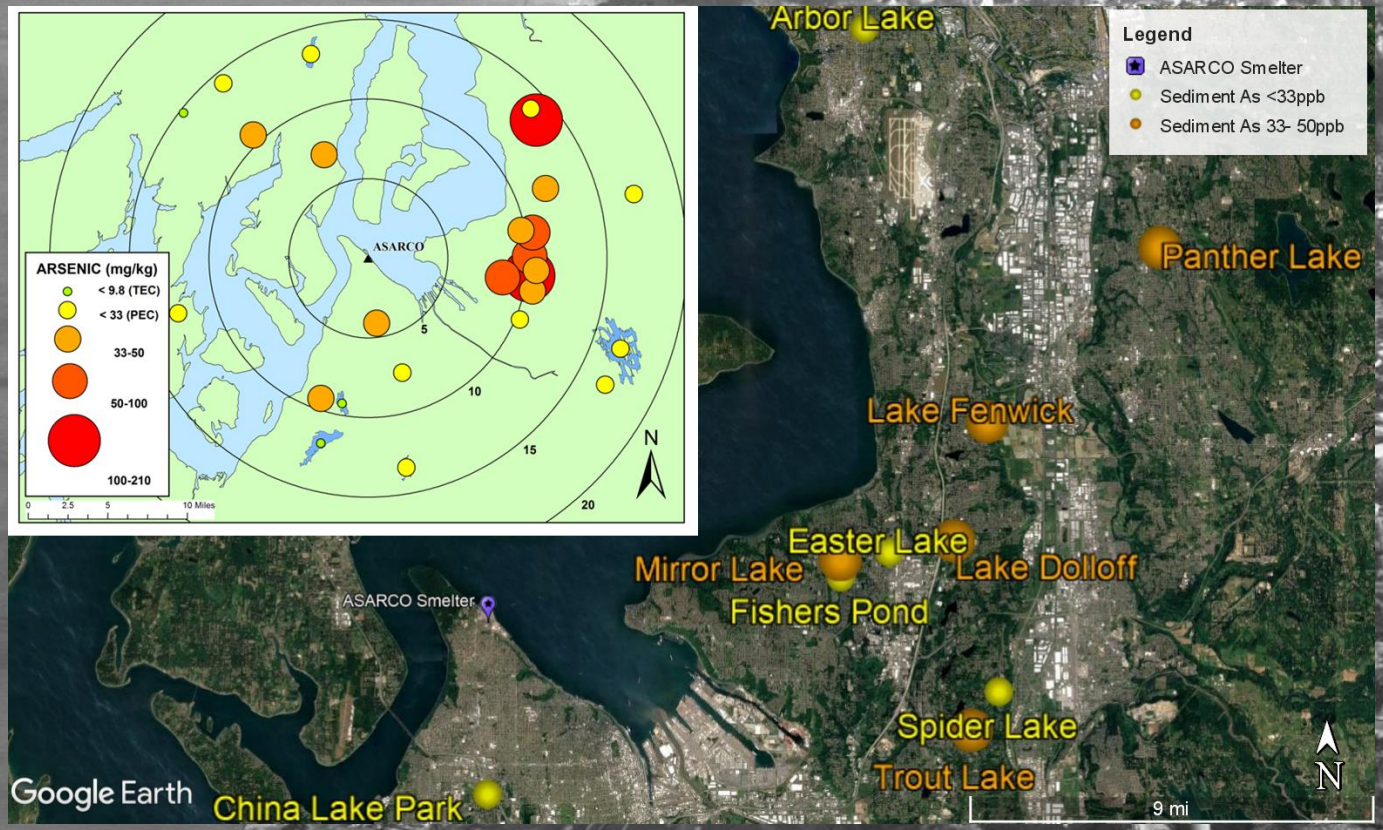
- Higher phosphate exchangeable (PE) and Fe-oxide reduction (CBD) fractions in Killarney in summer
- Organic matter concentration also higher in Killarney sediments (49.1% vs 27.5% in Angle)

Arsenic in Plankton






Bottom and surface waters mix daily
D_{mix}/H [-]
1
0.5
0
Bottom water DO remains fairly high
DO
DO Probe Temperature
DO [mg/L]
Water temp [Deg C]
12
10
8
6
4
2
0
30
25
20
15
10
5
0
Uniform distributions of mid-level As concentrations throughout water column
Depth [m]
0
1
2
3
Tot Arsenic [ppb]
120
100
80
60
40
20
Aug/17
Aug/24
Aug/31
Sep/07
Sep/14
Sep/21
Sep/28



New anecdotal evidence suggests **Killarney may have been treated with arsenic** – that may be why the As:Pb ratio is so different from other lakes

Lake Name	Max Depth (m)	Top Column As Conc. Avg (ppb)	Bottom As Conc. Avg (ppb)	Avg Sediment As Conc (ppb)
Panther	1.52	0.65	0.67	41.14
China	1.83	1.07	1.05	22.51
Spider	1.83	1.14	1.12	26.98
Fishers	1.83	0.82	0.81	17.31
Arbor	3.35	0.59	0.71	29.63
Easter	3.78	0.73	0.80	17.09
Dolloff	3.96	1.75	1.31	43.47
Mirror	6.48	1.43	2.69	44.94
Trout	7.82	1.52	1.91	53.07
Fenwick	8.66	1.03	2.14	41.14

Fed 2 different diets:

- High As (~100ppm)
- Low As (~20ppm)

Represent As levels in plankton of subject lakes

Melissa Driessnack and Julian Olden; UW Seattle

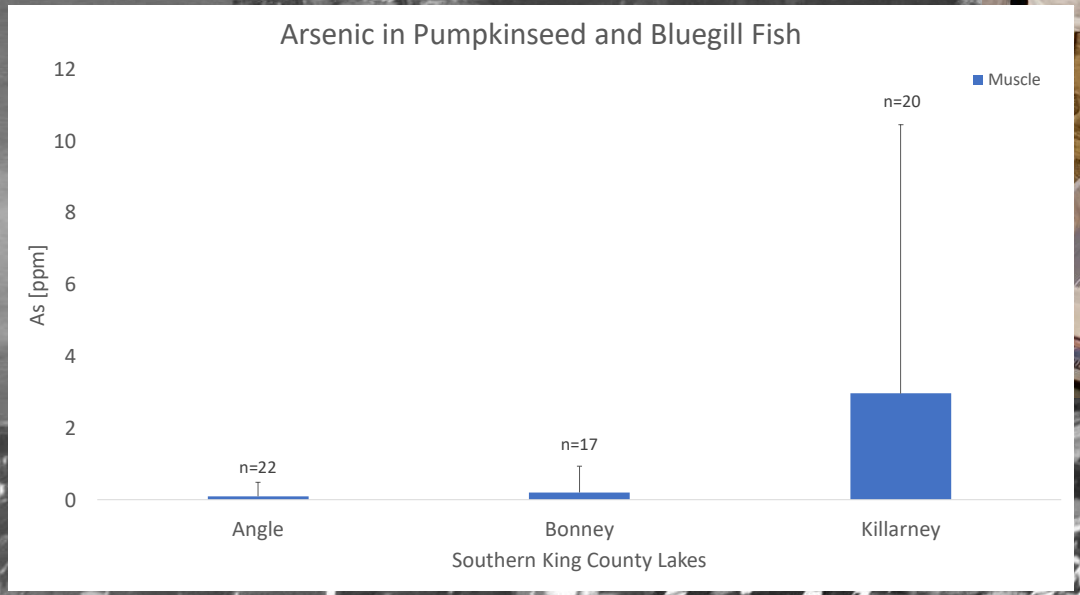
NOW: Looking for activation of genes for As-specific stress response with RNA-SEQ

ppm As	Control	20 ppm As	50 ppm As
gonad	1.57 ± 4.74[a]	0.77 ± 0.77[a]	0.45 ± 0.42[a]
gill	0.56 ± 0.56[a]	1.38 ± 3.48[a]	0.89 ± 1.13[a]
muscle	0.67 ± 0.59[a]	0.57 ± 0.42[a]	0.9 ± 1.04[a]
liver	0.85 ± 0.44[a]	1.39 ± 0.77[b]	1.63 ± 1.1[b]

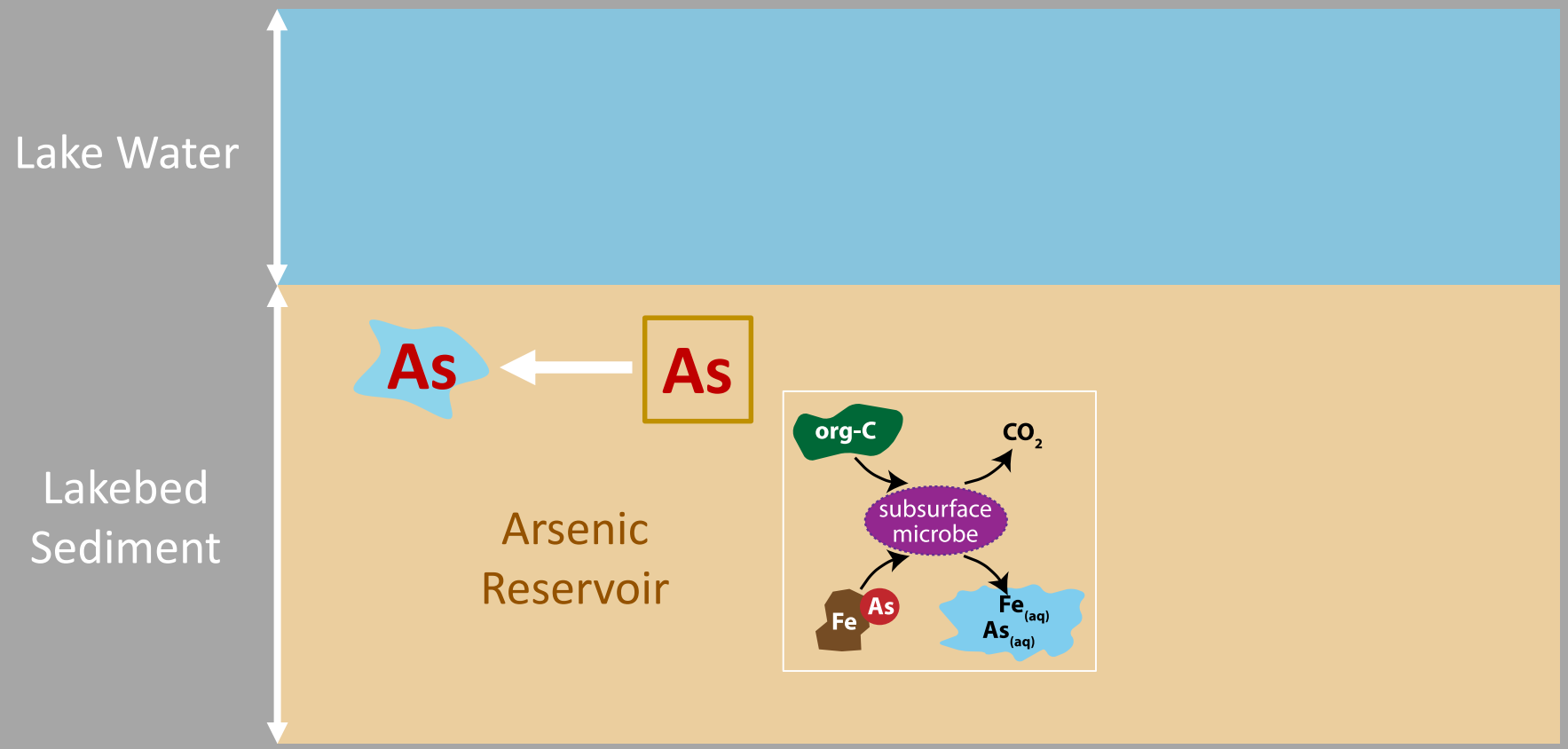
Pumpkinseed sunfish; *Lepomis gibbosus*

2019 Beach seining for pumpkinseed sunfish

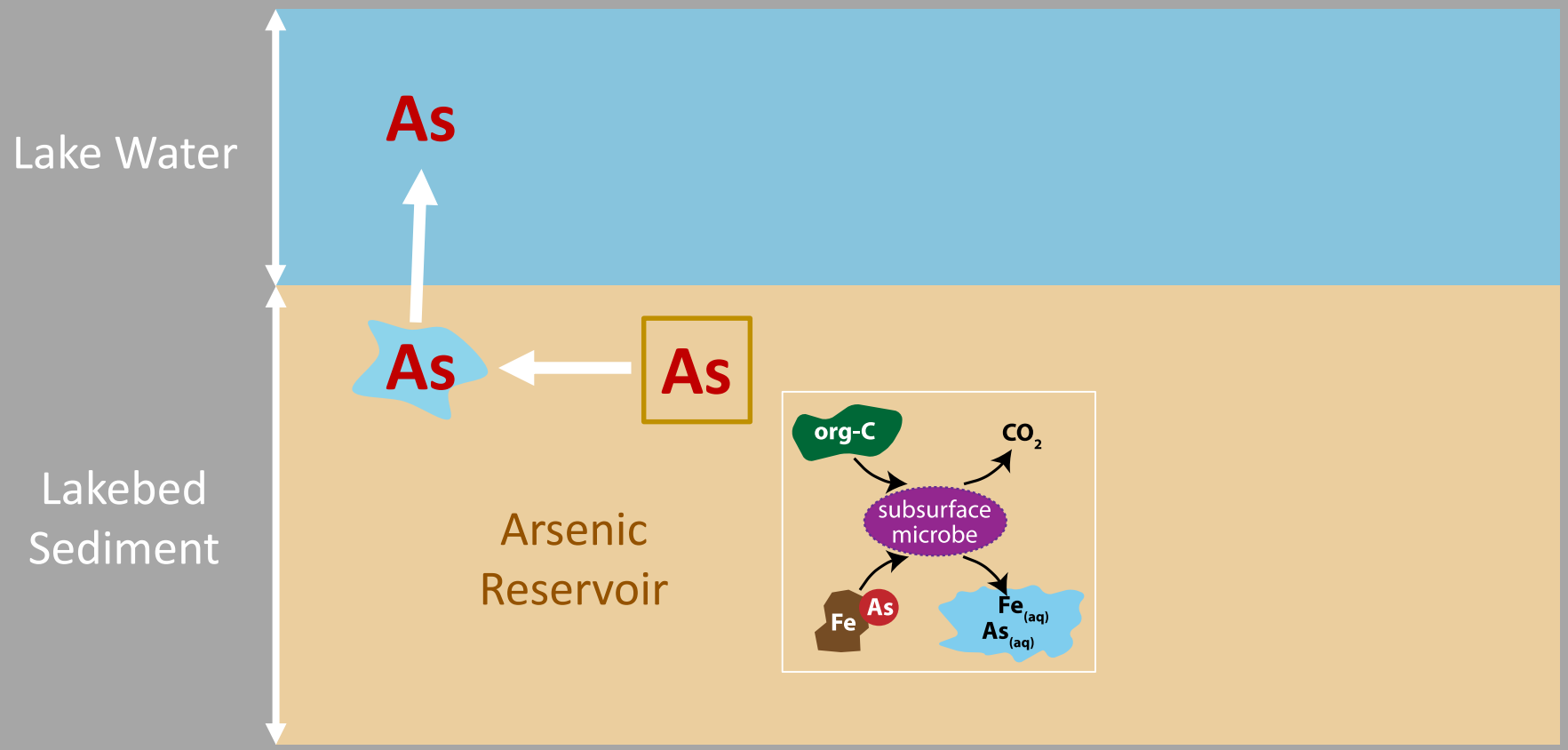
- PKS found in all but Lake Killarney
- Bluegill used instead with some overlap in other lakes
- Max As concentration in fish in Slovene lakes is less than 0.11 ppm



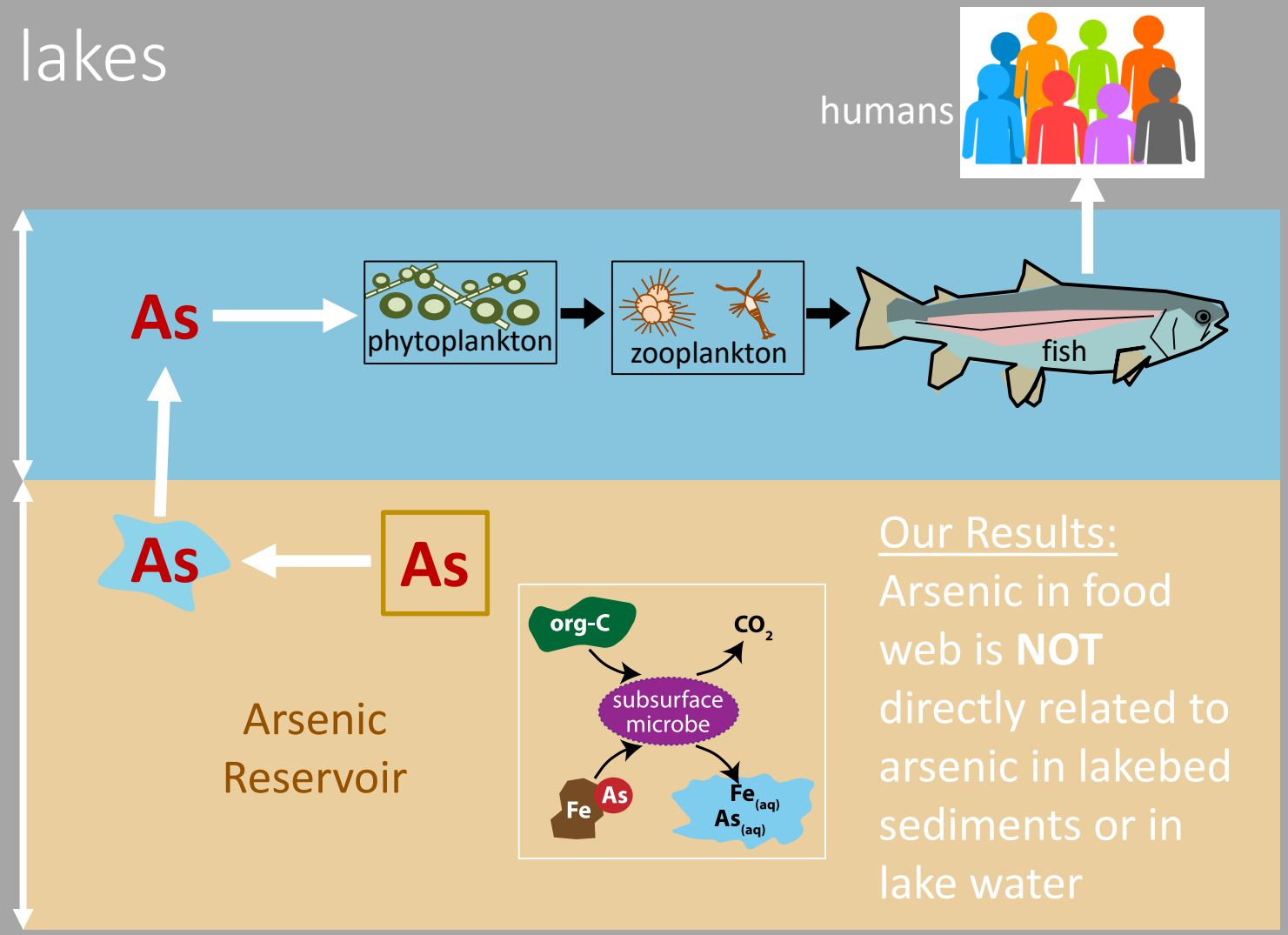
Arsenic in lakes



Arsenic in lakes



Arsenic in lakes



Our Results:
Arsenic in food web is **NOT** directly related to arsenic in lakebed sediments or in lake water