Western Airborne Selected Contaminants Assessment Program: Focus on GLAC Results Compared and Interpreted

Authors:, ***D. Landers, S. Simonich,*** D. Jaffe, L. Geiser, D. H. Campbell, A. Schwindt, C. Schreck, M. Kent, W. Hafner, H. E. Taylor, K. Hageman, ***S. Usenko***, ***L. Ackerman***, J. Schrulu, N. Rose, T. Blett, M. M. Erway

GLAC Workshop – Missoula, MT – 24 April 2008

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Key Issues for GLAC

- Highest % Ag Lands within 150 km of Park reflect among the highest CUPs and HUPs (SOCs) in snow, fish, etc.
- Clear East – West airshed differences: Old Man Lake generally highest in most SOCs with Snyder Lake by far highest in PAHs
- Fish exceed subsistence fisher thresholds for Dieldren and DDE
- Hg in fish among the lowest of all parks, piscivorous birds at risk from Hg in both lakes. MAs in kidney and spleen in both lakes correlated with age and Hg.
- One intersex fish found in Oldman Lake (East-side), both lakes had male fish with elevated estrogen-responsive protein, suggesting endocrine disruption.
- Along with SEKI and ROMO, GLAC is one of the three National Parks with highest SOCs
- High forest productivity could result in high loadings from litter fall and through fall.
- Lead, cadmium, and mercury profiles increase from approximately 1875 and decrease beginning in the 1960s. These profiles suggest a common historic source that may have been affected by reductions in emissions due to the Clean Air Act. This relationship is supported by the pattern observed in SCPs.

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How much of the contaminants in North America are from Trans-Pacific Transport?

In a new NASA study, researchers taking advantage of improvements in satellite sensor capabilities offer the first measurement-based estimate of the amount of pollution from East Asian forest fires, urban exhaust, and industrial production that makes its way to western North America. (Credit: Image courtesy of NASA/Goddard Space Flight Center)

"We used the latest satellite capabilities to distinguish industrial pollution and smoke from dust transported to the western regions of North America from East Asia. Looking at four years of data from 2002 to 2005 we estimated the amount of pollution arriving in North America to be equivalent to about 15 percent of local emissions of the U.S. and Canada,"

"Our study focused on East Asian pollution transport, but pollution also flows from Europe, North America, the broader Asian region and elsewhere

"So we should not simply blame East Asia for this amount of pollution flowing into North America."

http://www.sciencedaily.com/releases/2008/03/080317164336.htm

WHY WACAP?

1.3 million metric tons of pesticides used in China (WHO)

2600 metric tons of Hg emitted into the air from anthropogenic sources (UNEP)

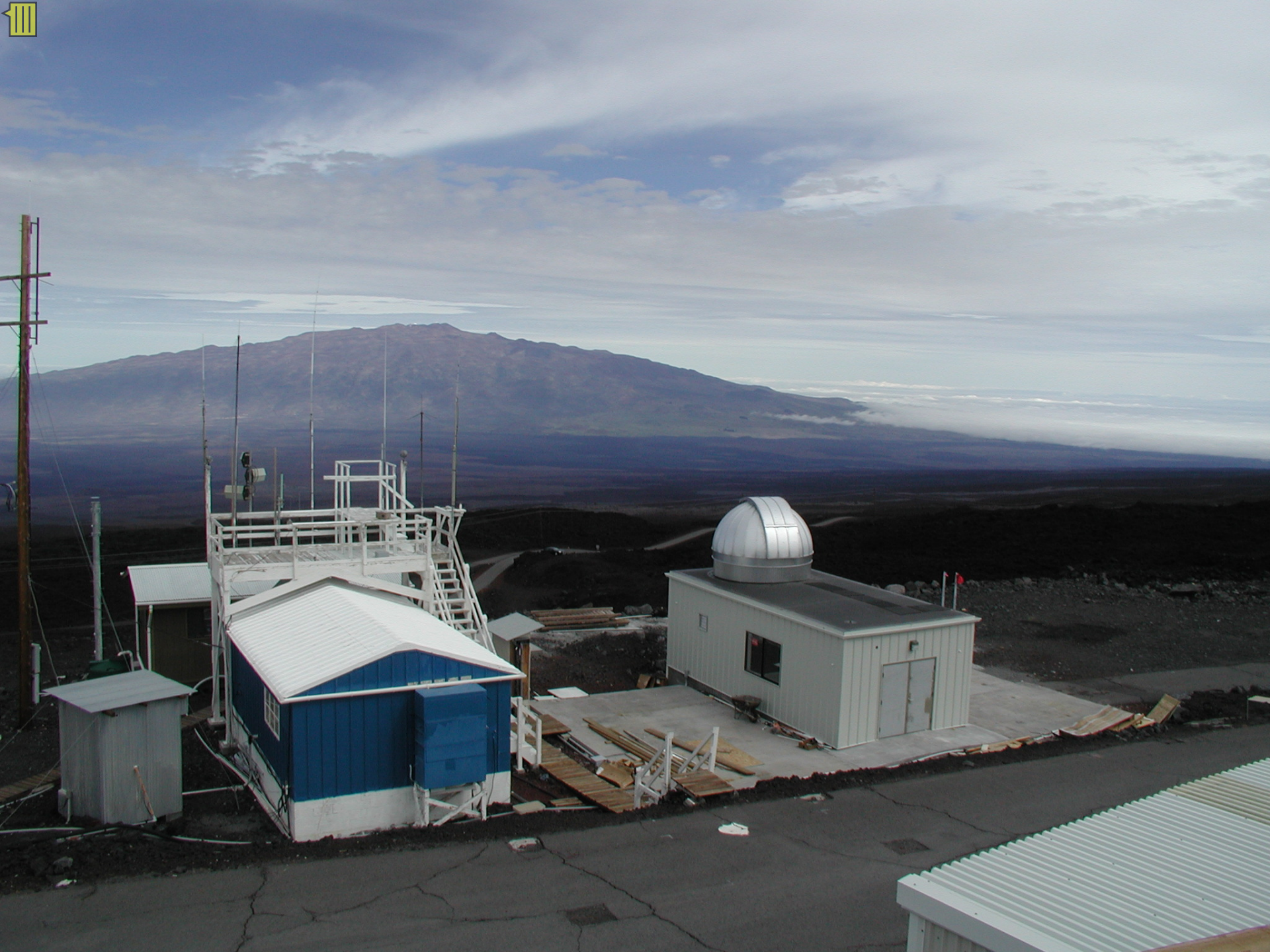
Between ‘92-’94 the US exported at least 30,000 metric tons of banned pesticides to the developing world (AAAS)

Flame retardants 10-1000 fold higher in American’s breast milk compared to Europeans (EHP)

300,000 American newborns at risk from Hg toxicity (CDC)

Presentation Outline

- Conclusions
- Brief WACAP Overview
- Results
- Discussion and Questions



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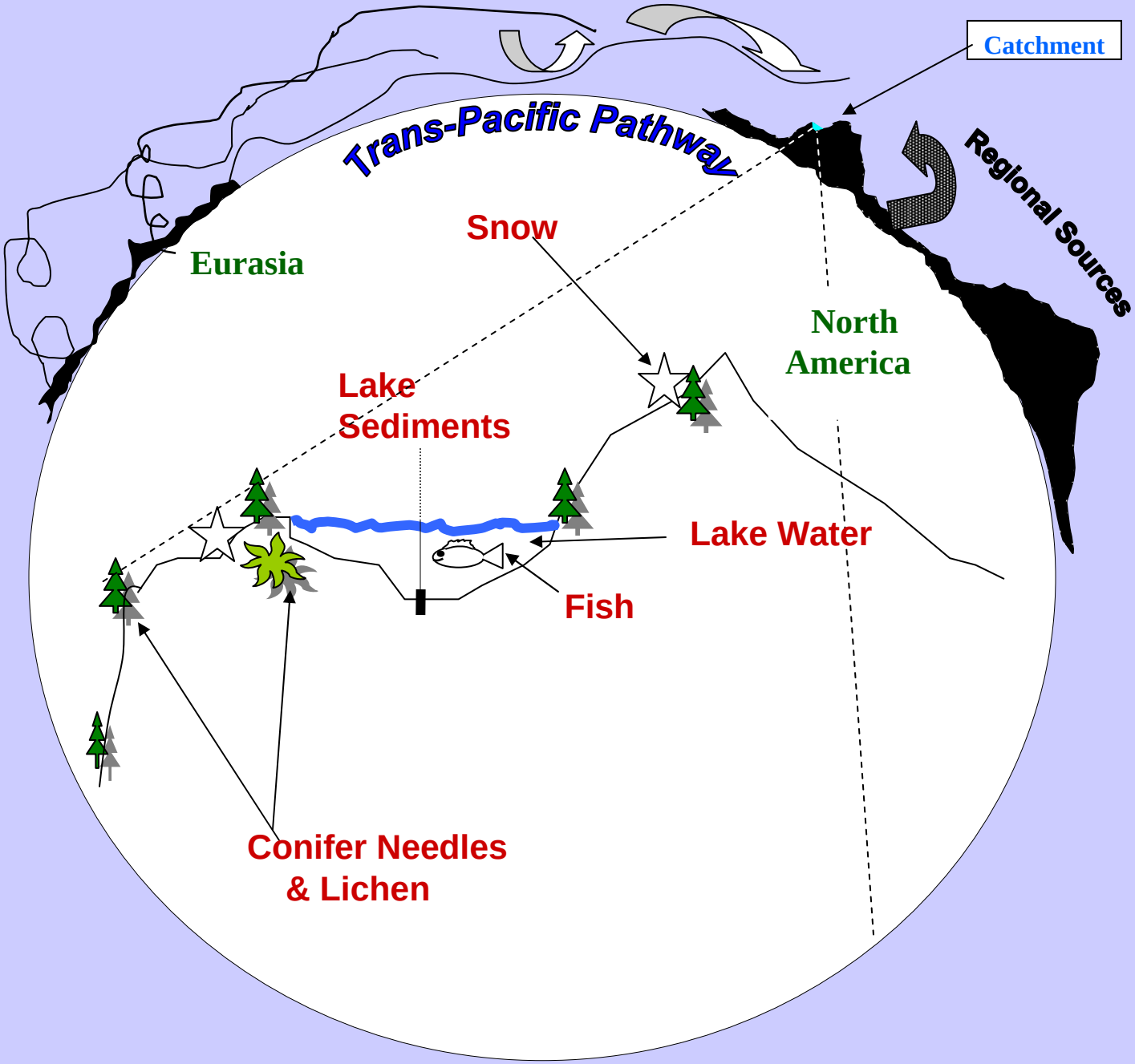
Oldman Lake, Glacier National Park
2026 masl



WACAP Indicators and Conceptual Diagram


Snow – annual flux SOCs and metals


Water - .003 – 25 yrs
Dissolved phase, SOCs, metals


Lake Sediment ~ 135 y
Chronology
SOCs, metals, SCPs


Conifer Needles – 2 yr
SOCs, metals
Lichen – unknown age
N, S, metals


Fish: 2 – 34 yr (selectable)
SOCs, metals, condition, enzymes, pathology

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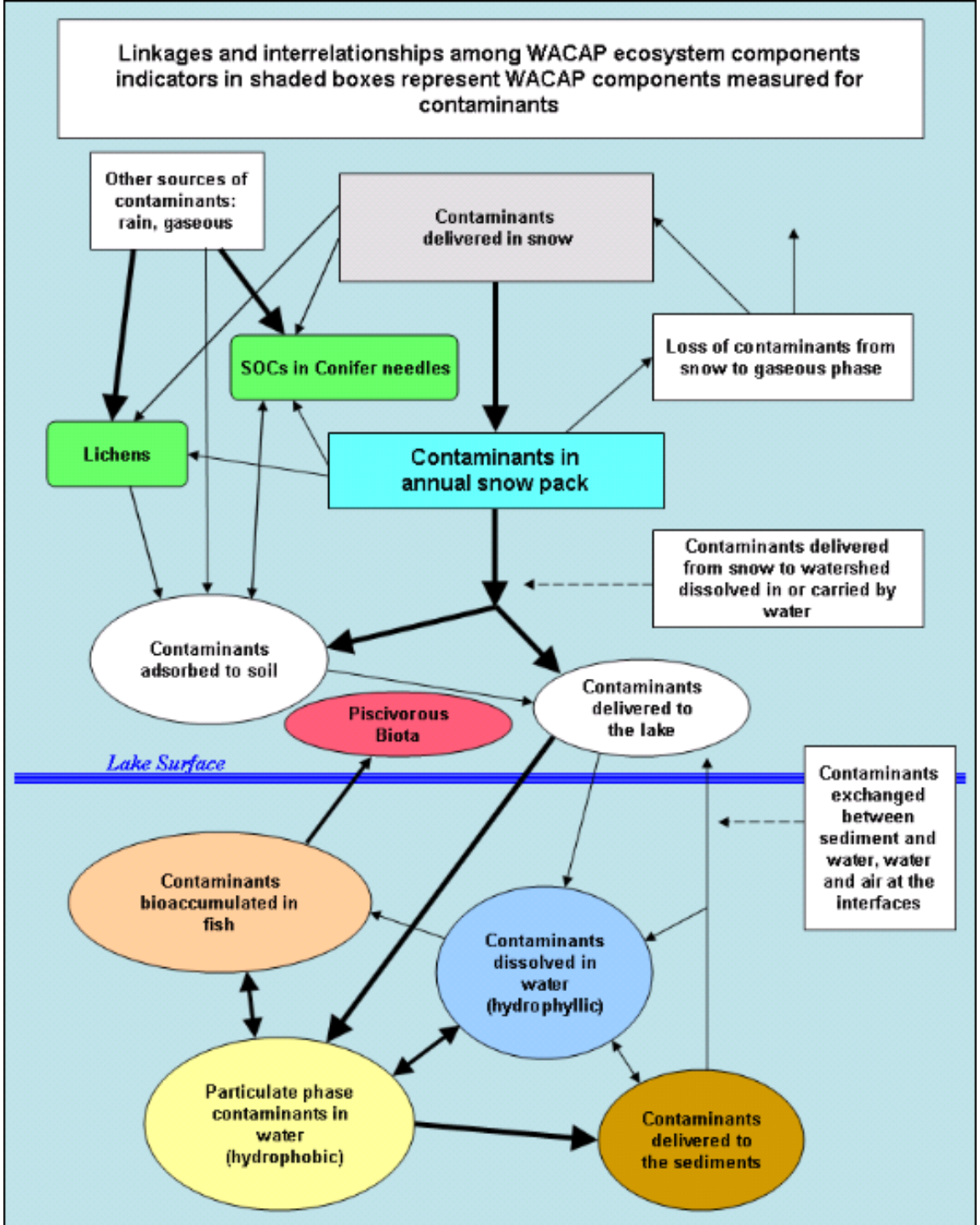

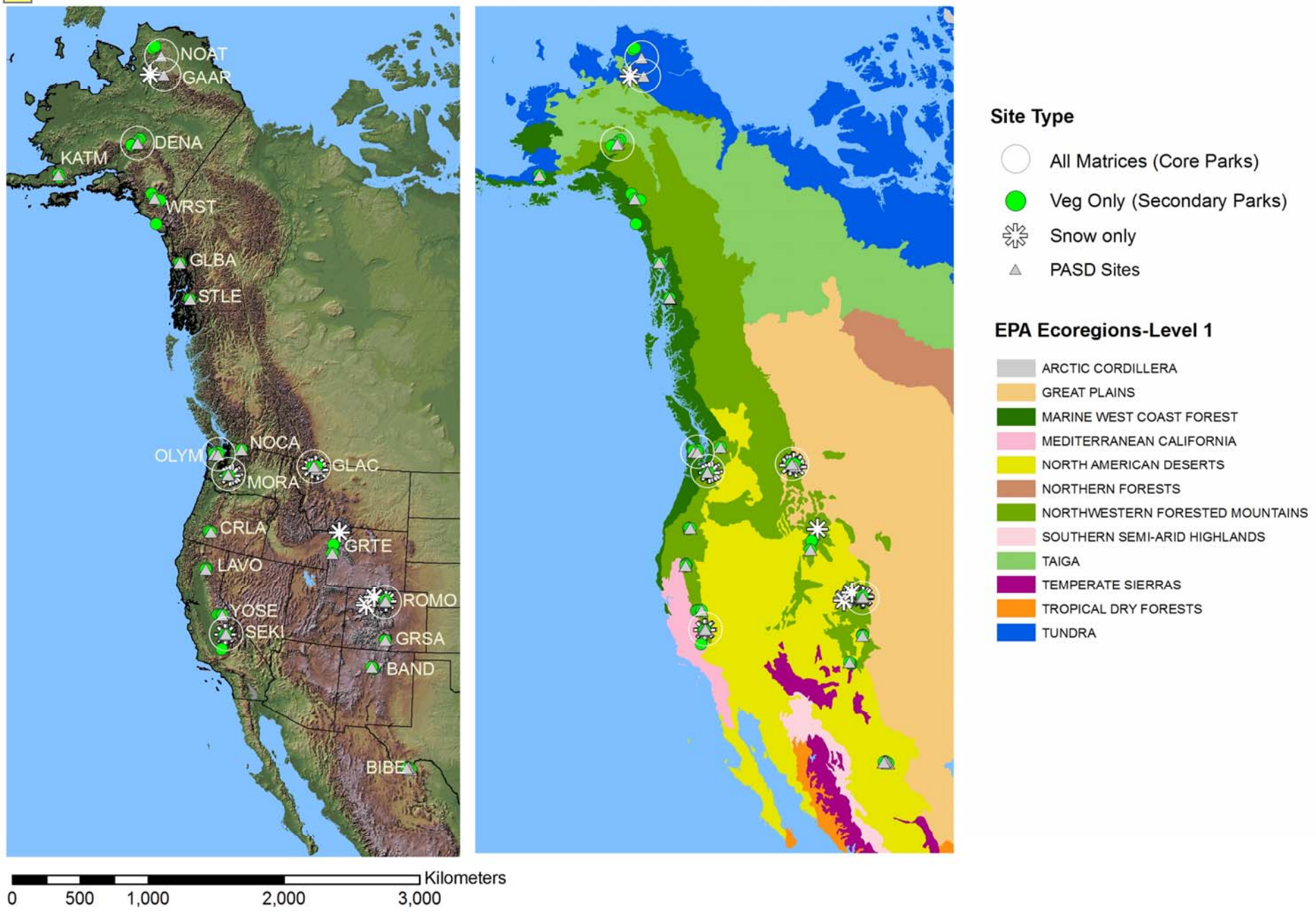

Figure 1. WACAP Sites mapped on North American shaded relief map and EPA Level 1 Ecoregions (Biomes).

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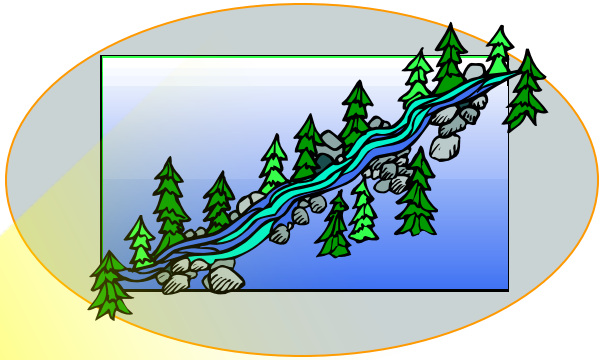
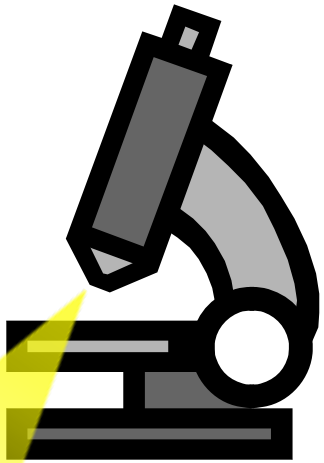
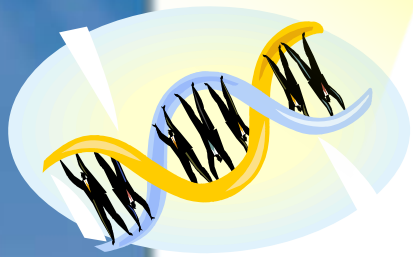


Macroscope



NEED TO UNDERSTAND AT MULTIPLE SCALES



Microscope

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2002

Pilot Studies:

Design & Methods

WESTERN AIRBORNE CONTAMINANTS ASSESSMENT PROJECT FINAL REPORT: VOLUME I

The Fate, Transport, and Ecological Impacts of Airborne Contaminants in Western National Parks (USA)

Burial Lake, Noatak National Preserve
Photo: Adam Schwindt

Dixon H. Landers, Staci Simonich, Daniel Jaffe, Linda Geiser, Donald H. Campbell, Adam Schwindt, Carl Schreck, Michael Kent, Will Hafner, Howard E. Taylor, Kimberly Hageman, Sascha Usenko, Luke Ackerman, Jill Schrlau, Neil Rose, Tamara Blett, Marilyn Morrison Erway

Technical Editor: Susan Christie

EPA/600/R-07/138
January 2008

WAC
http://

2007

Interpre-tive report and publica-tion..

We are Here!

What are the contaminant categories of potential risk to the Western National Parks due to the deposition of airborne contaminants?

- Current Use Pesticides (CUPS)
- Historic Use Pesticides (chemicals) HUPS (i.e. POPs)
- Mercury (and other metals)
- Emerging Chemicals (PBDE)

Target Semi-Volatile Organic Compounds

Electron Impact Ionization

PAHs: Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Retene, Chrysene, Triphenylene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[e]pyrene, Benzo[a]pyrene, Indeno[1,2,3-cd]pyrene, Dibenz[a,h]anthracene, Benzo[ghi]perylene

Combustion

Pesticides and degradation products:

o,p'-DDT*, p,p'-DDT, o,p'-DDD*, p,p'-DDD, o,p'-DDE, Endosulfan I, Endosulfan II, Endosulfan Sulfate, Ethion, Etradiazon, Hexazinone, Malathion, Methyl-Parathion, Parathion, Permethrin, Phorate, Trifluralin, Methoxychlor, Acetochlor*, Alachlor, Prometon, Pebulate, EPTC, Carbofuran, Carbaryl, Propachlor, Atrazine and degradation products, Simazine, Cyanazine

Agriculture

Surrogates: d_{10}-Fluorene, d_{10}-Phenanthrene, d_{10}-Pyrene, d_{12}-Triphenylene, d_{12}-Benzo[a]pyrene, d_{12}-Benzo[ghi]perylene, d_{14}-EPTC, d_{10}-Phorate, d_5-Atrazine, d_{10}-Diazinon, d_7-Malathion, d_{10}-Parathion, d_8-p,p'-DDE, d_8-p,p'-DDT, d_6-Methyl Parathion, d_{13}-Alachlor, d_{11}-Acetochlor

Internal Standards: d_{10}-Acenaphthene, d_{10}-Fluoranthene, d_{12}-Benzo[k]fluoranthene

Electron Capture Negative Ionization

PCBs:

PCB 74 (2,4,4',5-Tetrachlorobiphenyl),
PCB 101 (2,2',4,5,5'-Pentachlorobiphenyl),
PCB 118 (2,3',4,4',5-Pentachlorobiphenyl),
PCB 138 (2,2',3,4,4',5'-Hexachlorobiphenyl),
PCB 153 (2,2',4,4',5,5'-Hexachlorobiphenyl),
PCB 183* (2,2',3,4,4',5',6-Heptachlorobiphenyl),
PCB 187 (2,2',3,4',5,5',6-Heptachlorobiphenyl)

Industry

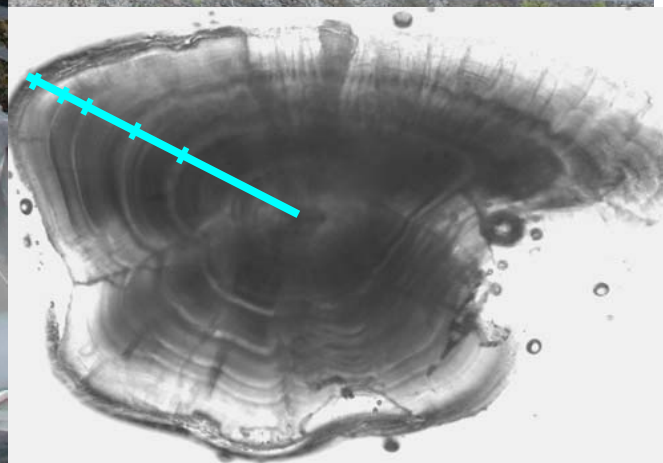
Pesticides and degradation products:

Hexachlorocyclohexanes (HCH) - α*, β, γ-(lindane), and δ, Chlordanes - cis*, trans*, oxy*, Nonachlor – cis, trans, Heptachlor, Heptachlor Epoxide, Endosulfans - I, II, and sulfan Sulfate, Mirex, Methoxychlor, Endrin Aldehyde, Hexachlorobenzene, Dacthal, Chlorothalonil, Chlorpyrifos and oxon, Trifluralin, Metribuzin, Triallate, Mirex

Agriculture

Surrogates: $^{13}C_{12}$ PCB 101 (2,2',4,5,5'-Pentachlorobiphenyl), $^{13}C_{12}$ PCB 180 (2,2', 3,4,4',5,5'-Heptachlorobiphenyl), d_{10} - Chlorpyrifos, $^{13}C_6$-HCB, d_6-γ-HCH, d_4-Endosulfan I, d_4-Endosulfan II, d_{14}-Trifluralin

Internal Standard: $^{13}C_{12}$ PCB 138 (2,2',3,4,4',4',5'-Hexachlorobiphenyl)



WACAP Results:

(With a **GLAC** perspective)

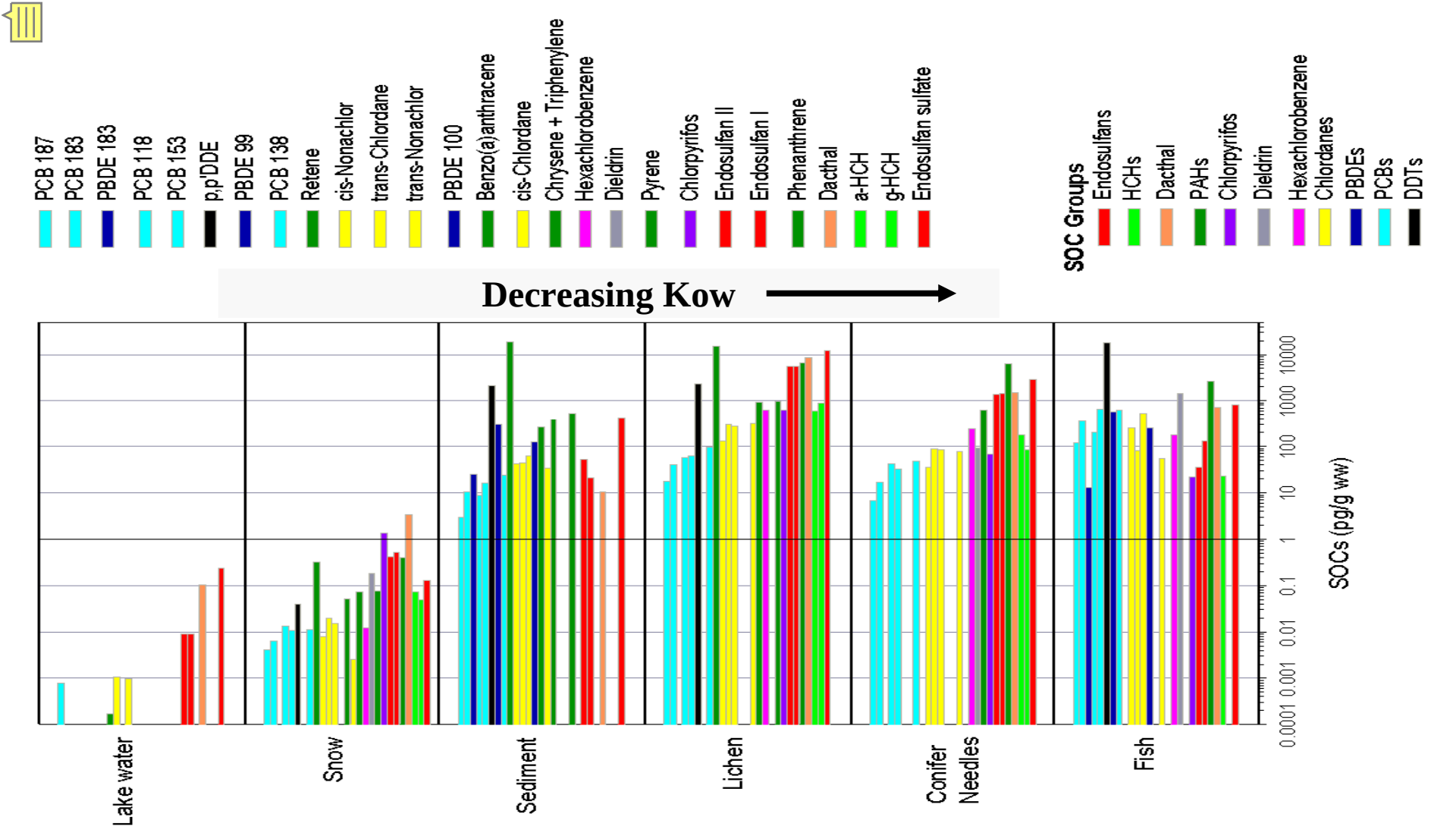


Figure 2. Mean SOC Concentrations (pg/g ww) in Lake Water, Snow, Sediments, Lichens, Conifer Needles, and Fish from Emerald Lake (SEKI). SOCs are ordered by increasing Kow, or decreasing polarity and solubility in water, color-coded by group. SOC concentrations were 3 to 7 orders of magnitude higher in sediments and biota relative to snow and water. SOC concentrations in water, snow, and vegetation, but not sediments and fish, generally decreased with decreasing polarity. Compared to vegetation, fish were better accumulators of PCBs and dieldrin and poorer accumulators of PAHs, endosulfans, HCHs, dacthal, and chlorpyrifos. If no data are shown, all samples were below detection limits; PBDEs were measured in sediments and fish only.

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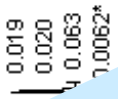
Current Use Pesti



Historic Use Pesticides

ES&T March 2008

Atmospherically Deposited PBDEs, Pesticides, PCBs, and PAHs in Western U.S. National Park Fish: Concentrations and Consumption Guidelines

LUKE K. ACKERMAN,†
ADAM R. SCHWINDT,‡ STACI L. MAS
SIMONICH,*,†,§ DAN C. KOCH
TAMARA F. BLETT,⊥ CARL B. S
MICHAEL L. KENT,‡ AND
DIXON H. LANDERS#

ES&T February 2008

Mercury Concentrations in Salmonids from Western U.S. National Parks and Relationships with Age and Macrophage Aggregates

ADAM R. SCHWINDT,
JOHN W. FOURNIE,
DIXON H. LANDERS,
CARL B. SCHRECK, AND
MICHAEL L. KENT

park for the tou
e pesticides.

Hageman, K. I. et al. 2006. Atmospheric Deposition of Current-U in Snow at National Parks in the Western United States. ES & T

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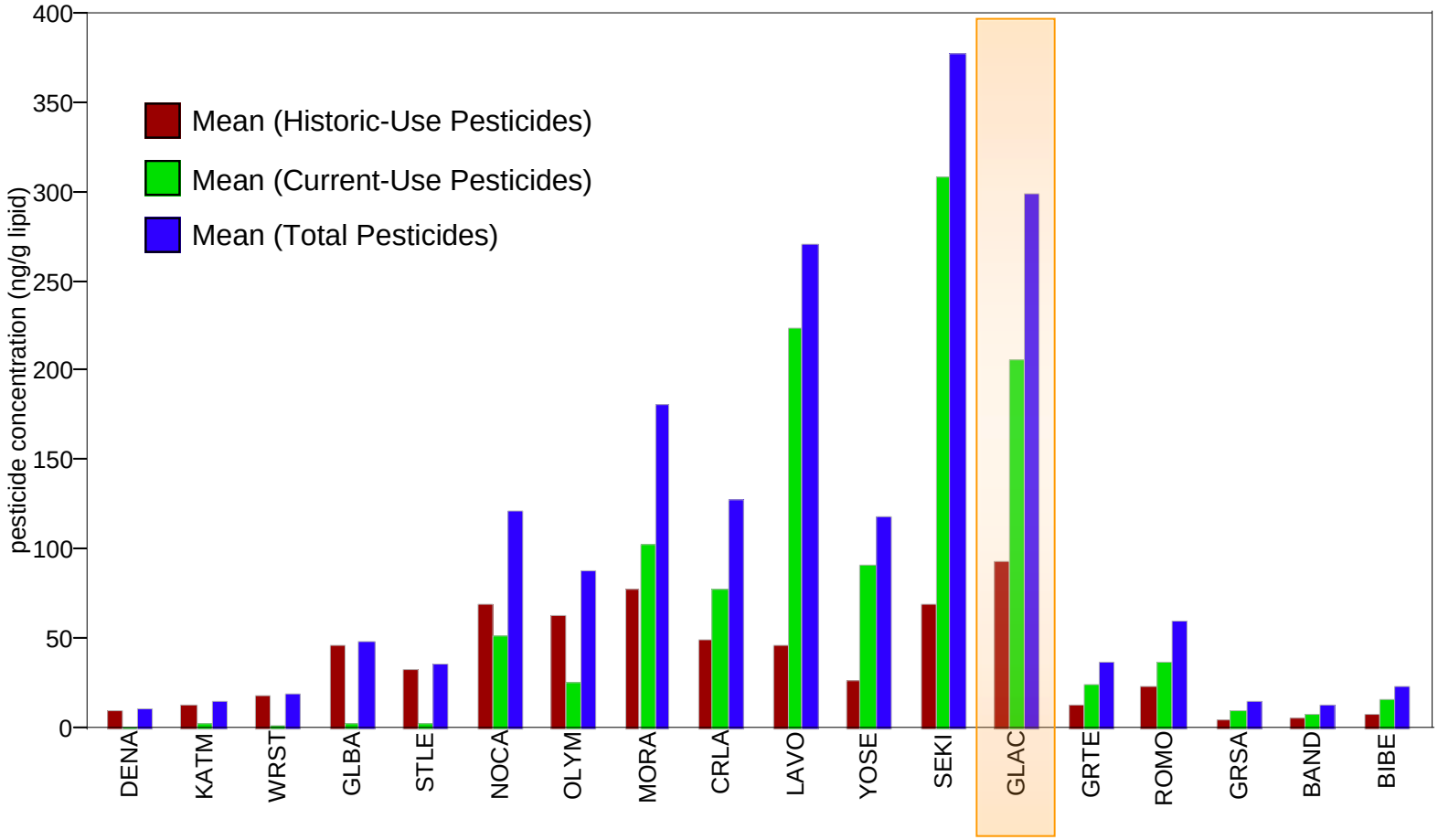

Figure 4-31. Mean Concentrations of Historic-Use (HCB, HCHs, Chlordanes, DDT, Dieldrin) and Current-Use (Trifluralin, Triallate, Chlorpyrifos, Dacthal, Endosulfans) Pesticides in Two-Year-Old Conifer Needles from WACAP Parks. Parks are ordered, left to right, from north to south along the Pacific Coast (DENA → SEKI), and from north to south in the Rocky Mountains (GLAC → BIBE). Current-use pesticides were not detected often in Alaska parks, comprised about one-third to one-half the total pesticide concentrations in northern Washington, and most of the pesticide burden elsewhere. Conifer needles were not sampled in NOAT and GAAR. Total pesticide burdens (current-use + historic-use) were highest in national parks of Washington, Oregon, California, and Montana.

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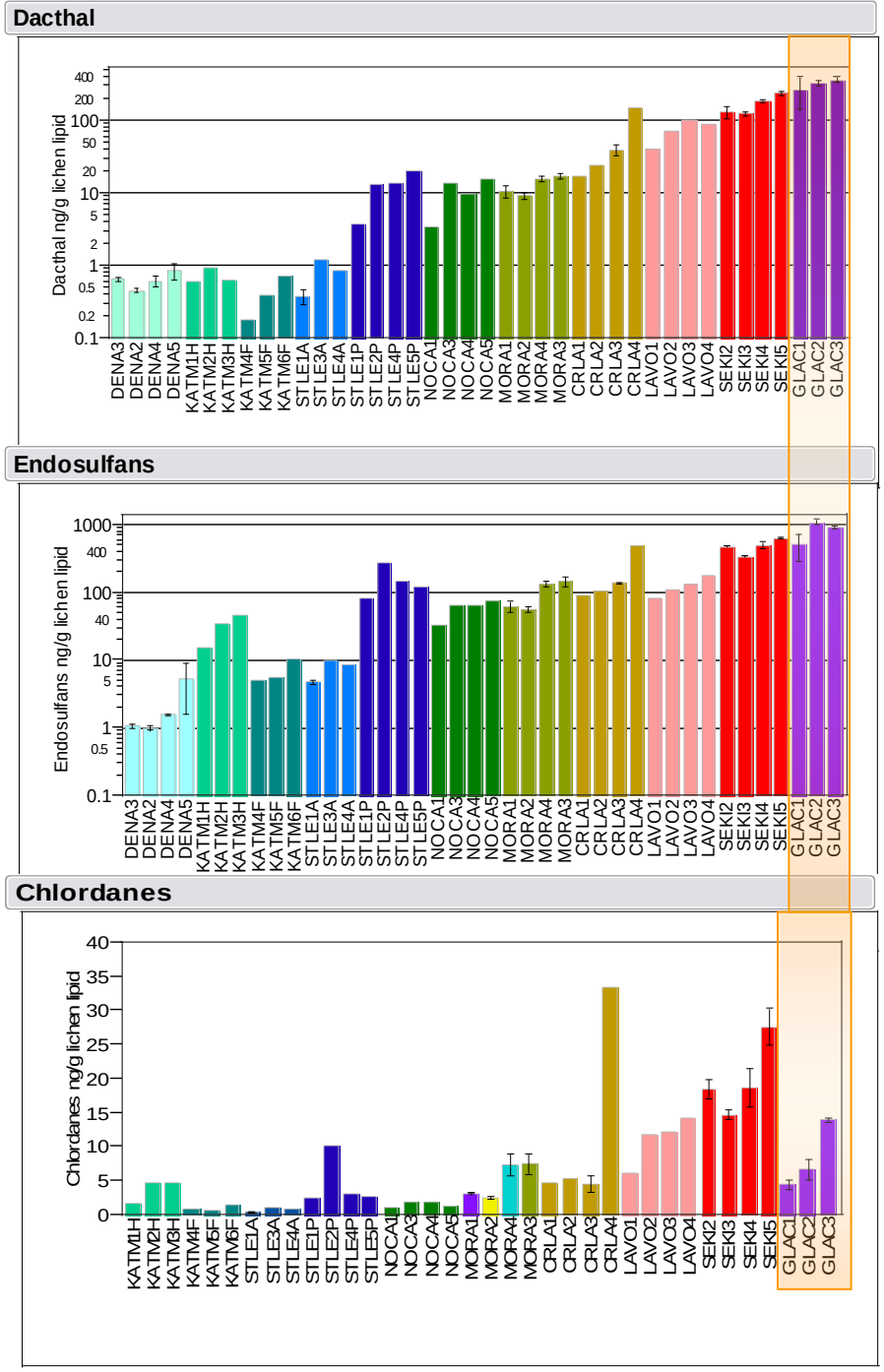

Lichens plotted by elevation within Parks

Figure 4-22. Elevational Gradients for Sum Dacthal, Sum Endosulfan, and Sum Chlordane Concentrations in Lichens. Within each park, sites are listed in order of increasing elevation. Codes H, F, A, and P refer to lichen species sampled (see Table 4-4). Bars show the standard error. Statistical analyses of elevational gradients are reported in Tables 4-3 and 4-4. Additional graphic displays for other SOCs are given in Appendix 4A.10. See Chapter 3 for data selection criteria for elevational trends analyses.

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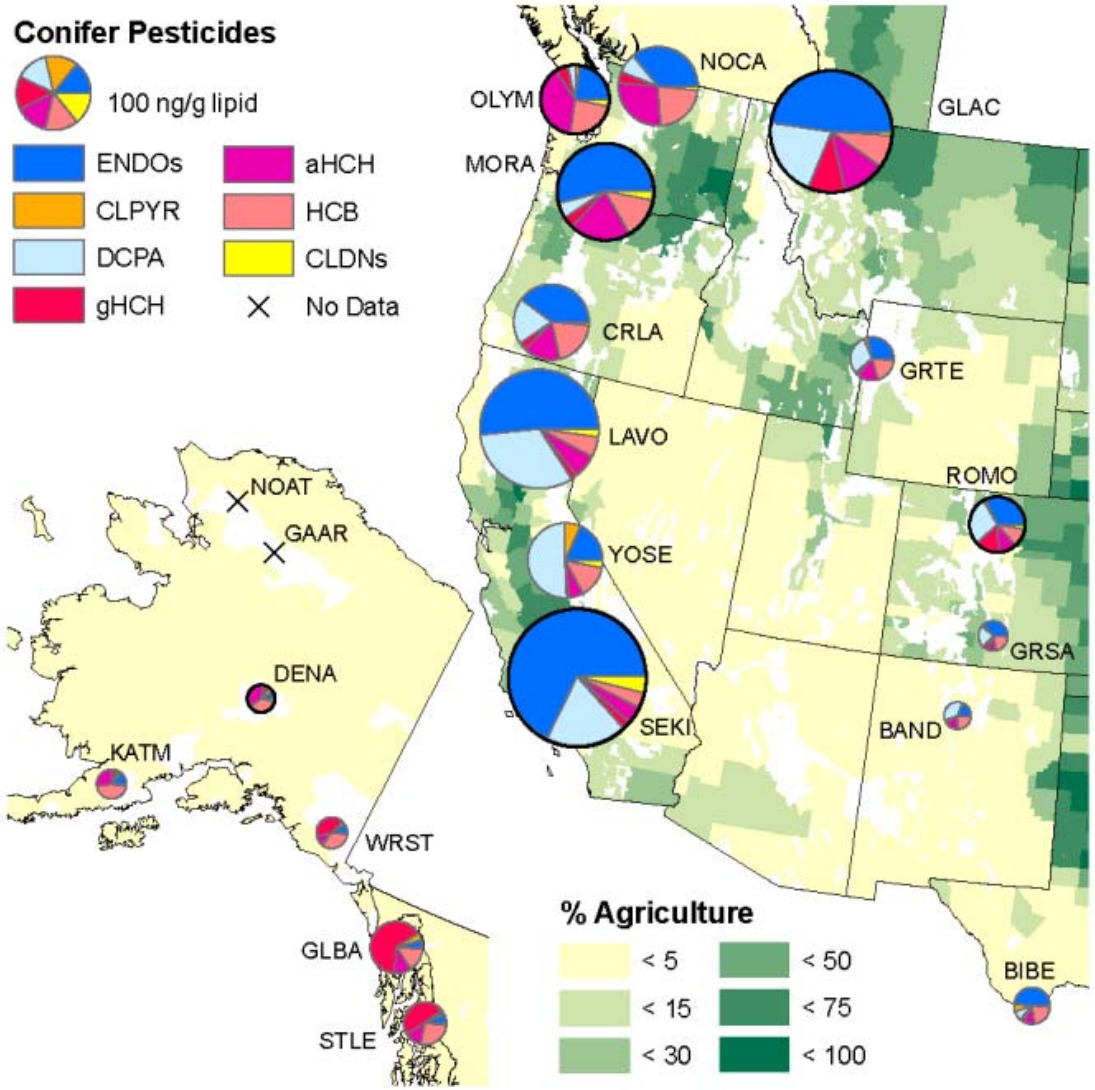

Figure 4-13. Pesticide Concentrations (ng/g lipid) in Conifer Needles from Core and Secondary WACAP Parks Overlaid on a Map of Agricultural Intensity (US Department of Agriculture, National Agriculture Statistics Service, 2002). Circle area is proportional to total pesticide concentration. Light to dark green shading indicates increasing agricultural intensity. White shading indicates national forests or parks. Current-use pesticides endosulfan and dacthal dominate pesticide concentrations in parks in the conterminous United States, where most agriculture occurs. Historic-use pesticides are relatively more important in Alaska, although total contaminant concentrations are lower. Conifers were not present in NOAT and GAAR. Sites outlined in black are the core parks. Pesticide coding is identical to that in Figure 4-12.

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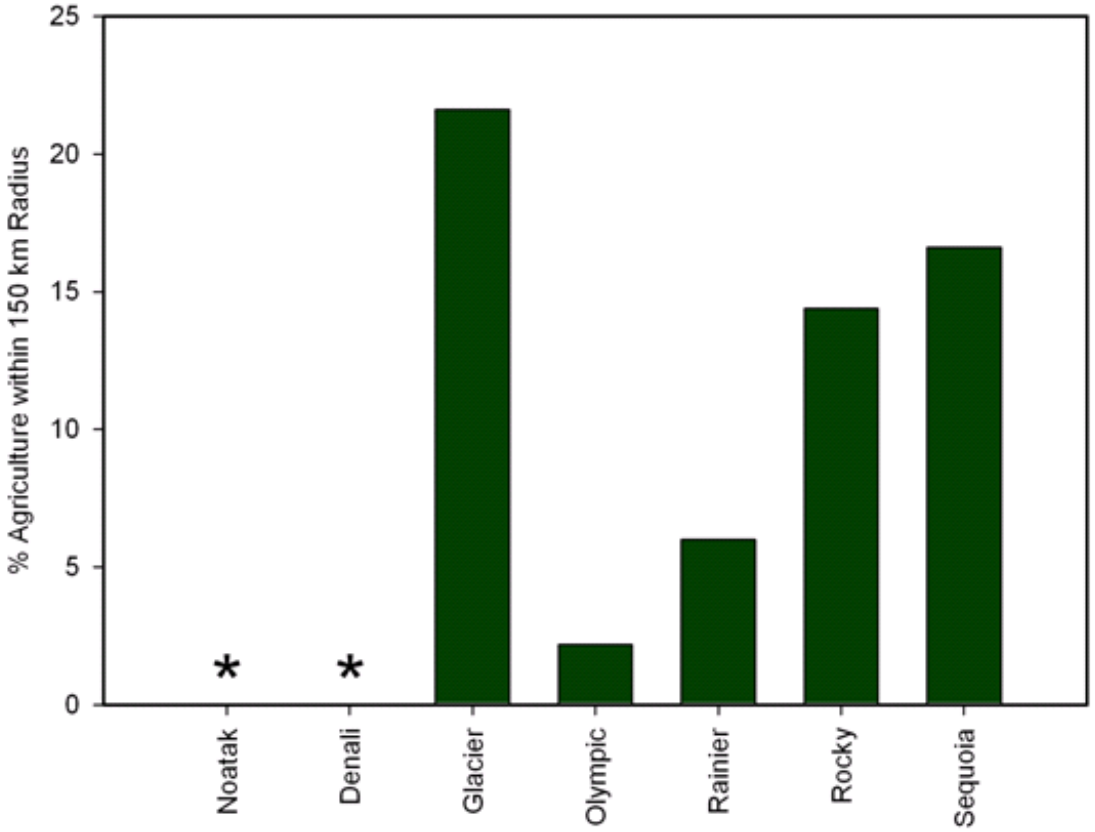

Figure 6: Percent of agricultural land within 150 km of the primary WACAP parks. Alaskan parks have no surrounding agriculture.

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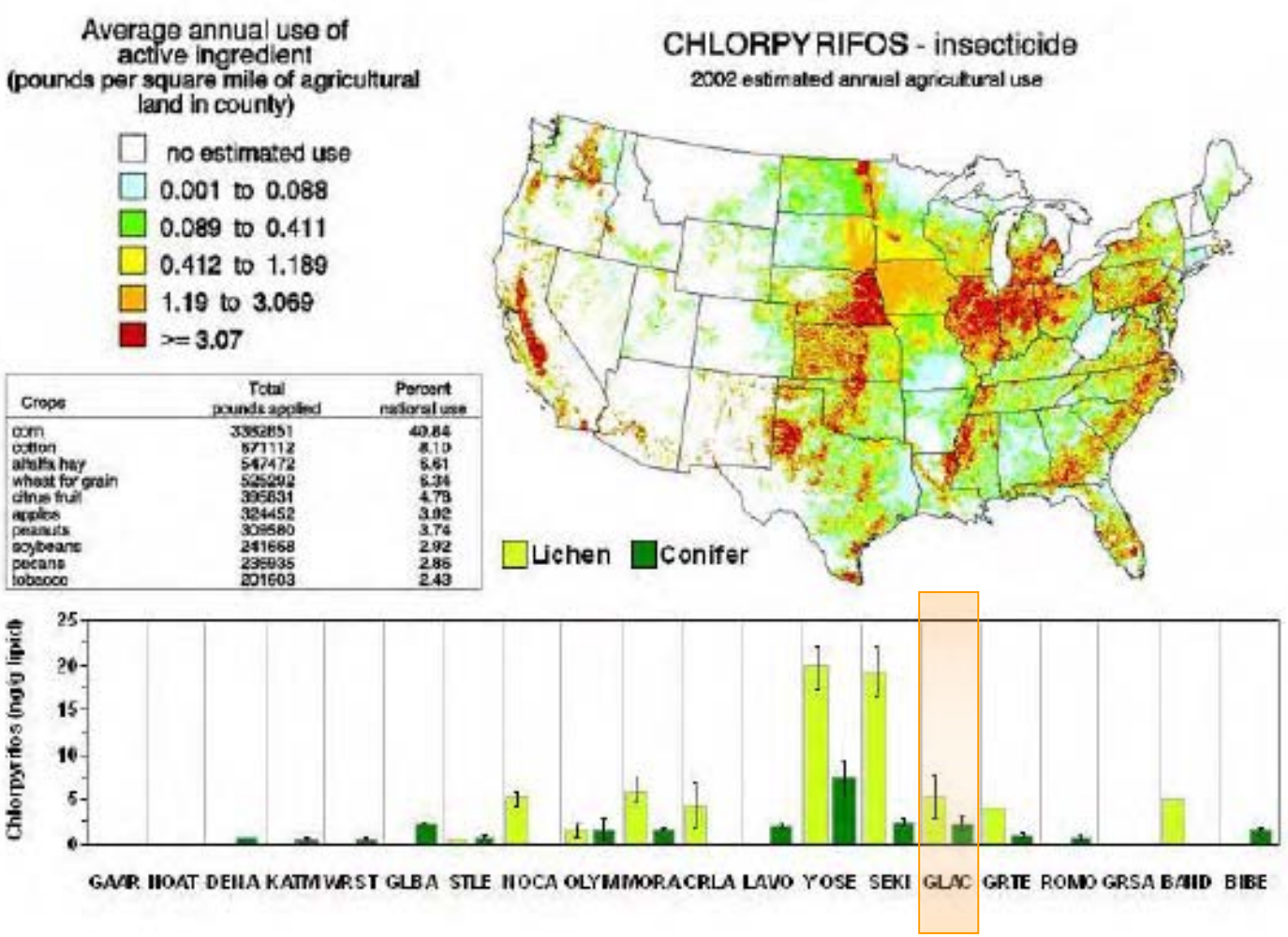

Figure 4-14. Uses and Estimated Application Intensity in 2002 of the Current-Use Insecticide Chlorpyrifos in the Conterminous 48 States vs. Mean Concentration in Vegetation (ng chlorpyrifos/g lipid conifer needles or lichens) from WACAP Parks. Chlorpyrifos were detected in vegetation in all parks except NOAT and GAAR, but highest concentrations were observed in SEKI and YOSE, close to the San Joaquin Valley in California, a particularly high use area. Error bars indicate one standard error.

Source of chlorpyrifos data:
http://ca.water.usgs.gov/pnsp/pesticide_use_maps/show_map.php?year=02&map=m6009.

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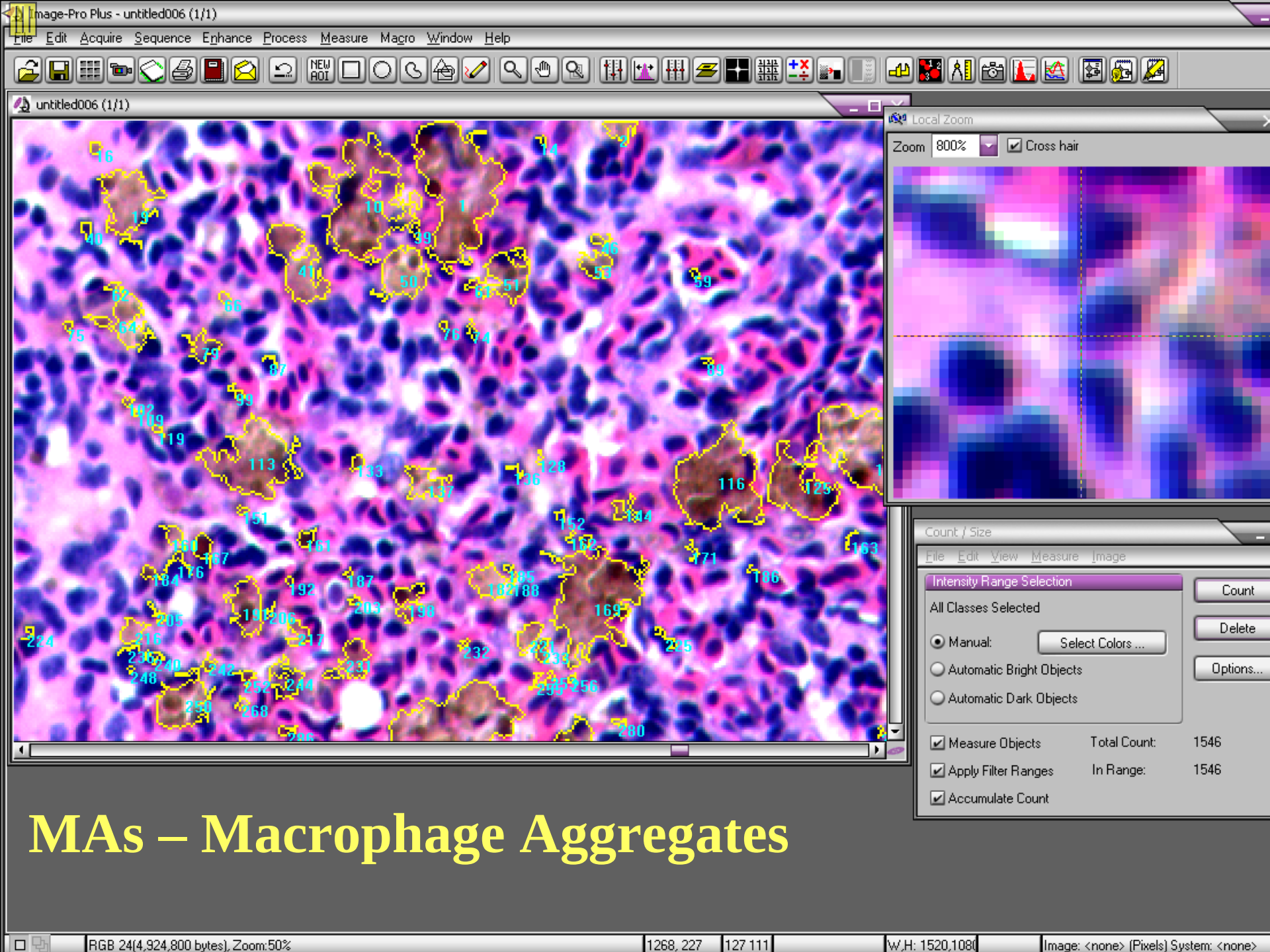
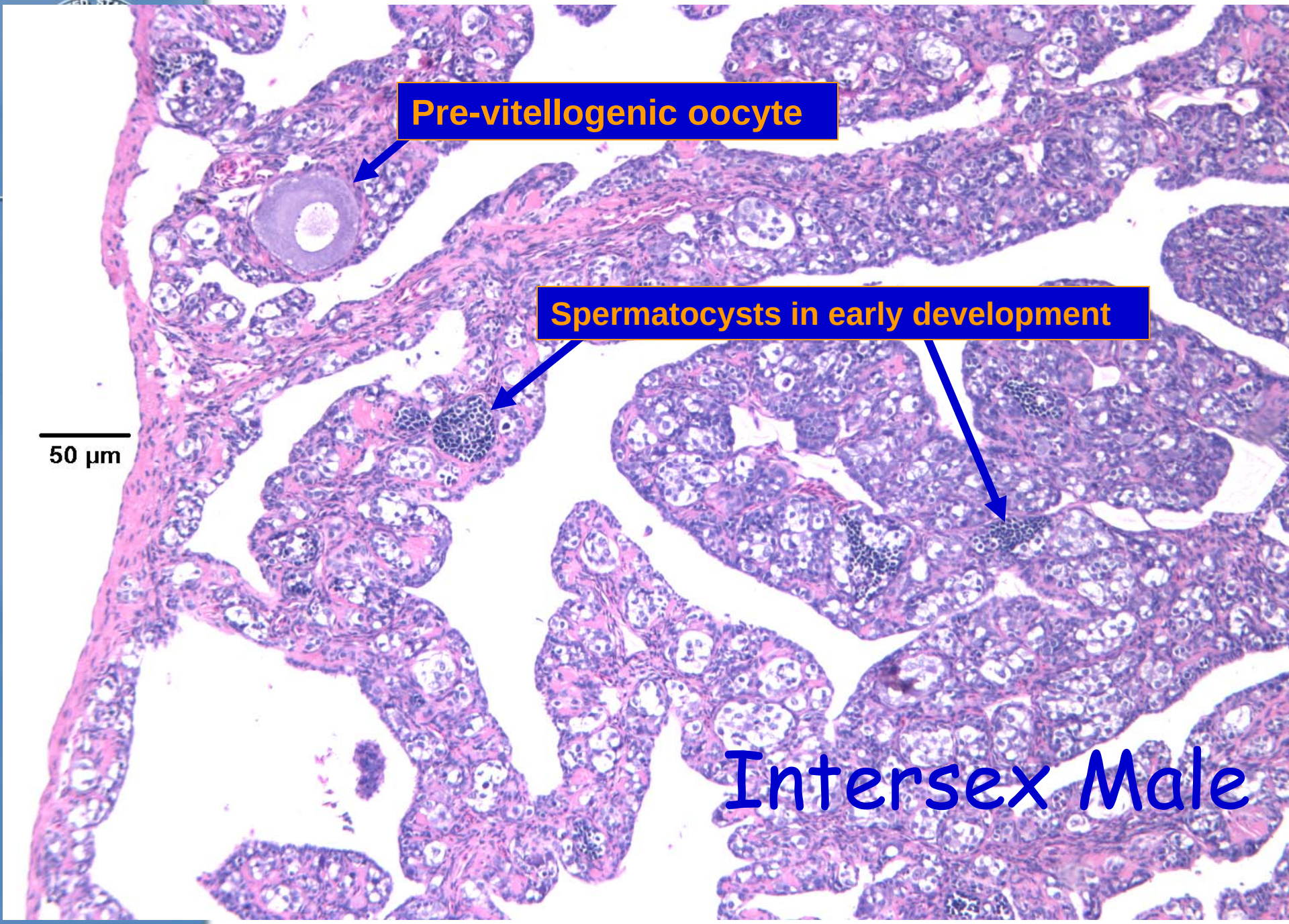
FISH STUDIES

Do Airborne Contaminants Affect Fish in the Lakes of Western US National Parks?



MAs – Macrophage Aggregates


Pre-vitellogenic oocyte
Spermatocysts in early development
50 µm
Intersex Male

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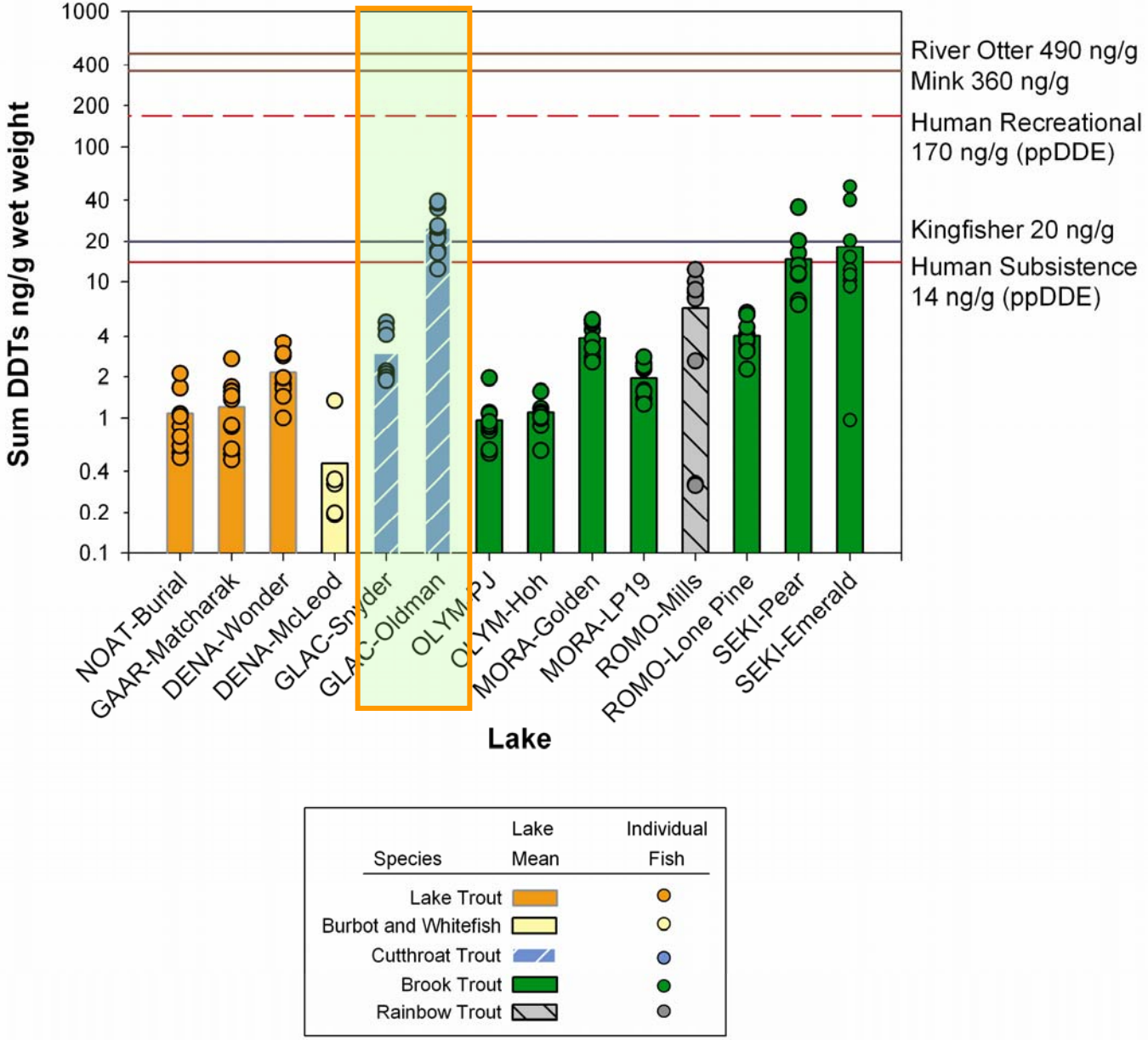
Mean Whole Body Fish Sum DDTs

Historic-Use



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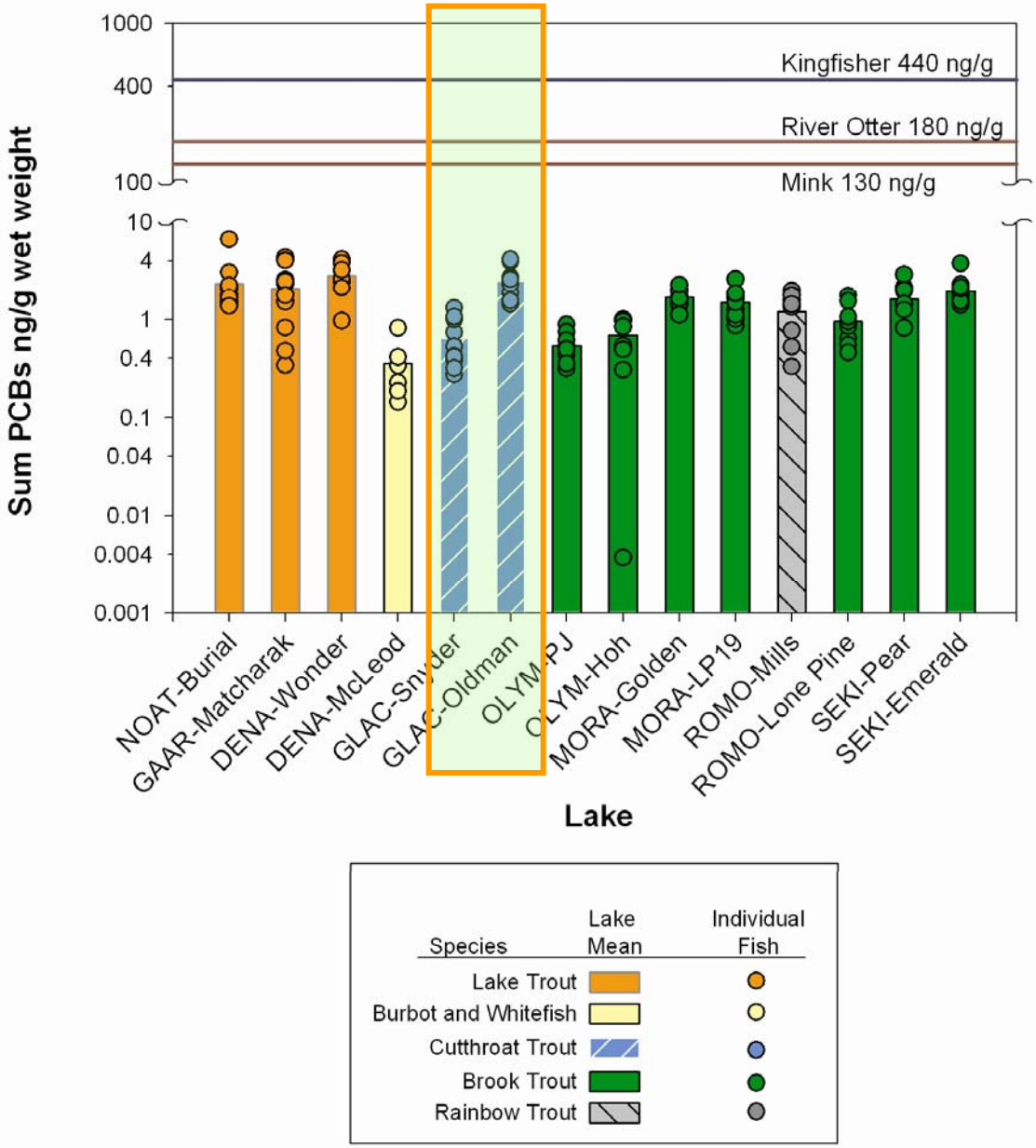
Mean Whole Body Fish Sum of PCBs

Historic-Use



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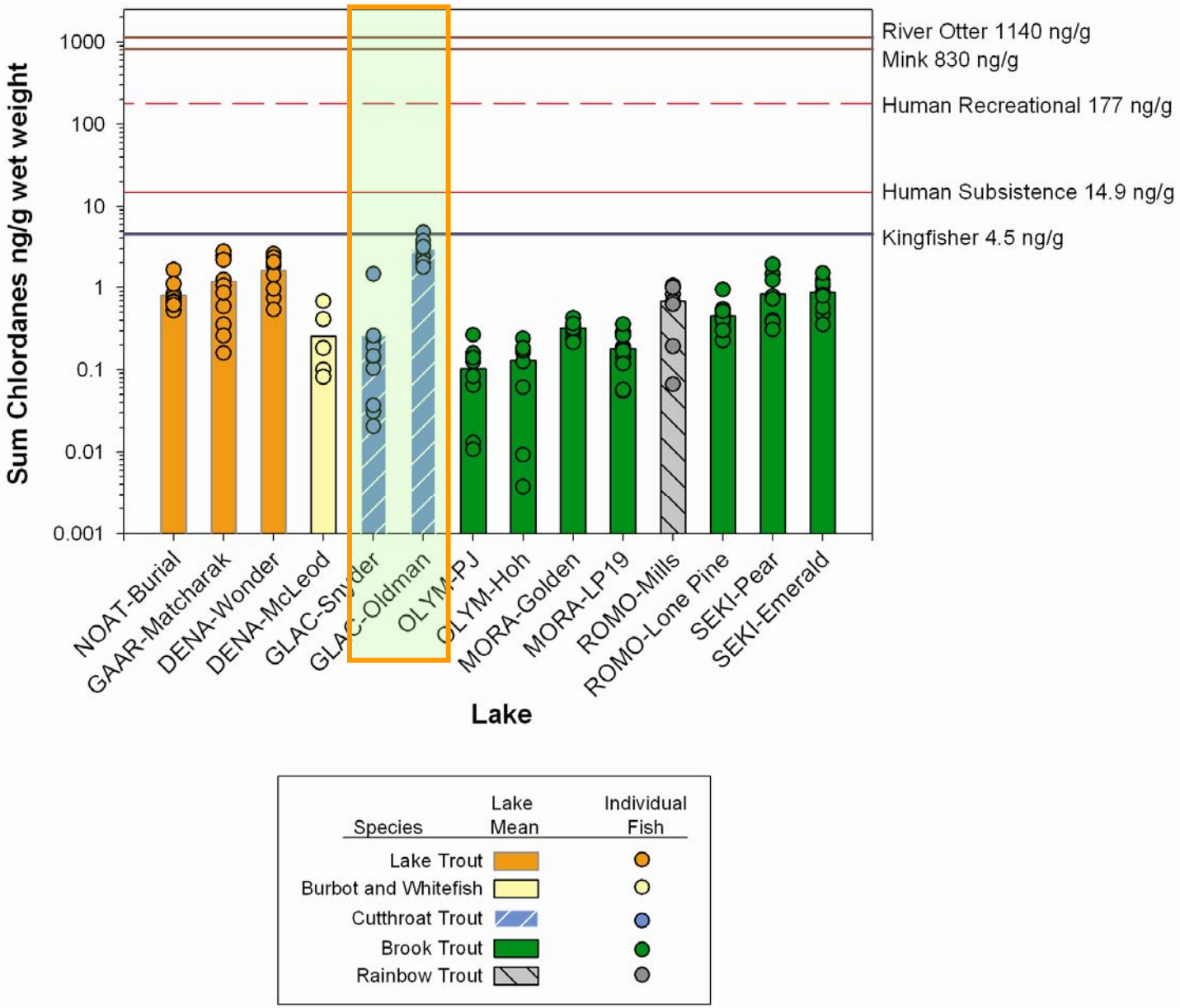
Mean Whole Body Fish Sum Chlordanes

Historic-Use



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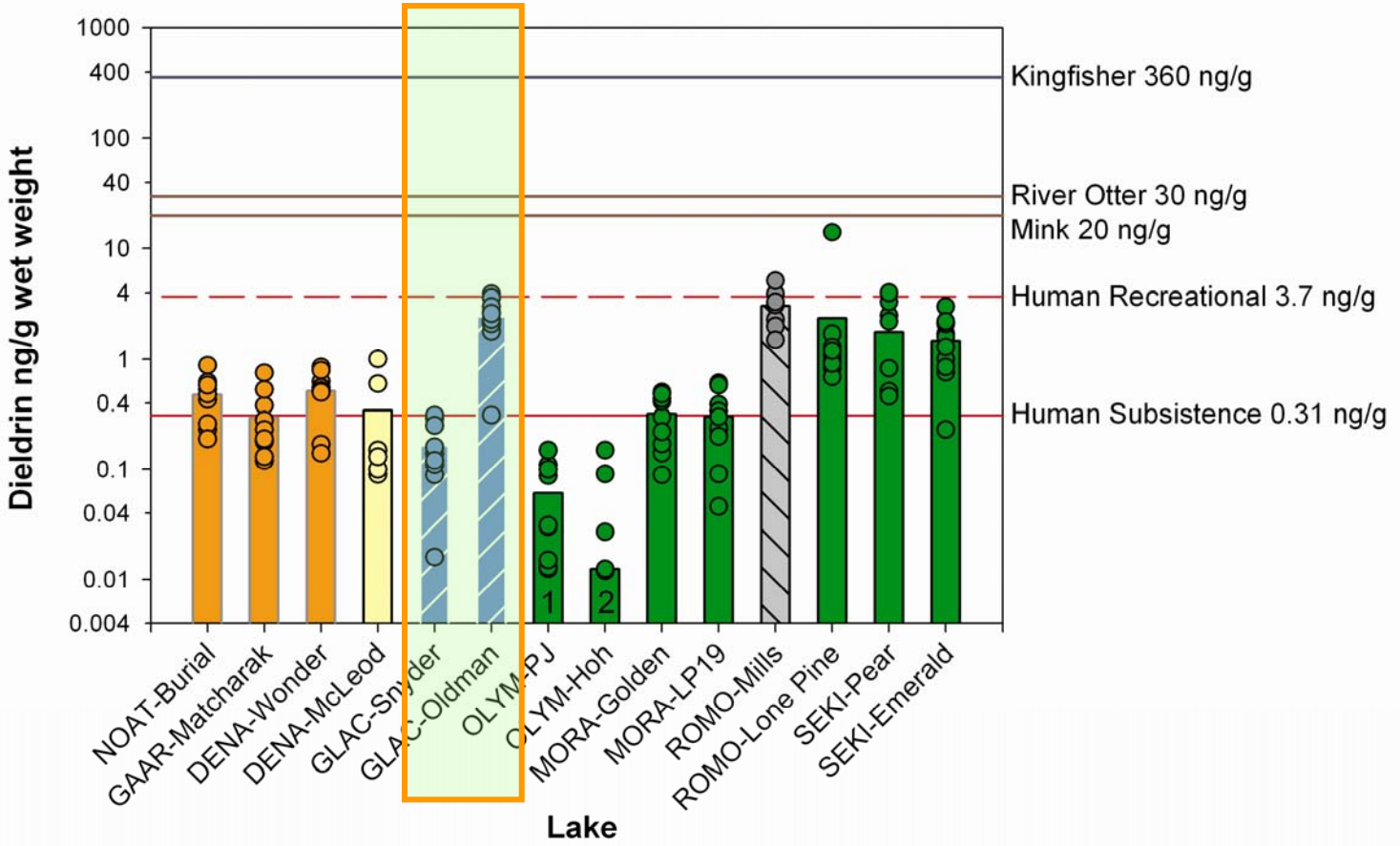
Mean Whole Body Fish Dieldrin

Historic-Use



Species	Lake Mean	Individual Fish
Lake Trout		
Burbot and Whitefish		
Cutthroat Trout		
Brook Trout		
Rainbow Trout		

If no label is present in the bar, the component was detected in at least 70% of the samples.
"1" indicates the analyte was detected in 50 - 70% of the samples,
"2" indicates the analyte was detected in less than 50% of the samples.

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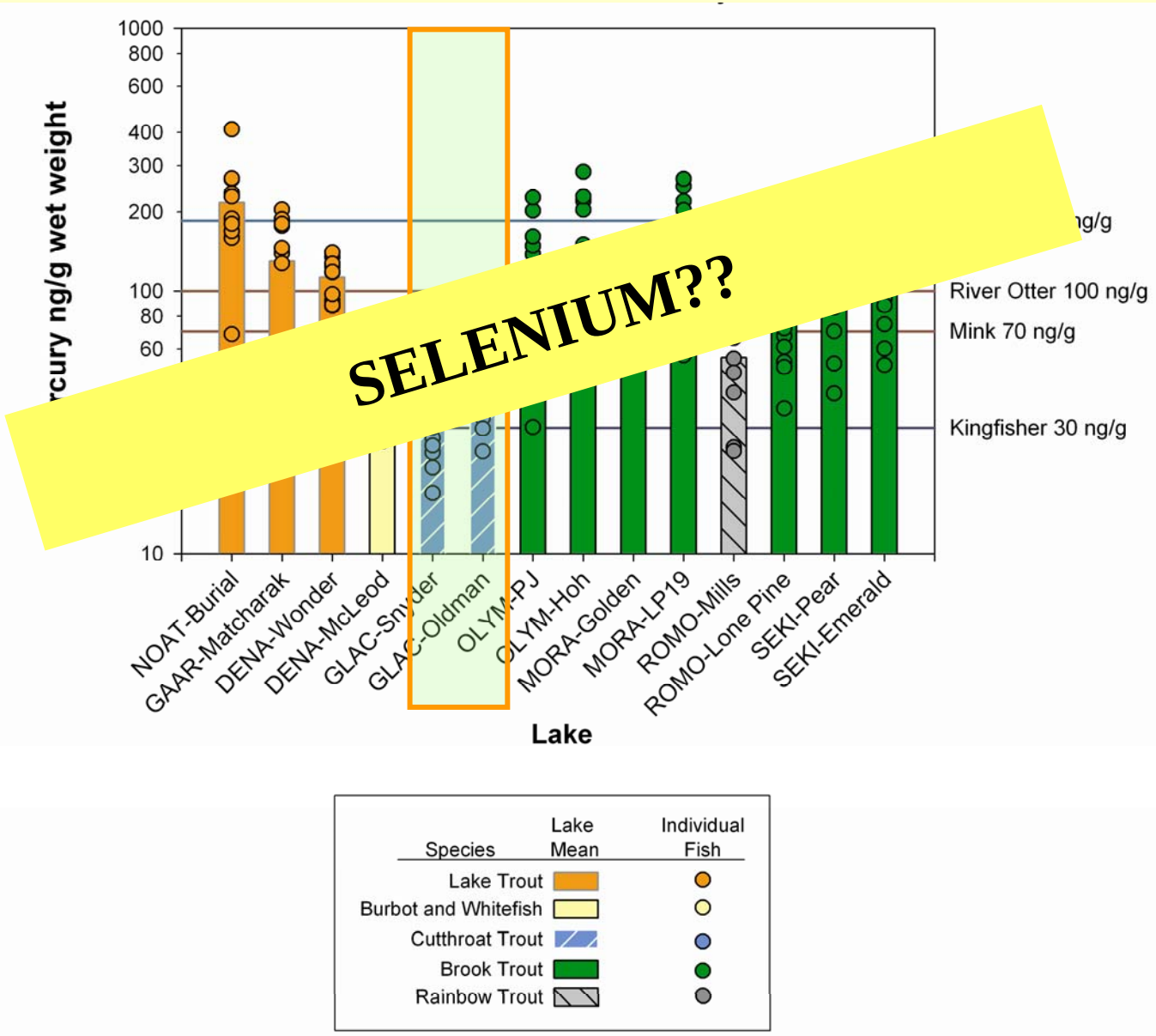
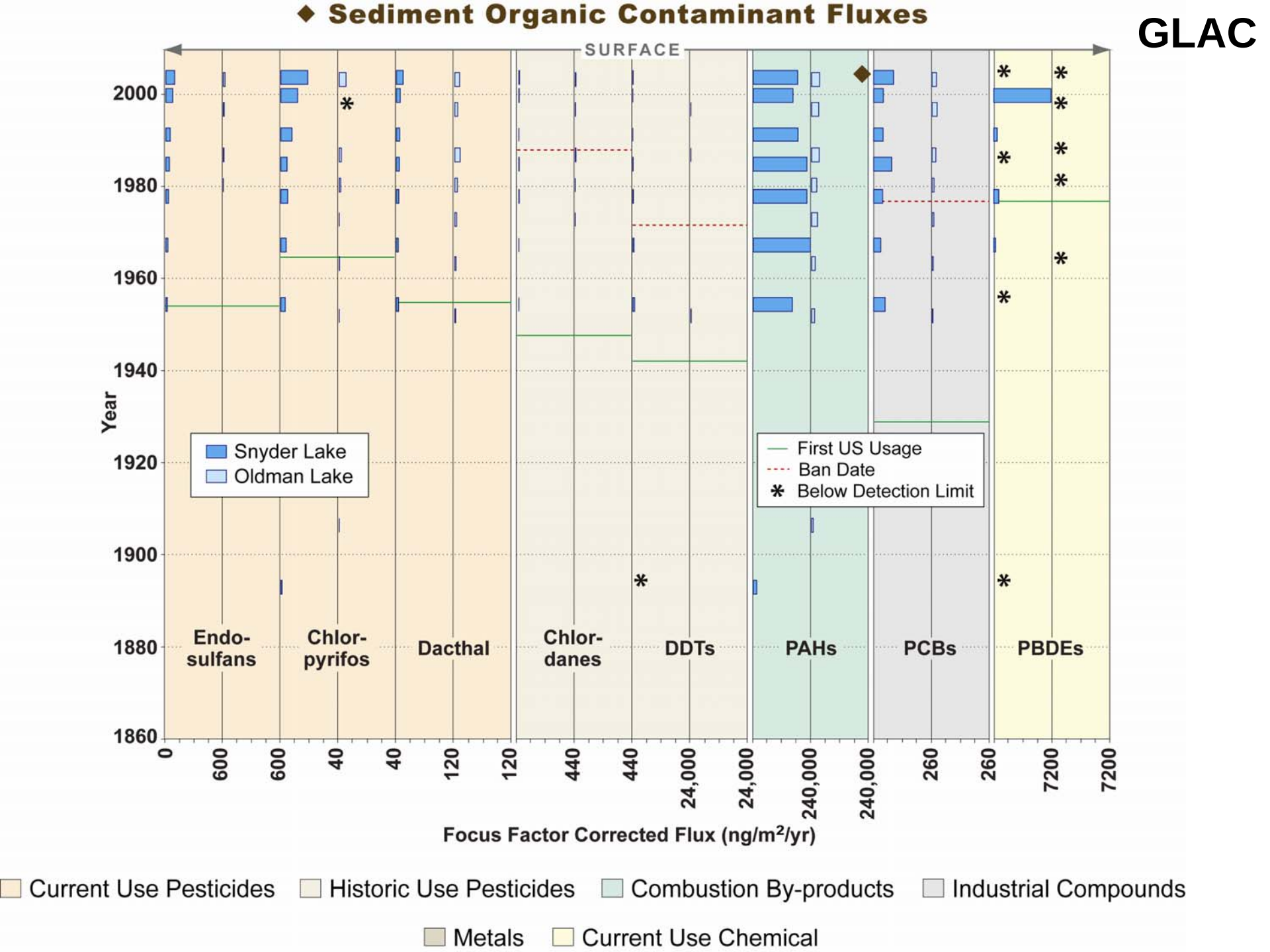
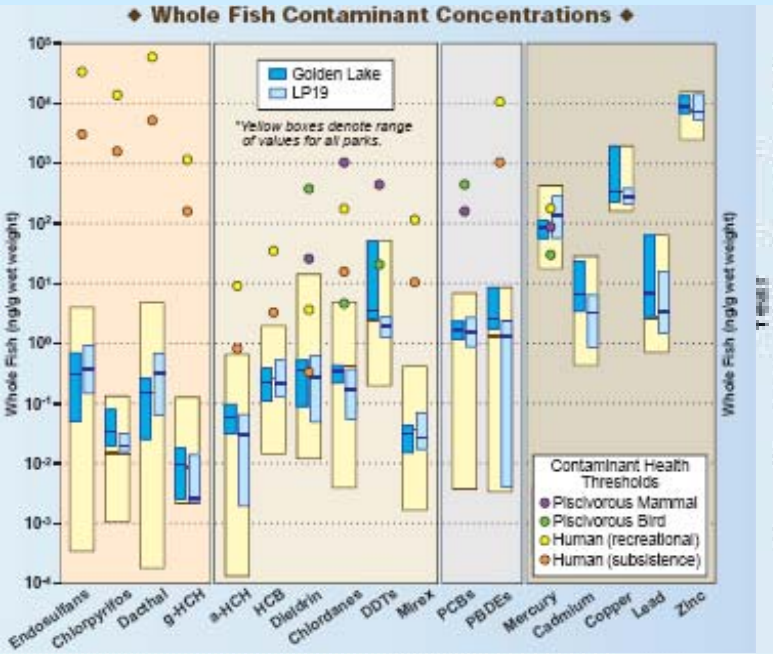
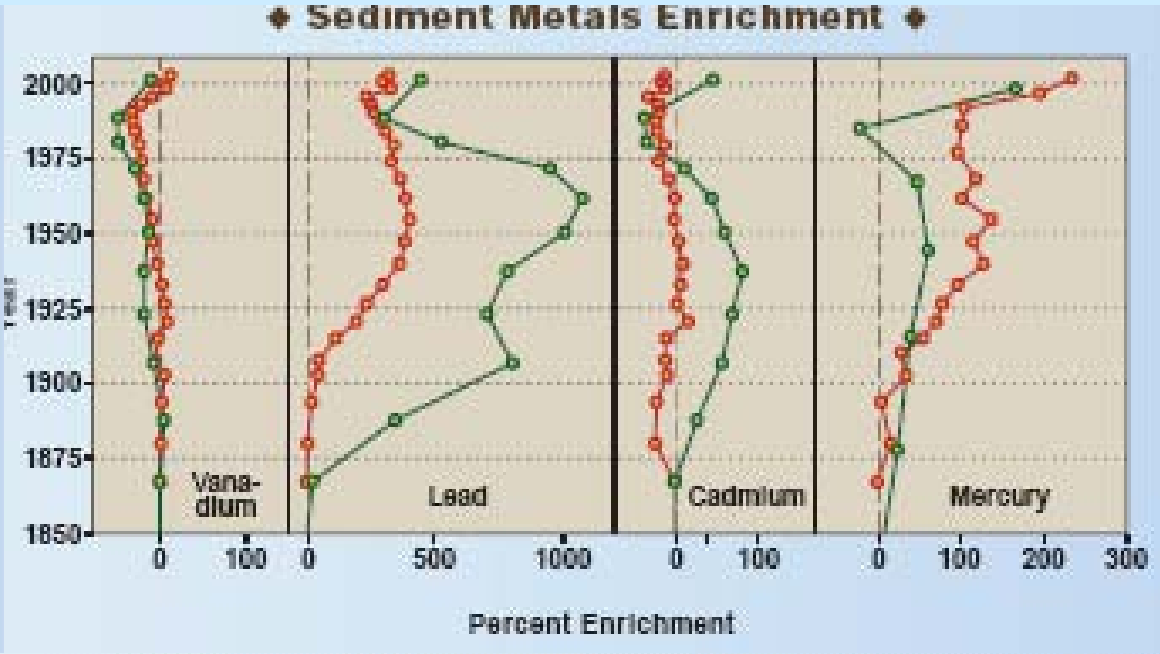
Mean Whole Body Fish Total Hg




◆ Sediment Organic Contaminant Fluxes
GLAC
SURFACE
Year
2000
1980
1960
1940
1920
1900
1880
1860
Snyder Lake
Oldman Lake
First US Usage
Ban Date
* Below Detection Limit
Endo-sulfans
Chlor-pyrifos
Dacthal
Chlor-danes
DDTs
PAHs
PCBs
PBDEs
0
600
600
40
40
120
120
440
440
24,000
24,000
240,000
240,000
260
260
7200
7200
Focus Factor Corrected Flux (ng/m²/yr)
Current Use Pesticides
Historic Use Pesticides
Combustion By-products
Industrial Compounds
Metals
Current Use Chemical

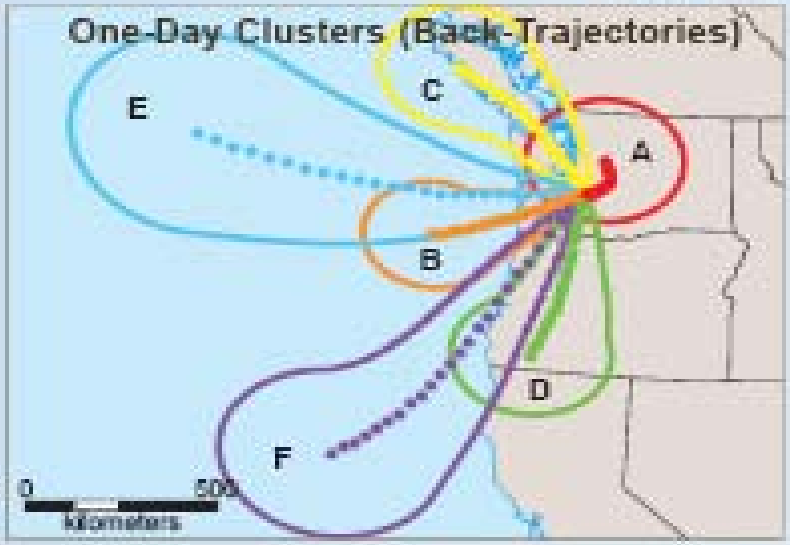
Mount Rainier National Park – Park Summary

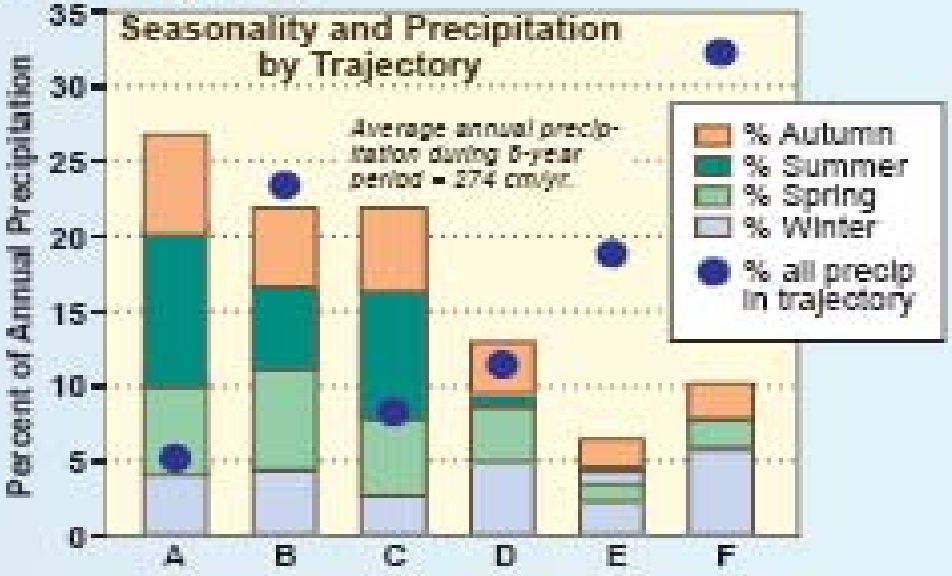




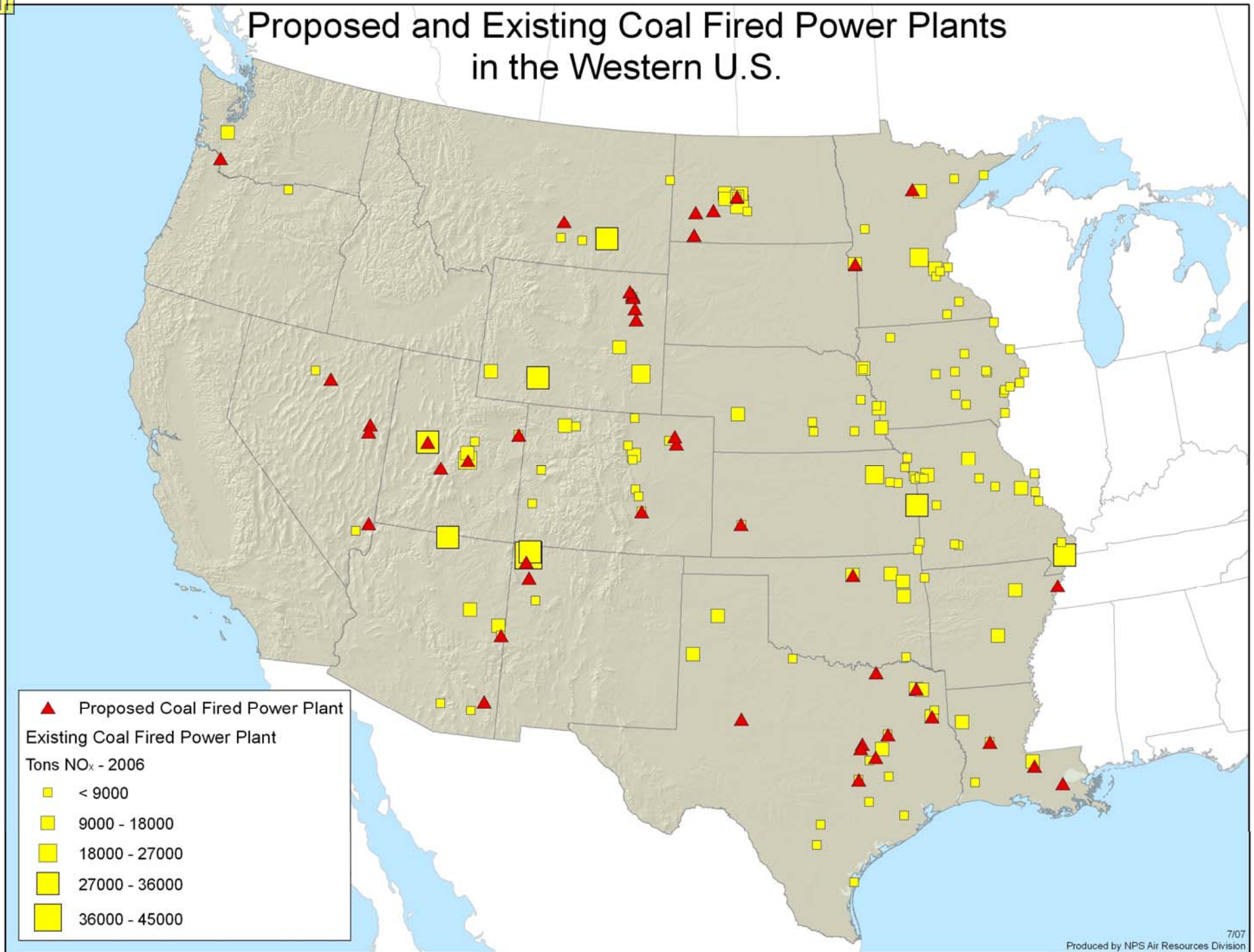
Current-Use Pesticides | Historic-Use Pesticides | Combustion By-products | Industrial Compounds | Metals | Current-Use Chemical

Atmospheric Transport





Proposed and Existing Coal Fired Power Plants in the Western U.S.



"Well, thank God we all made it out in time.
...'Course, now we're equally screwed."

You can
run,
But you
can't hide

Finish

WACAP WEB SITE:
http://www.nature.nps.gov/air/Studies/air_toxics/wacap.cfm

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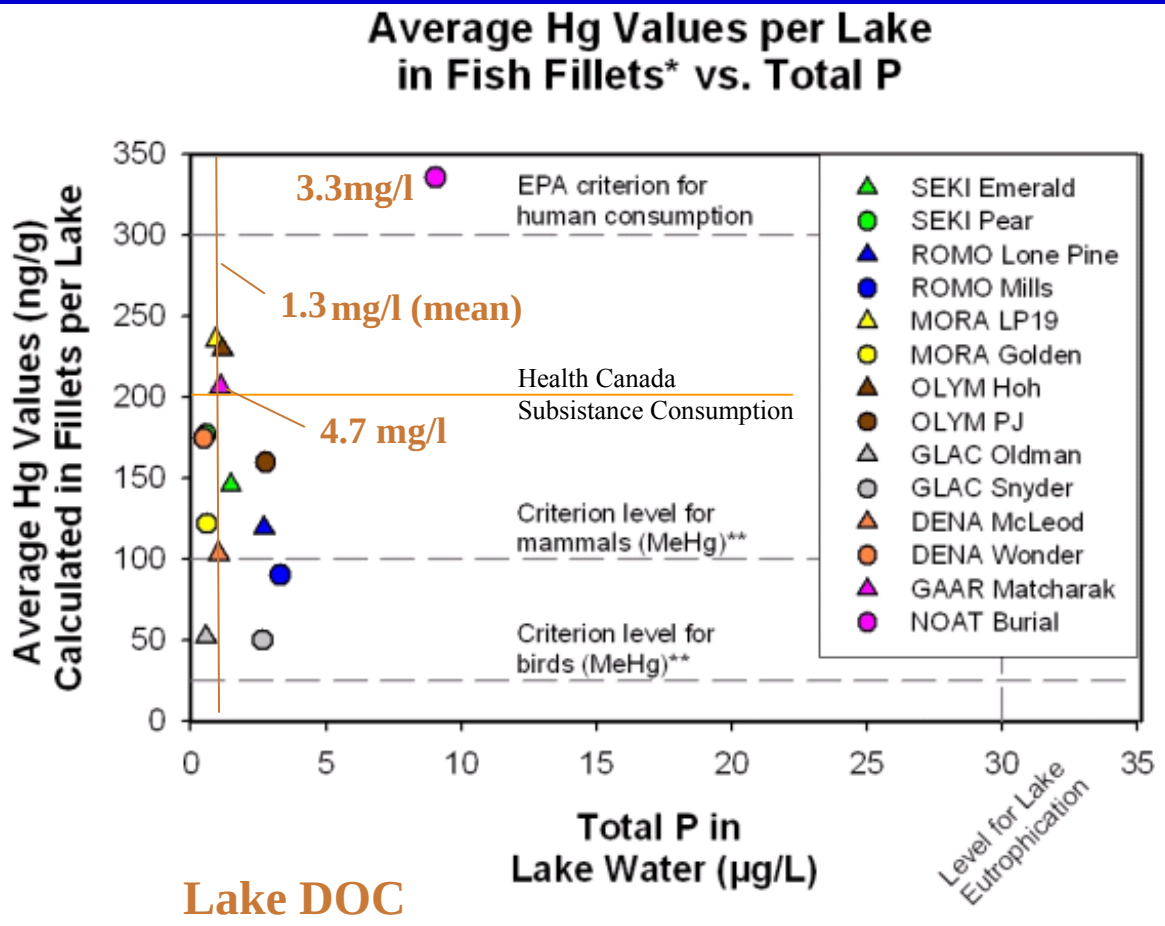
WACAP

Key WACAP Conclusion

- **Over half (77 of 136) of the individual fish from 11 of the 14 WACAP lakes, carried body burdens exceeding subsistence fishing limits for Dieldrin and/or p,p'-DDE; limits were calculated using US EPA guidelines. Exceeding a limit implies an increased risk (by at least 1 in 100,000) of developing cancer during a lifetime of frequent fish consumption.**



* Fillet concentrations calculated from whole fish Hg concentrations with method from Peterson *et al.* (2005), Arch Environ. Contam. Toxicol. 48:99-107

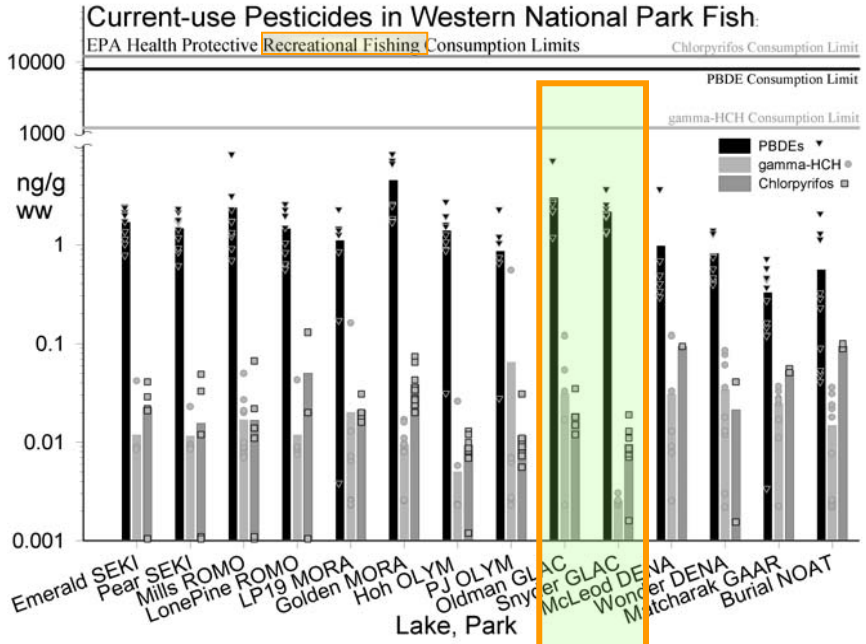
**MeHg limits taken from Yeardley et al., 1998 Environ. Toxicol. Chem. 17:1875-1884

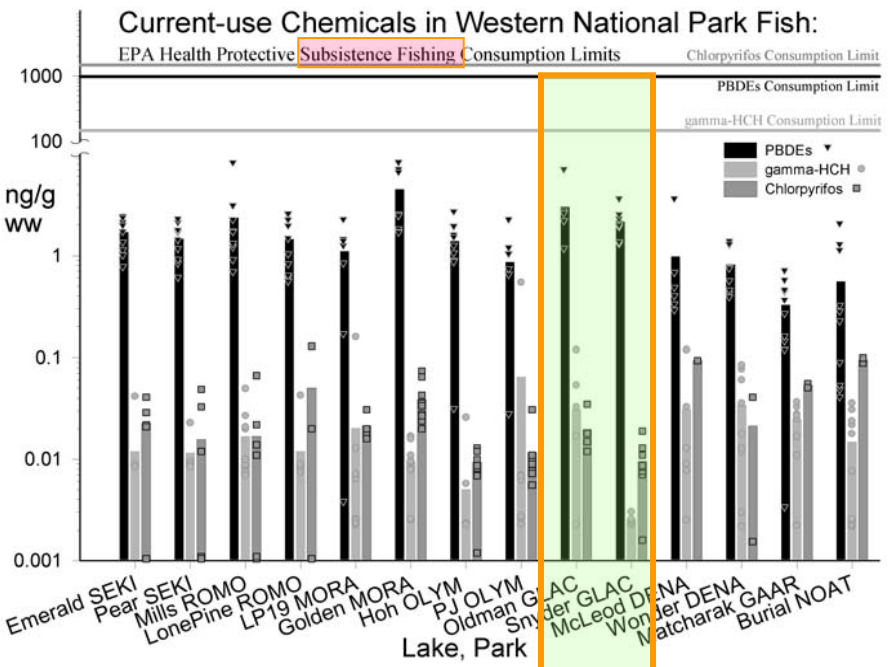
Conclusions Continued:

Vegetation. **Numbers and concentrations of PAHs detected were highest at GLAC compared to other parks. Close proximity to an aluminum smelter suggests a local source of PAHs contributing to the high concentrations. Other SOCs were in the mid to upper ranges compared to other parks such as: endosulfans, dacthal, DDTs, g-HCHs, a-HCH, HCB, triallate, chlorpyrifos, and PCBs. Dacthal, endosulfans, HCB, a-HCH, chlorpyrifos, DDTs, PCBs and PAHs were higher on the west side of the park, attributable to precipitation and temperature. Triallate, chlorpyrifos, and g-HCH were higher on the east side of the park, potentially attributable to agricultural intensity. Enhanced nitrogen and sulfur deposition related to regional agricultural intensity is of concern. Many rare but not highly toxic elements were high in lichen compared to other parks. Because forest productivity is high, pesticides scrubbed from the air by vegetation likely contribute significant contaminant loads to the ecosystem via canopy through-fall and needle litter-fall.**

Fish. **Pesticides (dacthal, g-HCH, HCB, dieldrin, and chlordanes) in Oldman Lake were higher than Snyder Lake possibly related to agricultural intensity. Oldman Lake fish exceeded contaminant health thresholds for otter, mink, and/or kingfishers for dieldrin, DDTs and PCBs. Fish in both lakes exceeded otter, mink, and/or kingfisher thresholds for chlordanes and Hg. Lake average dieldrin and p,p'-DDE fish concentrations in Oldman Lake exceeded contaminant health thresholds for subsistence fishers. Dieldrin concentrations in individual fish in Oldman Lake exceeded contaminant health thresholds for recreational fishers. Mercury was age-dependent at Snyder Lake. Kidney and/or spleen macrophage aggregates were significantly related to mercury and age at both lakes. All fish appeared reproductively normal, but elevated concentrations of estrogen-responsive protein were found in males from both lakes. One intersex male was found at Oldman Lake. These data are suggestive of endocrine disruption.**

Sediment. **SOC profiles were consistent with the first usage of these chemicals in the United States but, most have not decreased after use ceased. Snyder Lake profiles generally showed greater contaminant flux than Oldman Lake. PAHs in Snyder Lake indicate some decline in the recent sediments since approximately 1990. Lead, cadmium, and mercury profiles increase from approximately 1875 and decrease beginning in the 1960s. These profiles suggest a common historic source that may have been affected by reductions in emissions due to the Clean Air Act. This relationship is supported by the pattern observed in SCPs.**


Current-use Pesticides in Western National Park Fish:
EPA Health Protective Recreational Fishing Consumption Limits
Chlorpyrifos Consumption Limit
PBDE Consumption Limit
gamma-HCH Consumption Limit
PBDEs
gamma-HCH
Chlorpyrifos
ng/g ww
10000
1000
1
0.1
0.01
0.001
Emerald SEKI
Pear SEKI
Mills ROMO
LonePine ROMO
LP19 MORA
Golden MORA
Hoh OLYM
PJ OLYM
Oldman GLAC
Snyder GLAC
McLeod DENA
Wonder DENA
Matcharak GAAR
Burial NOAT
Lake, Park


Current-use Chemicals in Western National Park Fish:
EPA Health Protective Subsistence Fishing Consumption Limits
Chlorpyrifos Consumption Limit
PBDEs Consumption Limit
gamma-HCH Consumption Limit
PBDEs
gamma-HCH
Chlorpyrifos
ng/g ww
1000
100
1
0.1
0.01
0.001
Emerald SEKI
Pear SEKI
Mills ROMO
LonePine ROMO
LP19 MORA
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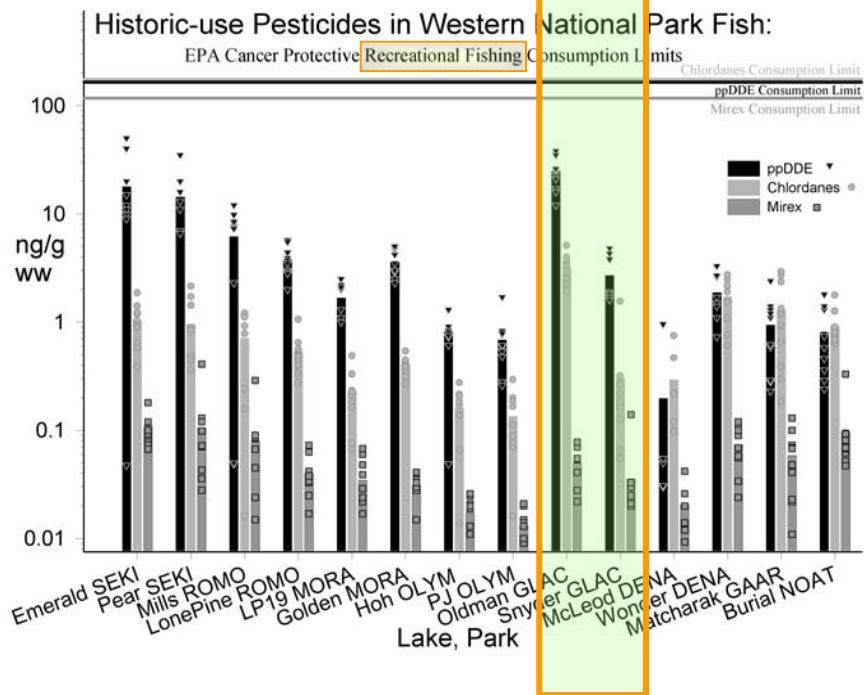
Historic-use Pesticides in Western National Park Fish:
EPA Cancer Protective Recreational Fishing Consumption Limits
Chlordanes Consumption Limit
ppDDE Consumption Limit
Mirex Consumption Limit
ppDDE
Chlordanes
Mirex
ng/g ww
100
10
1
0.1
0.01
Emerald SEKI
Pear SEKI
Mills ROMO
LonePine ROMO
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Golden MORA
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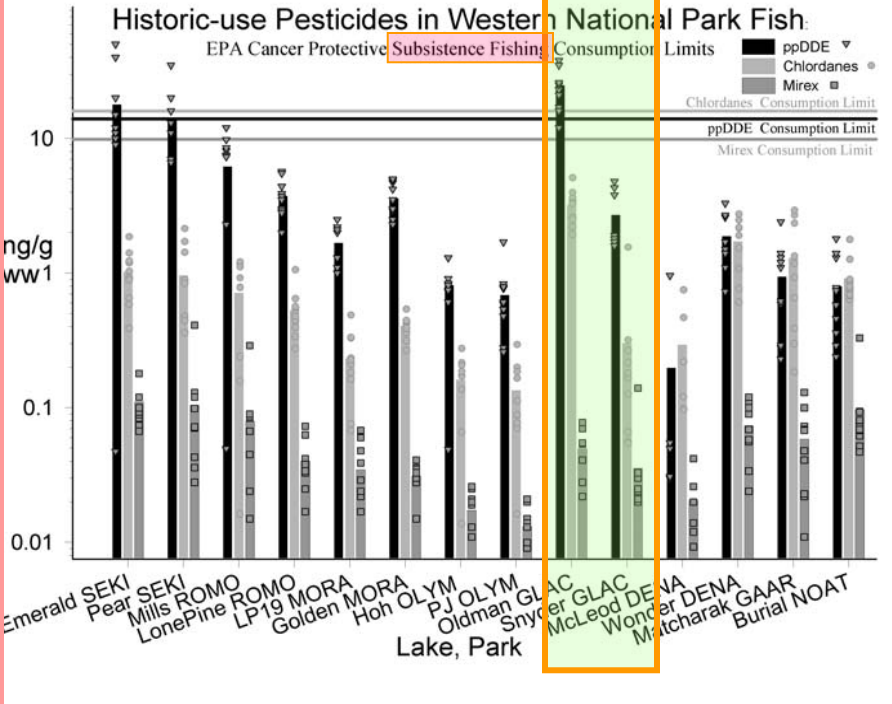
Historic-use Pesticides in Western National Park Fish:
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ppDDE
Chlordanes
Mirex
Chlordanes Consumption Limit
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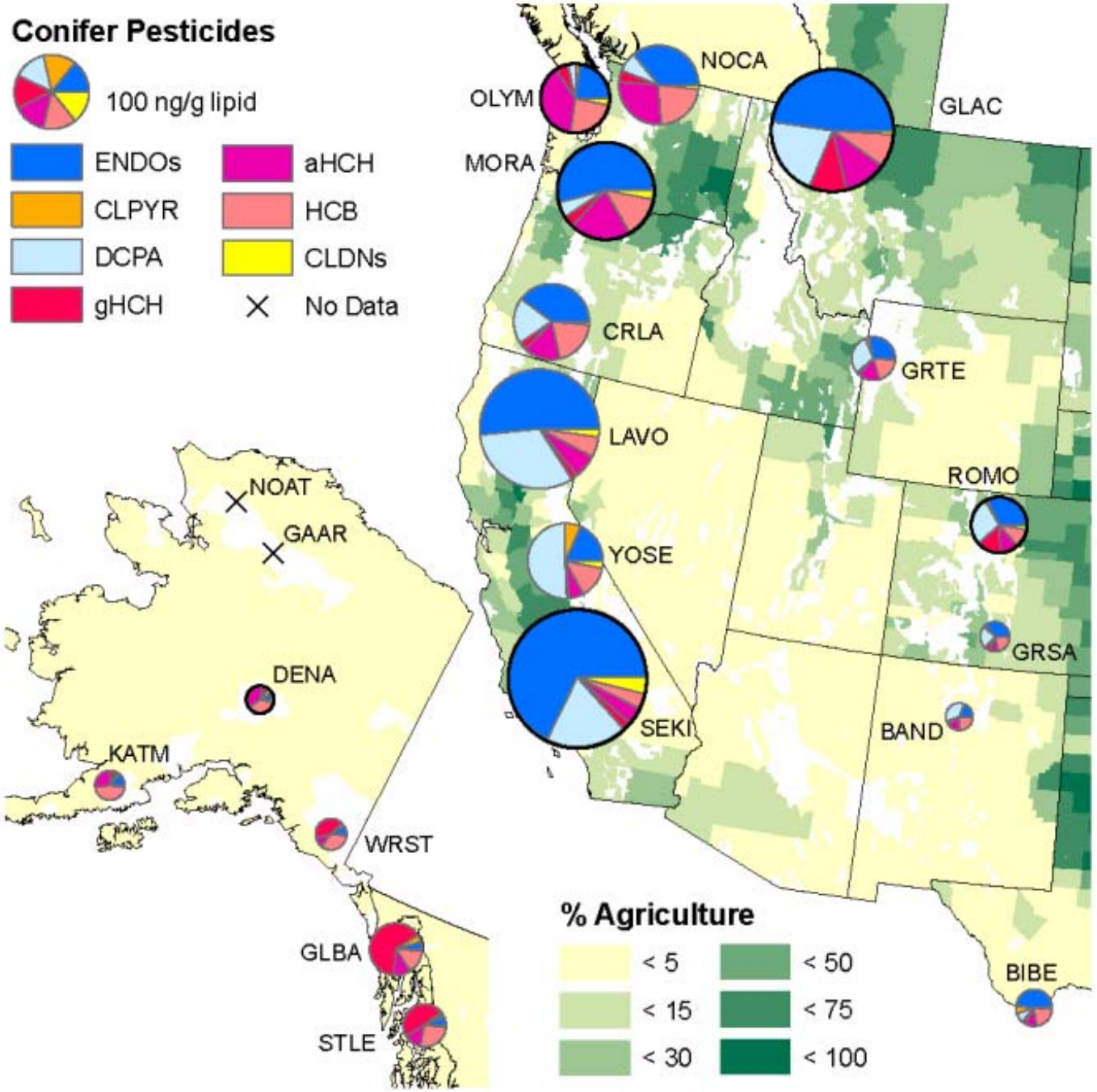
RESEARCH & DEVELOPMENT

Building a scientific foundation for sound environmental decisions

WACAP



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