Results of EPA’s Assessment of Fish Tissue from U.S. Rivers for Mercury and Persistent Organic Compounds with Implications for Aquatic Life and Human Health

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Fish Tissue Indicator
Mercury data- (n=541/542)
sites: 162 Urban sites
379 Non-urban

Legacy organic contaminant data-
(n=540/542):
163 Urban sites
377 Non-urban
National Rivers and Streams Assessment

• Out of 1,924 sites on rivers within the conterminous United States- 542 sampled that are 5\textsuperscript{th} order or greater in size

• Randomized site selection process yields nationally-representative weighted results

• Analyses are of sites where fish samples were collected and fillets analyzed (sampled population) which represent approximately 51,663 river miles.
National Rivers and Streams Assessment
Urban and Non-urban Sampling Locations by NARS Major Ecoregion

NRSA Sampling Locations $n = 542$

U.S. Environmental Protection Agency
Sample Collection

• Sampling conducted 2008-2009
• A single composite sample consisting of five adult fish of the same species and similar size (min>75% max) of fish was collected from each site.
• Target species: ubiquitous, abundant, easily identified, consumed by humans, large.
• Fillets were composited using the batch method.
## NRSA Analyses

### 541 sites: Mercury

**Direct Mercury Analyzer EPA method 7473**

(Journal article in preparation)

### Organic Analytes for 540 Sites

<table>
<thead>
<tr>
<th>21 PCB Congeners</th>
<th>20 Organochlorine pesticides</th>
<th>4,4'-DDT</th>
</tr>
</thead>
<tbody>
<tr>
<td>PCB 8-209</td>
<td>Aldrin</td>
<td>Dieldrin</td>
</tr>
<tr>
<td>8 PBDE Congeners:</td>
<td>alpha-BHC</td>
<td>Endosulfan II</td>
</tr>
<tr>
<td>BDE 47, 66, 99,100, 138, 153, 154, 183</td>
<td>gamma-BHC</td>
<td>Endrin</td>
</tr>
<tr>
<td>Method: GC-ECD</td>
<td>alpha-Chlordane</td>
<td>Heptachlor</td>
</tr>
<tr>
<td></td>
<td>gamma-Chlordane</td>
<td>Heptachlor epoxide</td>
</tr>
<tr>
<td></td>
<td>2,4'-DDD</td>
<td>Hexachlorobenzene</td>
</tr>
<tr>
<td></td>
<td>4,4'-DDD</td>
<td>Mirex</td>
</tr>
<tr>
<td></td>
<td>4,4'-DDE</td>
<td>cis-Nonachlor</td>
</tr>
<tr>
<td></td>
<td>2,4'-DDT</td>
<td>trans-Nonachlor</td>
</tr>
</tbody>
</table>

(Journal article in preparation)

8 PCB Congeners: 2,4'-DDD, 4,4'-DDD, 4,4'-DDE, 2,4'-DDT
Additional Fish Tissue Analytes not reported here

541 Sites:

Selenium: ICP-OES

Moisture: Karl Fisher titration

Lipids: Gravimetric method

163 urban sites:

13 PFCs (HPLC-MS/MS)

Poster here now

(Journal article in preparation)

4 synthetic musks and two of their metabolites

(Presented SETAC 2011)
Data are Nationally Representative

- Reporting today on 50 analytes x 540 sites = 27,000 data points
- Weighted data and means are nationally-representative and representative by:
  - Ecoregion (3); and
  - Urban and Non-urban sub-populations
- Mercury fillet tissue results converted to whole fish values for wildlife impact estimation
- Some unweighted site data depictions and analyses are not nationally representative
# Hg Statistics

<table>
<thead>
<tr>
<th>Statistic * (Hg ww ug/kg)</th>
<th>National n =541</th>
<th>Non-Urban n =379</th>
<th>Urban n=162</th>
<th>EHIGH n =190</th>
<th>PLNLOW n =280</th>
<th>WMTS n =71</th>
</tr>
</thead>
<tbody>
<tr>
<td>River Miles</td>
<td>51,663</td>
<td>40,752</td>
<td>10,911</td>
<td>14,738</td>
<td>29,739</td>
<td>7,186</td>
</tr>
<tr>
<td>River km</td>
<td>83,145</td>
<td>40,752</td>
<td>17,559</td>
<td>23,718</td>
<td>47,861</td>
<td>11,564</td>
</tr>
<tr>
<td>50th % ile*</td>
<td>175.6</td>
<td>170.5</td>
<td>200.6</td>
<td>176.0</td>
<td>180.1</td>
<td>125.3</td>
</tr>
<tr>
<td>95th % ile*</td>
<td>583.6</td>
<td>578.8</td>
<td>803.3</td>
<td>535.2</td>
<td>578.8</td>
<td><strong>854.1</strong></td>
</tr>
<tr>
<td>Mean*</td>
<td>228.9</td>
<td>223.3</td>
<td>250.2</td>
<td>210.0</td>
<td>231.4</td>
<td><strong>257.5</strong></td>
</tr>
<tr>
<td>Max. *</td>
<td>1,419</td>
<td>1,419</td>
<td>854</td>
<td>854</td>
<td>1,419</td>
<td>1,272</td>
</tr>
</tbody>
</table>
Hg Percentile Data by subgroup

- 120 ug/kg 1 meal/week threshold (Mink WV ~100 ug/kg)
- 300 ug/kg HH WQC, Eagle WV

 ug/kg (ppb)
(A.R. Olsen)
Hg subpopulations

Percentages of Samples in Subgroups <300 ug/kg and >300 ug/kg Hg
Standard error bars indicated

- National: 75% (>300), 25% (<300)
- EHIGH: 77% (>300), 23% (<300)
- PLNLOW: 78% (>300), 22% (<300)
- WMTS: 79% (>300), 21% (<300)
- Urban: 80% (>300), 20% (<300)
- NonUrban: 81% (>300), 19% (<300)
Mercury (Hg) Results

- All (100%) of the 541 fish fillet samples analyzed for Hg content >3.33 ug/kg (ppb) quantitation limit for the method.
- Weighted values for 137/541 samples (25.4%) exceeded the EPA HHWQC for Hg of 300 ug/kg, = 13,071 river miles (21,154 km)/ 51,663 miles (83,143 km) of sampled U.S. rivers (compare with 48.9% of lakes in NFLTS).
- Risk to piscivorous avian species (eagle) are similar to risks for humans at 300 ug/kg (@two meals per month level), whereas mink are more at risk than the avian species (and humans) @ the HH 1 meal per week (fillet) threshold of 120 ug/kg.
- No statistically-significant differences between non-urban/urban sites and among eco regions (EHIGH, PLNLOW, WMTS).
- Apparent higher 95th %ile, mean, and % of sites exceeding HHWQC in WMTS likely attributable to natural localized Hg source (geothermal area/Hg mining history).
Focus of organics analysis is on PCBs, PBDEs, Chlordane, and DDT, Dieldrin

PCBs

National Data
- Detected 505/540 sites = 93.5%
- National Mean = 32.7 ug/kg
- %>Screening Value (12ug/kg) = 48.0%

Non-Urban Sites (n=377)
- Detects Non-urban = 343
- Max Non-urban = 411.5 ug/kg=ppb
- Mean Non-urban = 26.9 ug/kg
- %>Screening value= 42.0%

Urban Sites (n=163)
- Detects Urban = 162
- Max Urban = 856.5 ug/kg
- Mean Urban = 54.2 ug/kg
- %>Screening value= 69.8%
Summed PCBs (21 of 209 congeners) in Fish Tissue Samples

All summed PCB fish tissue concentrations with Consumption frequency thresholds

(Cancer Endpoint Values 1/100,000)

Plotted values are unweighted site data.

Wildlife Value (WV) Mink (130 ug/kg)
No Consumption Level (94 ug/kg)
One meal/month Level (47 ug/kg)
One meal/week Level (12 ug/kg)

Note: Unweighted site data
Weighted Mean PCB Congener Concentrations in Fish Tissue from U.S. River Sites - Non-Urban and Urban
PBDEs

National Data
• Detected 497/540 sites = 92.0%

• National Mean = 11.6 ug/kg

• 1 Urban Site exceeded 210 ug/kg SV

Non Urban Waters (n=377)
• Detects Non-urban = 340
• Max Non-urban = 151.1 ug/kg=ppb
• Mean Non-urban = 8.6 ug/kg

Urban Waters (n=163)
• Detects Urban = 157
• Max Urban = 310.7 ug/kg
• Mean Urban = 22.5 ug/kg
PBDEs—pervasive, lower concentrations, lower toxicity

(Plotted values are unweighted site data)

CA Advisory Level

Mink WV
Comparison of Weighted Mean PBDE Compound Concentrations in Fish Tissue from Non-Urban and Urban Sites in U.S. Rivers
Total Chlordane and Summed DDT(s)

**Total Chlordane**
Detected 481/540 locations = 88.5%
National Mean = 6.3 ug/kg

- **Non Urban Waters (n=377)**
  - Detects Non-urban = 325
  - Max Non-urban = 87.1 ug/kg
  - Mean Non-urban = 5.1 ug/kg

- **Urban Waters (n=163)**
  - Detects Urban = 153
  - Max Urban = 311.4 ug/kg
  - Mean Urban = 10.8 ug/kg

**Summed DDT**
Detected 533/540 locations = 98.7%
National Mean = 13.8 ug/kg

- **Non Urban Waters (n=377)**
  - Detects Non-urban = 370
  - Max Non-urban = 170.3 ug/kg
  - Mean Non-urban = 12.3 ug/kg

- **Urban Waters (n=163)**
  - Detects Urban = 163
  - Max Urban = 294.3 ug/kg
  - Mean Urban = 19.0 ug/kg
Percentage Detections of Other Organic Pesticide Compounds in Fish Tissue Samples from Non-Urban and Urban Sites

- Aldrin
- Dieldrin
- Endrin
- Lindane
- Heptachlor
- Heptchl Epox.
- Mirex

Non-urban
Urban
### Human health screening values (SVs)- 1 meal/week

<table>
<thead>
<tr>
<th>Compound*</th>
<th>Non-cancer SV</th>
<th>Cancer SV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chlordane (total)</td>
<td>1200</td>
<td>67</td>
</tr>
<tr>
<td>Dieldrin</td>
<td>120</td>
<td>1.5</td>
</tr>
<tr>
<td>DDT (summed)</td>
<td>120</td>
<td>69</td>
</tr>
<tr>
<td>PCBs (summed)</td>
<td>47</td>
<td>12</td>
</tr>
<tr>
<td>PBDEs (summed)</td>
<td>210@</td>
<td>NA</td>
</tr>
</tbody>
</table>

(*ug/kg wet weight)

- @California sport fish advisory level
## Wildlife risk values (WVs)

<table>
<thead>
<tr>
<th>Compound</th>
<th>Mink WV</th>
<th>Kingfisher WV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chlordane</td>
<td>830</td>
<td>4.5</td>
</tr>
<tr>
<td>Dieldrin</td>
<td>20</td>
<td>360</td>
</tr>
<tr>
<td>DDT (total)</td>
<td>360</td>
<td>155</td>
</tr>
<tr>
<td>PCBs (total)</td>
<td>130</td>
<td>440</td>
</tr>
<tr>
<td>PBDEs@</td>
<td>32</td>
<td>13 (Kestrel)</td>
</tr>
</tbody>
</table>

(*ug/kg wet weight)


@Canadian Environmental Protection Act, 1999 Federal Environmental Quality Guidelines Feb. 2013
Time for some statistics: Weighted Data: Fish Tissue Concs. in Non-urban vs. Urban Sites

<table>
<thead>
<tr>
<th>Compound</th>
<th>Family</th>
<th>Mean Conc.(ug/kg)</th>
<th>Median Conc.(ug/kg)</th>
<th>St. Dev.(ug/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PCBs</td>
<td></td>
<td>26.9</td>
<td>54.2</td>
<td>8.6</td>
</tr>
<tr>
<td>PBDEs</td>
<td></td>
<td>8.6</td>
<td>22.5</td>
<td>3.6</td>
</tr>
<tr>
<td>Total Chlordane</td>
<td></td>
<td>5.1</td>
<td>10.8</td>
<td>1.6</td>
</tr>
<tr>
<td>DDTs</td>
<td></td>
<td>12.3</td>
<td>19.0</td>
<td>5.7</td>
</tr>
<tr>
<td>Dieldrin</td>
<td></td>
<td>2.6</td>
<td>3.8</td>
<td>0.4</td>
</tr>
</tbody>
</table>

**Bold** = greater in non-urban vs urban comparison
NARS Major Ecoregions
### Significance of Differences Between Subgroups

<table>
<thead>
<tr>
<th>Compound Family</th>
<th>Non-urban/Urban</th>
<th>EHIGH/PLNLOW</th>
<th>EHIGH/WMTS</th>
<th>PLNLOW/WMTS</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Greater mean</td>
<td>P value</td>
<td>Greater mean</td>
<td>P value</td>
</tr>
<tr>
<td>PCBs Urb. EHIGH</td>
<td>Urban</td>
<td>0.006</td>
<td>EHIGH</td>
<td>0.032</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PBDEs No E-R diff.</td>
<td>Urban</td>
<td>&lt;0.001</td>
<td>EHIGH</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chlordanes less WMTS</td>
<td>Urban</td>
<td>0.107</td>
<td>PLN LOW</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DDTs Urb.PLNLOW</td>
<td>Urban</td>
<td>0.017</td>
<td>PLN LOW</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dieldrin less WMTS</td>
<td>Urban</td>
<td>0.36</td>
<td>PLN LOW</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Comparisons based on Z-Tests using calculated weighted means and standard errors.
Top Quartile by Summed PCB Conc. of PCBs, PBDEs, Chlordanes, & DDTs in Fish Tissue from U.S. Rivers 5th order and Greater

(Site data depicted for illustrative purposes)
Co-occurrence of Total PCBs, PBDEs, Chlordanes, and DDTs in Fish Tissue at Concs. above Median Values

Synthesis 1- National Organics Data

- Dieldrin, chlordane, DDT, PCBs and PBDEs are pervasive in fish tissue samples collected from U.S. rivers-detects in fish tissue average 88.8% of river miles.

- The extent to which these compounds exceed risk-based screening values varies (weighted national data):
  - **PCBs 48% of sites**
  - PBDEs 0.26% (1 Site)
  - Summed DDTs 2.3% of sites
  - Total Chlordane 0.56% of sites
  - **Dieldrin 31.2% of sites**

- Extent of effect depends on conc. and SV or WV
Organics Synthesis 2

- Unlike Hg concentrations, PCBs, PBDEs, and DDT compounds occur at concentrations that are significantly higher in fish from urban sites nationally than from non-urban sites.

- PCBs concs. are highest in EHIGH, significantly higher in the PLNLOW and EHIGH ecoregions than in the WMTS.

- There is no significant difference in PBDE concentrations in fish tissue among eco-regions*

- DDT concentrations are significantly elevated in PLNLOW relative to EHIGH, but not to WMTS(CA ag?)

- Chlordane concentrations in fish tissue are significantly higher in samples from the PLNLOW and EHIGH than WMTS. No difference between non-urban and urban sites.
  - *(Previous analysis of POTWs in urban locations –SETAC 2012)
Conclusions

• Monitoring of fish tissue for assessment and for fish consumption advisories continues to be important for organic compounds as well as Hg
• Individual compounds seldom occur alone in fish tissue. Therefore, the presence and effects of any of these or other contaminants must be viewed in the context of co-occurring compounds
• Any new persistent organo-halogen compounds add to the existing overall organo-halogen burden in fish tissue potentially consumed by humans and wildlife.
Acknowledgements

- This assessment was the product of the combined efforts of many:
  - Sampling was conducted by state, federal agency, and contractor crews
  - The NRSA is operated by our colleagues in the EPA Office of Wetlands, Oceans, and Watersheds, and
  - The EPA Office of Research and Development, Western Ecology Division, are responsible for the sample design and for derivation of nationally-representative descriptive statistics
  - Mission support from Tetra Tech, CSC, and other contractors
Thanks for your interest