Willamette Mercury TMDL

Technical Approach

Willamette Mercury TMDL Advisory Committee Meeting
August 22, 2018

DEQ

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

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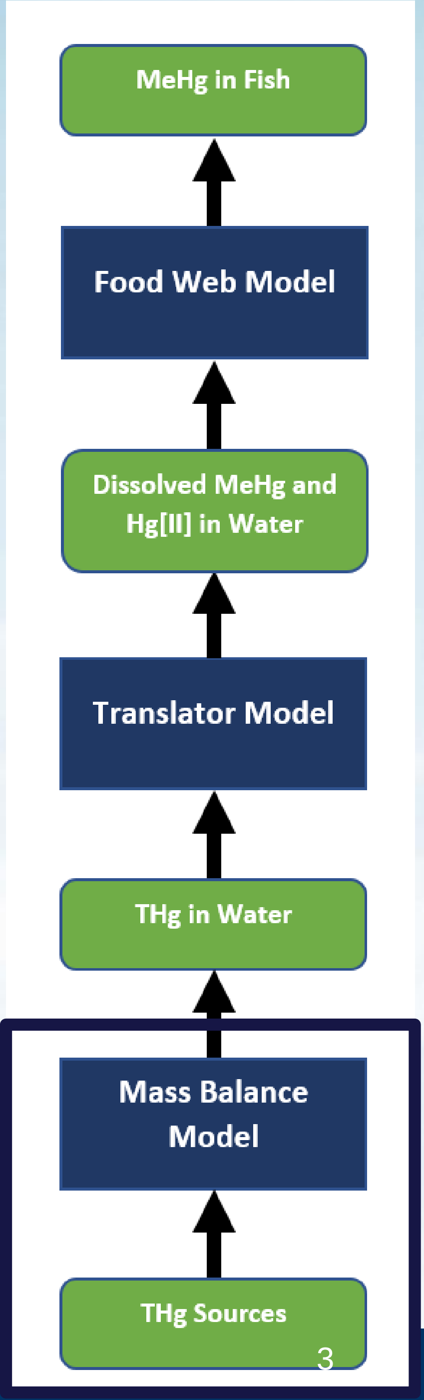
OVERVIEW OF TECHNICAL APPROACH

The 2006 TMDL Linkage Analysis

- ▶ Link sources of total mercury (THg) to methylmercury (MeHg) in fish
- ▶ Three components:

  1. **Mass Balance Model**: Link THg sources in the watershed to instream concentrations
  2. **Mercury Translator**: Link THg concentrations to MeHg and Hg[II] exposure concentrations
  3. **Food Web Model**: Link exposure concentrations of MeHg to fish tissue



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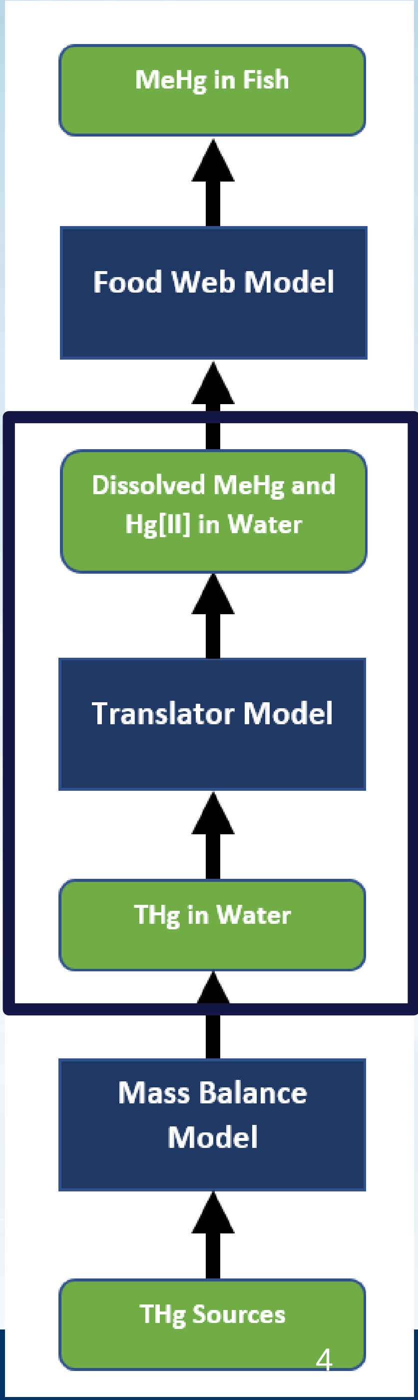

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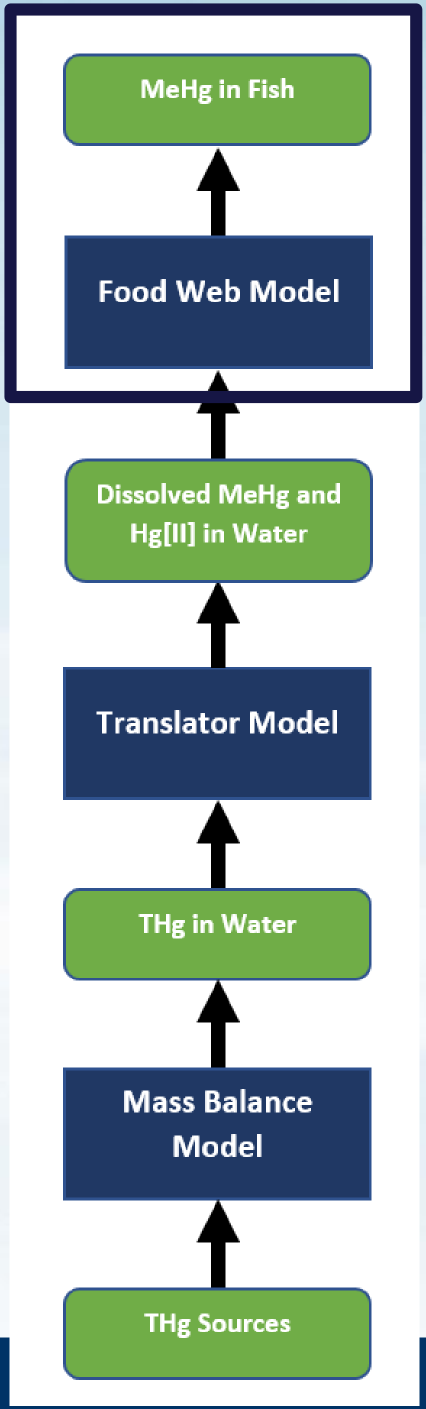

Required Reductions

- Percent reductions needed are calculated from:
  - Current water column THg concentrations (from monitoring data)
  - Needed water column THg concentrations to meet fish tissue standard
- Therefore, needed reductions do not depend on Mass Balance Model

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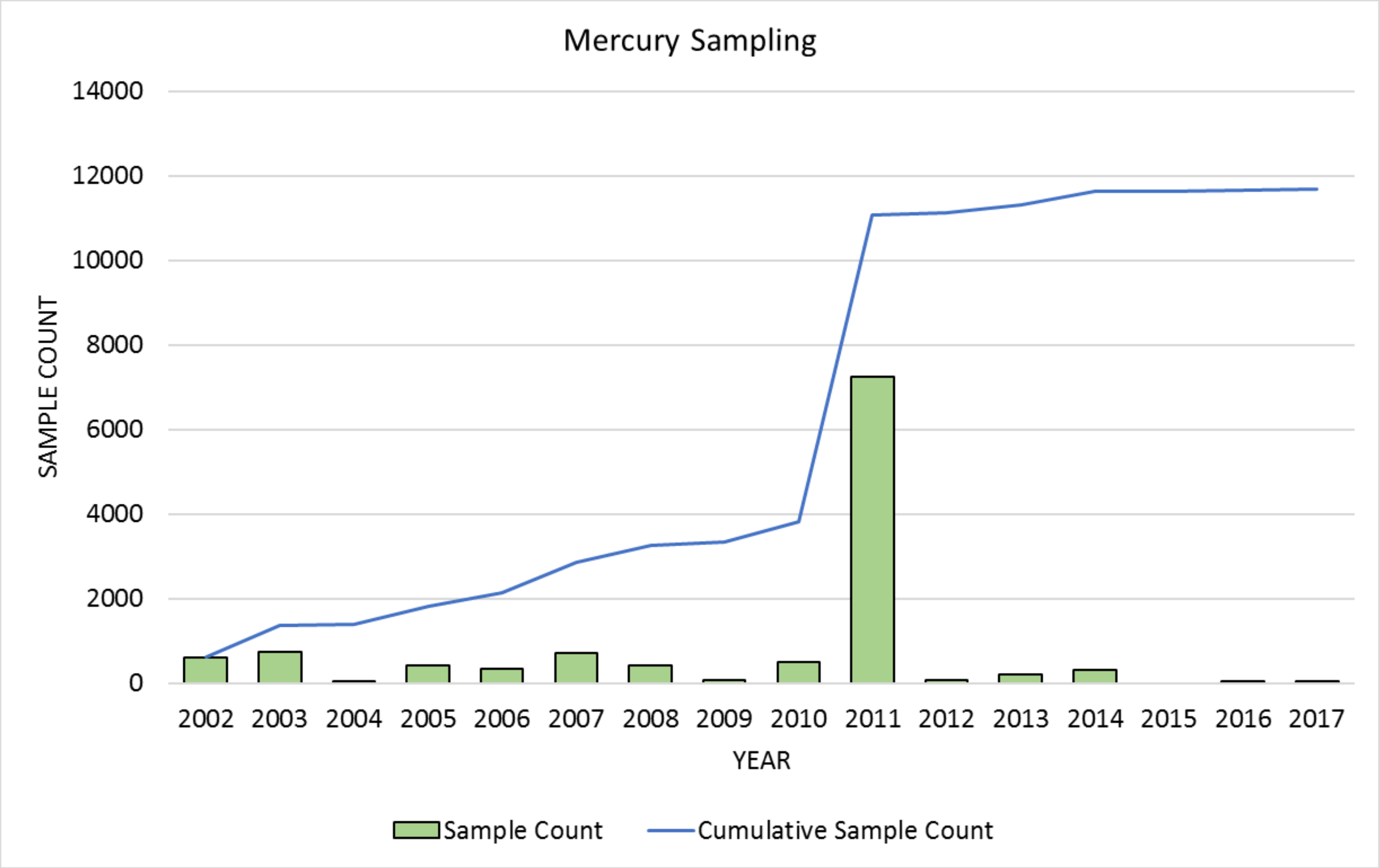
DATA SUMMARY

Data

- 2006 TMDL relied in large part on one year of MeHg sampling in 2002-2003
- Additional monitoring data has been collected since 2006 TMDL
- Watershed occupies 11,500 mi^2, so data availability varies spatially
- Even though mercury cycling is complex there is enough data available to support the TMDL

Temporal Distribution

- Monitoring studies have collected fish tissue, sediment and water column mercury samples – lots of data collected since the 2006 TMDL
- Analytical sampling methods have improved over time so use data from 2002-2017

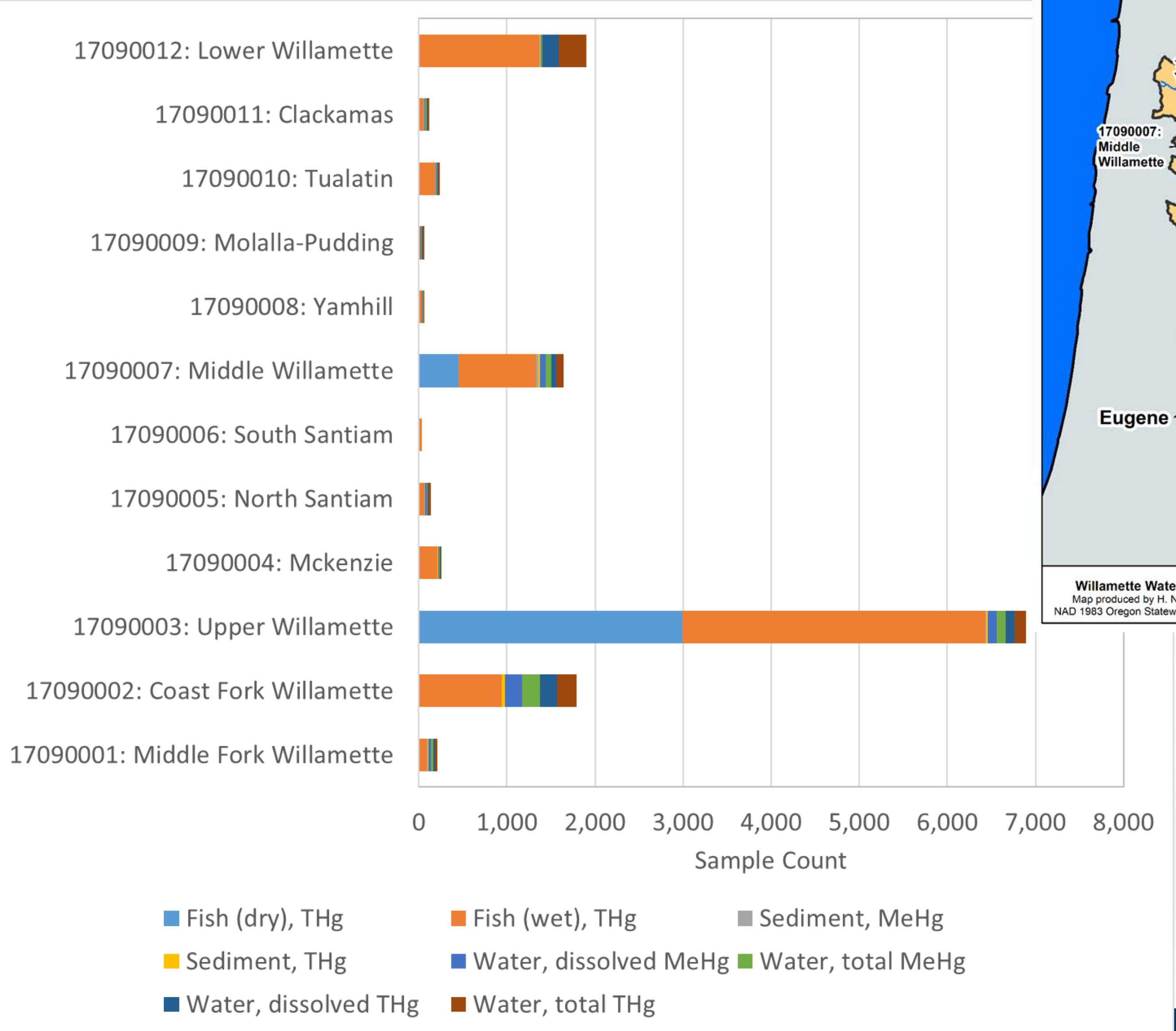


Mercury Data for TMDL Update

- Data collected by multiple agencies and studies were compiled for the update

Origin	Data Provider	Sampling Medium	Sample Dates
2006 TMDL Fish Data	ODEQ	Fish tissue	7/8/2003 – 9/2/2003
2008 Fish Sample Records from the DEQ Laboratory	ODEQ	Fish tissue	8/20/2008 – 10/28/2008
ARRA Willamette Mercury Monitoring Project	ODEQ	Water column, fish tissue, and sediment	8/23/2010 – 9/2/2010
Black Butte Mine Storm Sampling	EPA	Water column	1/7/2013 – 1/19/2017
Cottage Grove Analytical Reports	ODEQ	Fish tissue	6/2/2005 – 8/8/2005
Cottage Grove Reservoir Monitoring	EPA	Water column	3/8/2013 – 11/24/2014
DEQ Laboratory LASAR Database (Compilation of multiple sampling organizations)	ODEQ	Water column, fish tissue, and sediment	8/14/2002 – 3/30/2009
DEQ Toxics Monitoring Program	ODEQ	Fish tissue	8/20/2008 – 10/1/2010
EPA Mercury Database (Contains data from multiple states, agencies and studies compiled by Helen Rueda)	EPA	Fish tissue	7/8/1969 – 12/7/2010
NLA Lake Fish Tissue Mercury Data	EPA	Fish tissue	4/16/2014 – 10/17/2014
Portland Harbor Superfund Mercury Data	EPA	Water column and fish tissue	6/25/2002 – 9/5/2008
Smallmouth Bass Tissue Study	EPA	Fish tissue	8/27/2012 – 9/25/2012
USGS Mercury Data for Cottage Grove Lake and Coast Fork Willamette	EPA	Water column and sediment	7/13/1992 – 9/30/2014
USGS Willamette River Mercury Sampling	USGS	Fish tissue and water column	7/8/2011 – 8/26/2011

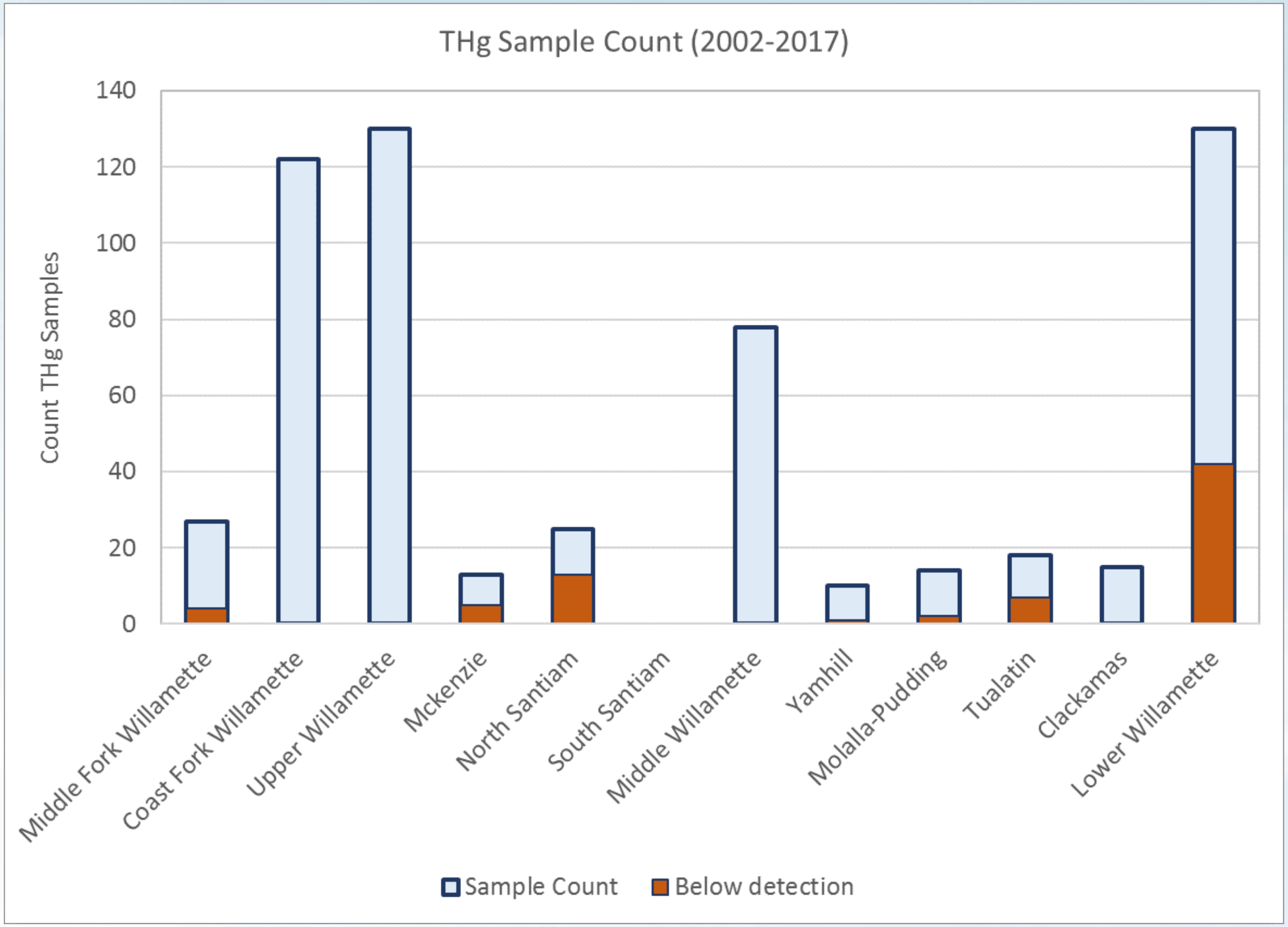
Data Availability by HUC8



- Samples are mostly from mainstem HUC8s and the Coast Fork Willamette HUC
- Mostly fish tissue samples

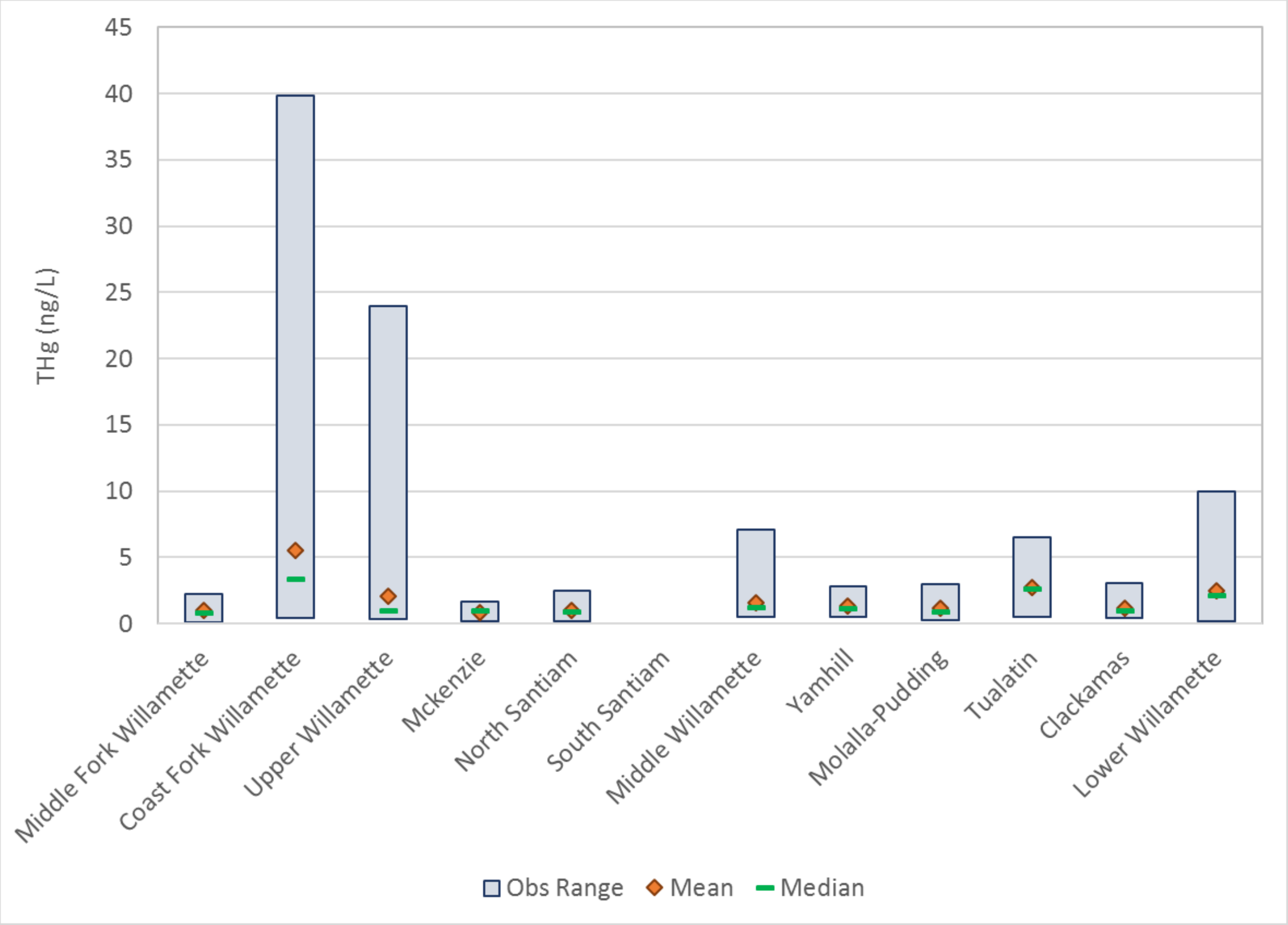
Water Column THg Data Availability

- 13% of samples are below the detection or reporting limit (i.e., censored)
- Using censored data directly would misrepresent dataset statistics
- Robust **R**egression on **O**rder **S**tatistics assumes the underlying distribution of the censored data is lognormal, and fits a regression
- Detected samples combined with estimated censored samples to calculate summary statistics



Water Column THg Concentrations

- Summary of water column THg with censored data corrected using ROS
- No data available for South Santiam
- 2006 THg Target
  - 0.92 ng/L
- Mean THg:
  - Lowest: McKenzie (0.81 ng/L; n=13)
  - Highest: Coast Fork (5.5 ng/L; n=122)
- Maximum THg:
  - Lowest: McKenzie (1.7 ng/L)
  - Highest: Coast Fork (40 ng/L)



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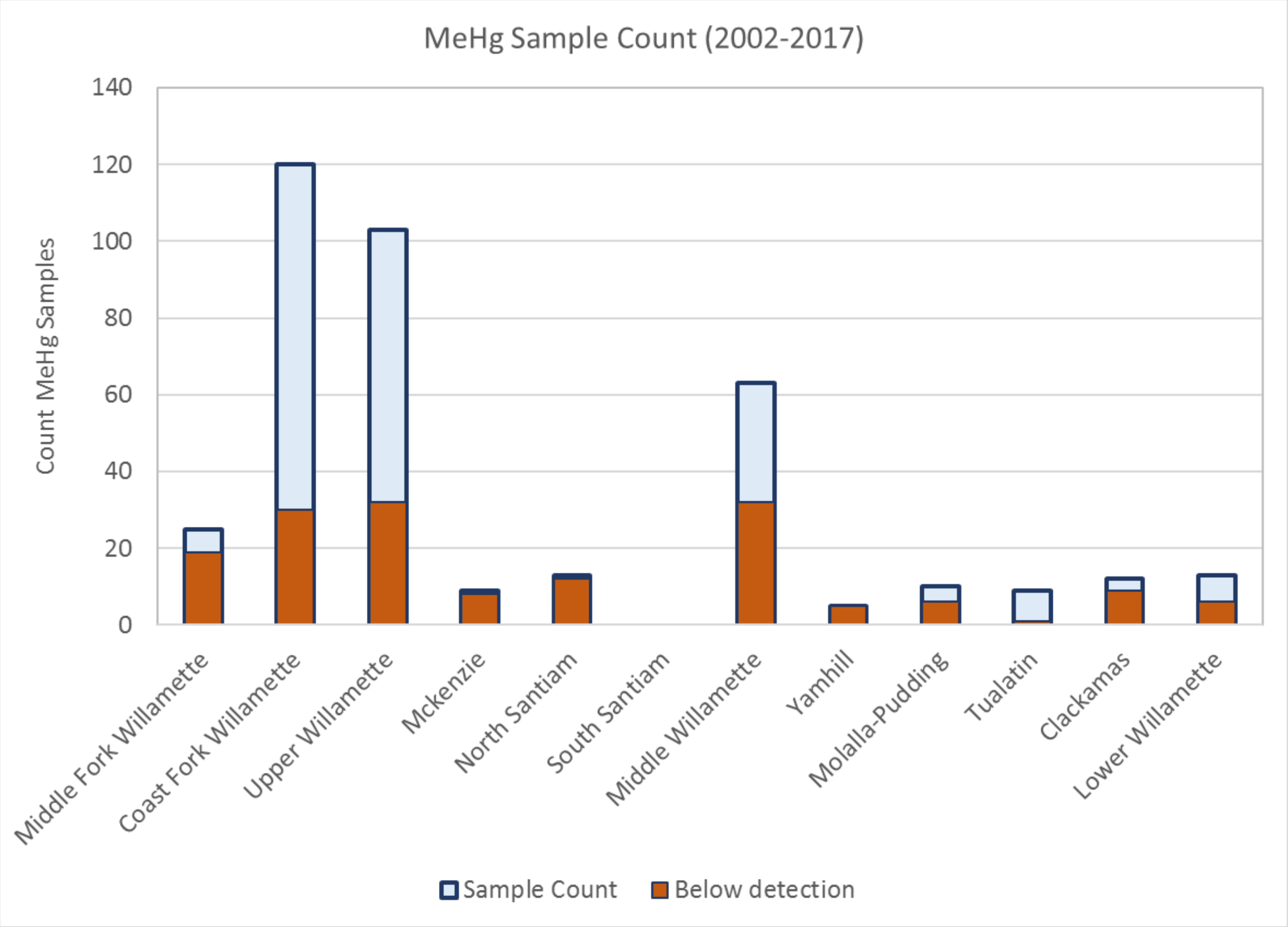
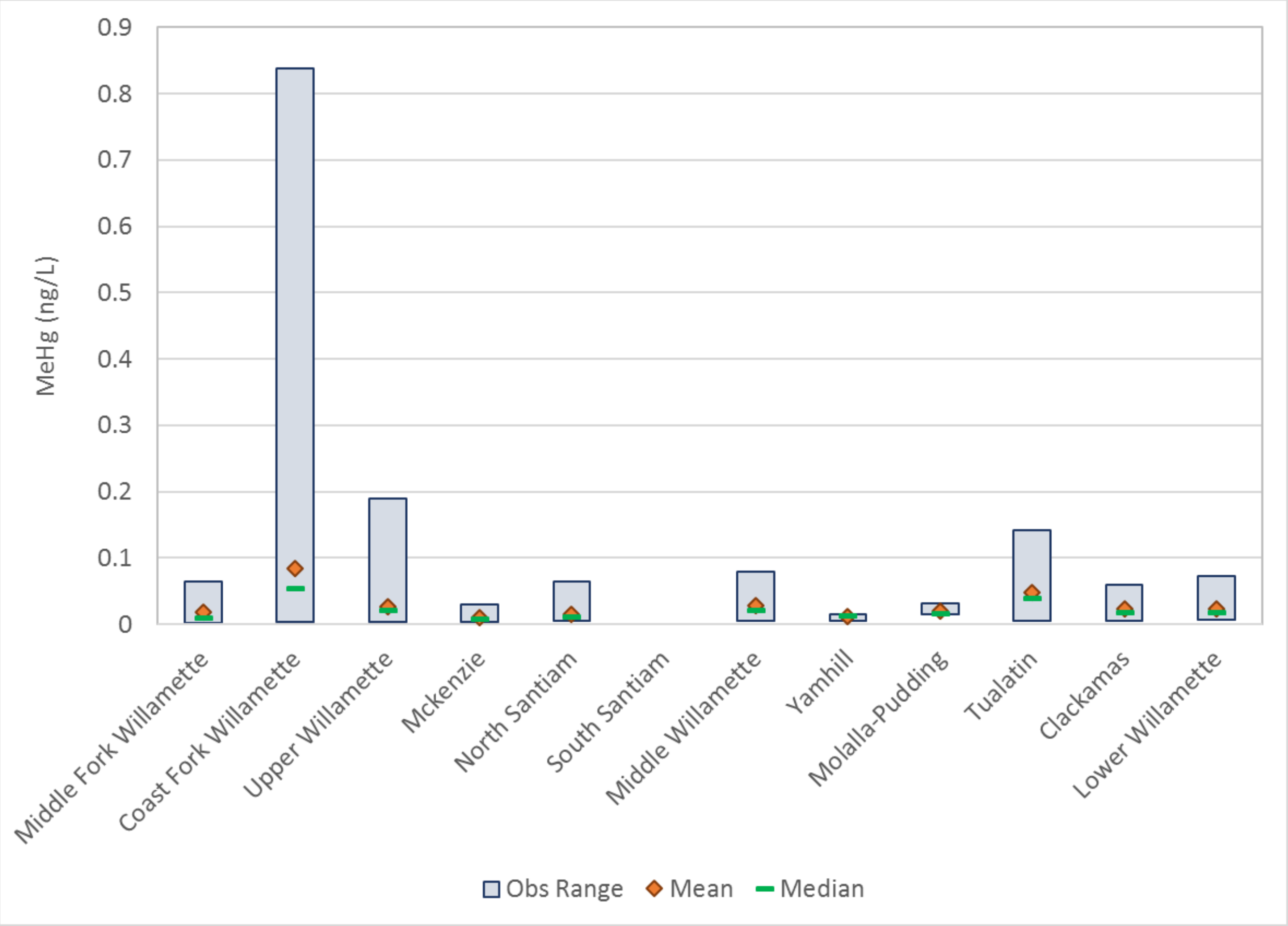
Water Column MeHg Data Availability

- More MeHg samples are censored (42%) compared to THg (13%)
- Detected samples combined with ROS estimated censored samples to calculate summary statistics



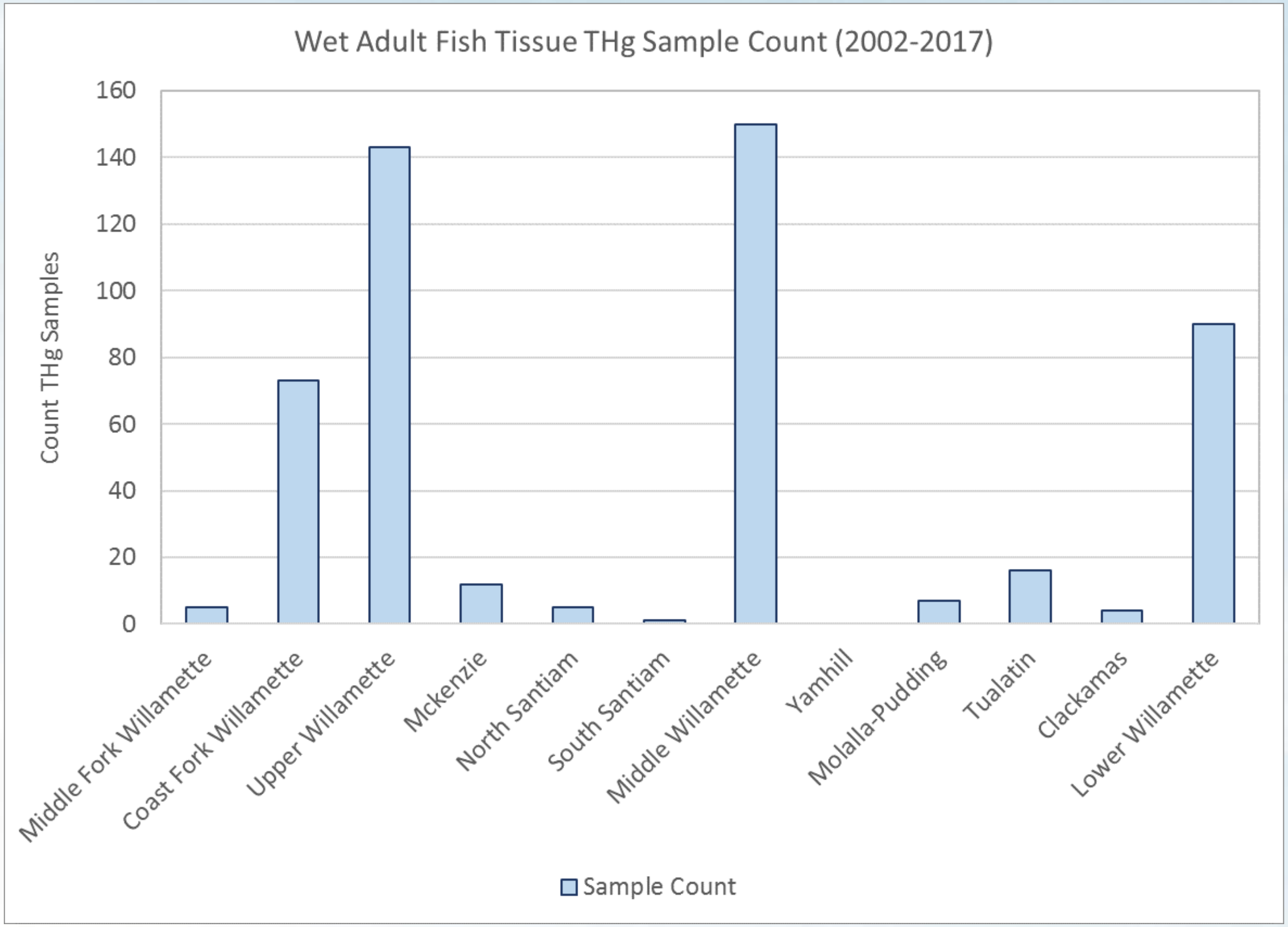
Water Column MeHg Concentrations

- Summary of water column MeHg with censored data corrected using ROS
- No data available for South Santiam
- Mean THg:
  - Lowest: McKenzie (0.01 ng/L; n=9)
  - Highest: Coast Fork (0.08 ng/L; n=120)
- Maximum THg:
  - Lowest: Yamhill (0.01 ng/L; n=5)
  - Highest: Coast Fork (0.84 ng/L)



Wet Adult Fish Tissue THg Data Availability

- Fish tissue samples primarily from mainstem HUCs
- No fish tissue samples available for Yamhill, and data are very limited in non-mainstem HUCs
- None of the fish tissue data are censored



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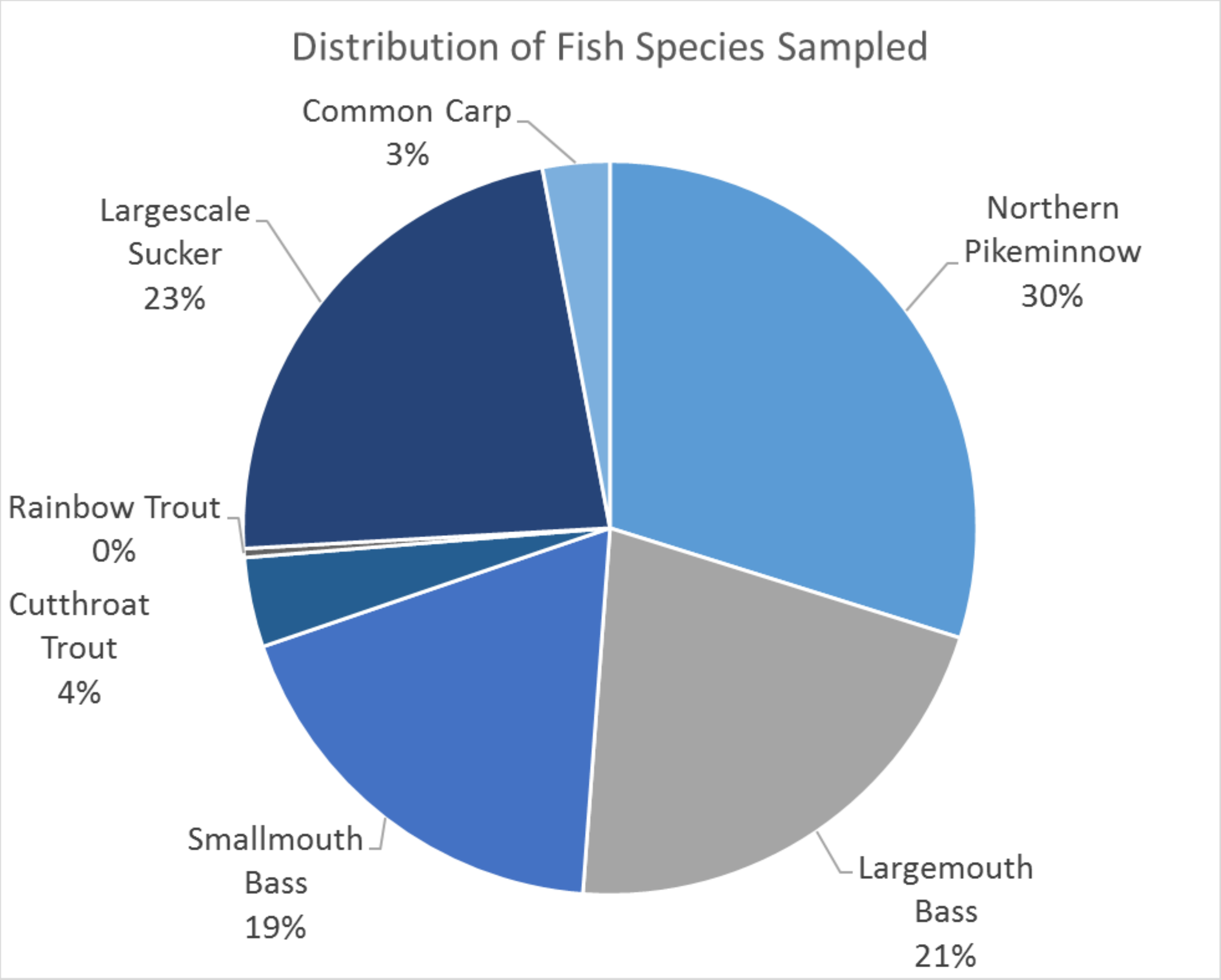
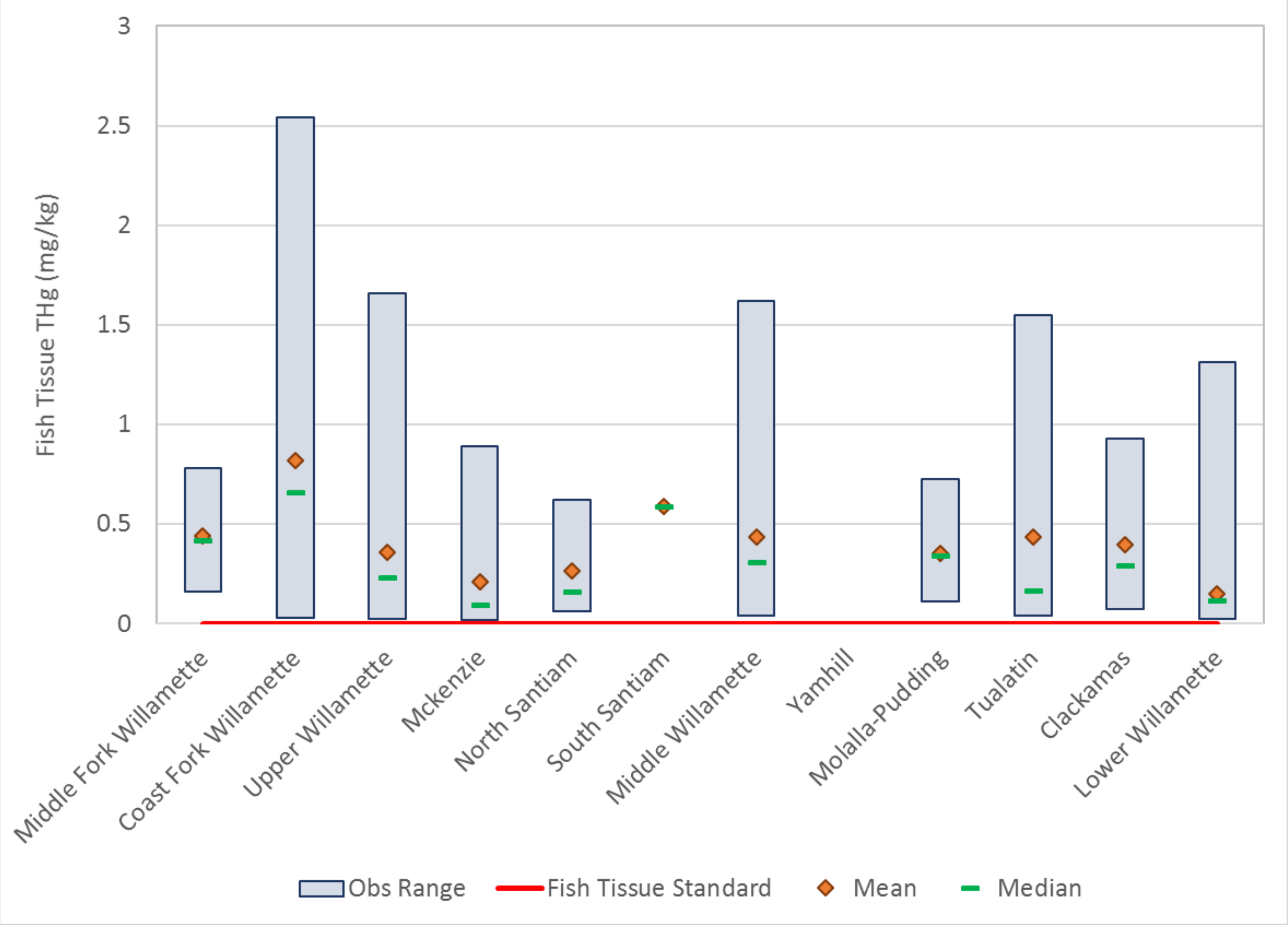
Wet Adult Fish Tissue THg Data Availability (continued)

- Most fish tissue samples are from Northern Pikeminnow, Largescale Sucker, Largemouth Bass, and Smallmouth Bass
- Fewer samples available for Common Carp, Cutthroat Trout, and Rainbow Trout

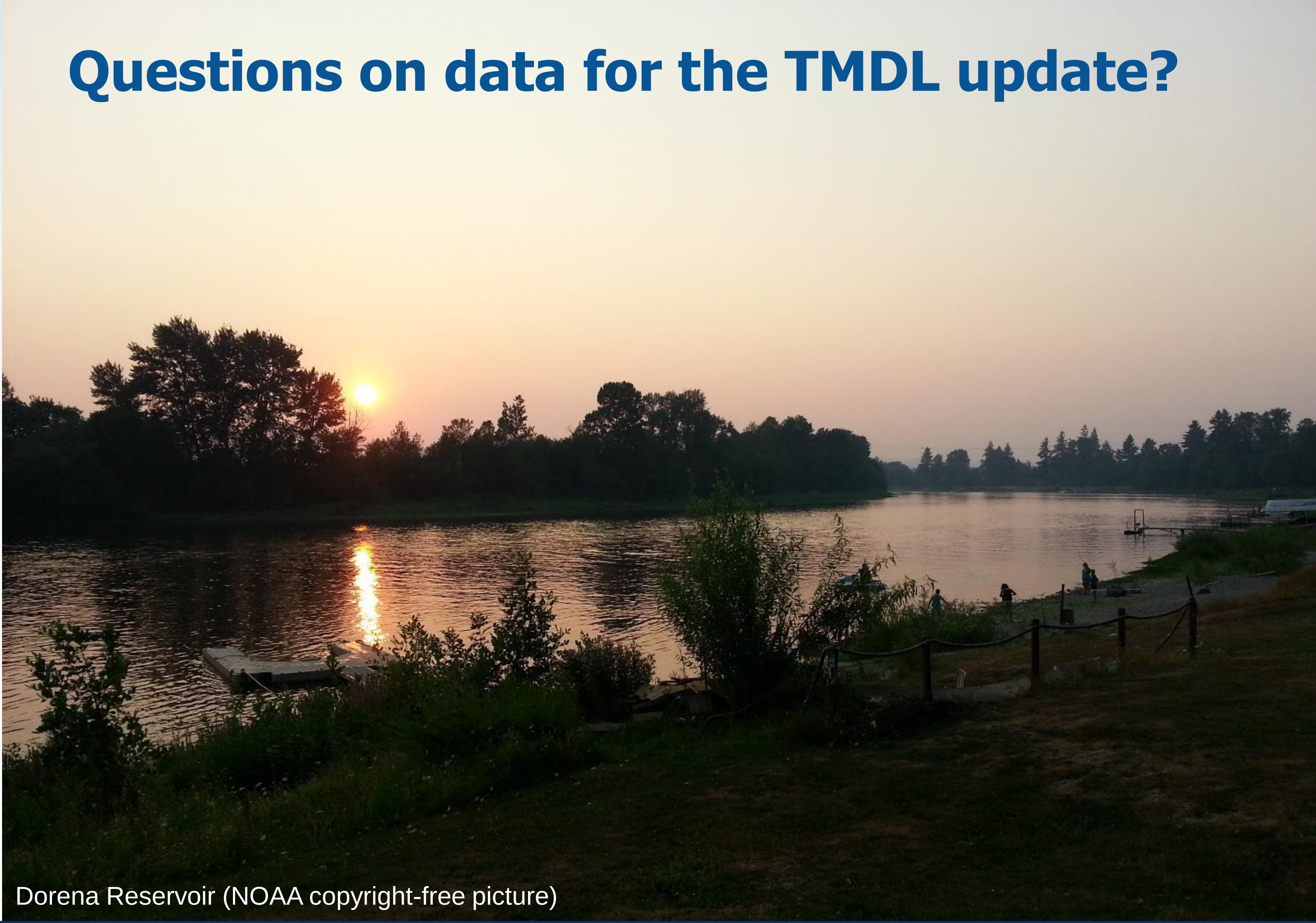


Fish Tissue THg Concentrations

- Fish tissue concentrations presented in units of mg-THg per kg-fish tissue
- Most mercury in fish is MeHg
- New fish tissue standard concentration
  - 0.04 mg/kg MeHg
  - Shown by red line
- Few samples collected meet new standard
- Mean THg:
  - Lowest: Lower Willamette (0.15 mg/kg; n=90)
  - Highest: Coast Fork (0.82 mg/kg; n=73)



Questions on data for the TMDL update?



Dorena Reservoir (NOAA copyright-free picture)

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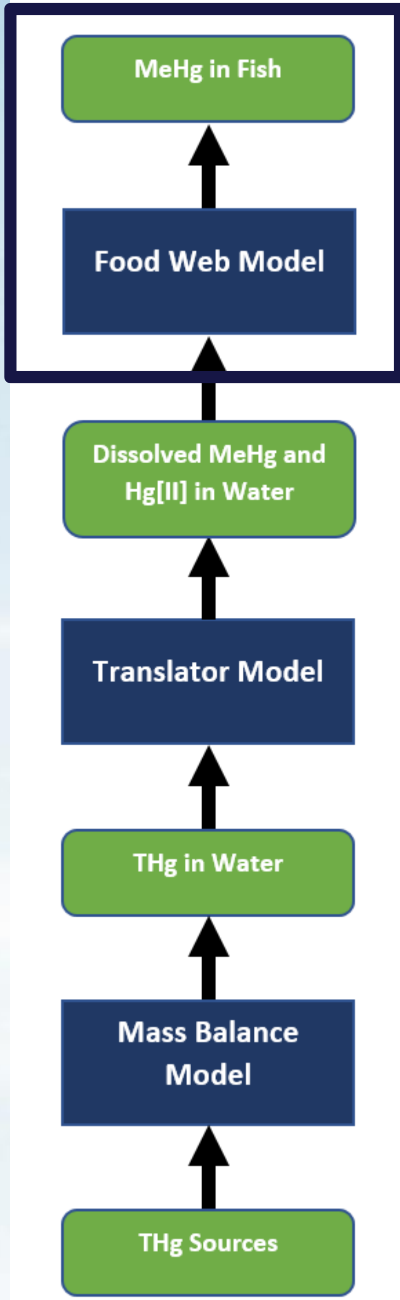
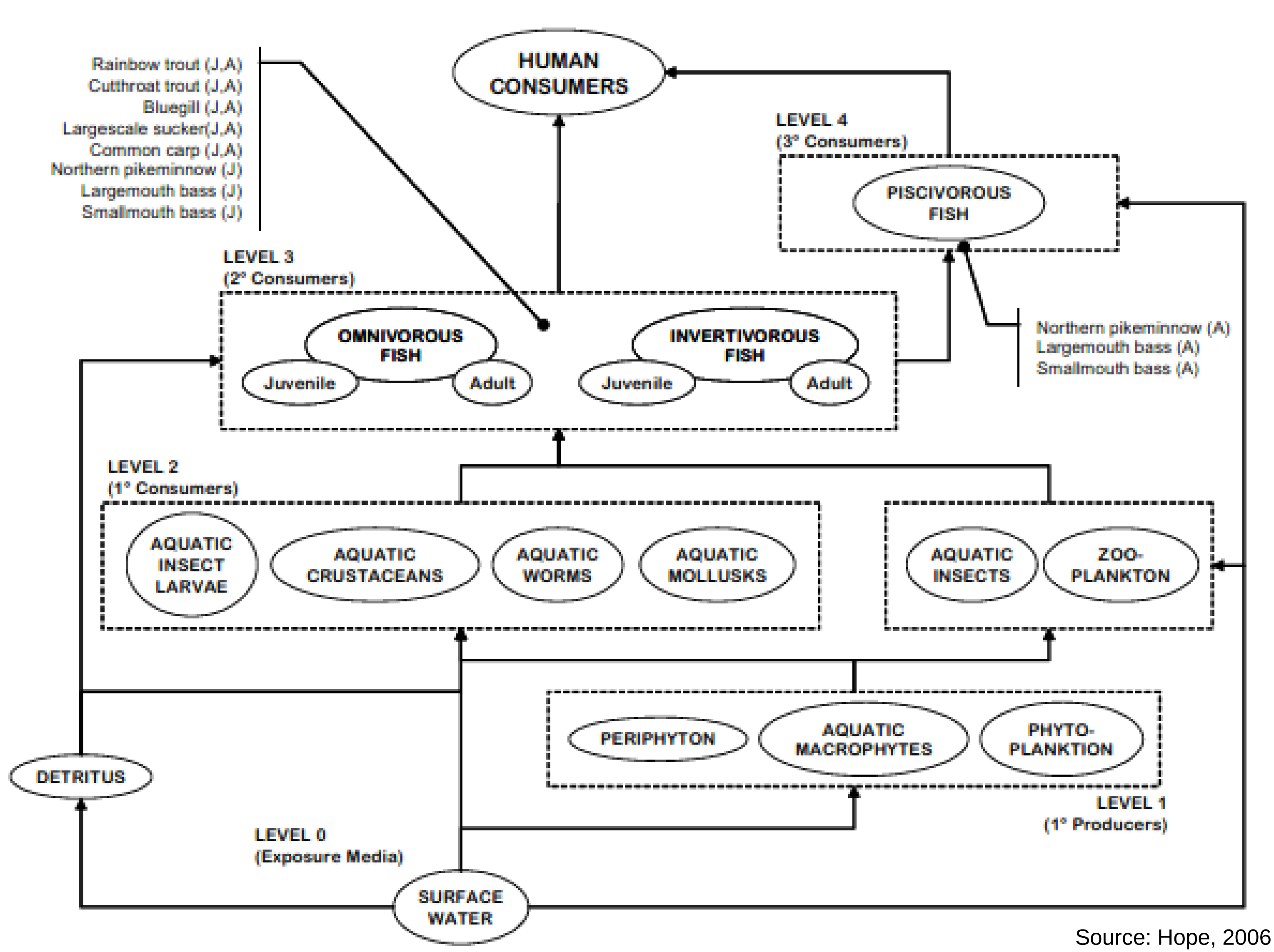
FOOD WEB MODEL



Purpose of the Food Web Model (FWM)

- Oregon fish tissue criterion: 0.04 mg/kg
- What are the water column THg exposure concentrations (MeHg and Hg[II]) needed to meet the fish tissue criterion?
- Preferable to use local data
- May vary for different species of interest
- Calibrated FWM simulates bioaccumulation
- Can use FWM to determine biomagnification factors

Feeding Relationships in the FWM



Source: Hope, 2006

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Who Eats What…

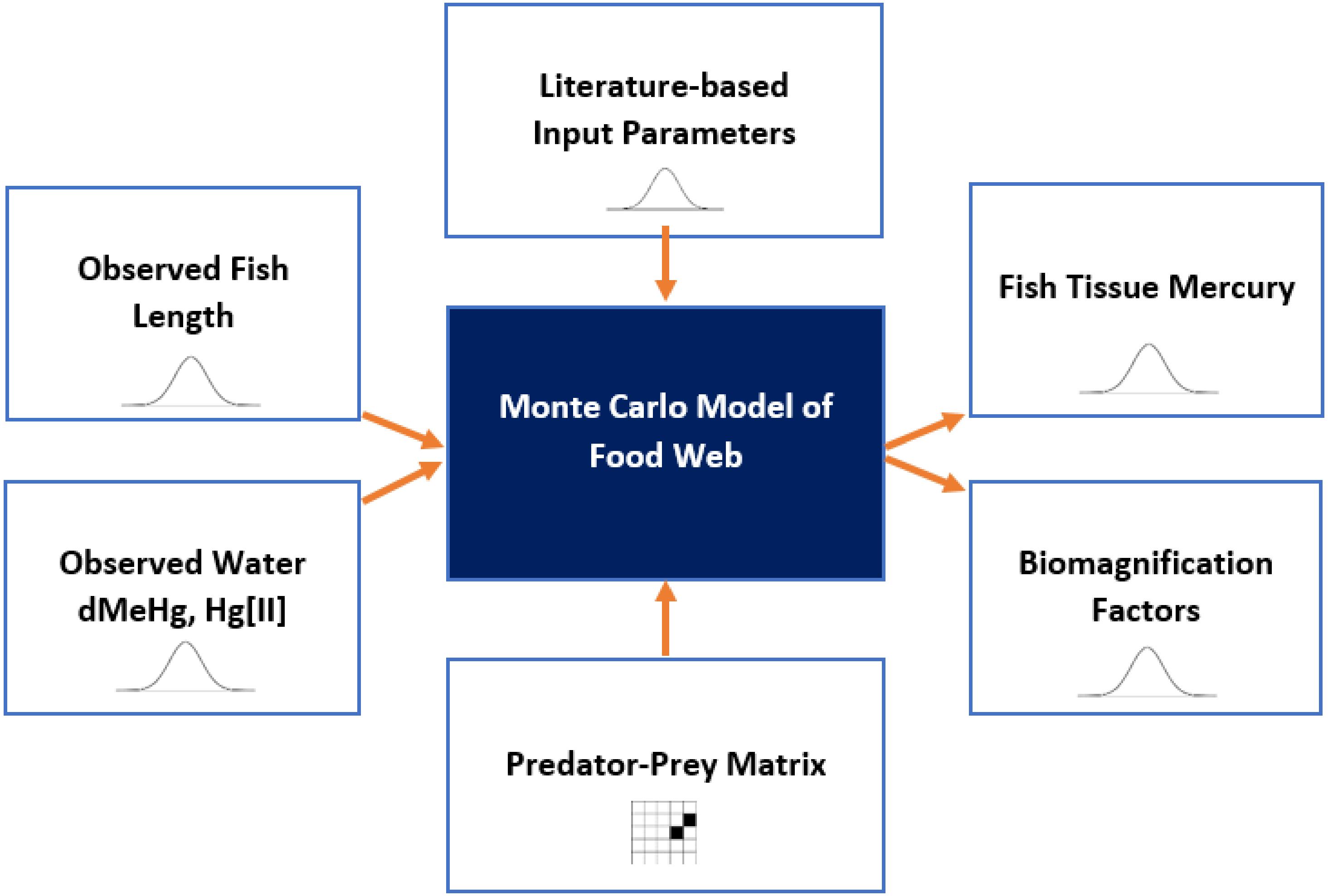
Table 5. Matrix of predator-prey interactions included in the model.

pred →												NPM		LMB		SMB		LSS		CAR		RBT		CTT		BLU	
prey ↓		DET	AQP	PHY	PER	ZOO	AQL	AQC	AQI	AQM	AQW	J	A	J	A	J	A	J	A	J	A	J	A	J	A	J	A
DET						●	●	●	●		●							●	●		●					●	●
AQP							●	●	●									●	●		●					●	●
PHY						●				●								●	●	●	●				●	●	●
PER							●	●			●							●	●	●	●					●	●
ZOO							●	●	●	●		●		●		●		●	●	●	●	●		●	●	●	●
AQL									●			●	●	●	●	●	●	●	●	●	●	●	●	●	●	●	●
AQC													●	●	●		●		●		●		●			●	●
AQI												●	●	●	●	●	●		●		●	●	●	●	●	●	●
AQM									●										●		●					●	●
AQW														●		●			●		●		●		●	●	●
NPM	J												●	●	●	●	●						●		●		●
	A														●		●										
LMB	J											●	●		●	●	●						●		●		●
	A												●				●										
SMB	J											●	●	●	●		●						●		●		●
	A												●		●												
LSS	J											●	●	●	●	●	●						●		●		●
	A												●		●		●										
CAR	J											●	●	●	●	●	●						●		●		●
	A												●		●		●										
RBT	J											●	●	●	●	●	●						●		●		●
	A												●		●		●										
CTT	J											●	●	●	●	●	●						●		●		●
	A												●		●		●										
BLU	J											●	●	●	●	●	●						●		●		●
	A												●		●		●										

Methods for Modeling the Food Web

- Monte Carlo model
  - Models a range of possible outcomes and associated probabilities
  - Represents any factor with inherent uncertainty with probabilistic distribution
  - Repeat runs over and over with stochastic selection of values from the input distributions
- Originally developed in Crystal Ball software; converted to R statistical programming language
- Steady-state approximation of complex and dynamic reality

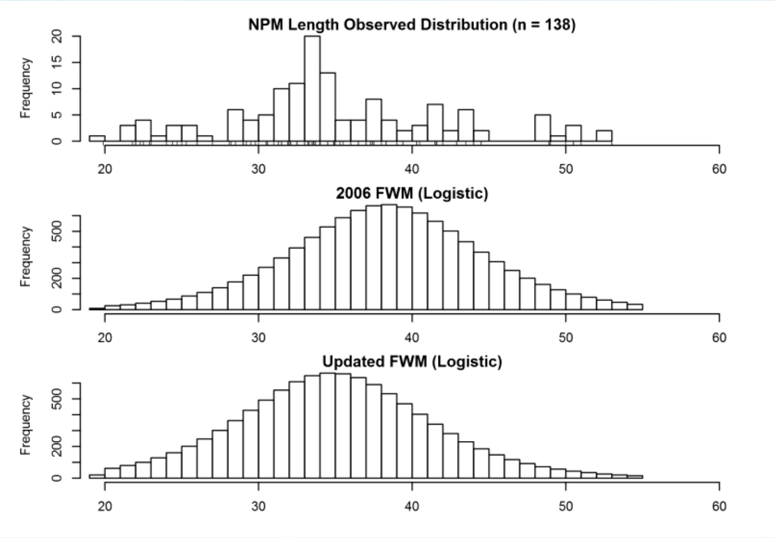
FWM Framework



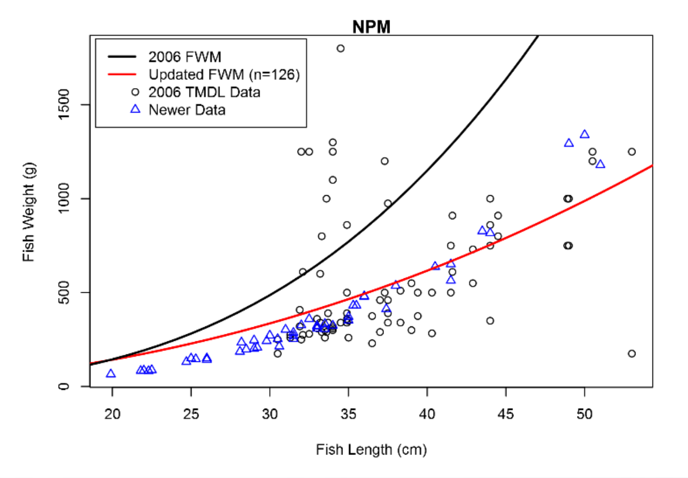
Updating the FWM

- ▶ Refit input distributions and model parameters
- ▶ Recalibrated model (e.g., observed fish tissue mercury)

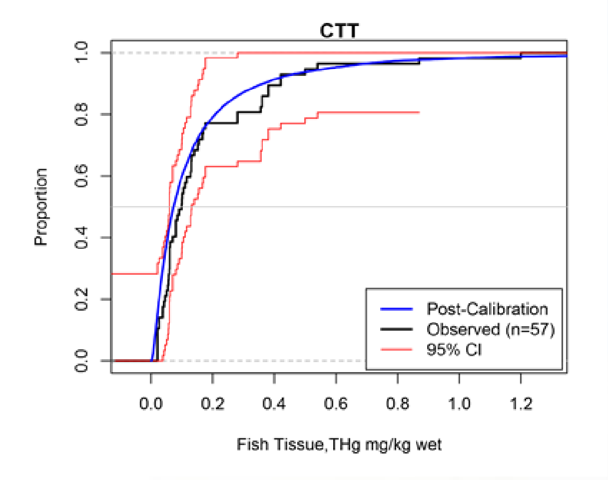
Updated Input Distributions



Updated Input Parameters



Post-calibration Fish Tissue THg



FWM Model Sensitivity Analysis

- Key factors contributing to variance in fish Hg:
  1. Diet specification
  2. MeHg elimination rate coefficients
  3. MeHg assimilation efficiency
  4. Adult body length (surrogate for weight/age)
  5. MeHg distribution
- Item 1 represented stochastically
- Additional data to specify 4 and better fit 2 and 3

Biomagnification Factors from FWM

- Relate fish tissue mercury concentration to the water column exposure concentrations (dissolved MeHg)
- Species specific (tropic level III and IV fish)

$$TL_n = \left[\frac{TC}{BMF_{ME,n} \cdot \Omega}\right] \cdot CF$$

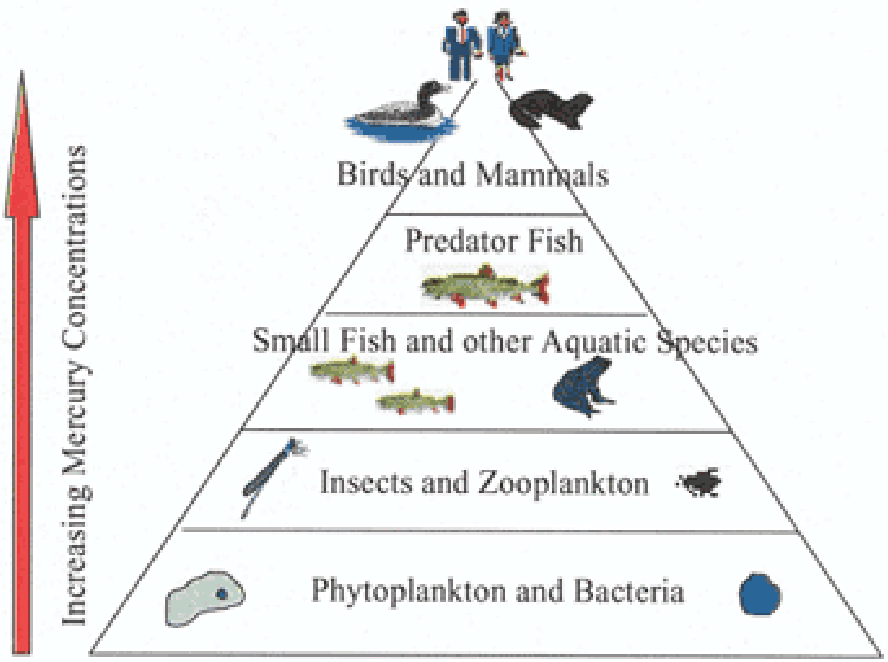
TL_n is the total mercury target level for the n^{th} fish species (ng/L),
TC is the revised fish tissue criterion for MeHg in fish (0.040 mg/kg),
$BMF_{ME,\,n}$ is the biomagnification factor for the n^{th} fish species (L/kg),
Ω represents the Mercury Translator, and
CF is a conversion factor ($1 \cdot 10^6$ ng/mg).

Biomagnification Factors from FWM (continued)

- Presented as a probabilistic distribution
- Still need to determine necessary instream THg concentrations

Fish Species	Mean	Standard Deviation	5th %ile	Median	95th %ile
Bluegill	1.22E+07	1.94E+07	1.43E+06	6.39E+06	2.76E+07
Common Carp	7.78E+06	8.35E+06	1.49E+06	5.48E+06	1.56E+07
Cutthroat Trout	4.81E+06	6.05E+06	4.59E+05	2.94E+06	1.08E+07
Largemouth Bass	2.74E+07	5.46E+07	2.16E+06	1.36E+07	5.71E+07
Largescale Sucker	7.69E+06	8.10E+06	1.53E+06	5.44E+06	1.55E+07
Northern Pikeminnow	3.26E+07	6.50E+07	2.63E+06	1.78E+07	7.01E+07
Rainbow Trout	7.59E+06	1.25E+07	5.78E+05	4.04E+06	1.68E+07
Smallmouth Bass	9.31E+06	1.25E+07	9.92E+05	5.73E+06	2.00E+07

Questions on the FWM?



Accumulation of mercury in the food chain.

Image source: Clean the Rain, Clean the Lakes: National Wildlife Federation, 2000.

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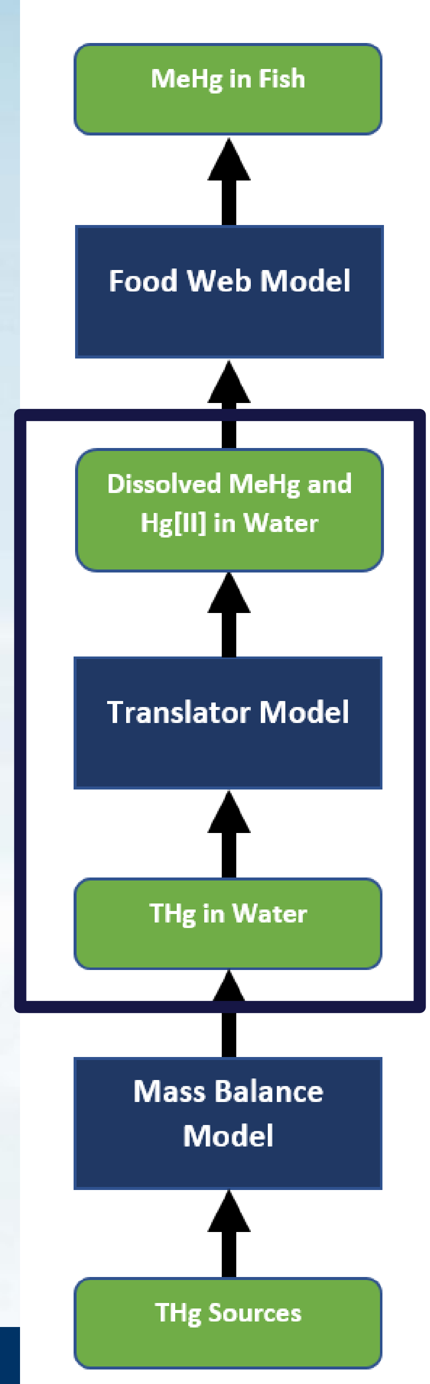
MERCURY TRANSLATOR MODEL



MeHg Production

- Most Hg in environment is in inorganic forms
- Converted to MeHg by bacteria under low oxygen conditions in saturated soils, sediment, or lake bottom water
- Non-linear process that depends on temperature, carbon, sulfur, and reduction/oxidation conditions
- Limited data to mechanistically model this process

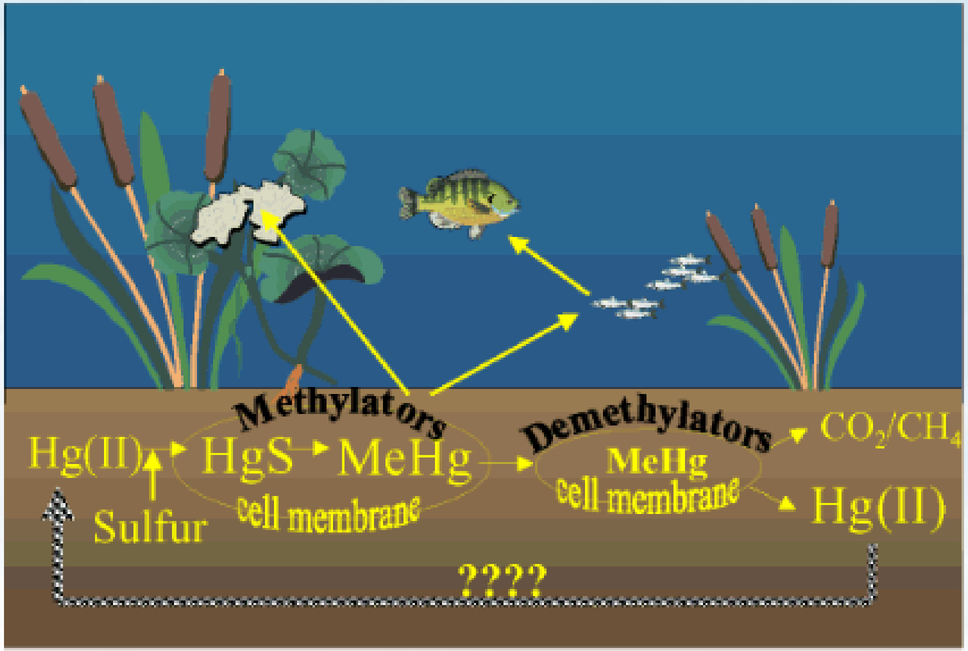


Image source: South Florida Restoration Science Forum (https://sofia.usgs.gov/sfrsf/rooms/acme_sics/acme/)

Mercury Translator Model (Ω)

- *Purpose*: Convert dissolved MeHg [dMeHg] target exposure concentrations from FWM to corresponding THg concentration targets in water
- Translator is an empirical approximation of the complex relationships that determine Hg solubility and methylation

$$\Omega = \frac{dMeHg}{THg}$$

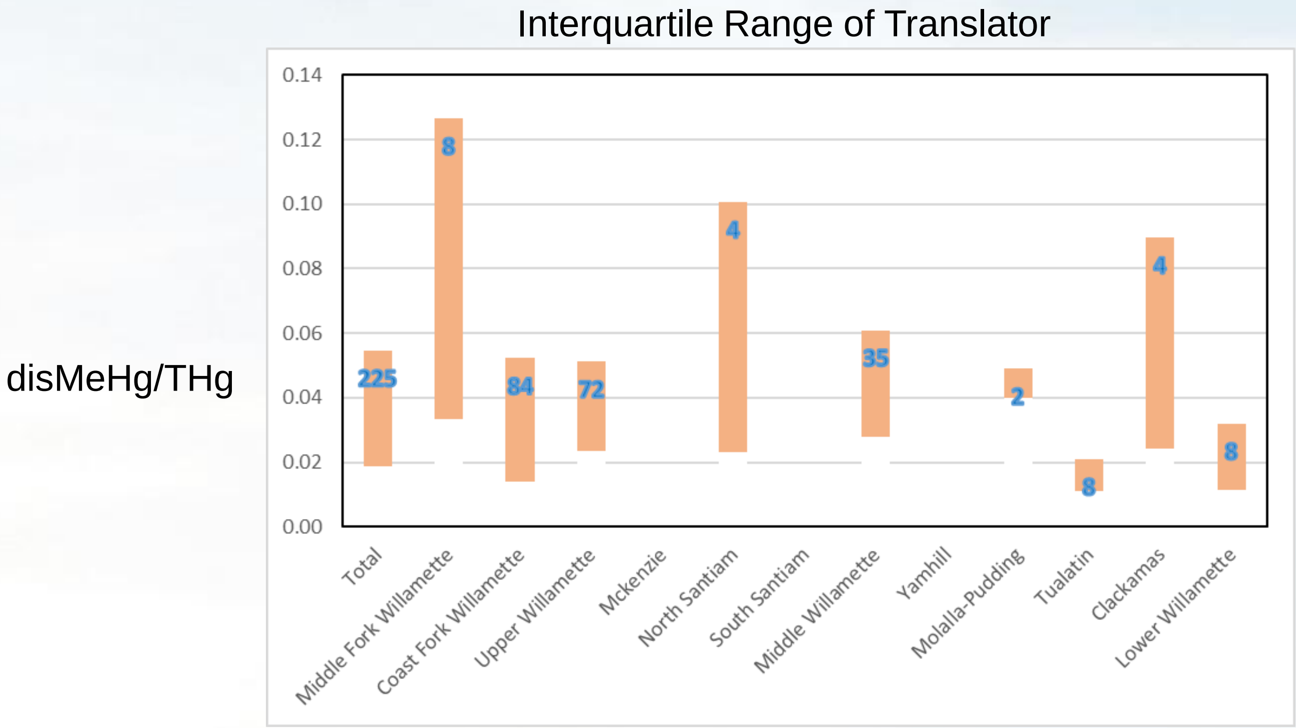
- Input data: paired dissolved MeHg and THg samples
  - *Paired means that dissolved MeHg and THg were sampled at the same time and location*

Refining the Translator

- Large amounts of paired data now available
- Ω may vary according to local biochemical conditions
- Key assumptions
  - THg in the water column is indicator of mercury available for methylation through equilibrium at the water-sediment interface
  - Central tendency reflective of relationship between THg supply and methylation rate

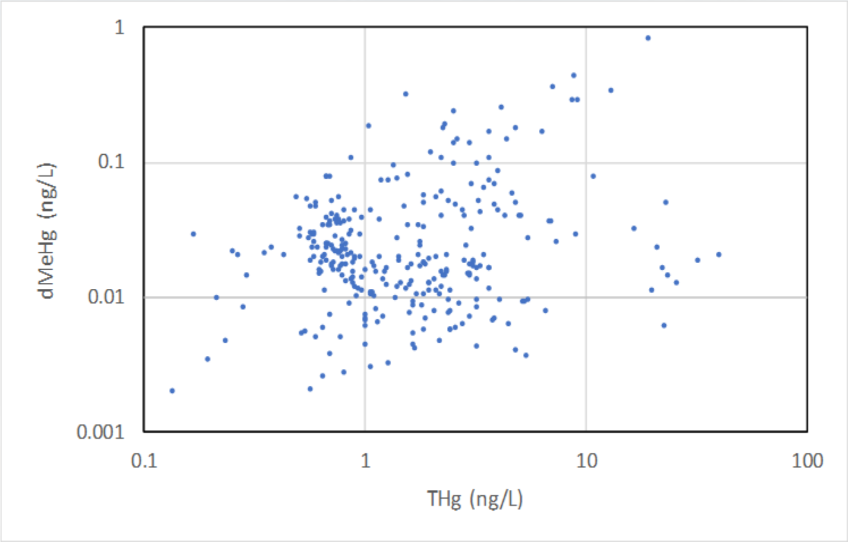
Spatial Variation

- HUCs with larger sample sizes exhibit similar ratios
- HUCs that significantly differ have small sample sizes, and using separate translators may be unreliable

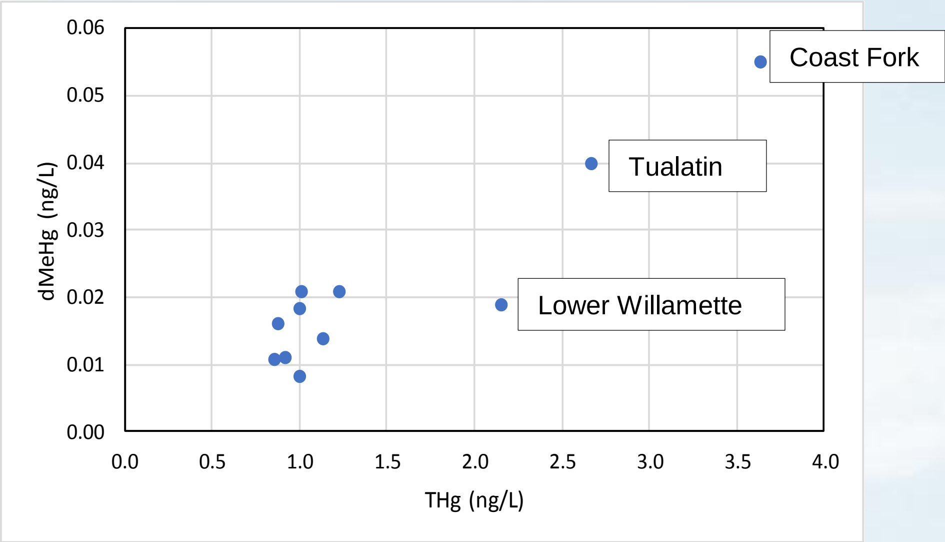


Pairing versus Aggregating Observed Data

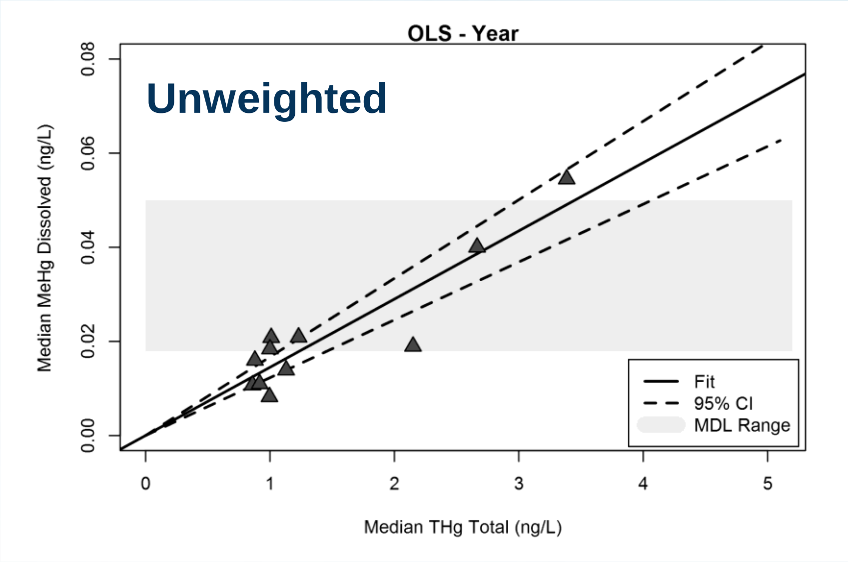
- Paired samples
  - Weak predictive relationship

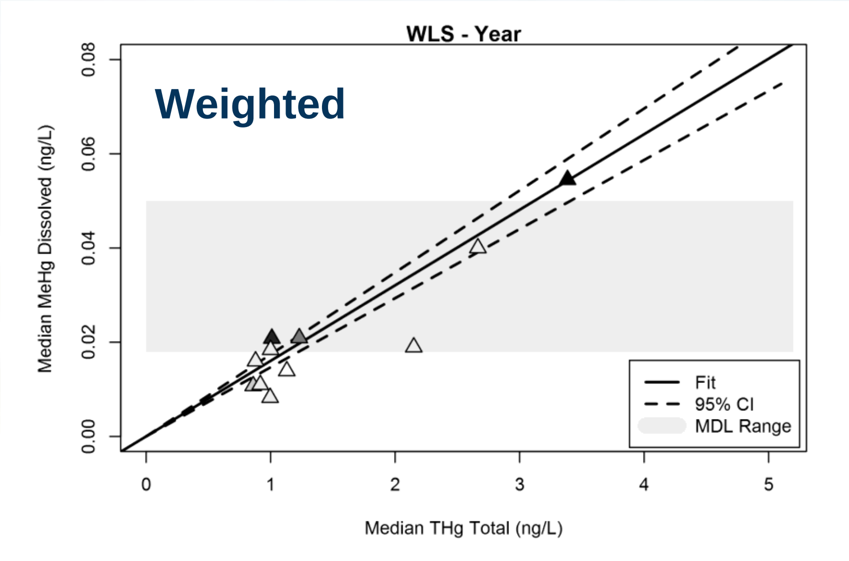


- Aggregated by HUC8
  - Strong predictive relationship



Weighted by Sample Count versus Unweighted





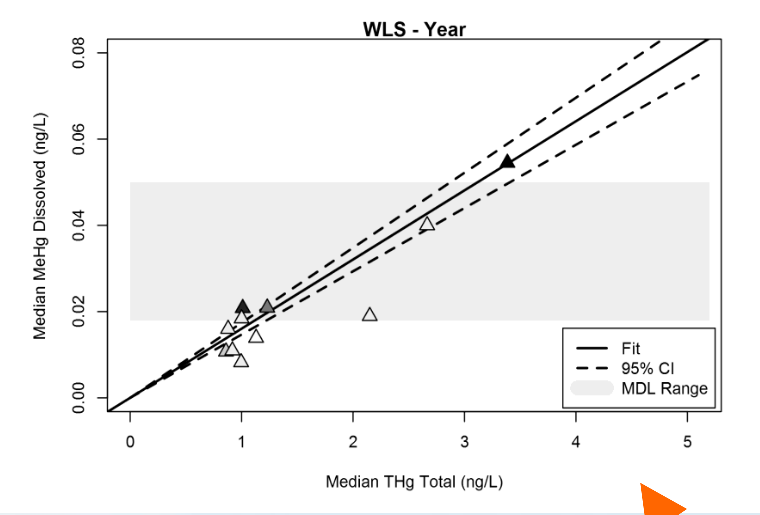
- Findings
  - Slope similar
  - Translator not biased by weighting
  - Both good models

Sample Counts for Weighted Translator

Watershed	Sample Count
Middle Fork Willamette	17
Coast Fork Willamette	71
Upper Willamette	95
McKenzie	9
North Santiam	9
South Santiam	0
Middle Willamette	55
Yamhill	5
Molalla-Pudding	10
Tualatin	9
Clackamas	8
Lower Willamette	9
Total	**97**

Variation in Ω

- Assessed performance of
  - Seasonal translators
  - Weighted versus unweighted translators
  - Translators with and without Coast Fork data



Scenario	Season	Slope	Slope SE	Slope P-value	Lower 95%CL	Upper 95%CL	R^2
WLS, All Data	Year	0.0160	0.0006	<0.0001	0.0147	0.0174	0.99
	Summer	0.0347	0.0021	<0.0001	0.0300	0.0393	0.96
	Winter	0.0070	0.0006	<0.0001	0.0057	0.0083	0.93
OLS, All Data	Year	0.0145	0.0010	<0.0001	0.0123	0.0167	0.96
	Summer	0.0260	0.0038	<0.0001	0.0175	0.0346	0.82
	Winter	0.0086	0.0010	<0.0001	0.0063	0.0109	0.87
WLS, No Coast Fork	Year	0.0164	0.0013	<0.0001	0.0136	0.0193	0.95
	Summer	0.0305	0.0038	<0.0001	0.0220	0.0391	0.88
	Winter	0.0075	0.0011	0.0001	0.0050	0.0101	0.83
OLS, No Coast Fork	Year	0.0145	0.0012	<0.0001	0.0118	0.0172	0.94
	Summer	0.0219	0.0038	0.0003	0.0134	0.0305	0.79
	Winter	0.0101	0.0013	<0.0001	0.0071	0.0131	0.86

THg Water Column Targets

Biomagnification Factors

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Revised Fish Tissue Criterion

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Mercury Translator

→

Water Column THg Targets

$$TL_n = \left[\frac{TC}{BMF_{ME,n} \cdot \Omega}\right] \cdot CF$$

TL_n is the total mercury target level for the n^{th} fish species (ng/L),
TC is the revised fish tissue criterion for MeHg in fish (0.040 mg/kg),
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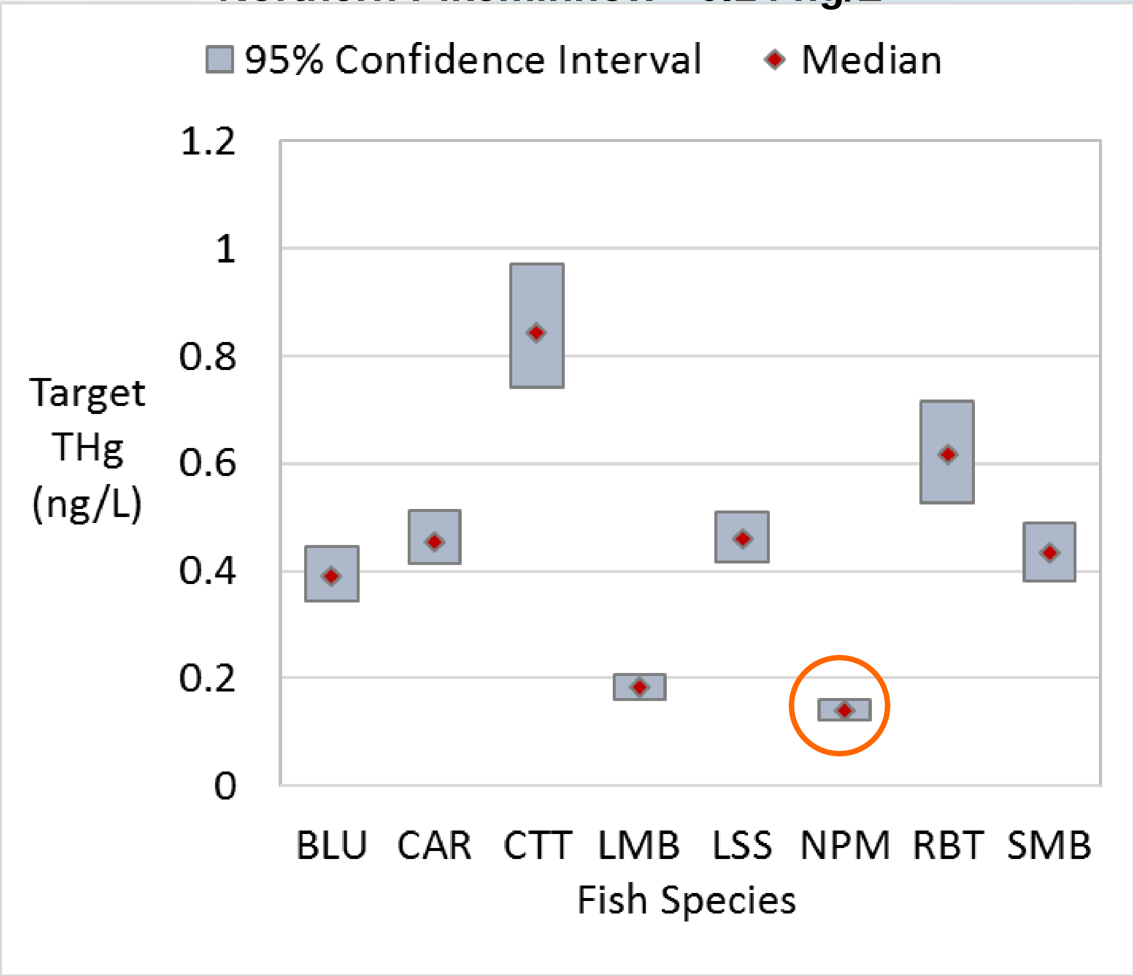
Ω represents the Mercury Translator, and
CF is a conversion factor ($1 \cdot 10^6$ ng/mg).

THg Water Column Targets (continued)

Most conservative target:
Northern Pikeminnow - 0.14 ng/L



Questions on the mercury translator model?

Cottage Grove Reservoir (Image credit: Liam Schenk, USGS)

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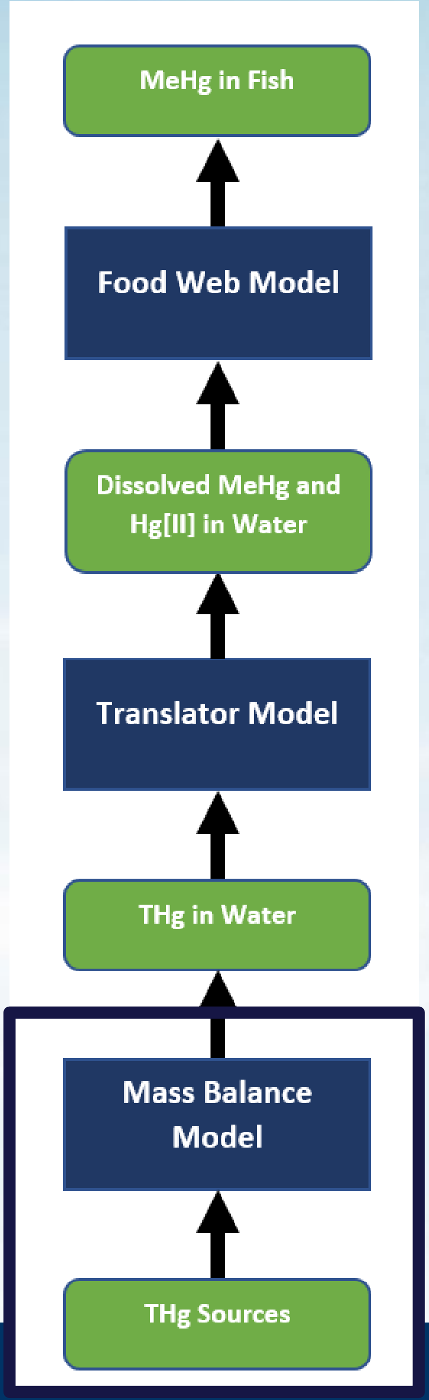
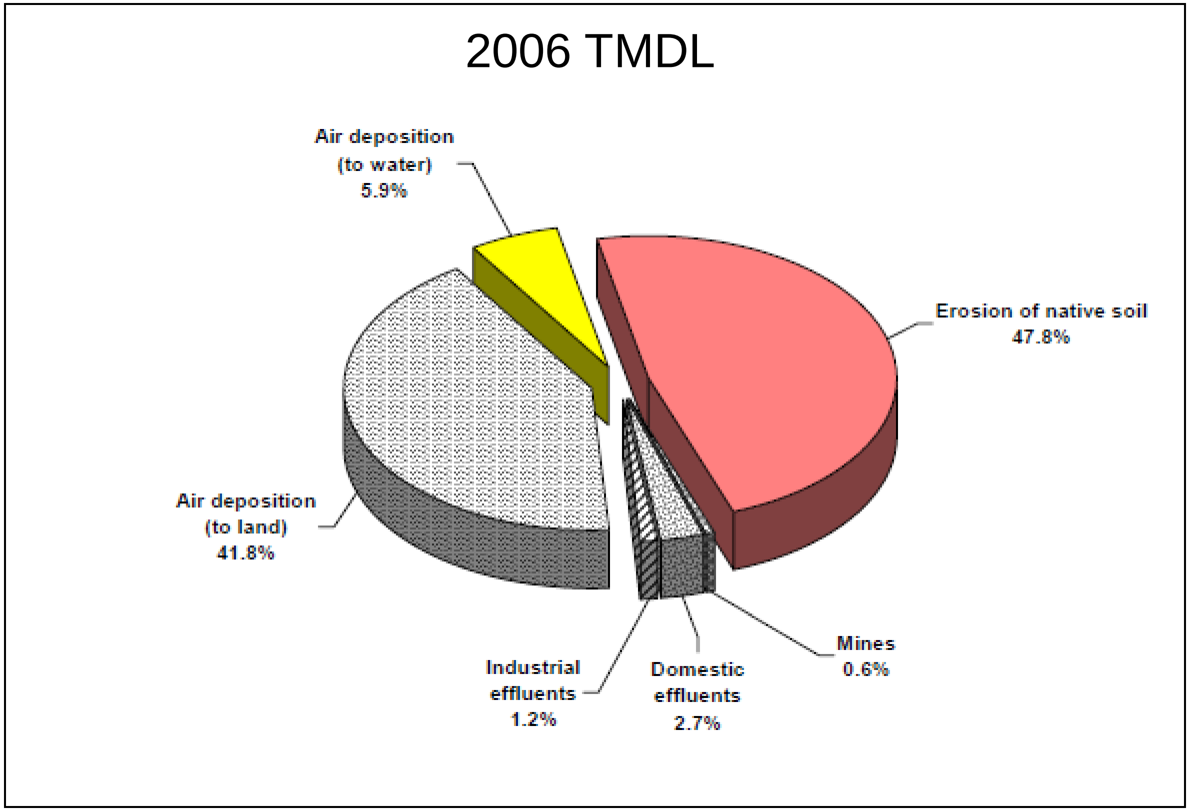
MASS BALANCE MODEL



Mass Balance Model (MBM)

- *Purpose*: Connect sources of THg to ambient THg concentrations in the river network

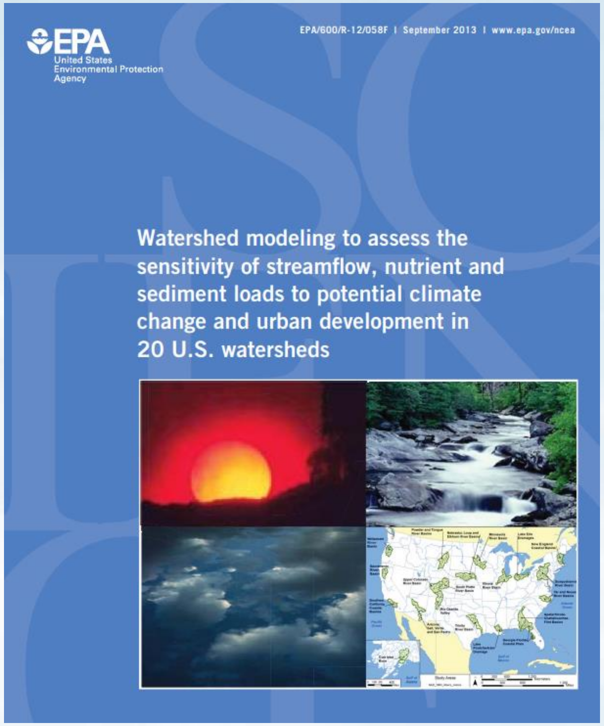


Areas for Improvement of 2006 MBM

- ►Used USLE soil erosion, single, uniform soil THg concentration, and generic delivery ratio
- ►Required delivery ratio estimate for atmospheric deposition
- ►Limited data for characterizing mine and point source loads
- ►Focus on load at mouth – but THg concentration predictor had R^2 of only 20%
- ►Improvements can be made to the 2006 TMDL MBM through the use of a watershed model and additional data

Improving the Mass Balance Model

- ► Improved data availability across categories
- ► Use existing Hydrologic Simulation Program – FORTRAN (HSPF) watershed model that mechanistically represents flow and sediment loading/transport
- ► Developed by Tetra Tech and AQUA TERRA to support an EPA climate study
- ► Report Available: https://cfpub.epa.gov/ncea/global/recordisplay.cfm?deid=256912

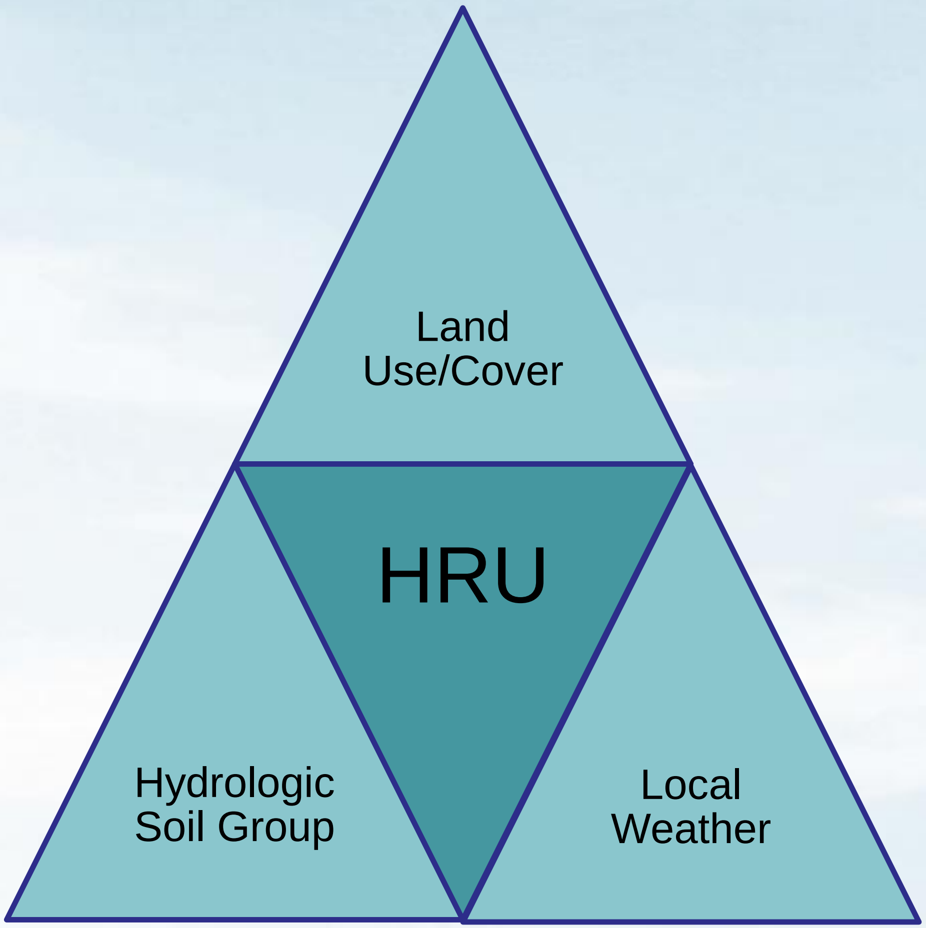


Willamette River Basin HSPF Model

- Simulation uses hourly time step
- Calibrated for flow and sediment
- Incorporates weather zones and land cover types combined with soil information and imperviousness
- Subwatersheds at approximately HUC10 scale
- Not a mercury model
- It's still useful for characterizing long-term average results for unit area land cover
  - Surface and subsurface flow components
  - Sediment erosion and delivery to and through stream network, including reservoir trapping

HSPF Upland Representation

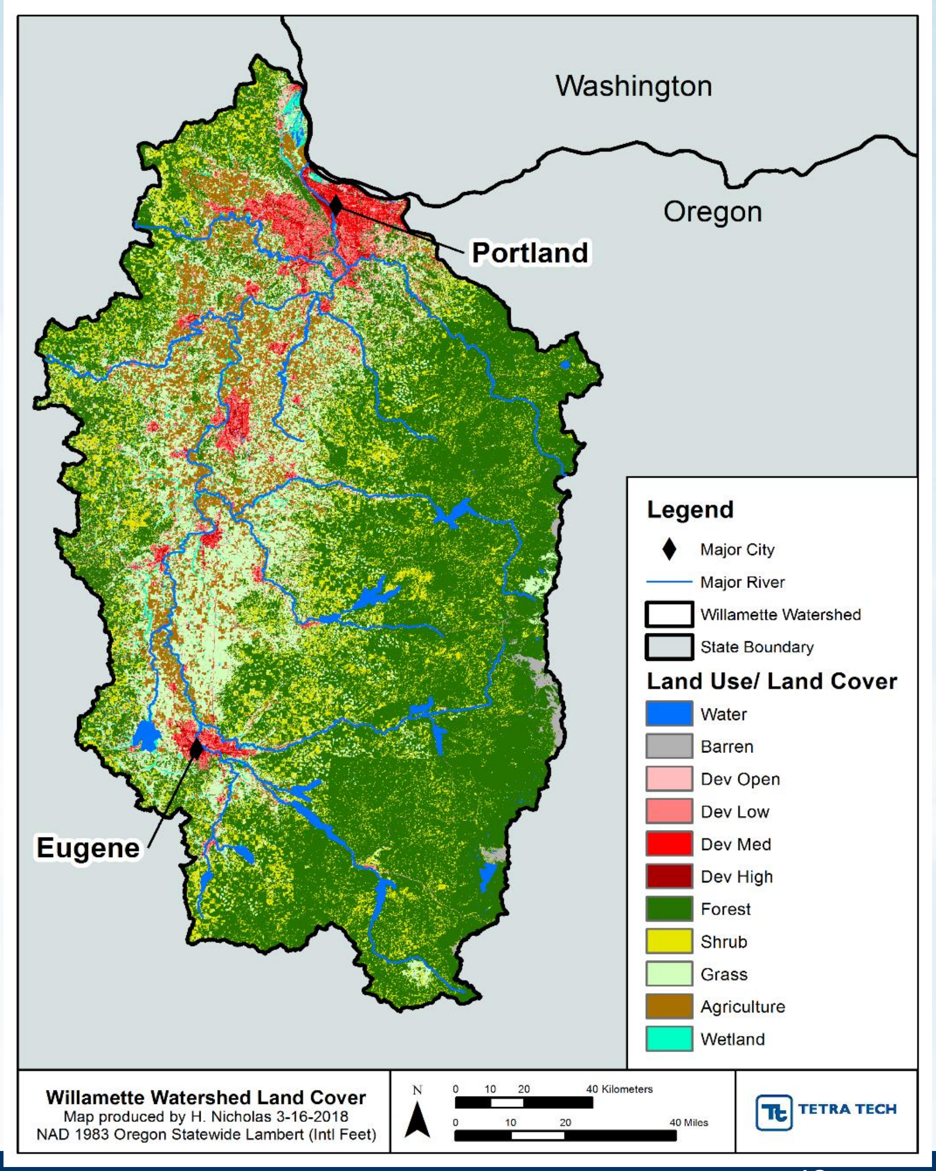
- Upland processes simulated at the Hydrologic Response Unit (HRU) level
- HRUs represent diverse combinations of land use, soil, and weather
- Provide useful information about flow and sediment transport across the landscape



WRB HSPF Model Land Use/Cover

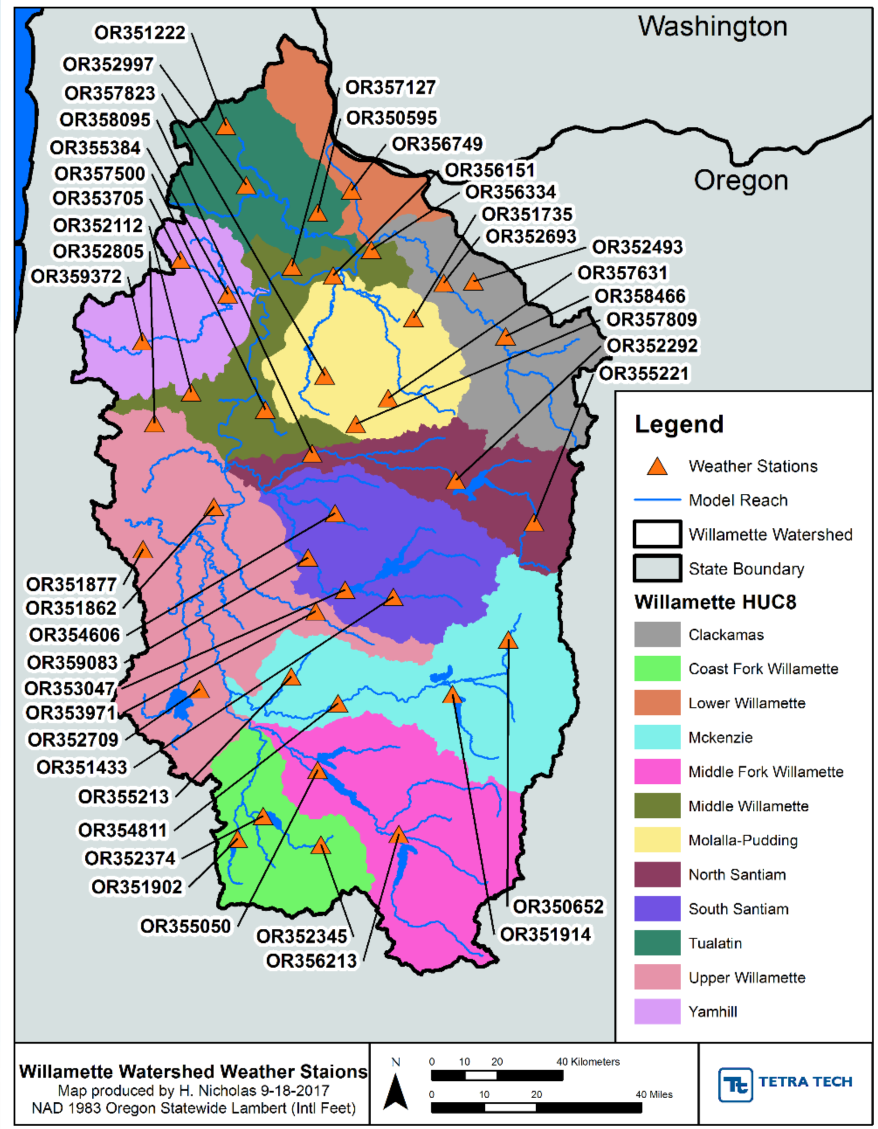
- Land use originally developed with National Land Cover Database (NLCD) 2001
- Updated with NLCD 2011

Land Cover	Total Area (mi^2)
Agriculture	912
Barren	102
Developed-High Density	81
Developed-Medium Density	204
Developed-Low Density	333
Developed-Open	305
Forest	5,920
Grassland	1,902
Shrub	1,412
Water	103
Wetland	192
Total	11,466

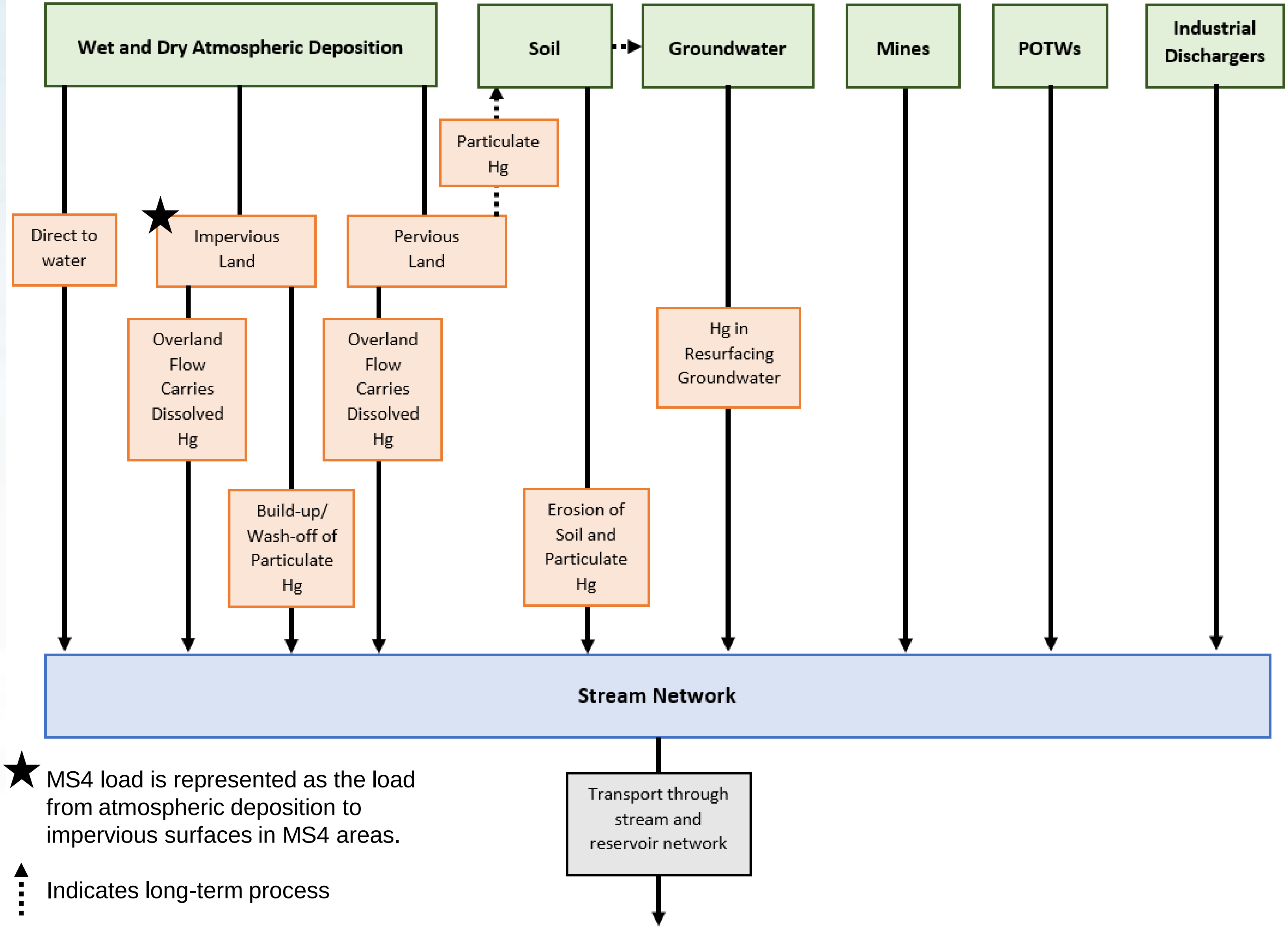


WRB HSPF Soils and Weather Data

- Hydrologic Soil Group (HSG) from gridded STATSGO coverages
- Weather data from BASINS4 (comprehensive source for meteorological data)



Mass Balance Model Framework

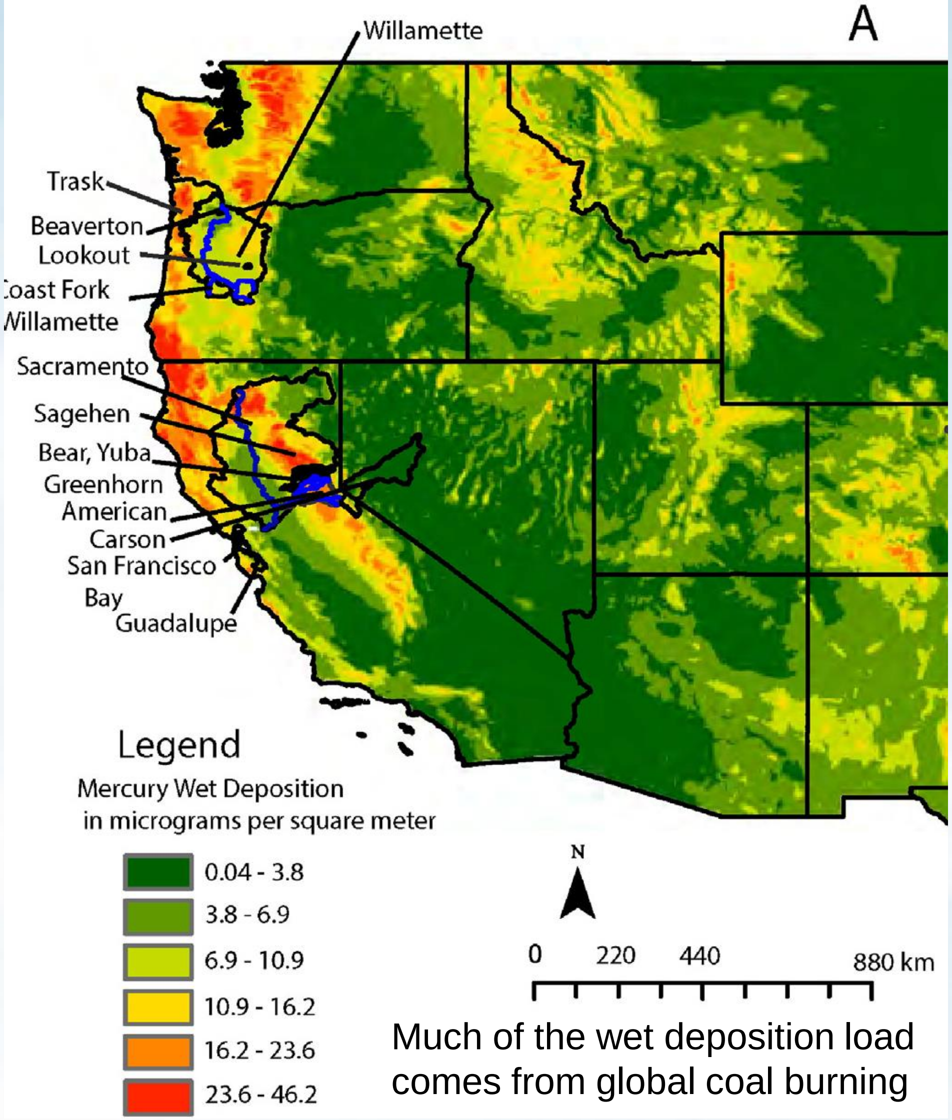


★ MS4 load is represented as the load from atmospheric deposition to impervious surfaces in MS4 areas.

Indicates long-term process

Atmospheric Deposition Fluxes

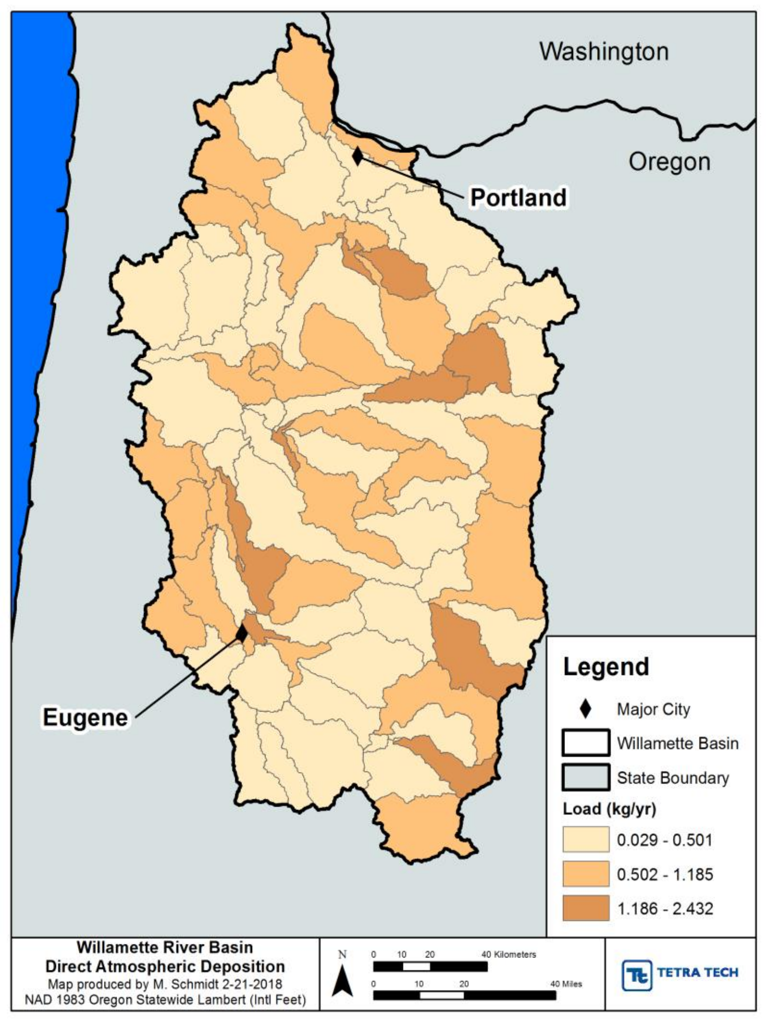
- ► Flux: The rate that a mass of mercury moves from the atmosphere to the landscape
- ► Summarized for western U.S. by Domagalski et al., 2016 (*Science of the Total Environment* 568: 638-650.)
  - ▪ Wet deposition
    - • National Atmospheric Deposition Program
    - • Annual average flux: 9.62 µg/m^2/yr
  - ▪ Dry Deposition
    - • Community Multiscale Air Quality Model
    - • Annual average flux: 4.24 µg/m^2/yr



Much of the wet deposition load comes from global coal burning

Atmospheric Deposition Load to Streams

- ► Combine fluxes with information from HSPF
  - ▪ Wet deposition load estimated from deposition rate grids, fraction of precipitation that becomes runoff, and land area
  - ▪ Dry deposition to impervious surfaces using a buildup-washoff model
- ► Dry deposition to pervious land and wet deposition that infiltrates is accounted for in soil erosion component
- ► Include mercury deposition direct to water surface

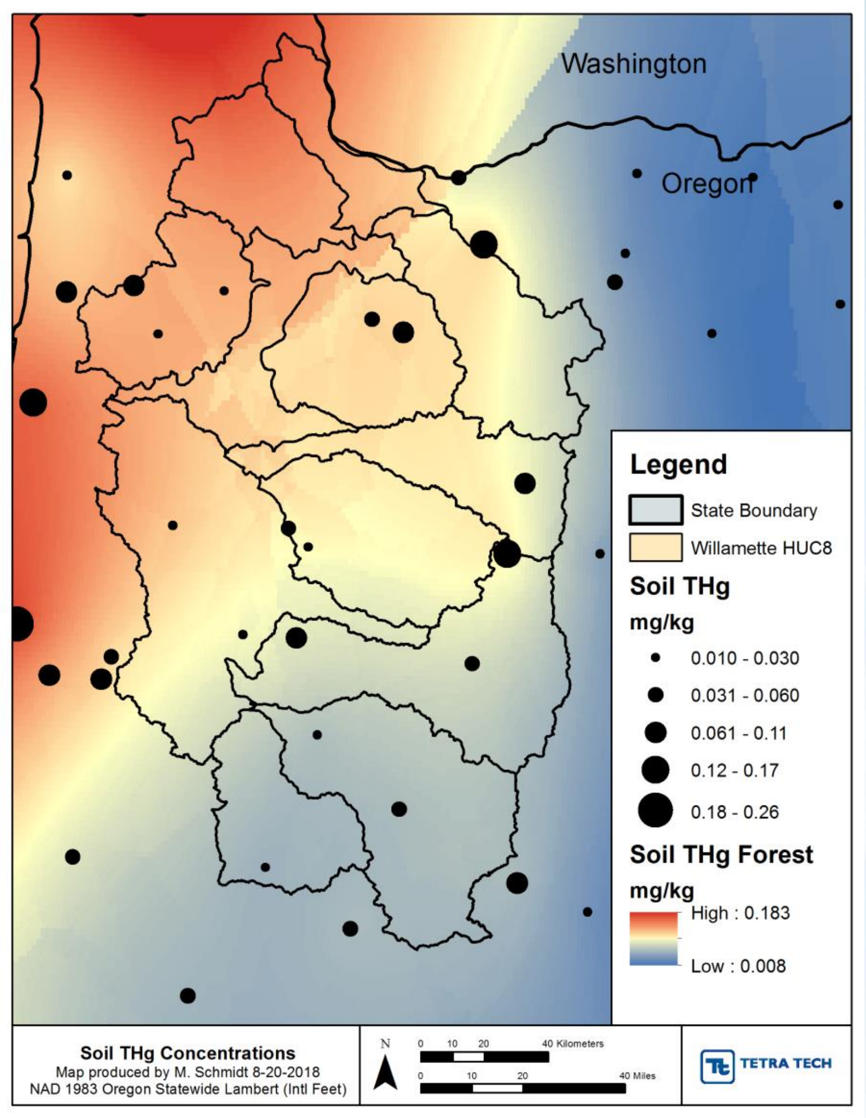


Soil Erosion

- ► Mercury stored in soil comes from recent atmospheric deposition, legacy emissions, plant litter, geology
- ► Sheet and rill erosion and gully erosion can transport particulate-mercury to waterbodies
- ► Previously used a statewide-NRCS erosion rate, enrichment factor, and single delivery factor
- ► Differs based on cover, soil type, rainfall patterns, and slope – characterize with HSPF model

Soil Matrix THg Concentration

- ► Soil THg potency factors expected to differ by geology and land use/cover
- ► Gridded data indicates potency ranges from 0.01-0.20 mg-THg/kg-soil in the WRB
  - Smith, D.B., et al. 2013. Geochemical and Mineralogical Data for Soils of the Conterminous United States. U.S. Geological Survey Data Series 801.
- ► Performed spatial interpolation by land use



Groundwater

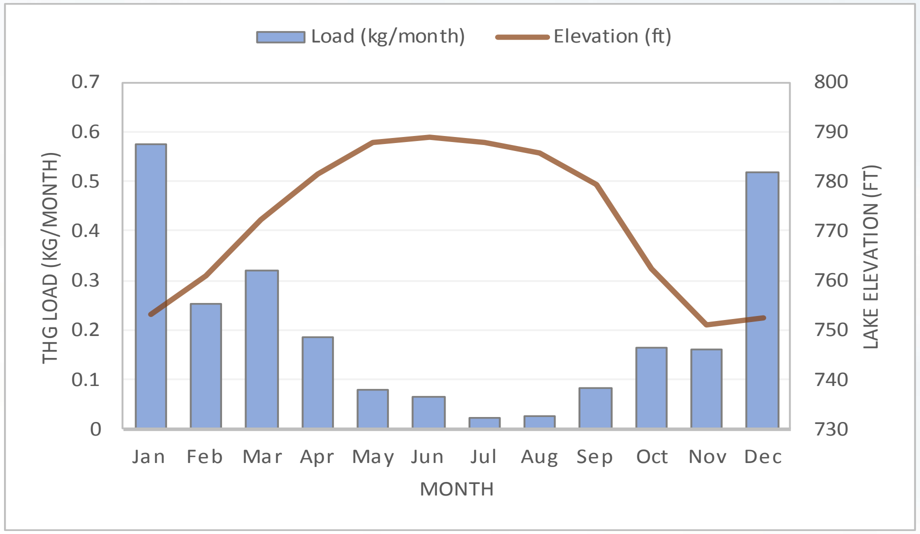
- Dissolved mercury leaches through surface soils and enter streams with resurfacing groundwater
- Only a few well samples available
- Groundwater mercury concentration studies:
  - A lake study in Wisconsin: 2 – 4 ng/L (Krabbenhoft and Babiarz, 1992)
  - Forested Minnesota watershed: 0.9 ng/L (Grigal et al., 2000)
  - Groundwater sampling near Black Butte Mine
    - Samples in the vicinity of the mine from 1998 all non-detect but limit of 200 ng/L (Oregon Health Authority, 2013)
    - Two samples collected in 2013 for background groundwater quality upstream of the mine as part of the remediation investigation were below detection (5 ng/L limit) and a third was 1.19 ng/L
- Likely low concentration, but high flow volume since groundwater is the primary flow pathway

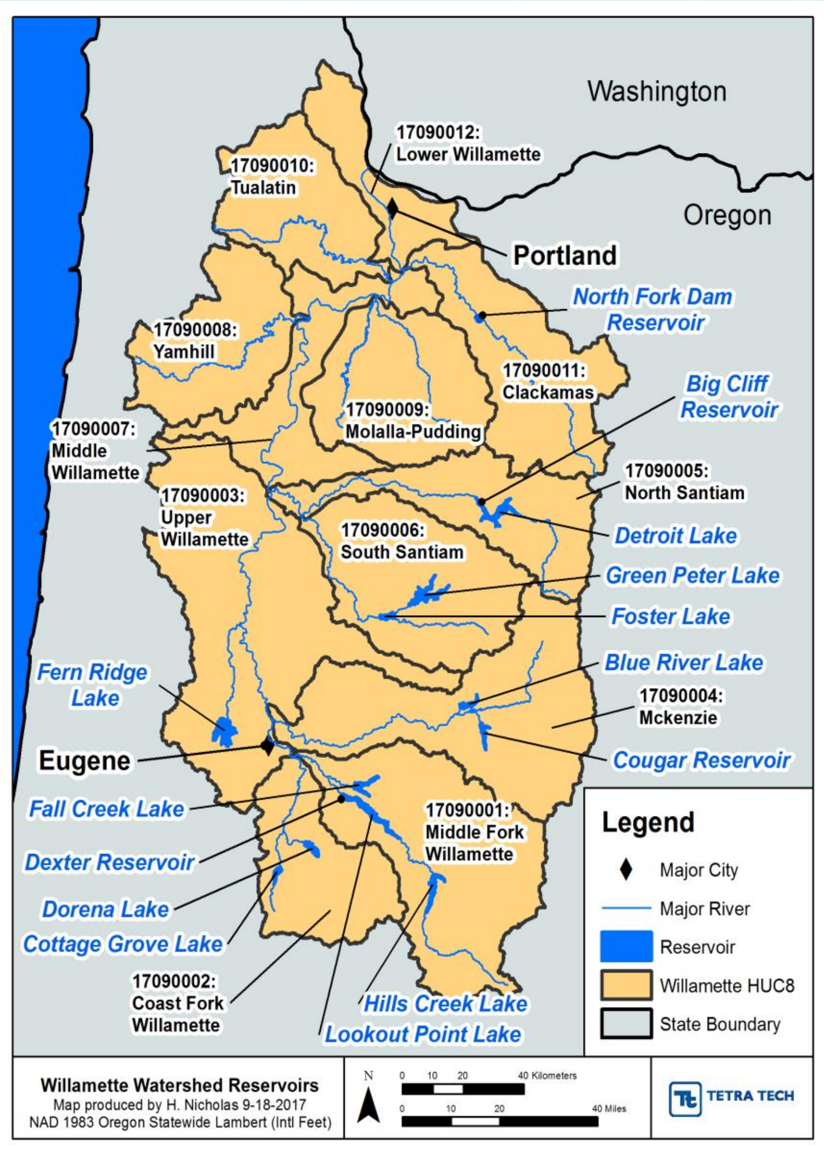
Mines

- Very limited mine data for 2006 TMDL
- Use empirical approach to calculate loads
- Mines in the Coast Fork watershed
  - Black Butte Mine
    - Historic Hg mine upstream of Cottage Grove Reservoir
    - Flows to Dennis and Furnace Creeks
  - Bohemia District
    - Historic gold mine that used mercury amalgamation
    - Along Upper Row River, above Dorena Reservoir
  - Loads leaving the downstream reservoirs also modeled
- Data still limited for other mines in the basin

Reservoirs

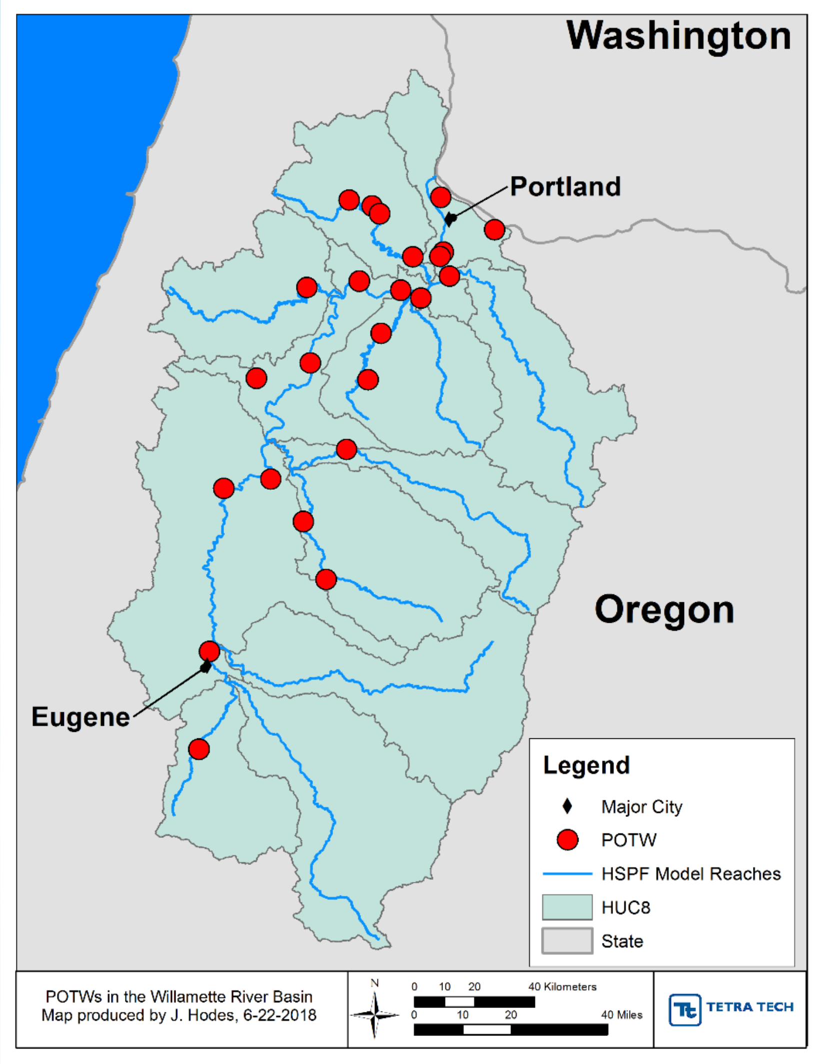
- Reservoirs trap sediment and associated Hg – but can provide an ideal location for creation of MeHg
- Use empirical analysis where data is available to estimate net change in THg (e.g., USGS LOADEST software)





POTW Discharges

- POTW effluent may contain Hg from multiple sources (e.g., dental amalgams)
- Flow and mercury self-monitoring data available to calculate loads for most permitted POTWs
- THg Concentrations
  - NPDES-DOM-A: 0.3 - 25 ng/L (n=227)
  - NPDES-DOM-B: 1.7 - 6.8 ng/L (n=67)
  - NPDES-DOM-C: 1.4 – 30 ng/L (n=61)



Permitted Industrial Process Wastewater Discharges

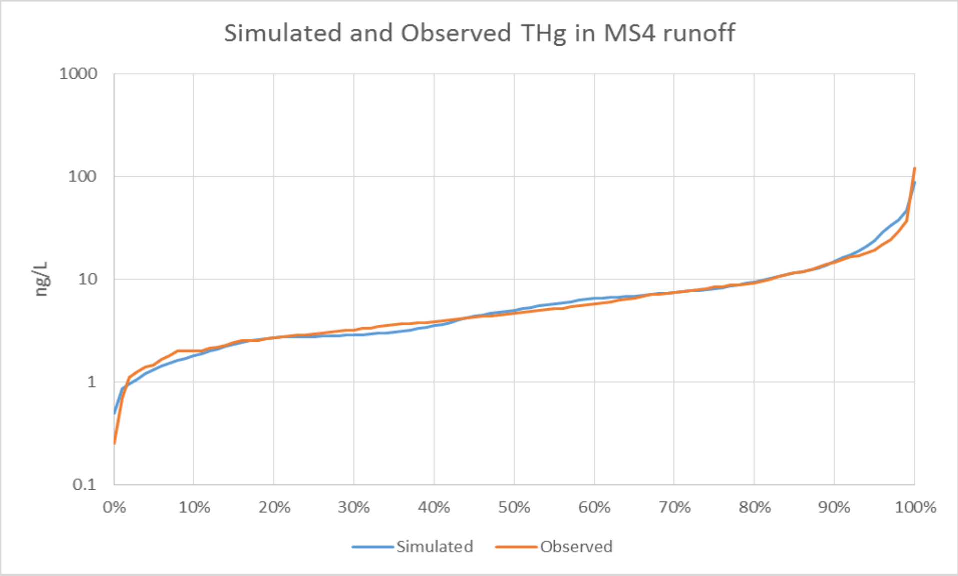
- Industrial sources can be significant mercury sources because of potentially high concentrations (even though flows often low)
- No data available for 2006 TMDL
- Data sources:
  - Self-monitoring of THg by permit holders
  - Loads from EPA Toxics Release Inventories
  - Discharge Monitoring Reports (e.g., monthly flow records)
  - Permit application and renewal documents

SIC Code	Categorical Description	Average THg Concentration (ng/L)
24xx	Timber products	5.5 (n=9)
26xx	Paper products	9.1 (n=8)
33xx	Primary metal industries	10 (n=1)

Urban Stormwater

- Municipal Separate Storm Sewer Systems (MS4s) are subject to discharge permits and can be sources of mercury
- Load calculated as atmospheric deposition to effective impervious MS4 areas
- Effective impervious area is the impervious area that is hydrologically connected to the storm sewer system

- MS4 stormwater monitoring (n=655)
  - Range: 0.25 – 120 ng/L
  - First Quartile: 2.94 ng/L
  - Median: 4.62 ng/L
  - Third Quartile: 8.31 ng/L



Summary

- ► Provide technical analyses to support TMDL to meet Court requirements
- ► Apply and build-on technical framework used for 2006 TMDL
  - Apply new fish tissue criterion
  - Incorporate new data across source categories
  - Make use of existing watershed model
- ► Three modeling components:

  1. Mass Balance Model: Link THg sources in the watershed to instream concentrations

  2. Mercury Translator: Link THg concentrations to MeHg and Hg[II] exposure concentrations

  3. Food Web Model: Link exposure concentrations of MeHg to fish tissue

Questions?

Willamette River near Portland (Image credit: Stuart Seeger, Flickr)