Modeling the Atmospheric Transport and Deposition of Mercury

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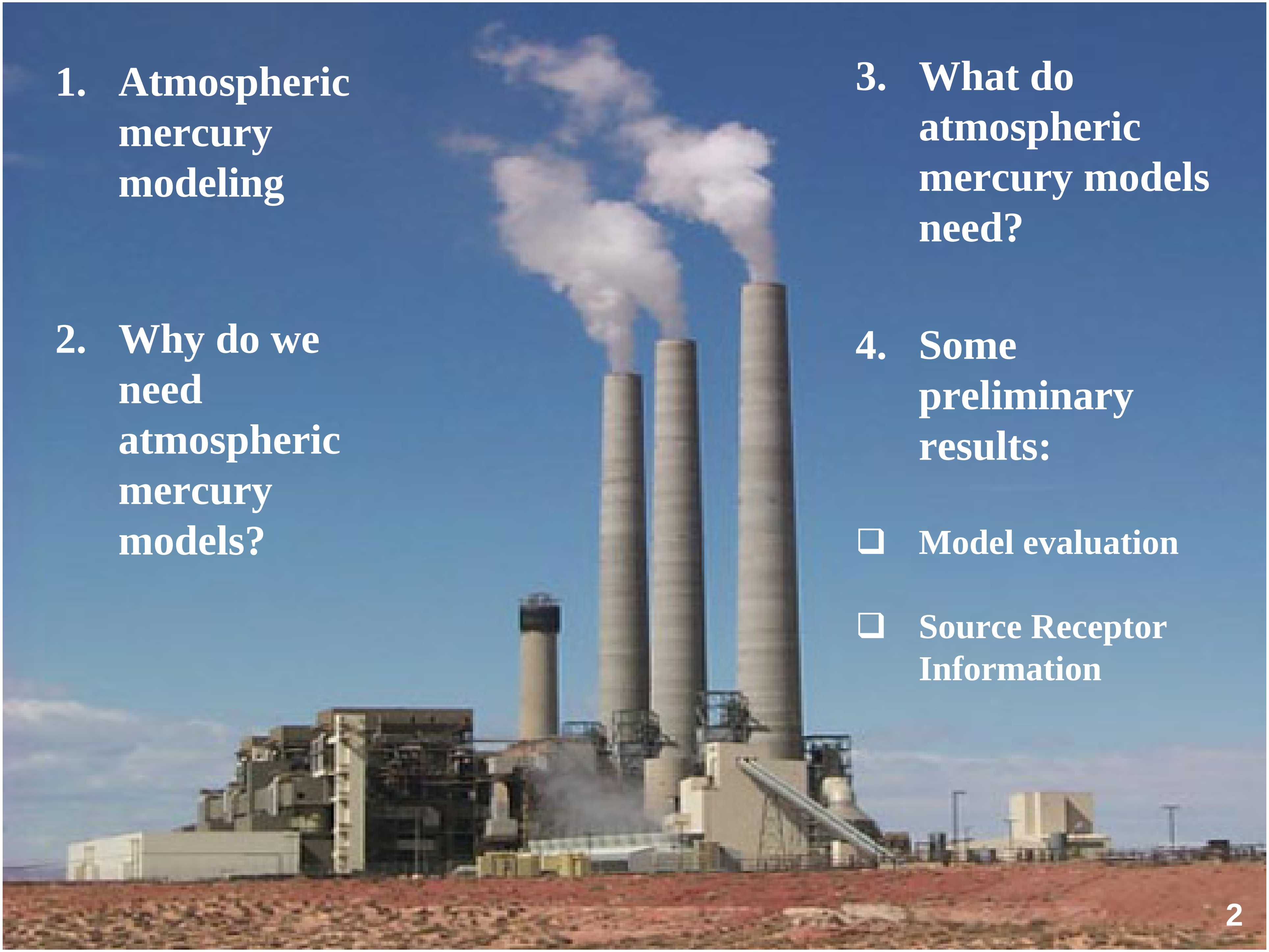
Materials assembled for
"Mercury in Maryland" Meeting, Appalachian Lab,
Univ. of Maryland Center for Environmental Science
301 Braddock Road, Frostburg MD, Nov 2-3, 2005



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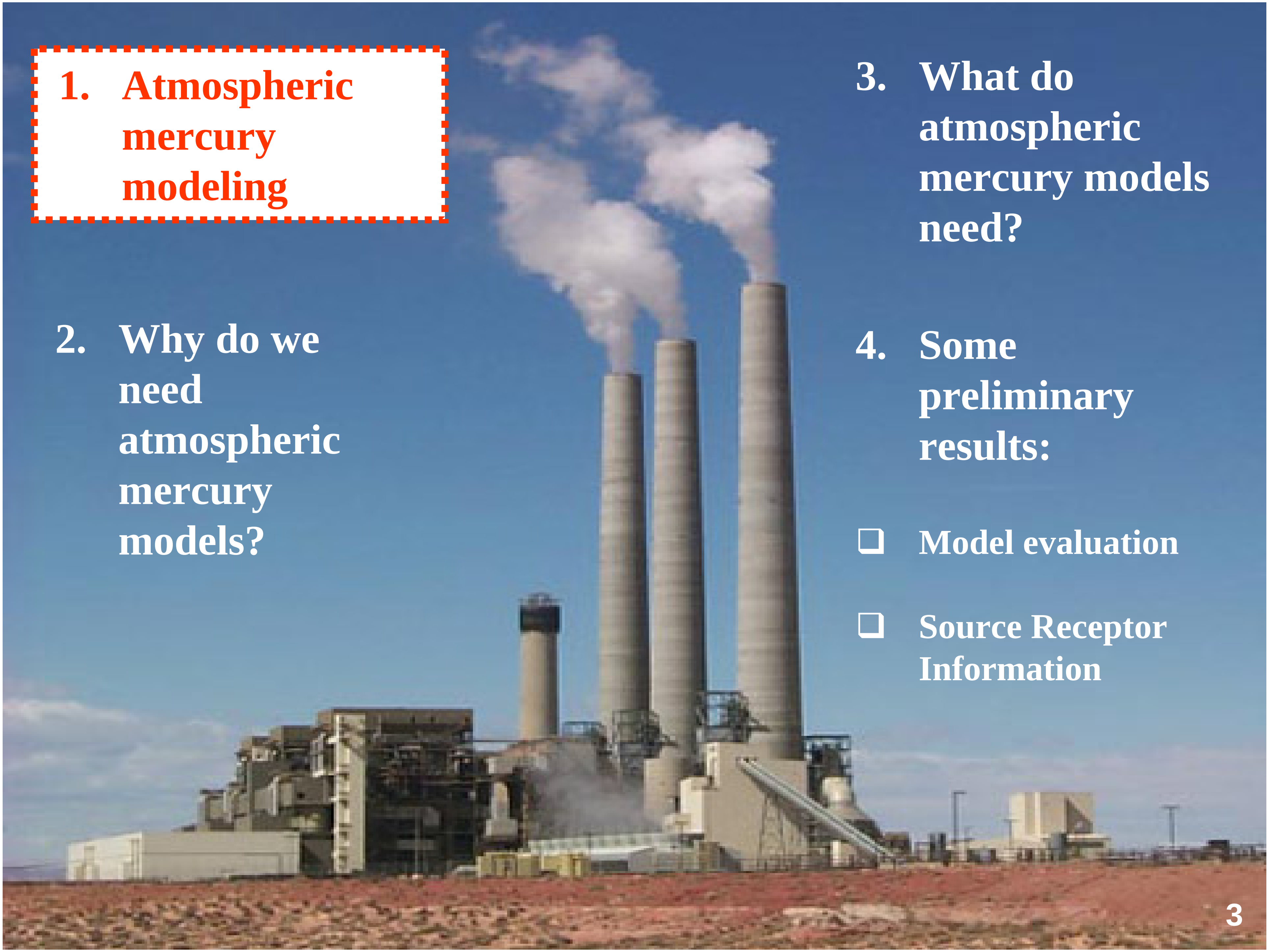
2. Why do we need atmospheric mercury models?
3. What do atmospheric mercury models need?
4. Some preliminary results:

- ❑ Model evaluation
- ❑ Source Receptor Information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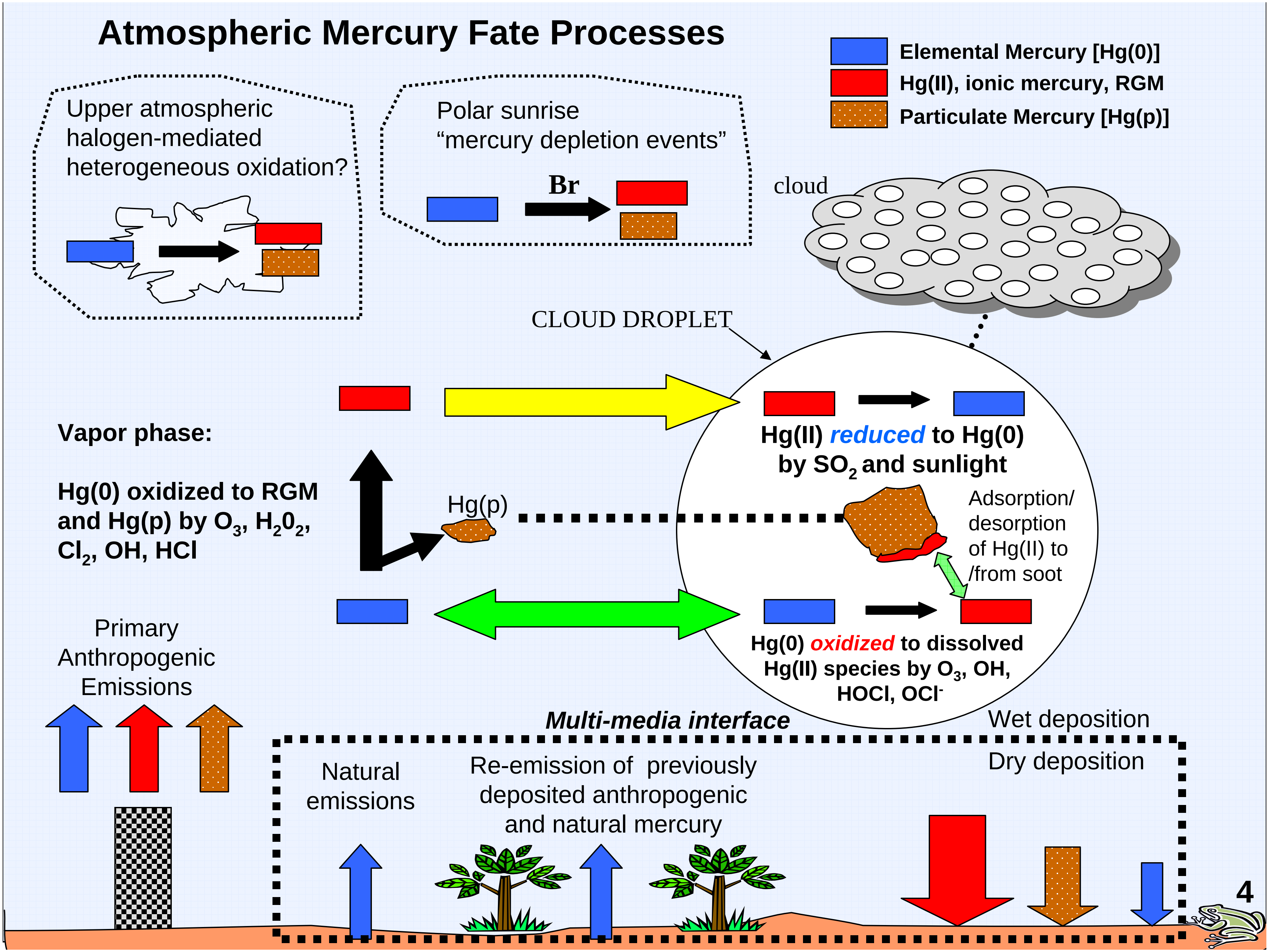
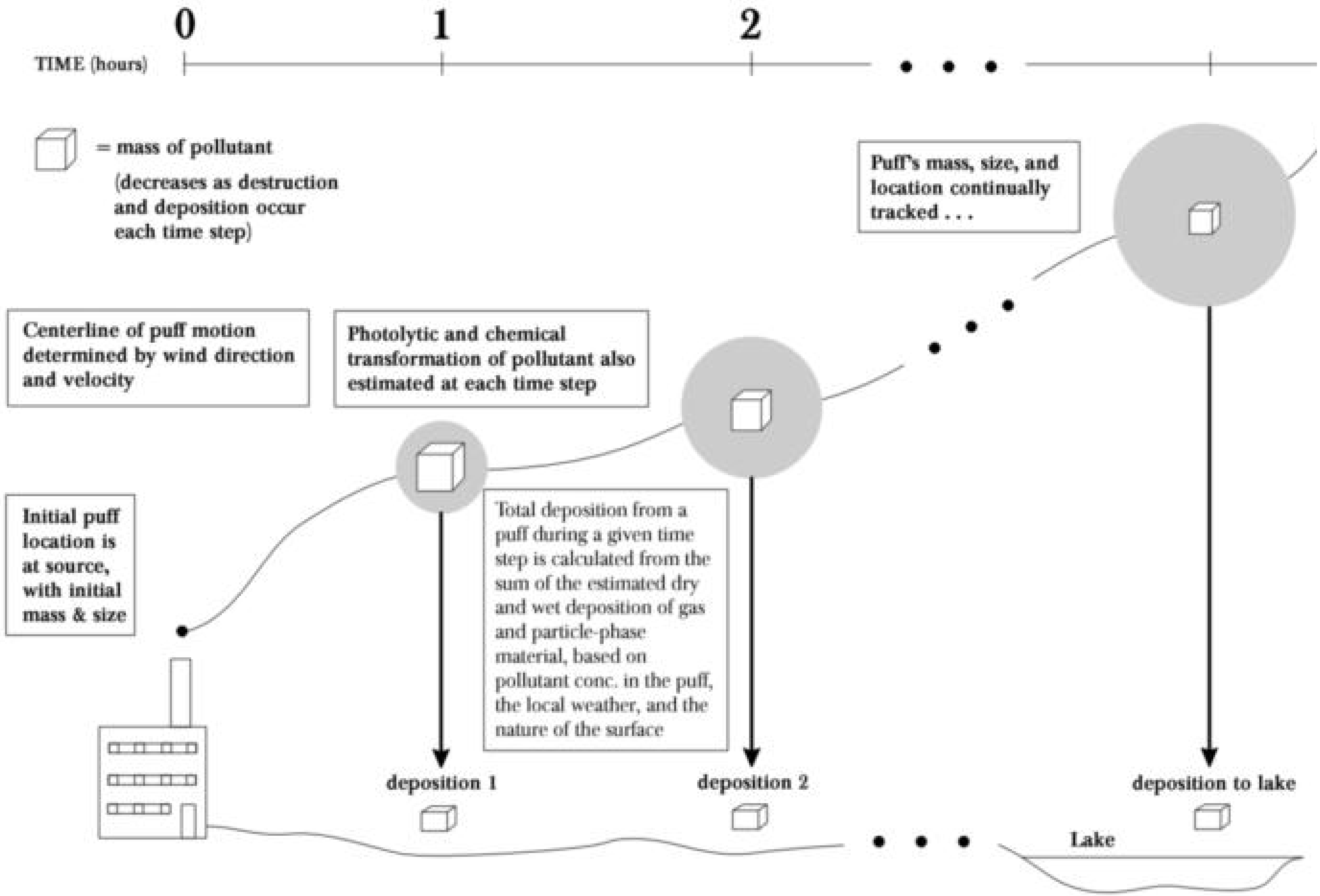
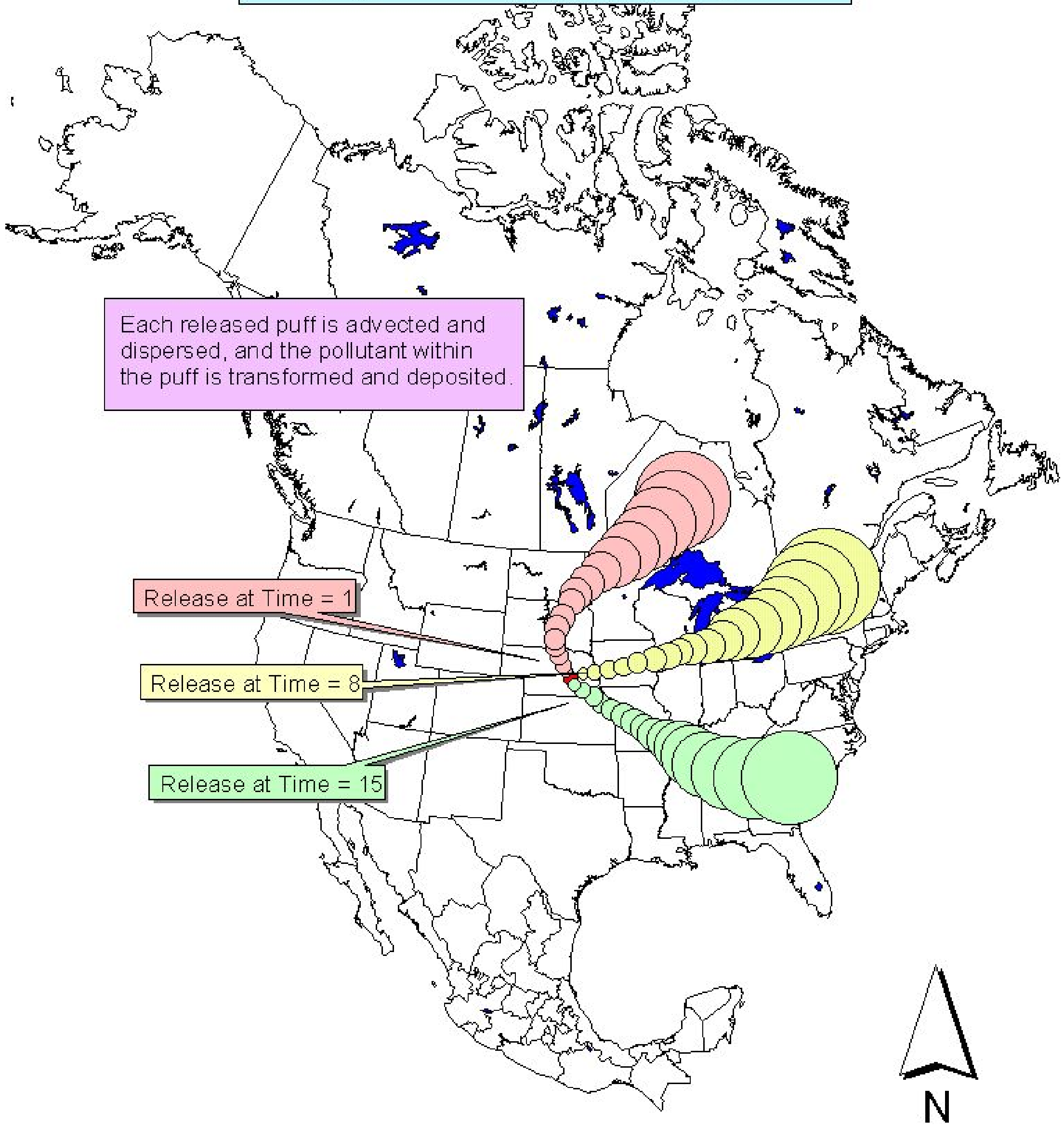

Atmospheric Mercury Fate Processes
Elemental Mercury [Hg(0)]
Hg(II), ionic mercury, RGM
Particulate Mercury [Hg(p)]
Upper atmospheric halogen-mediated heterogeneous oxidation?
Polar sunrise "mercury depletion events"
Br
cloud
CLOUD DROPLET
Vapor phase:
Hg(0) oxidized to RGM and Hg(p) by O_3, H_2O_2, Cl_2, OH, HCl
Hg(p)
Hg(II) *reduced* to Hg(0) by SO_2 and sunlight
Adsorption/ desorption of Hg(II) to /from soot
Hg(0) *oxidized* to dissolved Hg(II) species by O_3, OH, HOCl, OCl^-
Primary Anthropogenic Emissions
Multi-media interface
Wet deposition
Dry deposition
Natural emissions
Re-emission of previously deposited anthropogenic and natural mercury
4

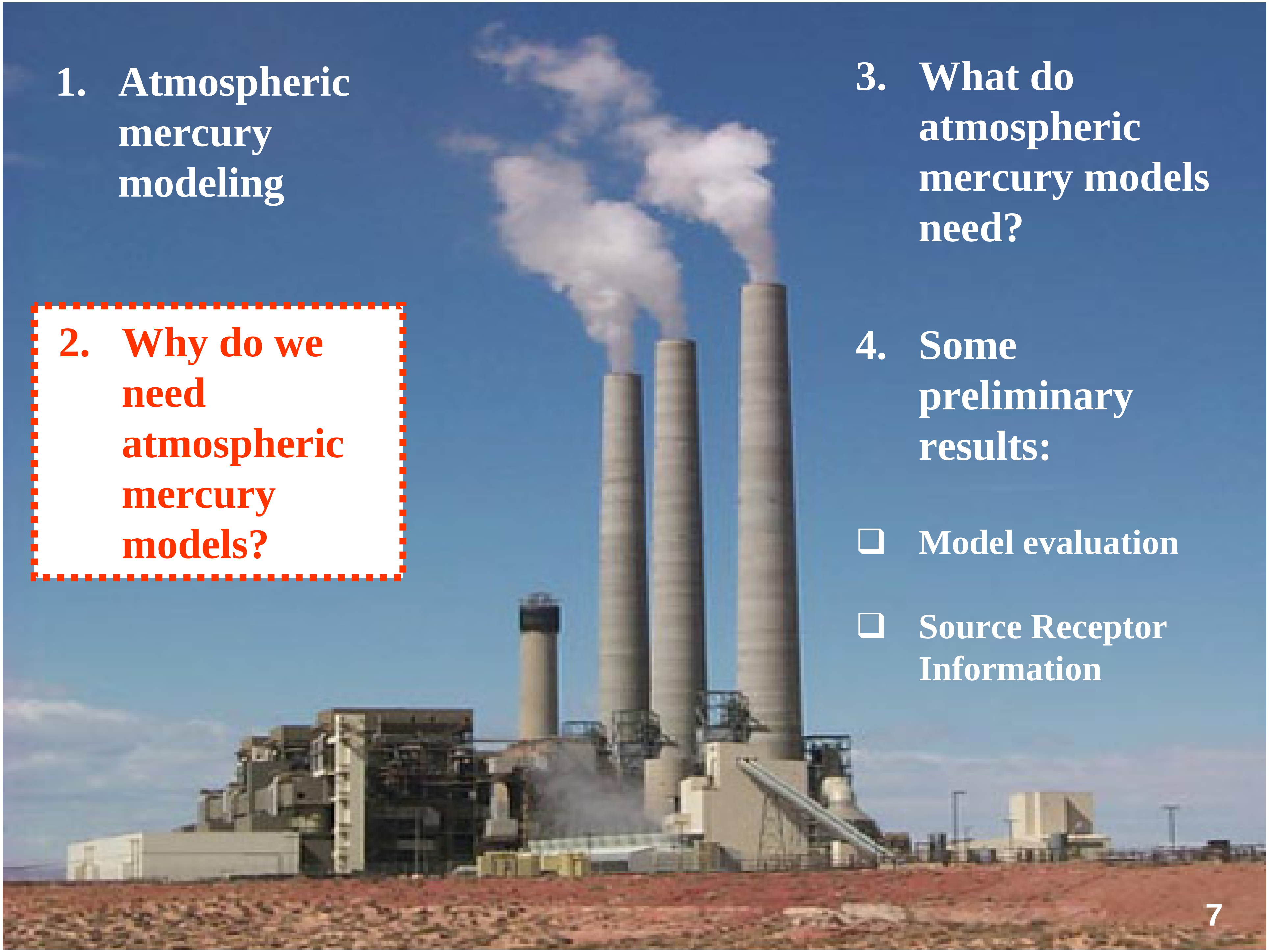
NOAA HYSPLIT MODEL

Lagrangian Puff Air Transport and Deposition Model



Over the entire modeling period (e.g., one year), puffs are released at periodic intervals (e.g., once every 7 hours).





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Why do we need atmospheric mercury models?

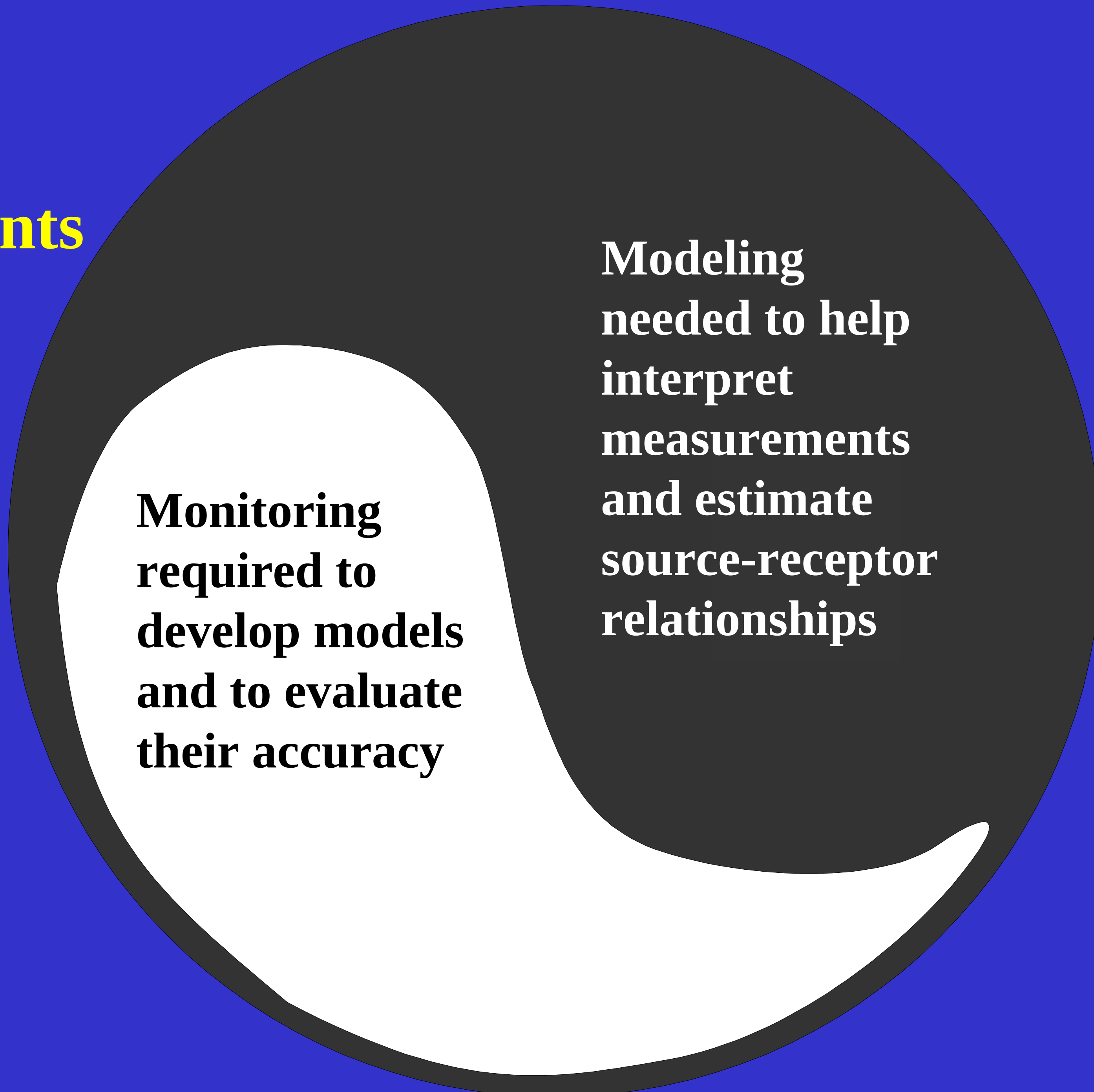
- to get *comprehensive source attribution* information --- **we don't just want to know how much is depositing at any given location, we also want to know where it came from…**

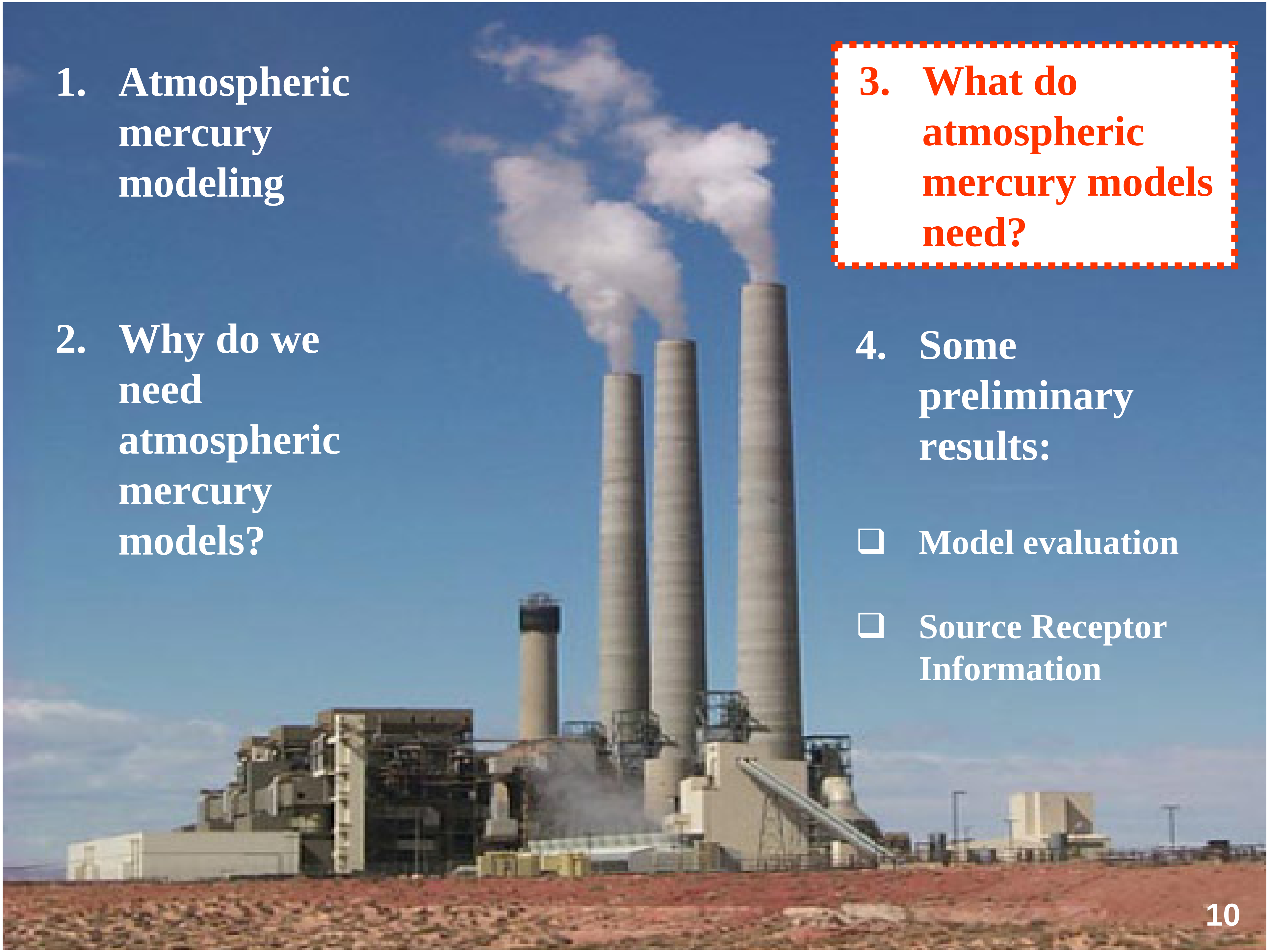
- to estimate *deposition over large regions,* …because deposition fields are highly spatially variable, and one can't measure everywhere all the time…

- to estimate *dry deposition*

- to evaluate *potential consequences* of alternative future emissions scenarios

But models must have measurements





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2. Why do we need atmospheric mercury models?

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What do atmospheric mercury models need?

Emissions Inventories

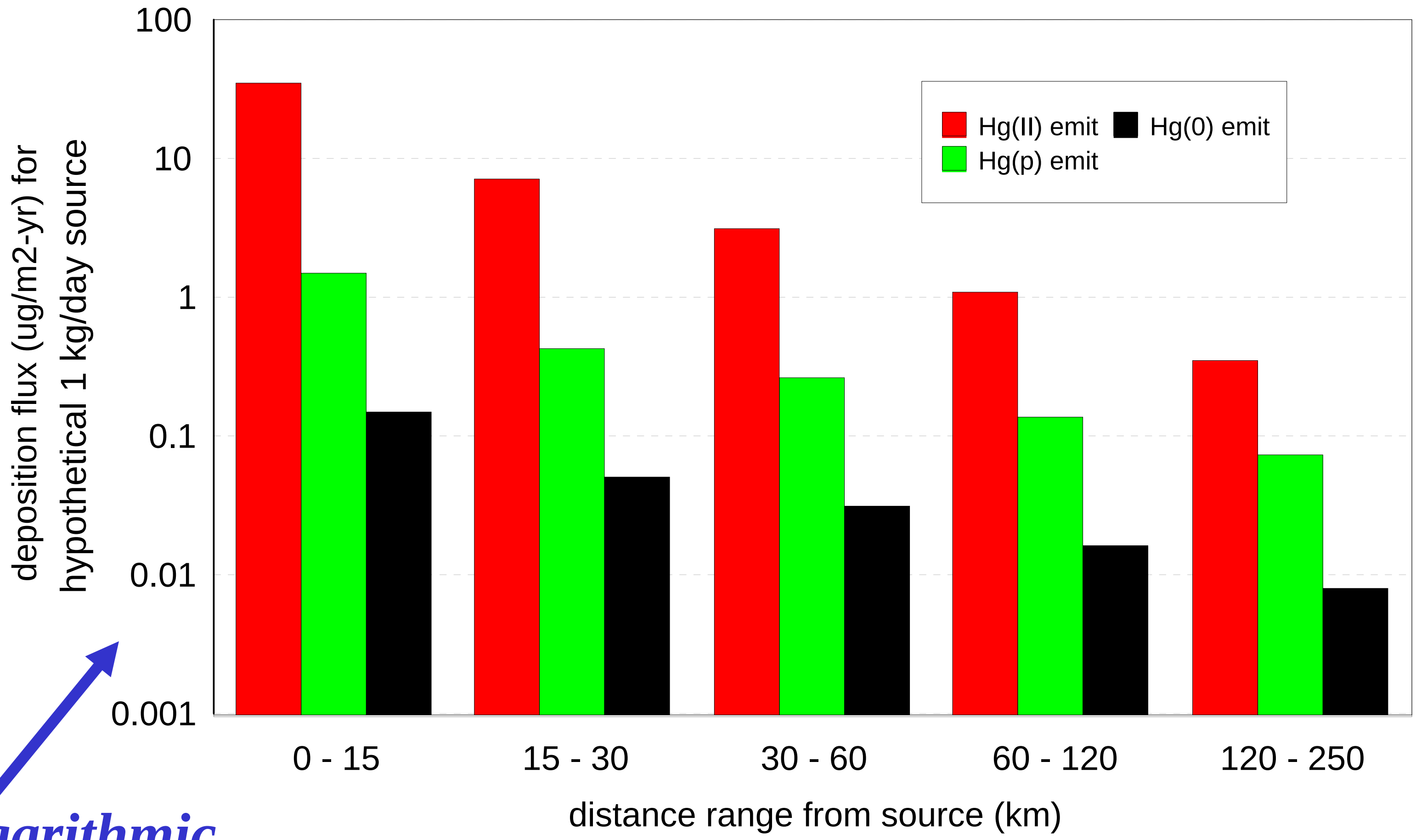
Meteorological Data

Scientific understanding of phase partitioning, atmospheric chemistry, and deposition processes

Ambient data for comprehensive model evaluation and improvement

	some challenges facing mercury modeling
emissions inventories	• need *all* sources • accurately divided into *different Hg forms* • **U.S. 1996, 1999, 2003** / **CAN 1995, 2000, 2005** • *temporal* variations (e.g. shut downs)
meteorological data	• precipitation not well characterized
scientific understanding	• what is RGM? what is Hg(p)? • accurate info for known reactions? • do we know all significant reactions? • natural emissions, re-emissions?
ambient data for model evaluation	• Mercury Deposition Network (MDN) is great, but: • also need RGM, Hg(p), and Hg(0) concentrations • also need data above the surface (e.g., from aircraft) • also need source-impacted sites (not just background)

Why is emissions speciation information critical?



Logarithmic

Hypothesized rapid reduction of Hg(II) in plumes?
If true, then dramatic impact on modeling results…

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Atmospheric Chemical Reaction Scheme for Mercury

Reaction	Rate	Units	Reference
GAS PHASE REACTIONS			
$Hg^0 + O_3 \rightarrow Hg(p)$	3.0E-20	cm^3/molec-sec	Hall (1995)
$Hg^0 + HCl \rightarrow HgCl_2$	1.0E-19	cm^3/molec-sec	Hall and Bloom (1993)
$Hg^0 + H_2O_2 \rightarrow Hg(p)$	8.5E-19	cm^3/molec-sec	Tokos et al. (1998) (upper limit based on experiments)
$Hg^0 + Cl_2 \rightarrow HgCl_2$	4.0E-18	cm^3/molec-sec	Calhoun and Prestbo (2001)
$Hg^0 + OHC \rightarrow Hg(p)$	8.7E-14	cm^3/molec-sec	Sommar et al. (2001)
AQUEOUS PHASE REACTIONS			
$Hg^0 + O_3 \rightarrow Hg^{+2}$	4.7E+7	$(molar-sec)^{-1}$	Munthe (1992)
$Hg^0 + OHC \rightarrow Hg^{+2}$	2.0E+9	$(molar-sec)^{-1}$	Lin and Pehkonen(1997)
$HgSO_3 \rightarrow Hg^0$	$T*e^{((31.971*T)-12595.0)/T)}$ [T = temperature (K)]	sec^{-1}	Van Loon et al. (2002)
$Hg(II) + HO_2C \rightarrow Hg^0$	~ 0	$(molar-sec)^{-1}$	Gardfeldt & Jonnson (2003)
$Hg^0 + HOCl \rightarrow Hg^{+2}$	2.1E+6	$(molar-sec)^{-1}$	Lin and Pehkonen(1998)
$Hg^0 + OCl^{-1} \rightarrow Hg^{+2}$	2.0E+6	$(molar-sec)^{-1}$	Lin and Pehkonen(1998)
$Hg(II) \leftrightarrow Hg(II)_{(soot)}$	9.0E+2	liters/gram; t = 1/hour	eqlbrm: Seigneur et al. (1998) rate: Bullock & Brehme (2002).
$Hg^{+2} + h< \rightarrow Hg^0$	6.0E-7	$(sec)^{-1}$ (maximum)	Xiao et al. (1994); Bullock and Brehme (2002)

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- **Data availability**
- **Simple vs. Complex Measurements**

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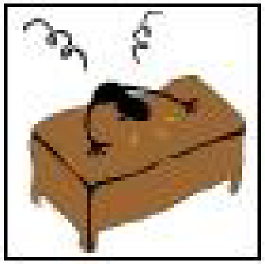
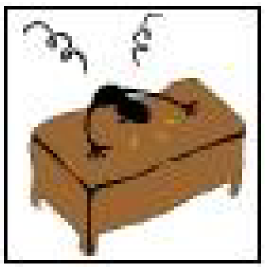
Data availability



A major impediment to evaluating and improving atmospheric Hg models has been the lack of speciated Hg air concentration data



There have been very few measurements to date, and these data are rarely made available in a practical way (timely, complete, etc.)

The data being collected at Piney Reservoir could be extremely helpful!

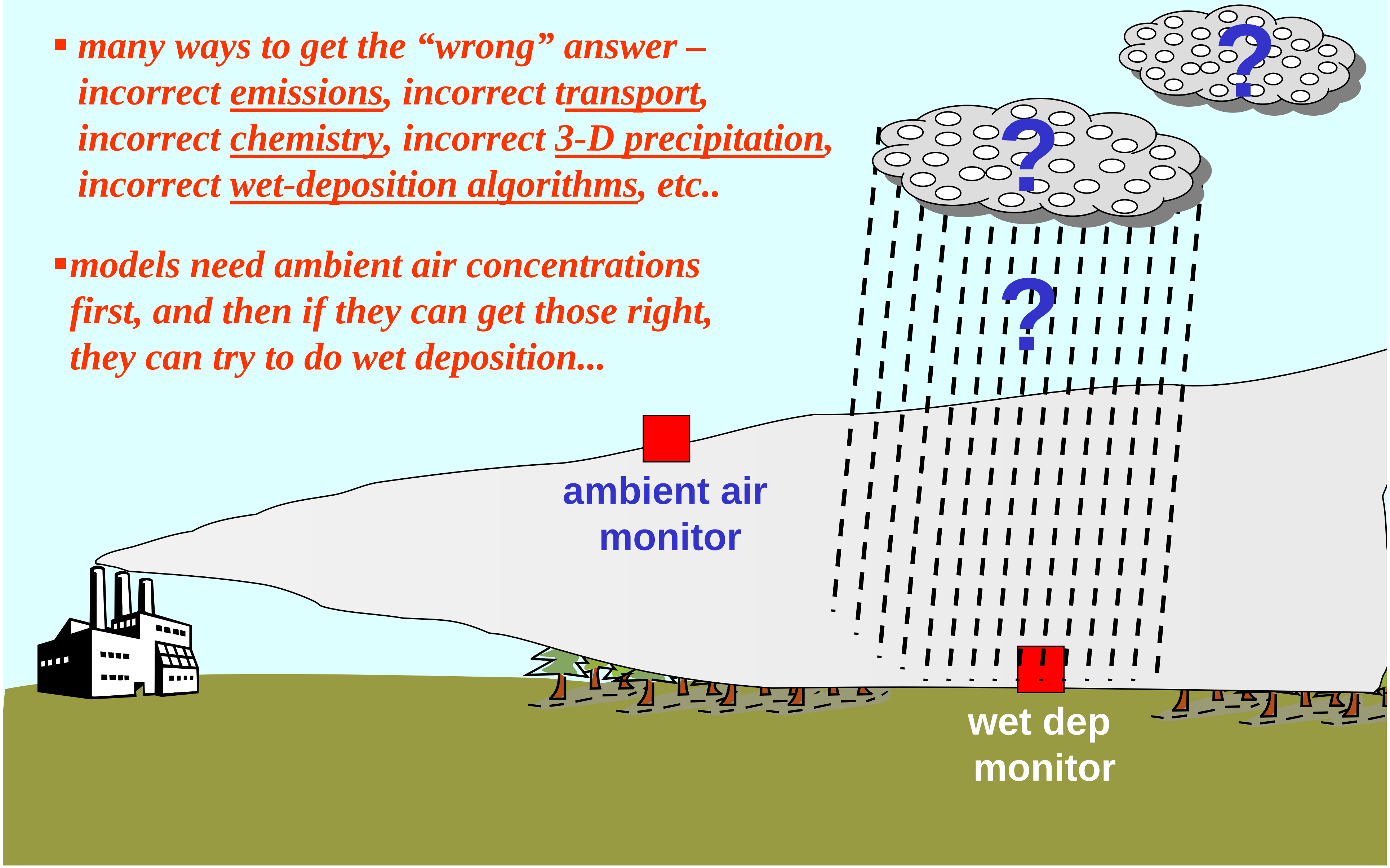
Some Additional Measurement Issues (from a modeler's perspective)

- **Data availability**
- **Simple vs. Complex Measurements**

Simple vs. Complex Measurements:
1. Wet deposition is a very complicated phenomena...

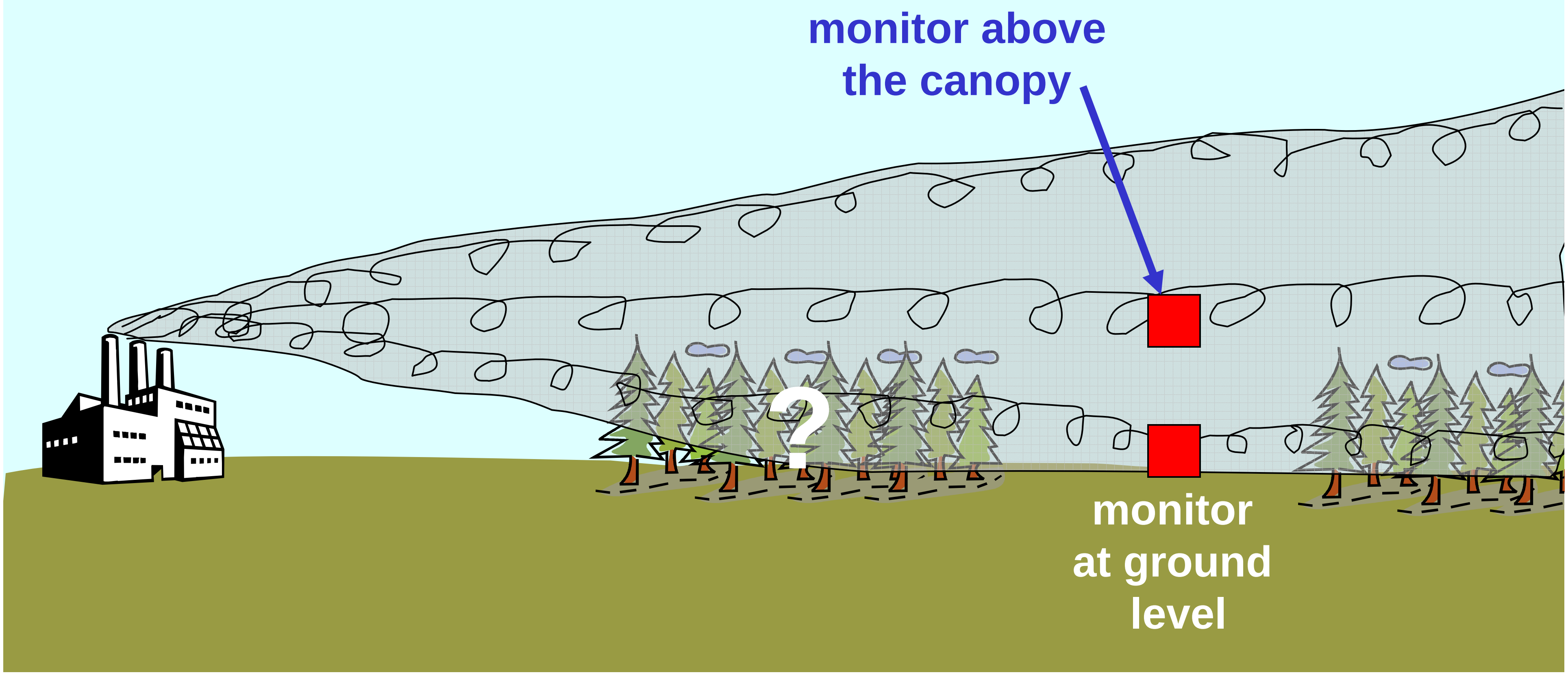
- *many ways to get the "wrong" answer – incorrect emissions, incorrect transport, incorrect chemistry, incorrect 3-D precipitation, incorrect wet-deposition algorithms, etc..*

- *models need ambient air concentrations first, and then if they can get those right, they can try to do wet deposition...*



Simple vs. Complex Measurements:
2. Potential complication with ground-level monitors...
(“fumigation”, “filtration”, etc.)...

- *atmospheric phenomena are complex and not well understood;*
- *models need “simple” measurements for diagnostic evaluations;*
- *ground-level data for rapidly depositing substances (e.g., RGM) hard to interpret*
- *elevated platforms might be more useful (at present level of understanding)*



Simple vs. Complex measurements - 3. Urban areas:

a. **Emissions inventory poorly known**
b. **Meteorology very complex (flow around buildings)**
c. **So, measurements in urban areas not particularly useful for current large-scale model evaluations**

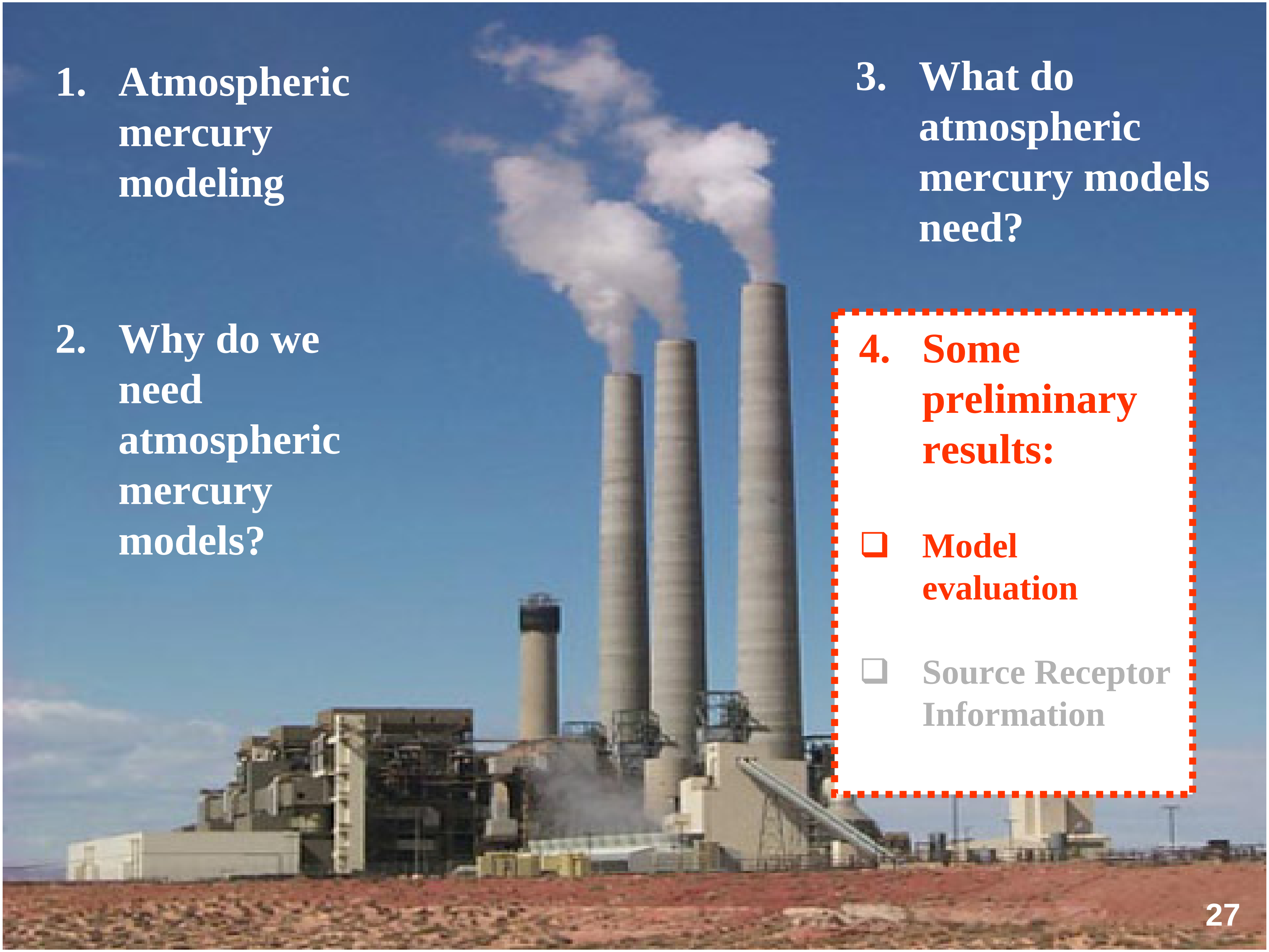
Simple vs. Complex Measurements – 4: extreme near-field measurements

- Sampling near intense sources?
- Must get the fine-scale met “perfect”

Ok, if one wants to develop hypotheses regarding *whether or not this is actually a source* of the pollutant *(and you can’t do a stack test for some reason!).*

Complex vs. Simple Measurements – 5: Need some source impacted measurements

- **Major questions regarding plume chemistry and near-field impacts (are there “hot spots”?)**
- **Most monitoring sites are designed to be “regional background” sites (e.g., most Mercury Deposition Network sites).**
- **We need some source-impacted sites as well to help resolve near-field questions**
- ***But not too close – maybe 20-30 km is ideal (?)***



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Participants

D. Syrakov ………………………………. **Bulgaria….NIMH**

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J. Munthe, I. Wängberg ………………... **Sweden….. IVL**

R. Bullock ………………………………**USA………EPA**

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A. Ryaboshapko, I. Ilyin, O.Travnikov…**EMEP……MSC-E**

Intercomparison Conducted in 3 Stages

I. **Comparison of chemical schemes for a cloud environment**

II. **Air Concentrations in Short Term Episodes**

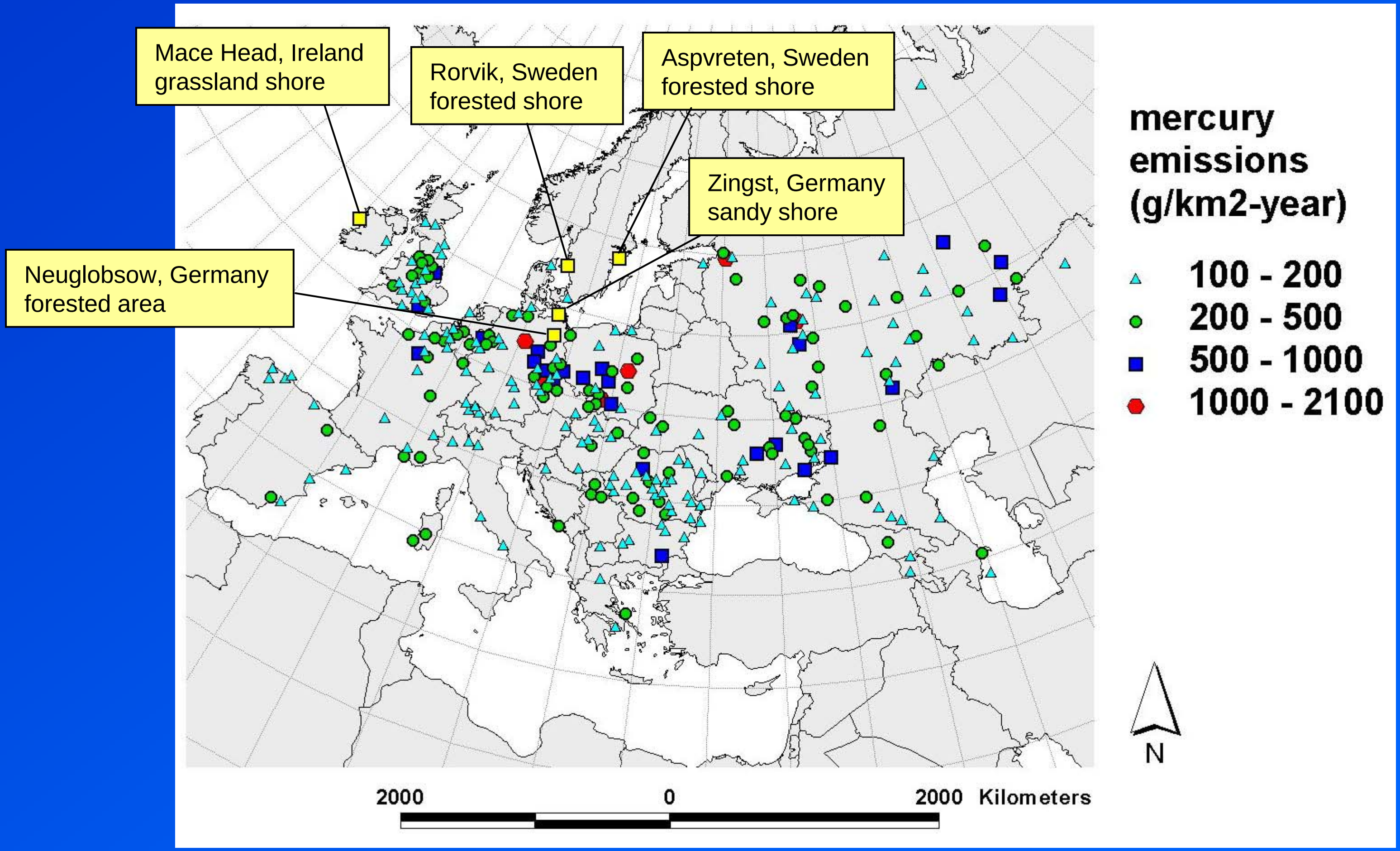
III. **Long-Term Deposition and Source-Receptor Budgets**

Participating Models

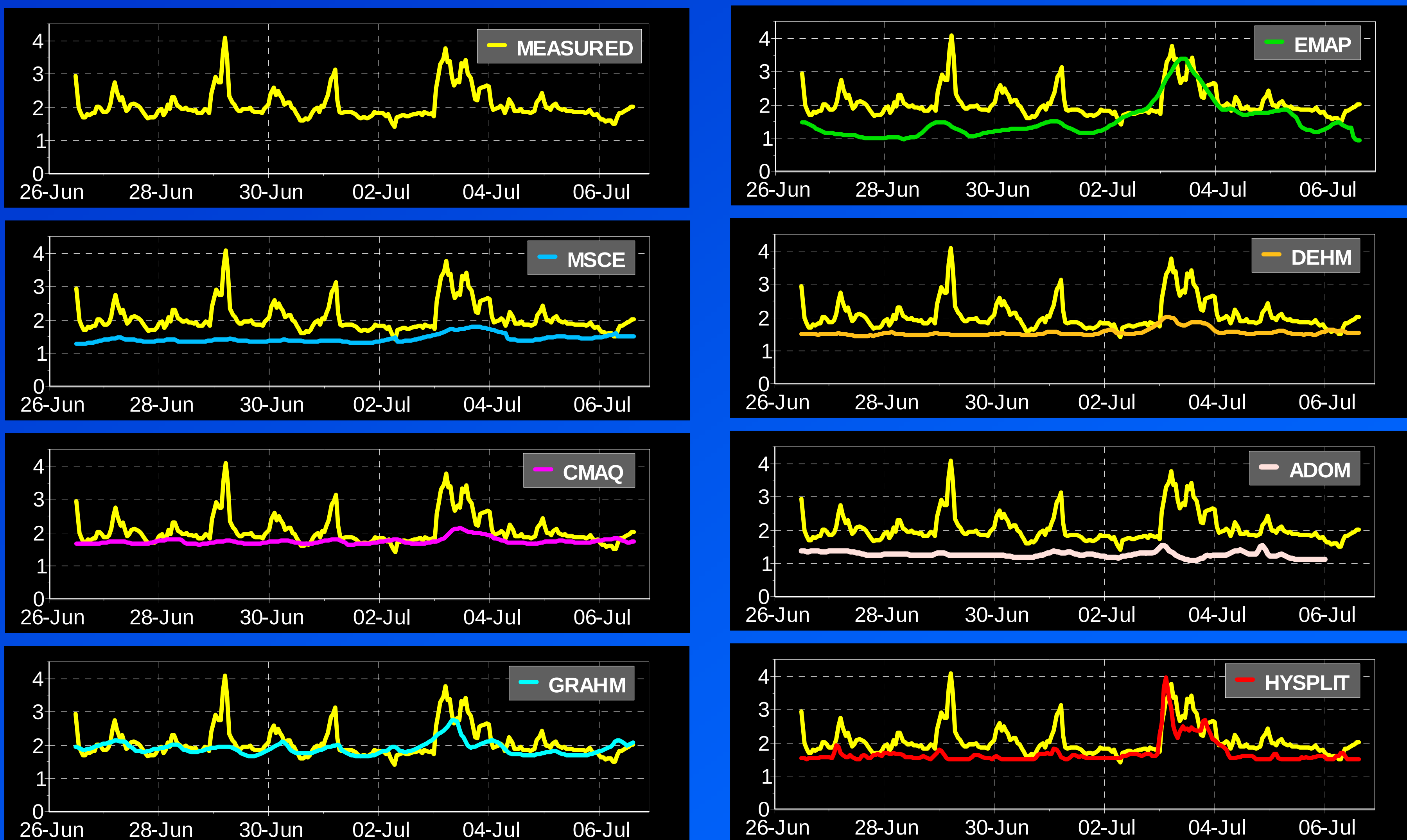
Model Acronym	***Model Name*** **and Institution**	**Stage**		
		I	**II**	**III**
CAM	*Chemistry of Atmos. Mercury model*, Environmental Institute, Sweden			
MCM	*Mercury Chemistry Model*, Atmos. & Environmental Research, USA			
CMAQ	*Community Multi-Scale Air Quality model*, US EPA			
ADOM	*Acid Deposition and Oxidants Model*, GKSS Research Center, Germany			
MSCE-HM	*MSC-E heavy metal regional model*, EMEP MSC-E			
GRAHM	*Global/Regional Atmospheric Heavy Metal model*, Environment Canada			
EMAP	*Eulerian Model for Air Pollution*, Bulgarian Meteo-service			
DEHM	*Danish Eulerian Hemispheric Model*, National Environmental Institute			
HYSPLIT	*Hybrid Single Particle Lagrangian Integrated Trajectory model*, US NOAA			
MSCE-HM-Hem	*MSC-E heavy metal hemispheric model*, EMEP MSC-E			

Anthropogenic Mercury Emissions Inventory and Monitoring Sites for Phase II

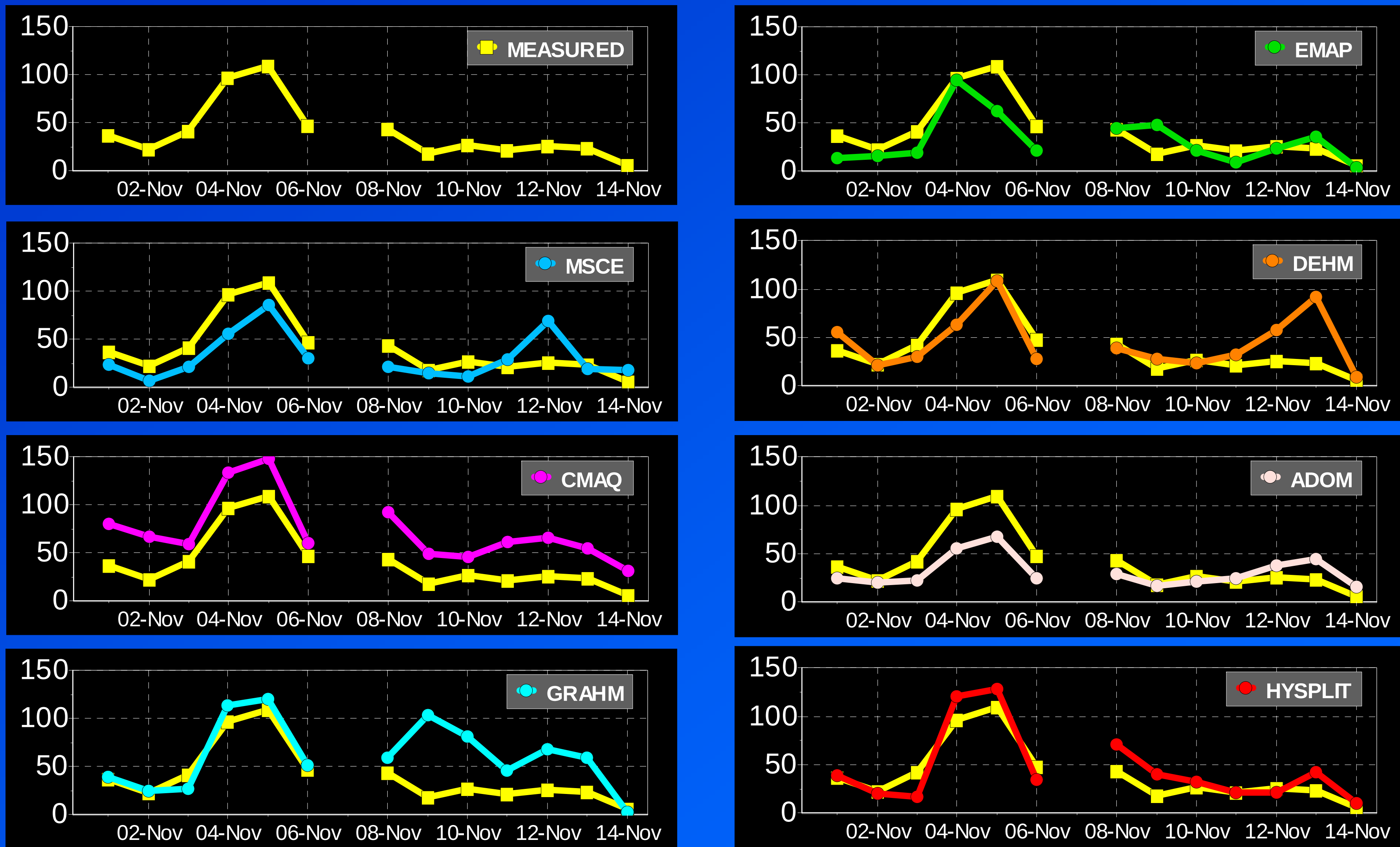
(note: only showing largest emitting grid cells)



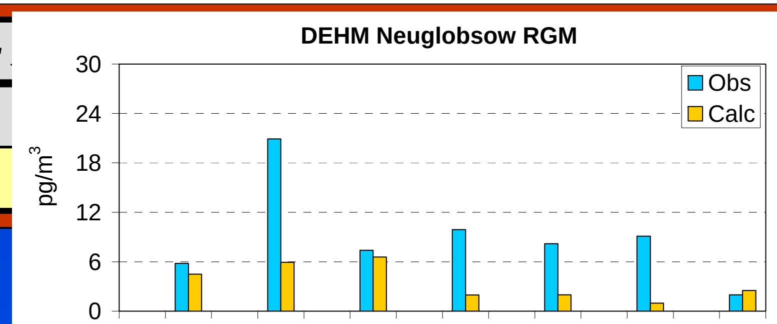
Total Gaseous Mercury (ng/m^3) at Neuglobsow: June 26 – July 6, 1995

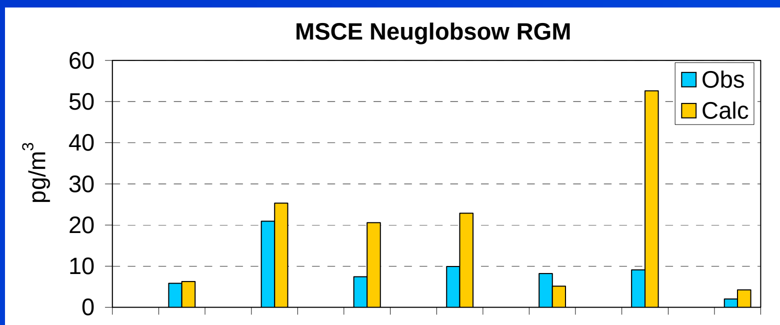


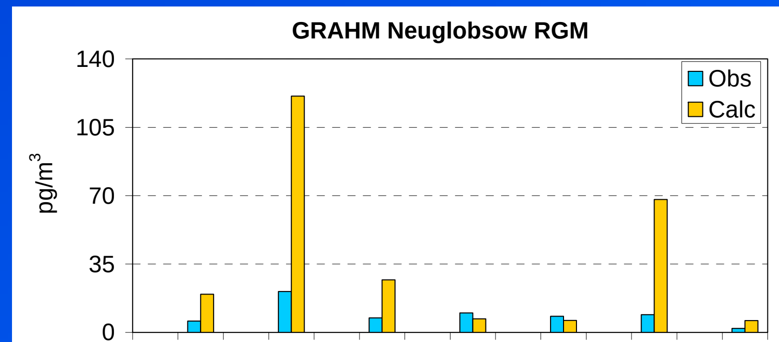
Total *Particulate* Mercury (pg/m^3) at Neuglobsow, Nov 1-14, 1999

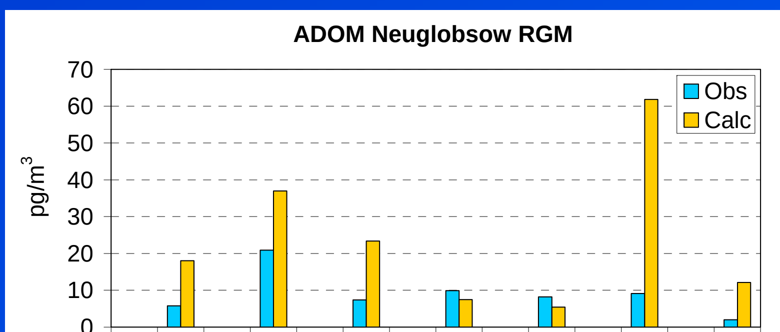


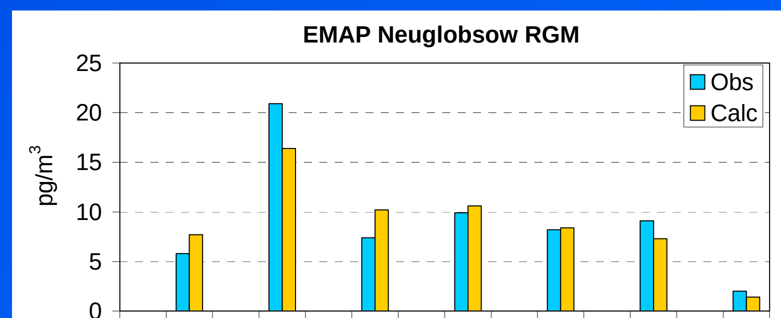
Reactive Gaseous Mercury at Neuglobsow, Nov 1-14, 1999

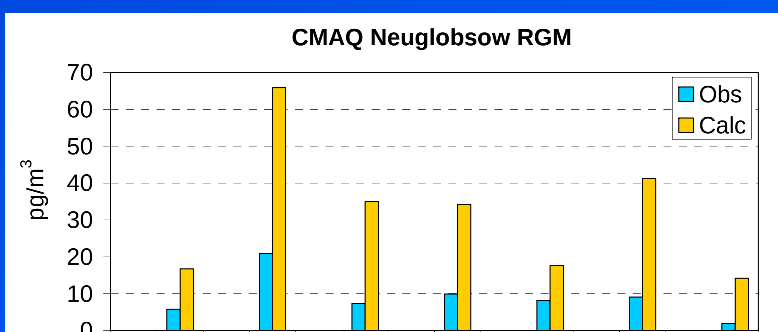


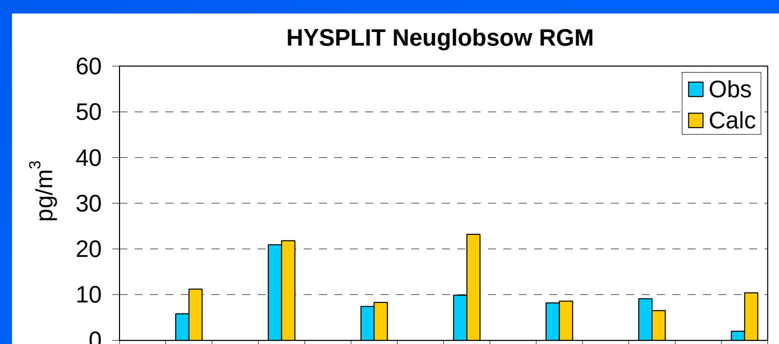


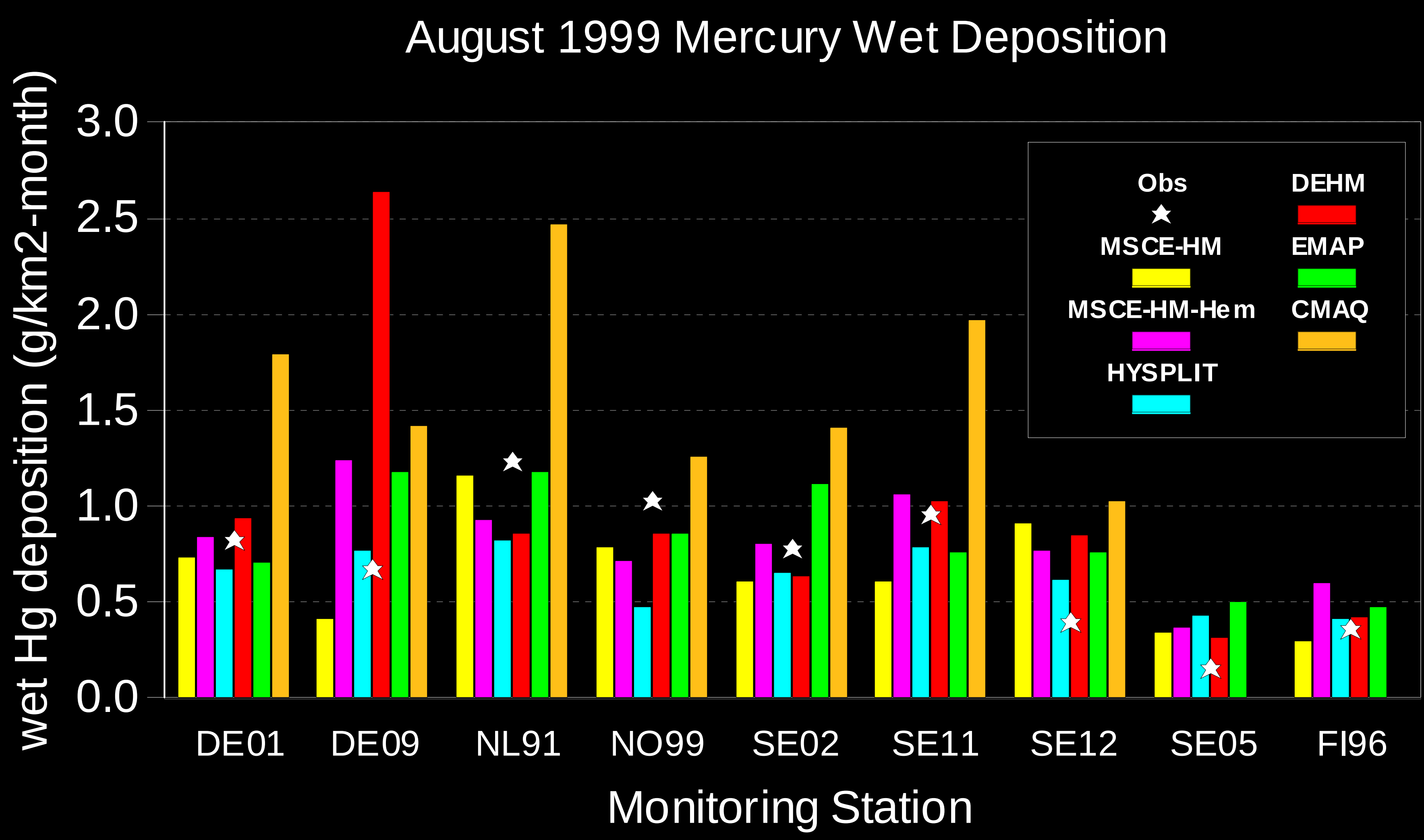


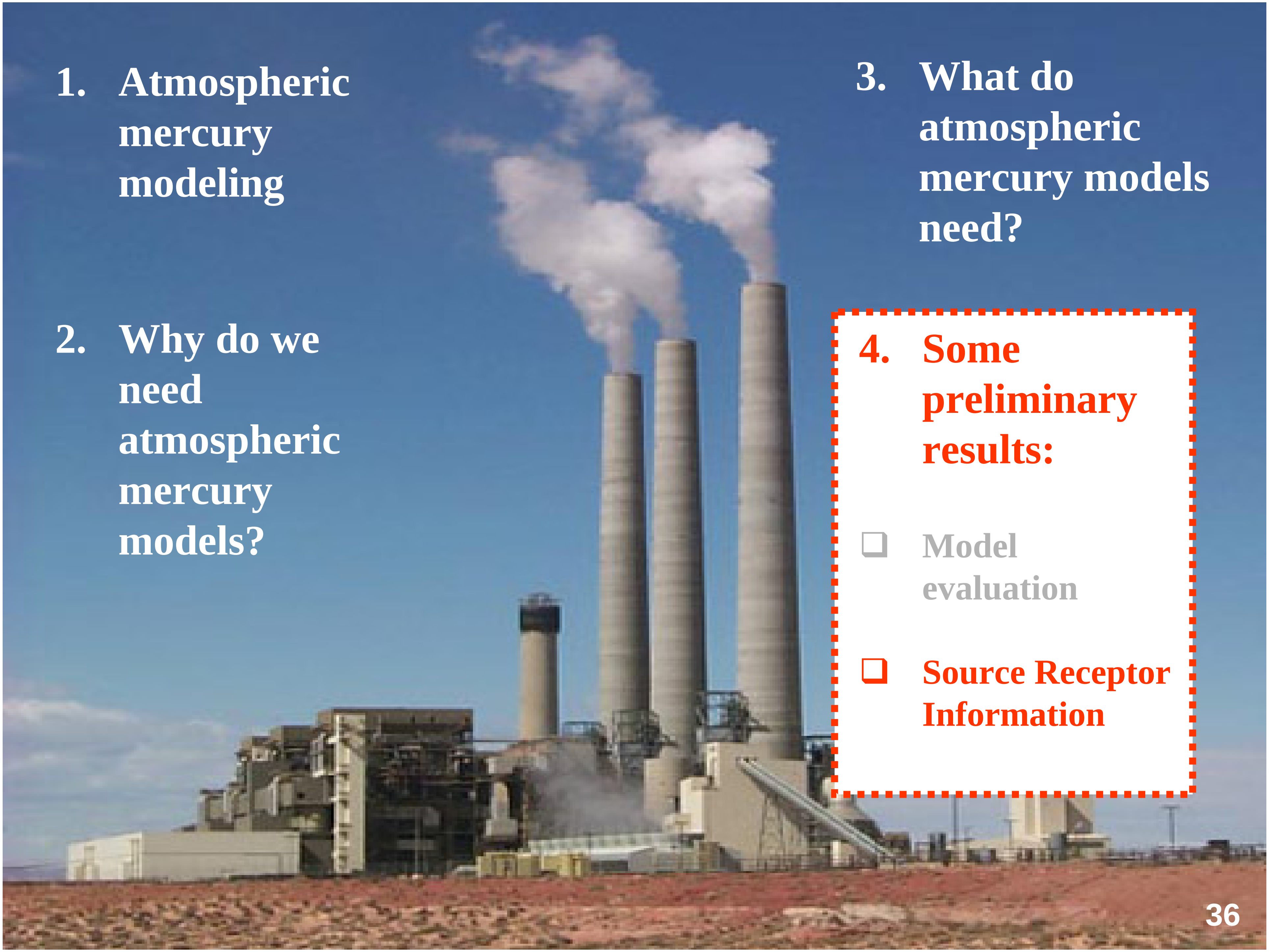









August 1999 Mercury Wet Deposition
wet Hg deposition (g/km2-month)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
Obs
DEHM
MSCE-HM
EMAP
MSCE-HM-Hem
CMAQ
HYSPLIT
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station



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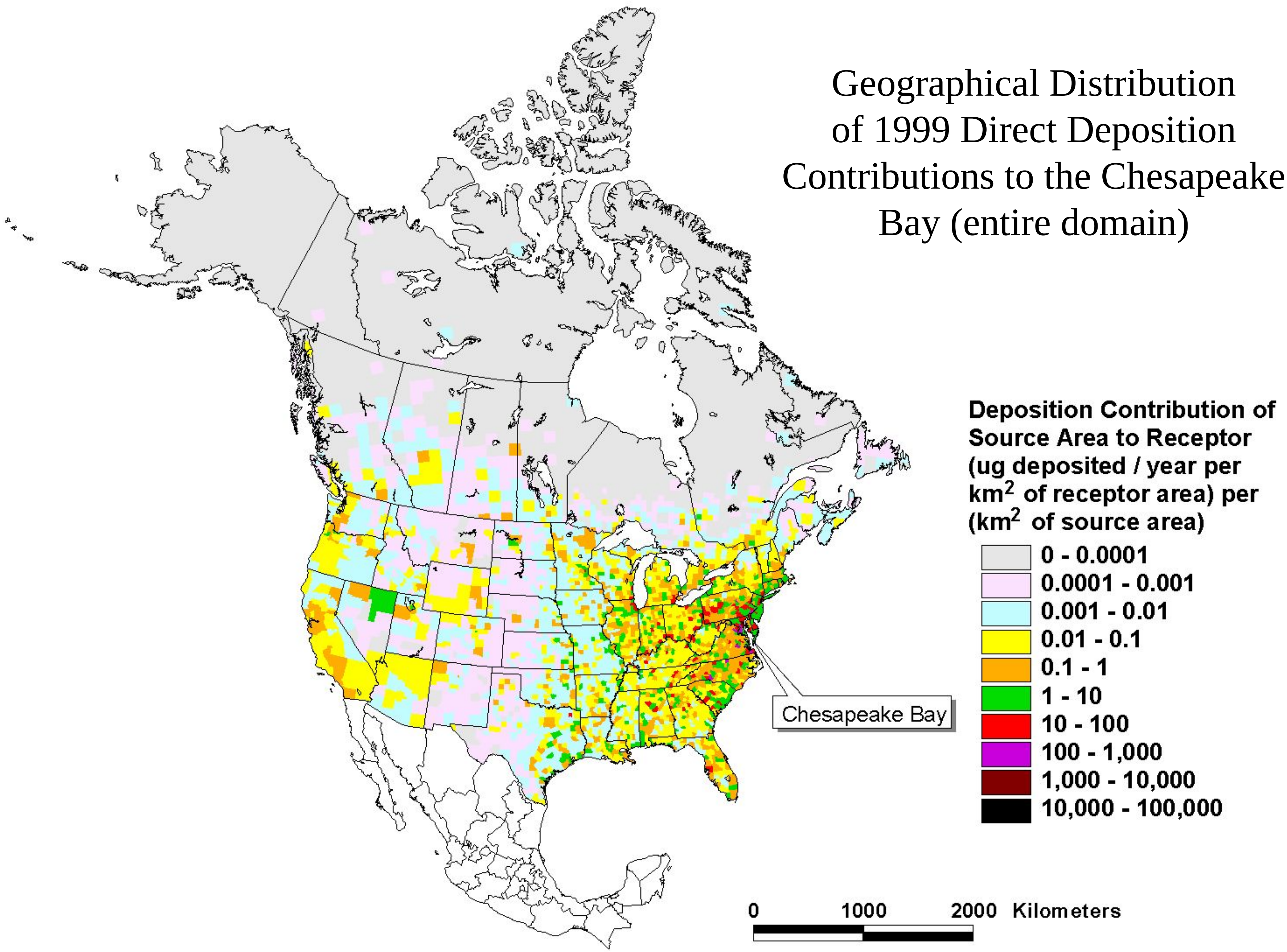
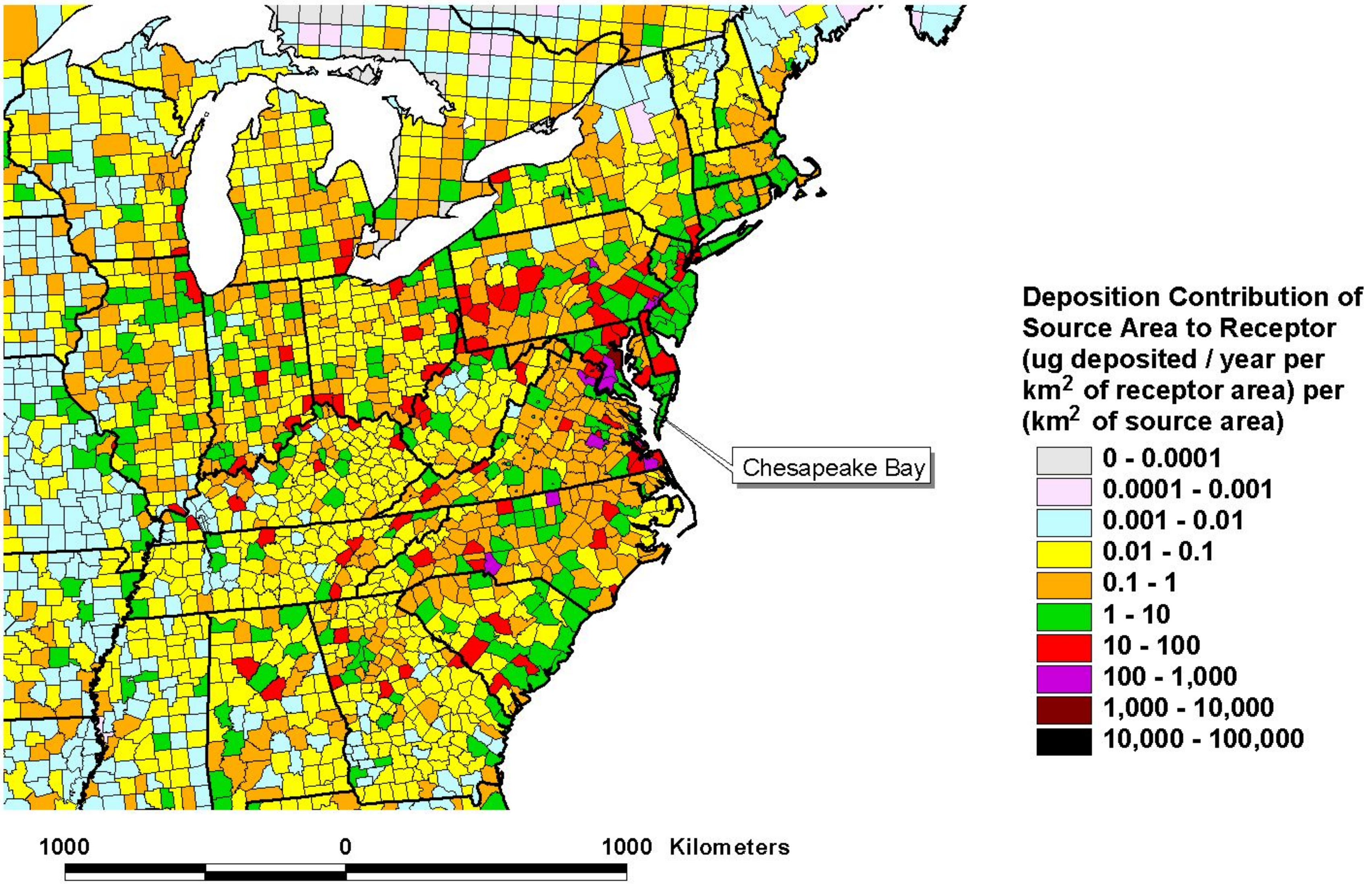
Example of Detailed Results: 1999 Results for Chesapeake Bay

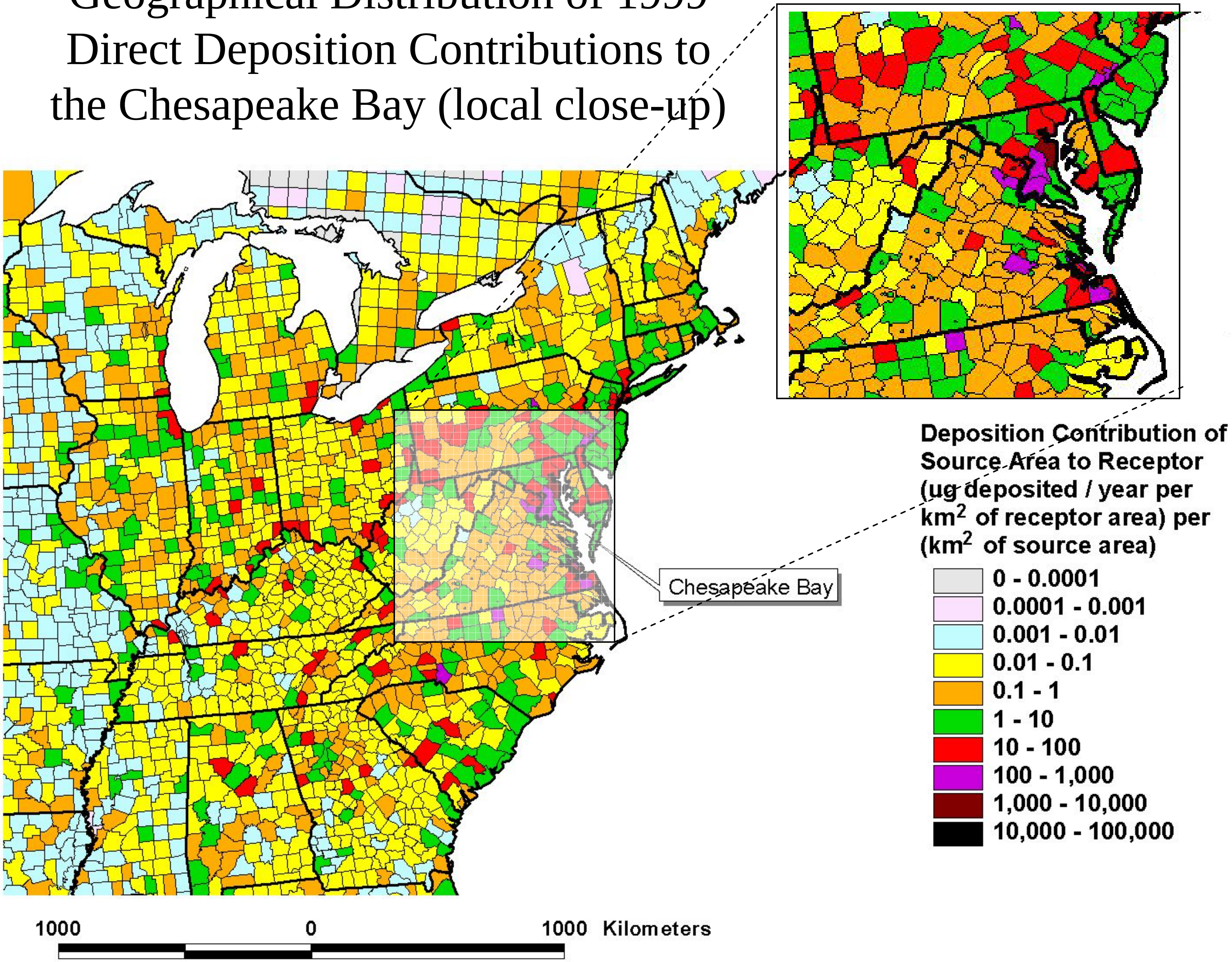
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (entire domain)



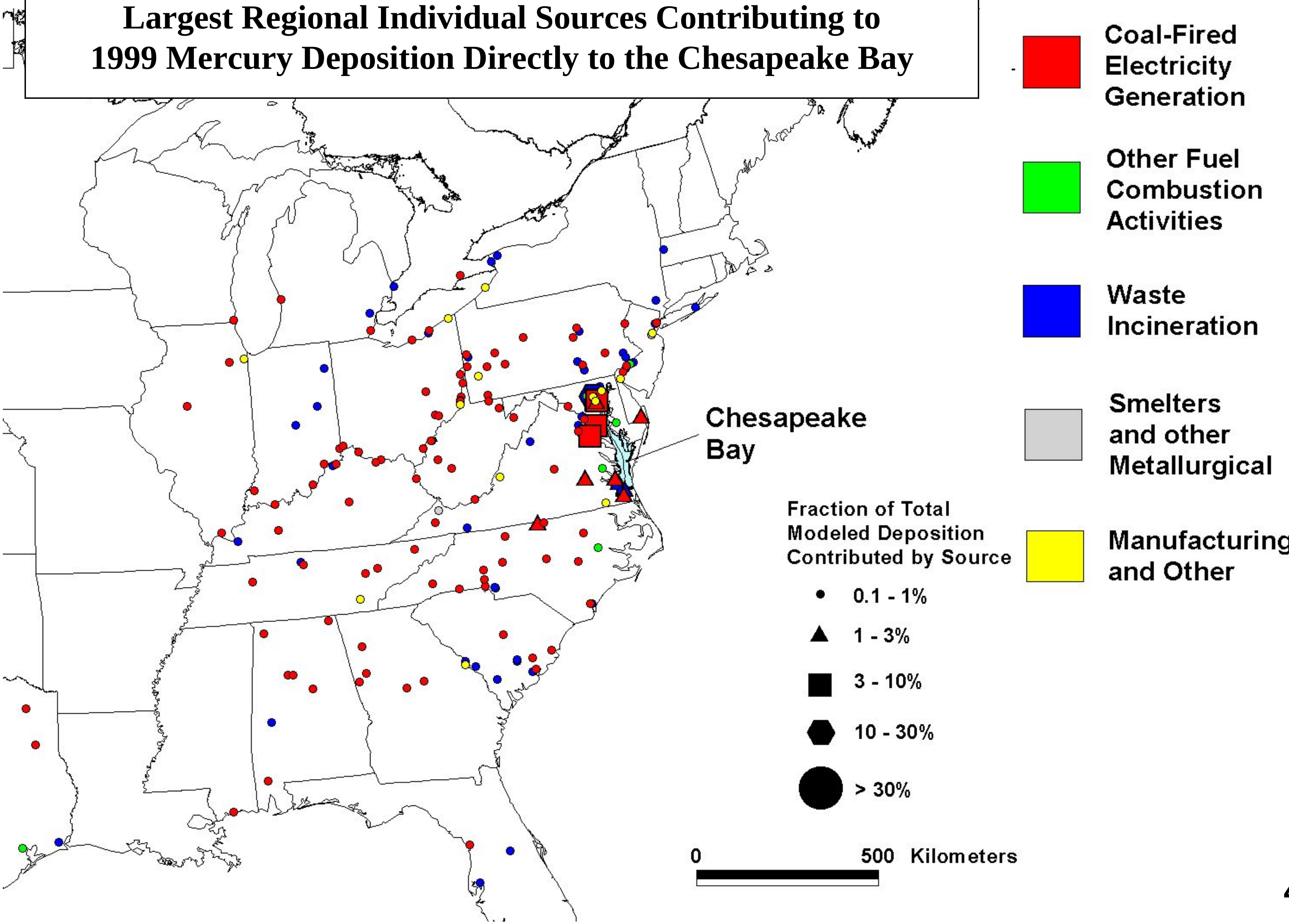
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (regional close-up)



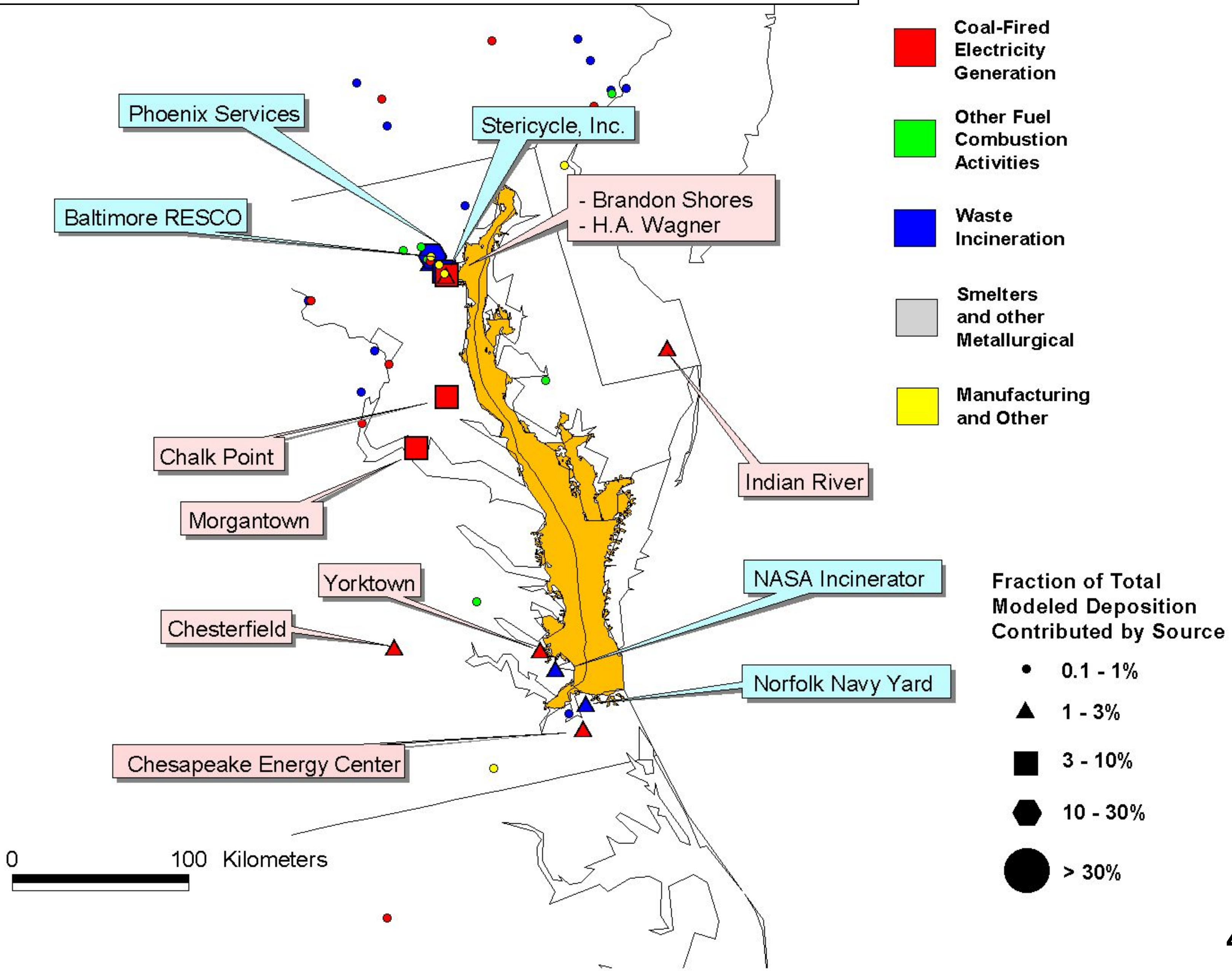
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (local close-up)



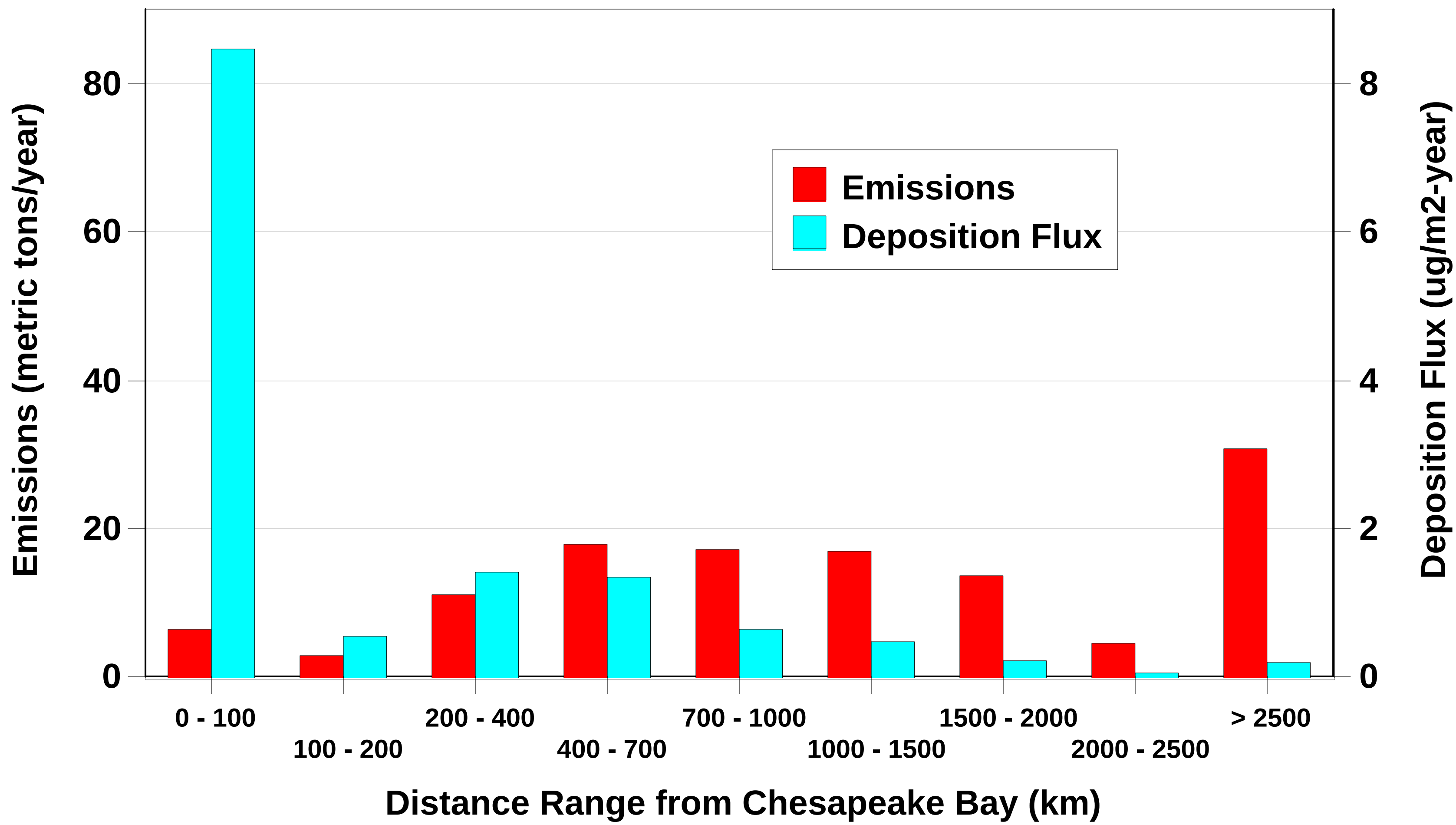
Largest Regional Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay



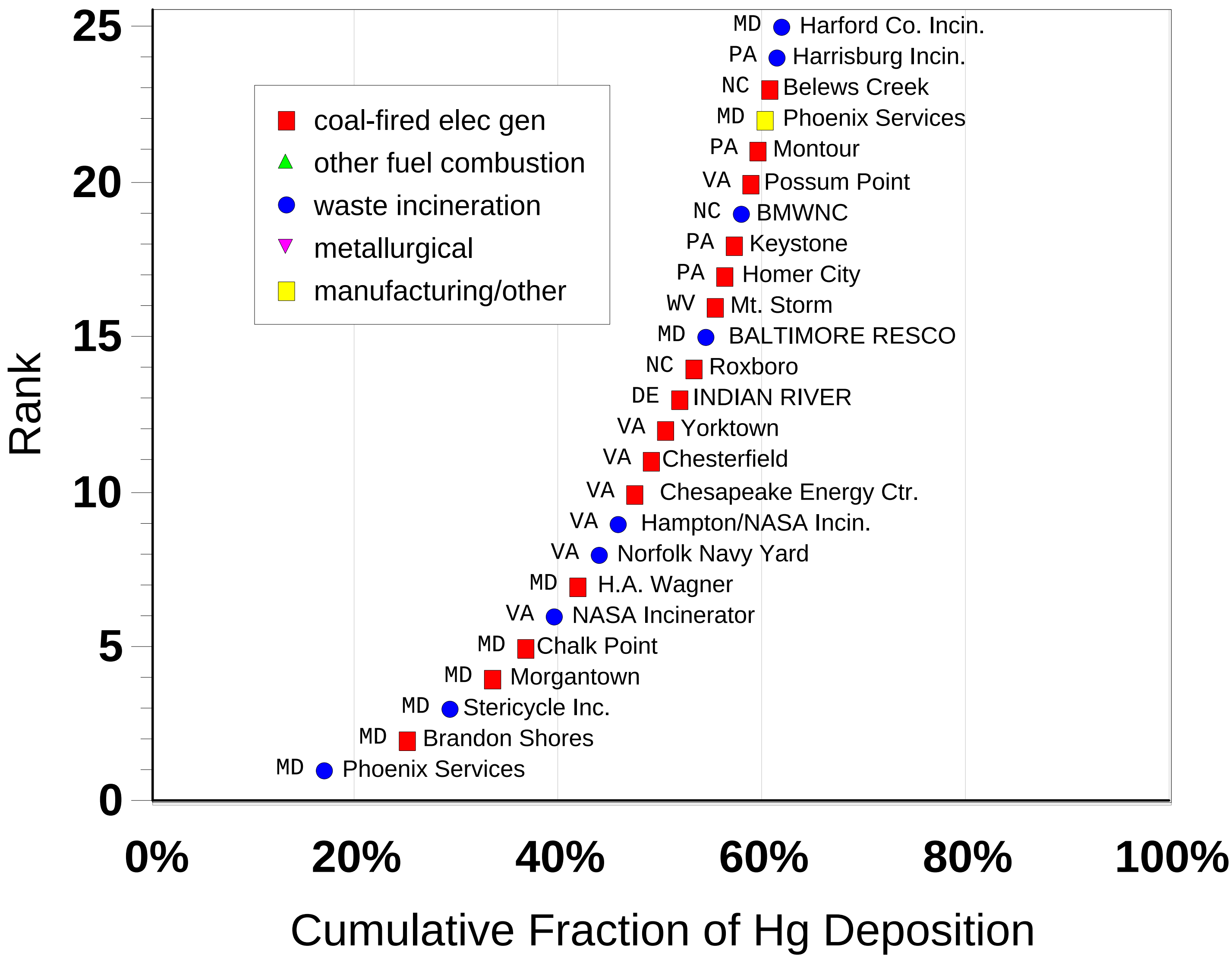
Largest Local Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay



Emissions and Direct Deposition Contributions from Different Distance Ranges Away From the Chesapeake Bay

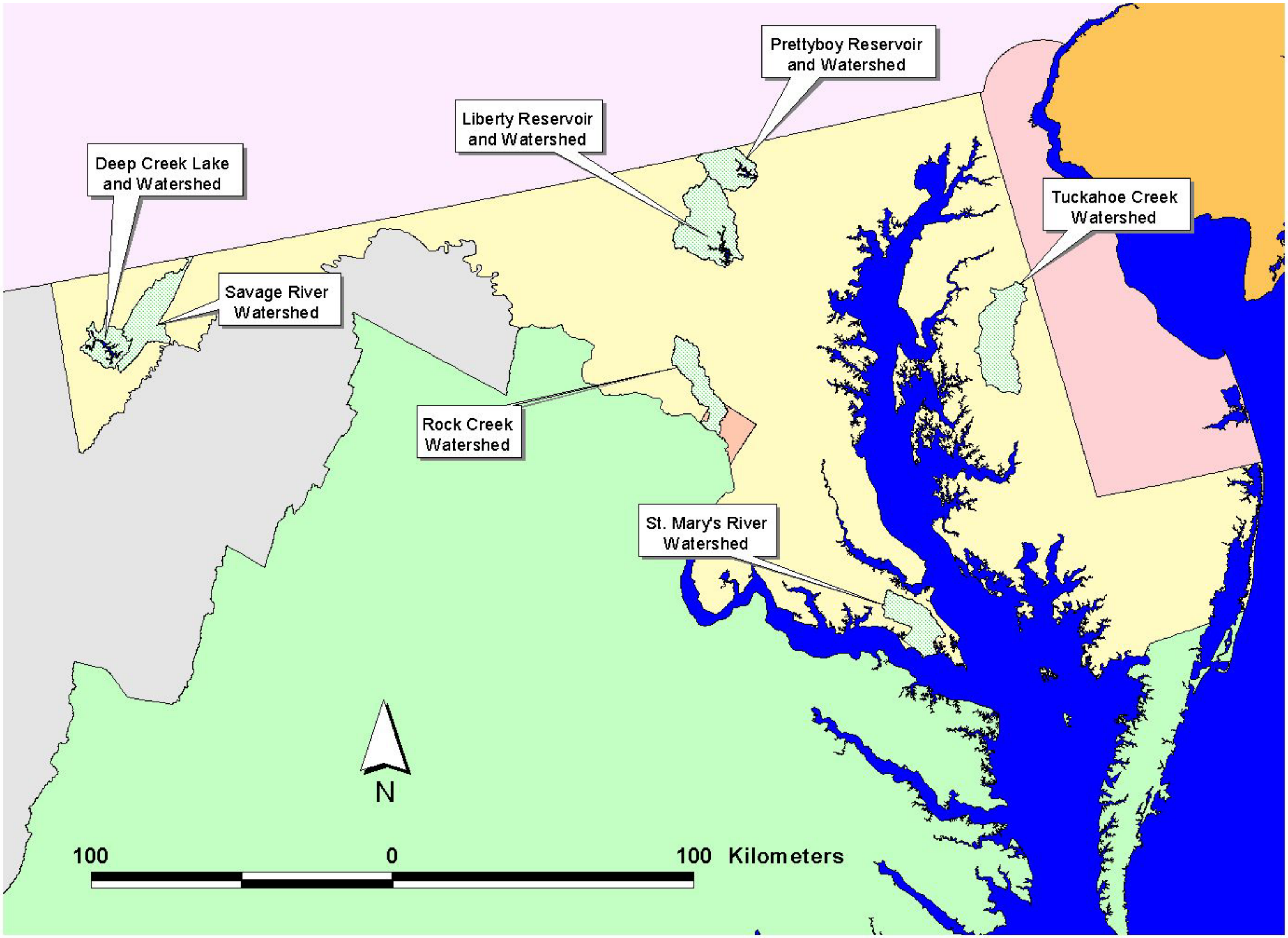


Top 25 Contributors to 1999 Hg Deposition Directly to the Chesapeake Bay

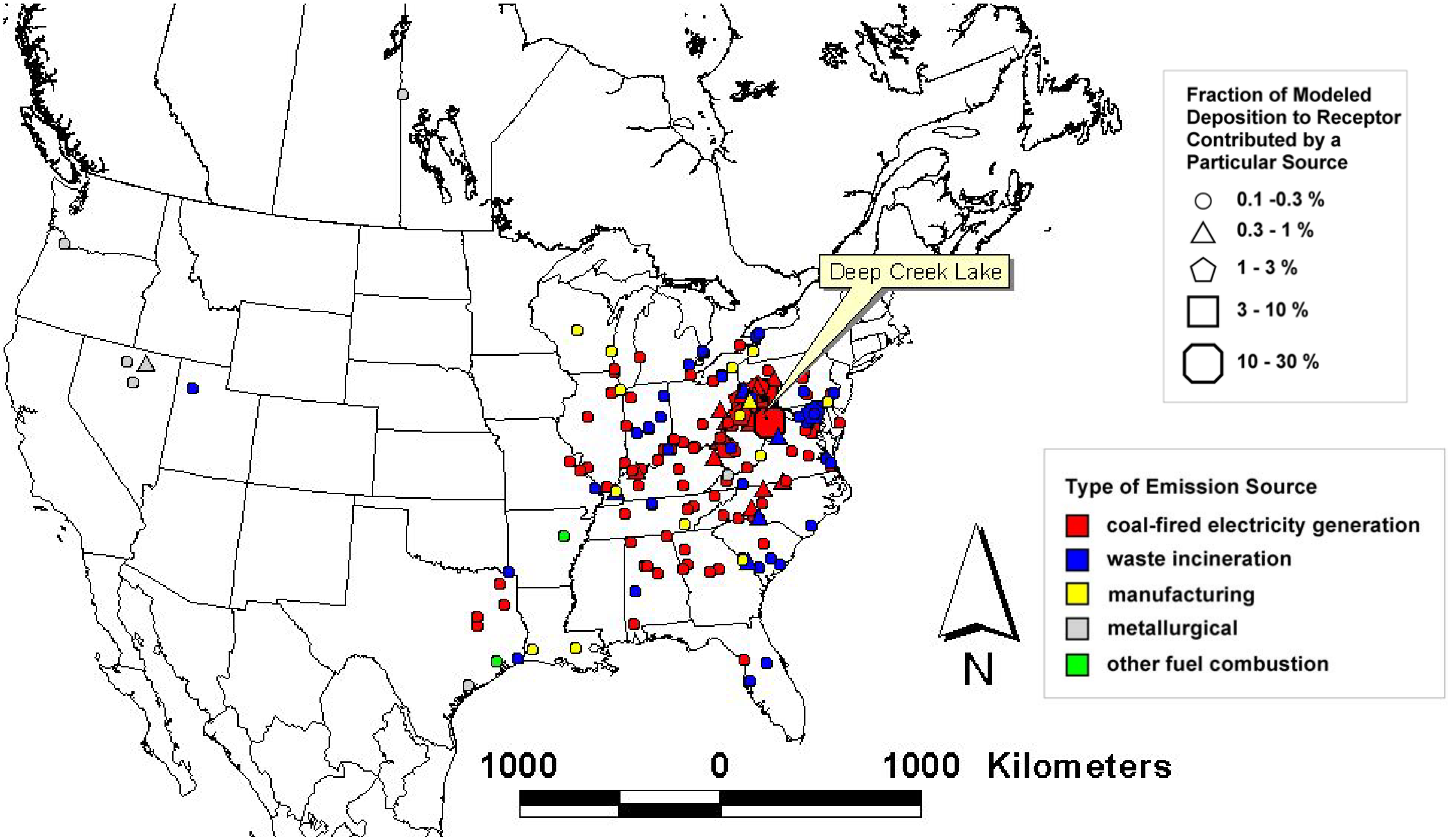


Preliminary Results for other Maryland Receptors

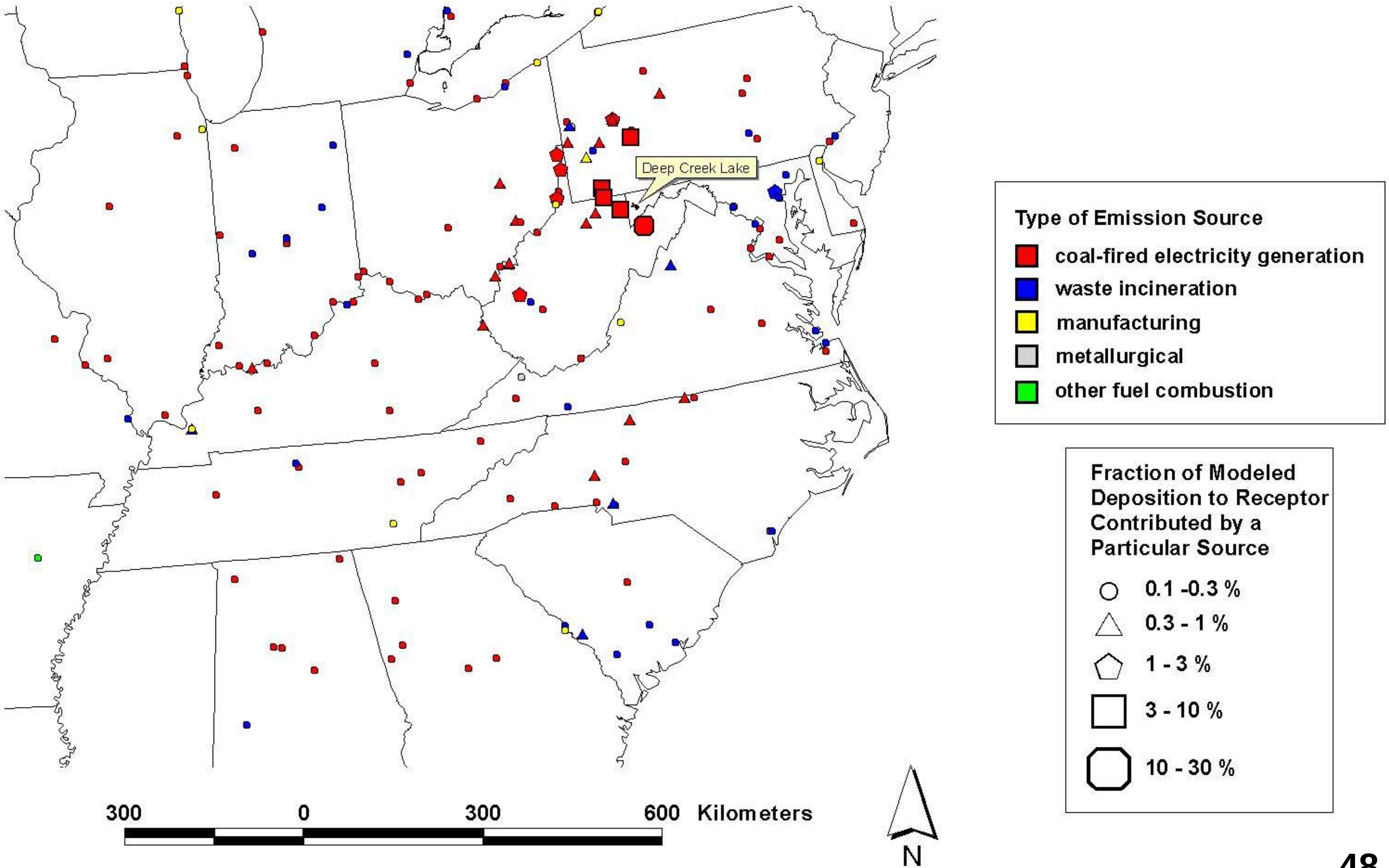
Maryland Receptors Included in Recent Preliminary HYSPLIT-Hg modeling *(but modeling was not optimized for these receptors!)*



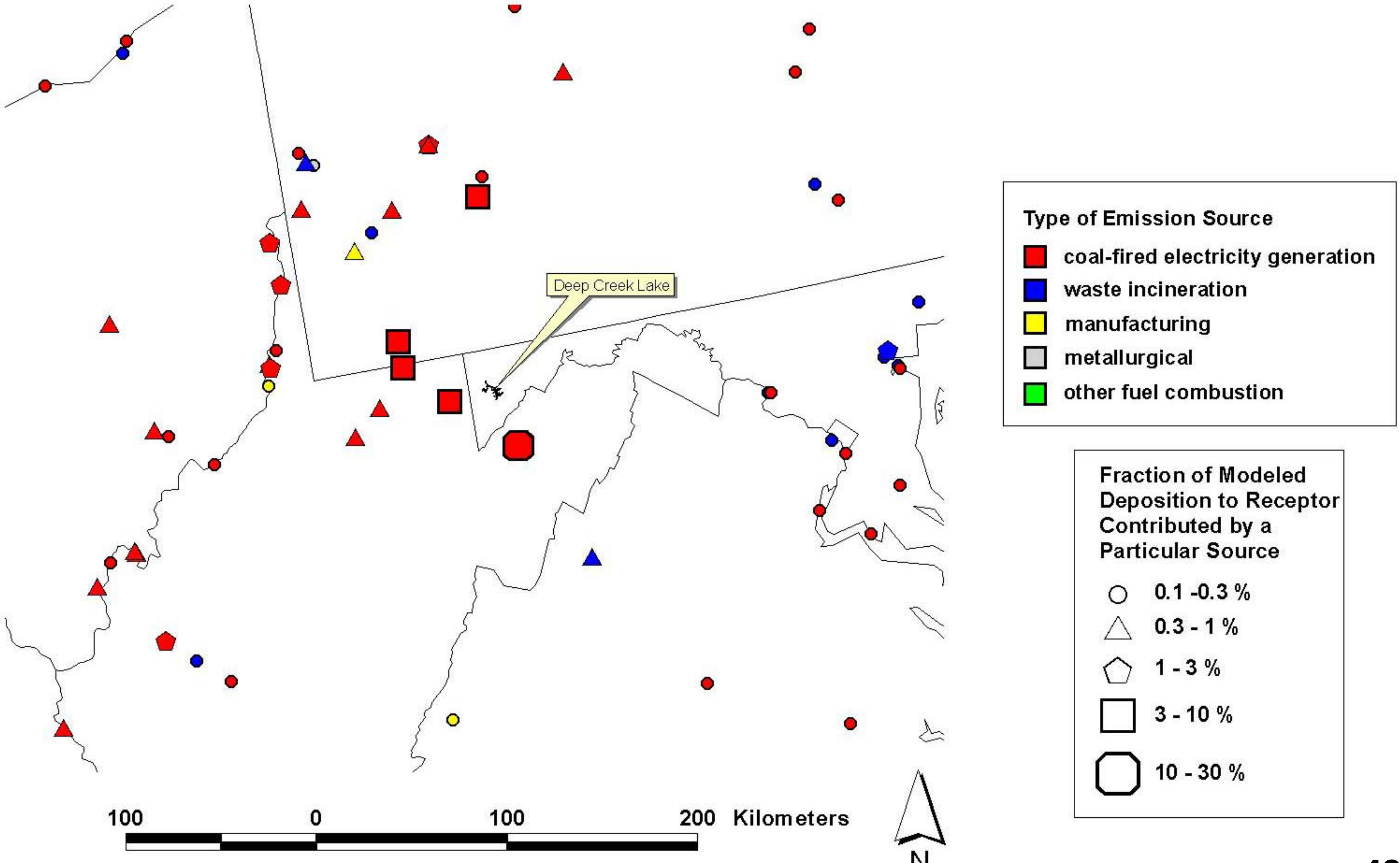
Largest Modeled Atmospheric Deposition Contributors Directly to Deep Creek Lake based on 1999 USEPA Emissions Inventory (national view)



Largest Modeled Atmospheric Deposition Contributors Directly to Deep Creek Lake based on 1999 USEPA Emissions Inventory (regional view)



Largest Modeled Atmospheric Deposition Contributors Directly to Deep Creek Lake based on 1999 USEPA Emissions Inventory (close-up view)



Some Next Steps

- Use more highly resolved meteorological data grids
- Expand model domain to include global sources
- Simulate natural emissions and re-emissions of previously deposited Hg
- Additional model evaluation exercises ... more sites, more time periods, more variables
- Sensitivity analyses and examination of atmospheric Hg chemistry (e.g. marine boundary layer, upper atmosphere)
- Dynamic linkage with ecosystem cycling models

Conclusions

- Models needed for source-receptor and other info
- At present, many model uncertainties & data limitations
- Monitoring data required to evaluate and improve models
- For this, simple may be better than complex measurements
- Some useful model results appear to be emerging
- Future is much brighter because of this coordination!

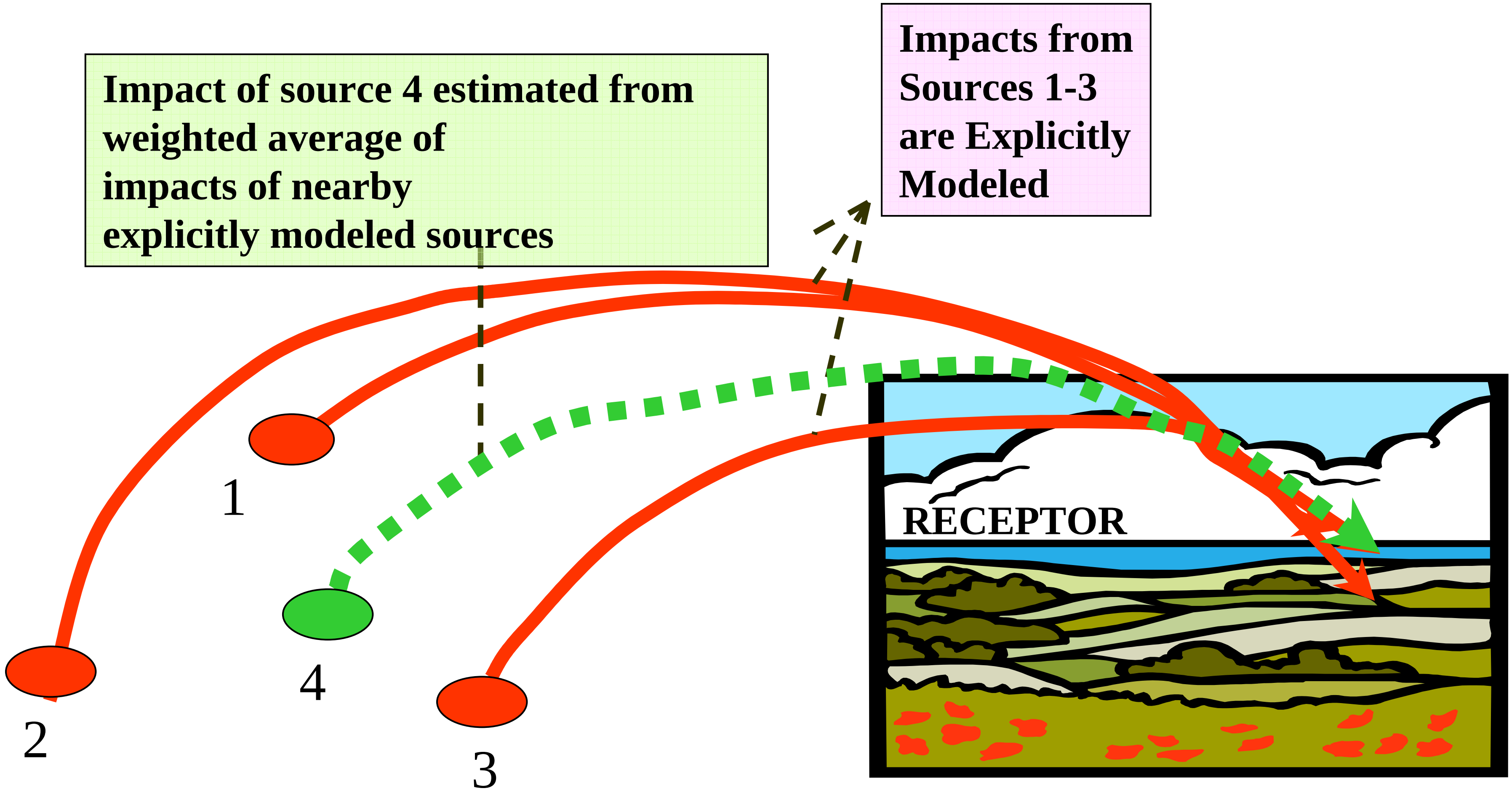
Thanks

EXTRA SLIDES

Why might the atmospheric fate of mercury emissions be essentially linearly independent?

- **Hg is present at extremely trace levels in the atmosphere**
- **Hg won't affect meteorology (can simulate meteorology independently, and provide results to drive model)**
- **Most species that complex or react with Hg are generally present at *much* higher concentrations than Hg**
- **Other species (e.g. OH) generally react with many other compounds than Hg, so while present in trace quantities, their concentrations cannot be strongly influenced by Hg**
- **Wet and dry deposition processes are generally 1st order with respect to Hg**
- **The current "consensus" chemical mechanism (equilibrium + reactions) does not contain any equations that are not 1st order in Hg**

Spatial interpolation

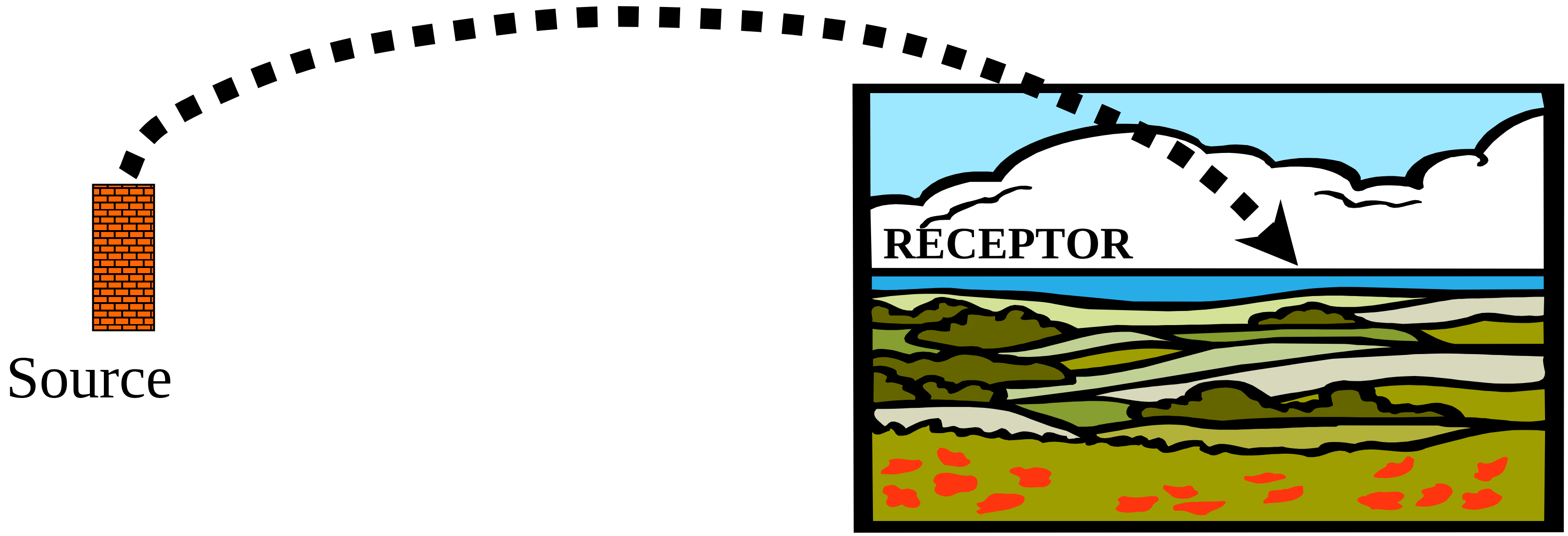


- **Perform separate simulations at each location for emissions of pure Hg(0), Hg(II) and Hg(p)**

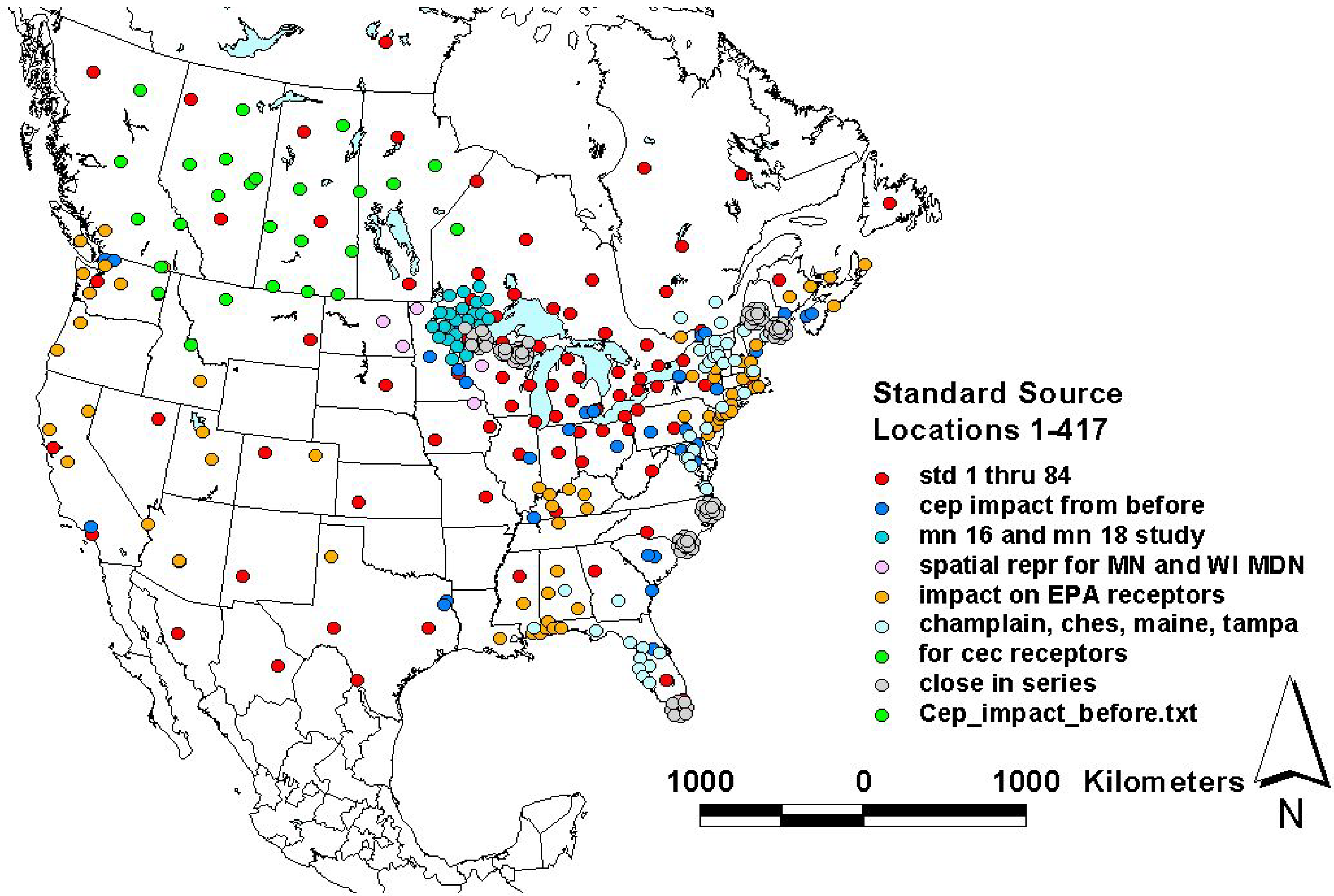
  [*after emission, simulate transformations between Hg forms*]

- **Impact of emissions mixture taken as a linear combination of impacts of pure component runs on any given receptor**

"Chemical Interpolation"

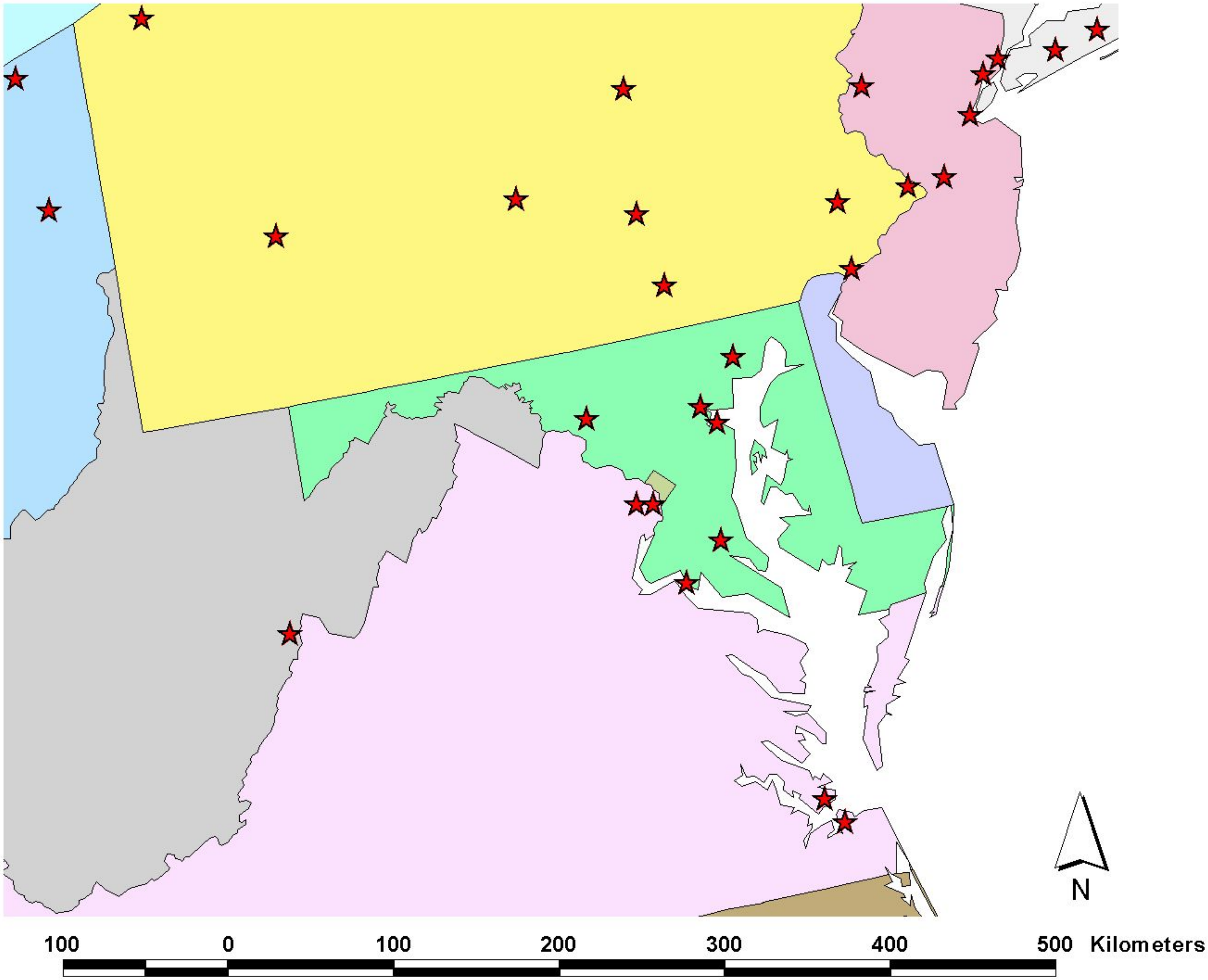


Impact of Source Emitting 30% Hg(0) 50% Hg(II) 20% Hg(p)	**=**	**0.3 x**	**Impact of Source Emitting Pure Hg(0)**
			+
		0.5 x	**Impact of Source Emitting Pure Hg(II)**
			+
		0.2 x	**Impact of Source Emitting Pure Hg(p)**

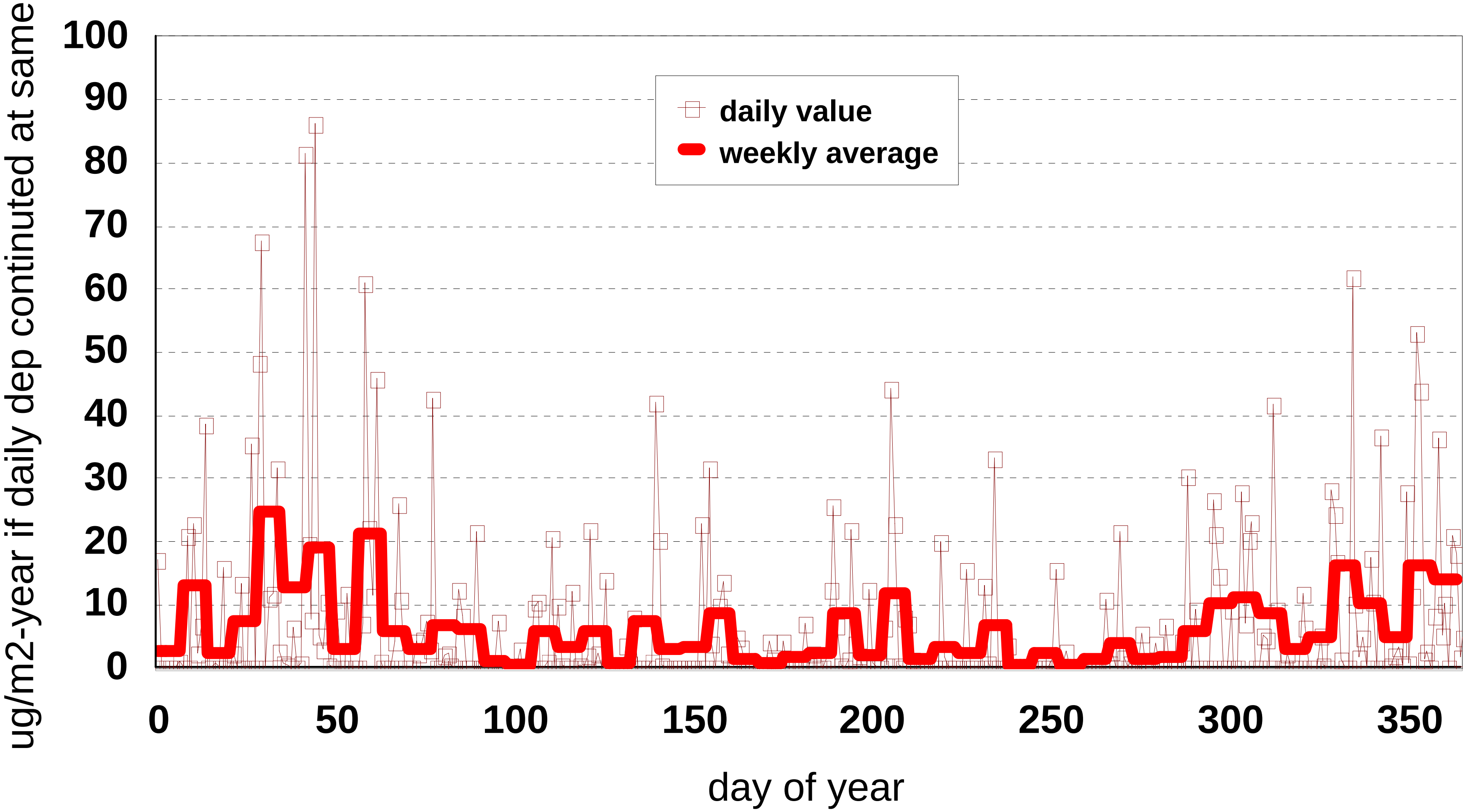


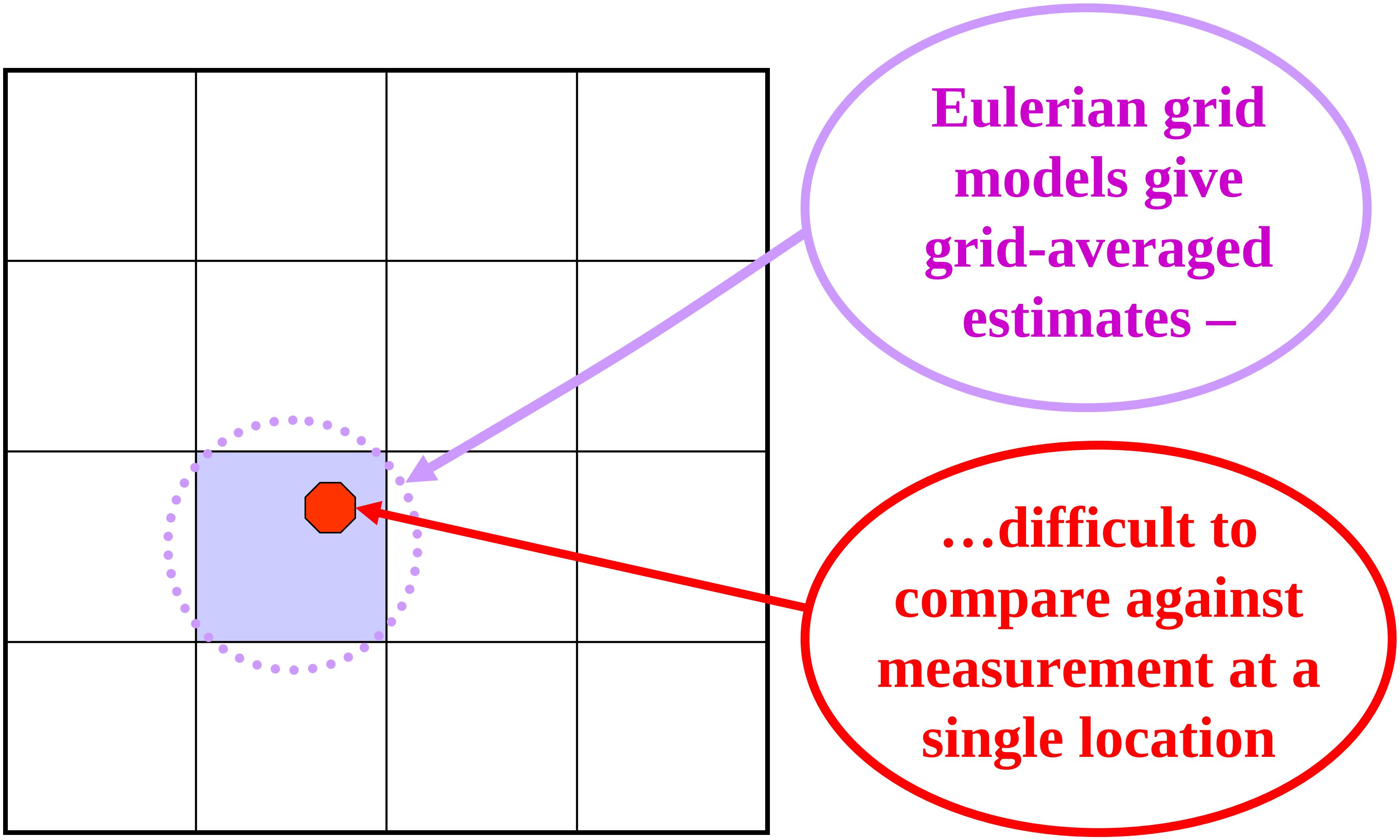
Standard Source
Locations 1-417
std 1 thru 84
cep impact from before
mn 16 and mn 18 study
spatial repr for MN and WI MDN
impact on EPA receptors
champlain, ches, maine, tampa
for cec receptors
close in series
Cep_impact_before.txt
1000
0
1000
Kilometers
N

Standard Source Locations in Maryland region during recent simulation



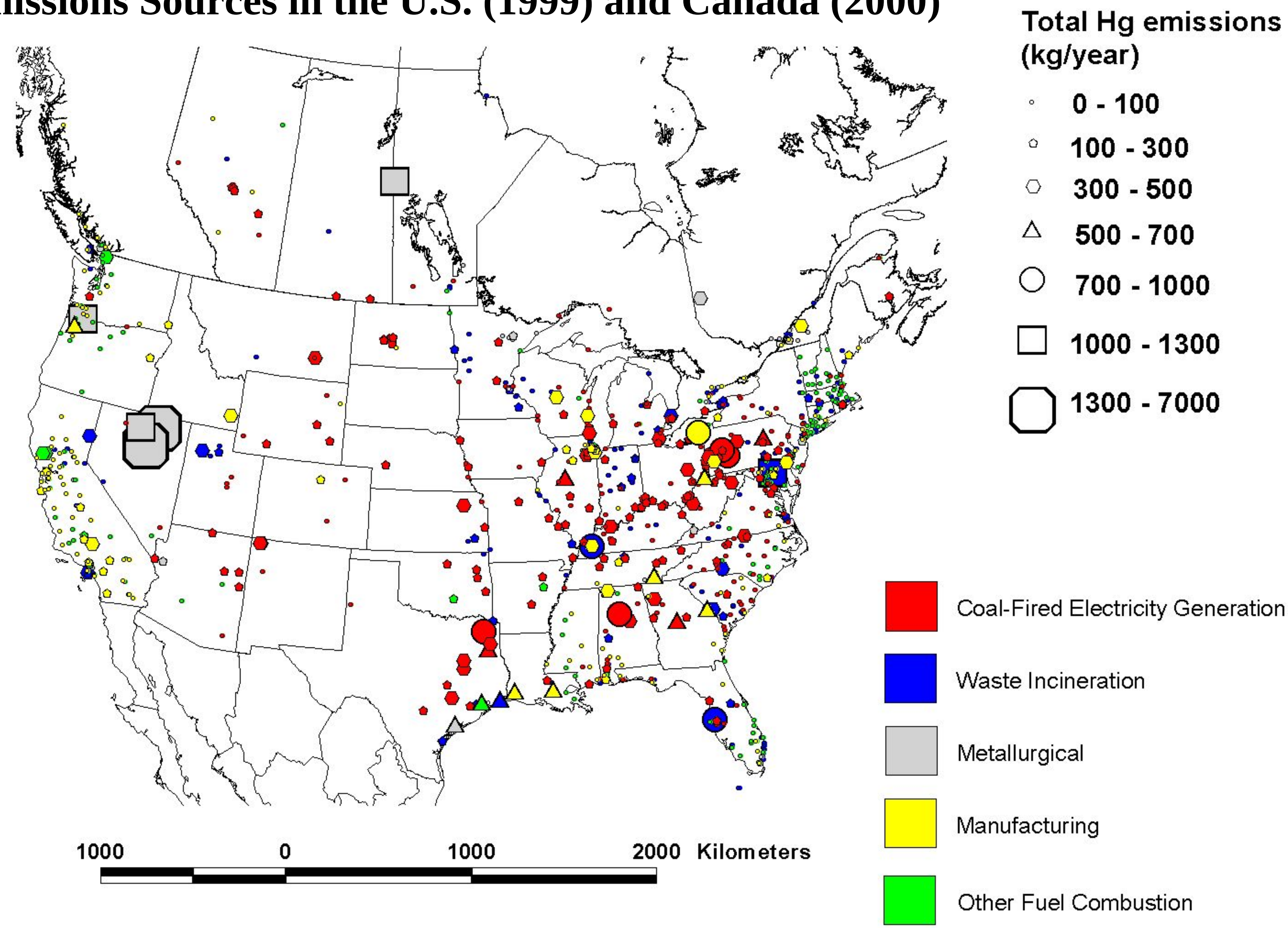
Illustrative example of total deposition at a location ~40 km "downwind" of a 1 kg/day RGM source

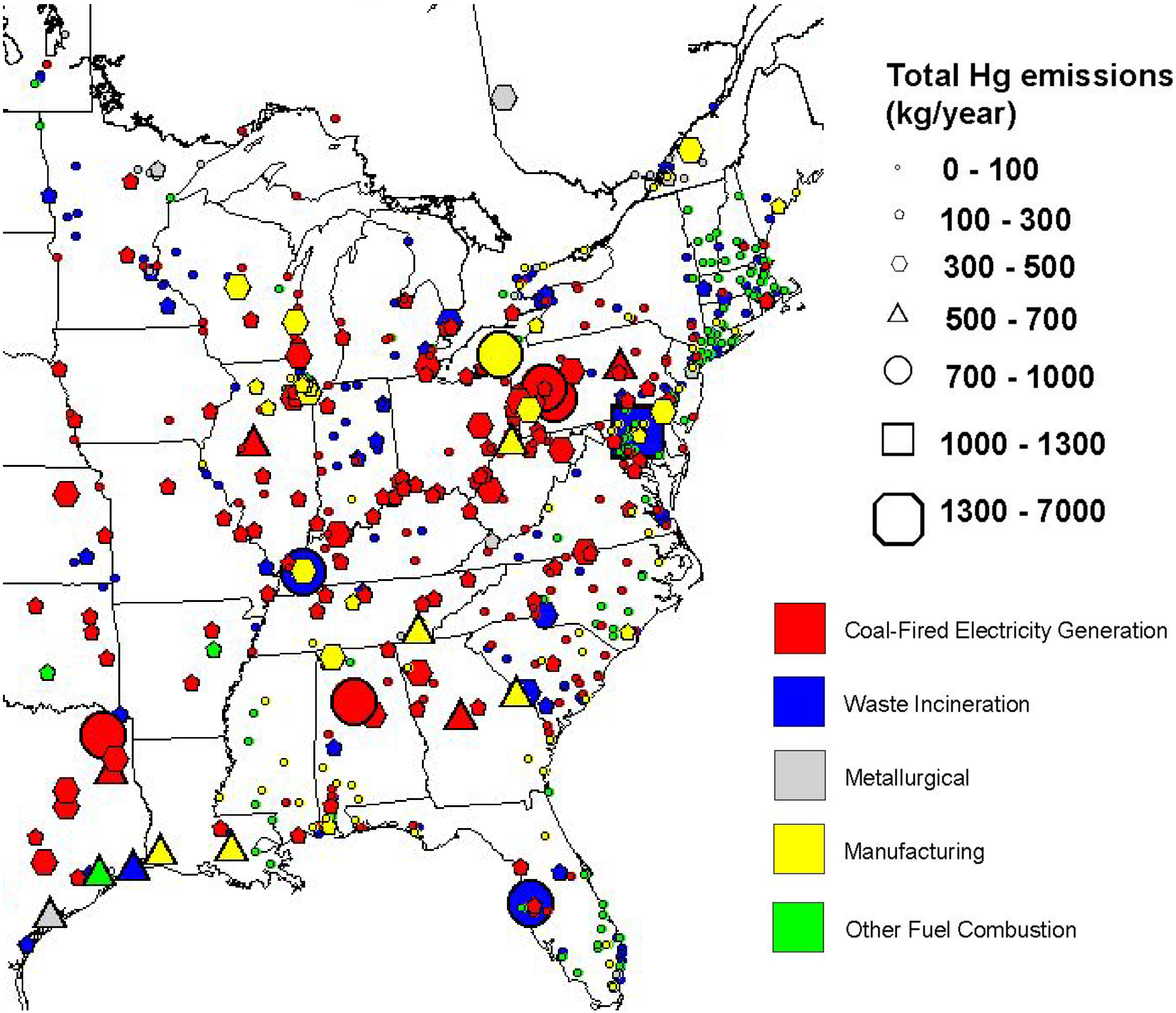




Eulerian grid models give grid-averaged estimates –
…difficult to compare against measurement at a single location

Geographic Distribution of Largest Anthropogenic Mercury Emissions Sources in the U.S. (1999) and Canada (2000)




Total Hg emissions
(kg/year)
0 - 100
100 - 300
300 - 500
500 - 700
700 - 1000
1000 - 1300
1300 - 7000
Coal-Fired Electricity Generation
Waste Incineration
Metallurgical
Manufacturing
Other Fuel Combustion

- **In principle, we need do this for each source in the inventory**

- **But, since there are more than 100,000 sources in the U.S. and Canadian inventory, we need shortcuts…**

- **Shortcuts described in Cohen *et al***
  Environmental Research **95**(3), 247-265, 2004

Cohen, M., Artz, R., Draxler, R., Miller, P., Poissant, L., Niemi, D., Ratte, D., Deslauriers, M., Duval, R., Laurin, R., Slotnick, J., Nettesheim, T., McDonald, J. "Modeling the Atmospheric Transport and Deposition of Mercury to the Great Lakes." *Environmental Research* **95**(3), 247-265, 2004.

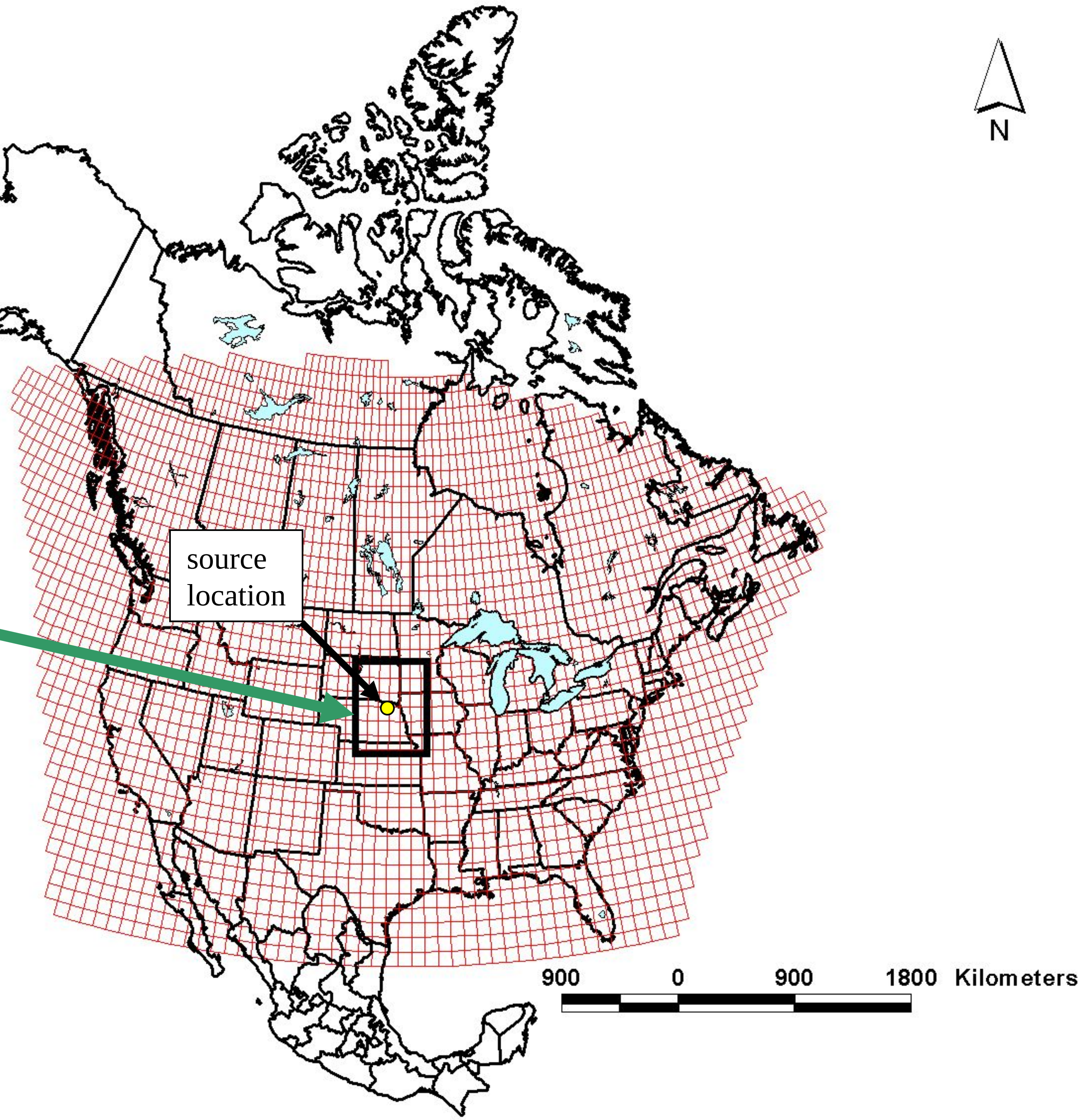
Note: Volume 95(3) is a Special Issue: "An Ecosystem Approach to Health Effects of Mercury in the St. Lawrence Great Lakes", edited by David O. Carpenter.

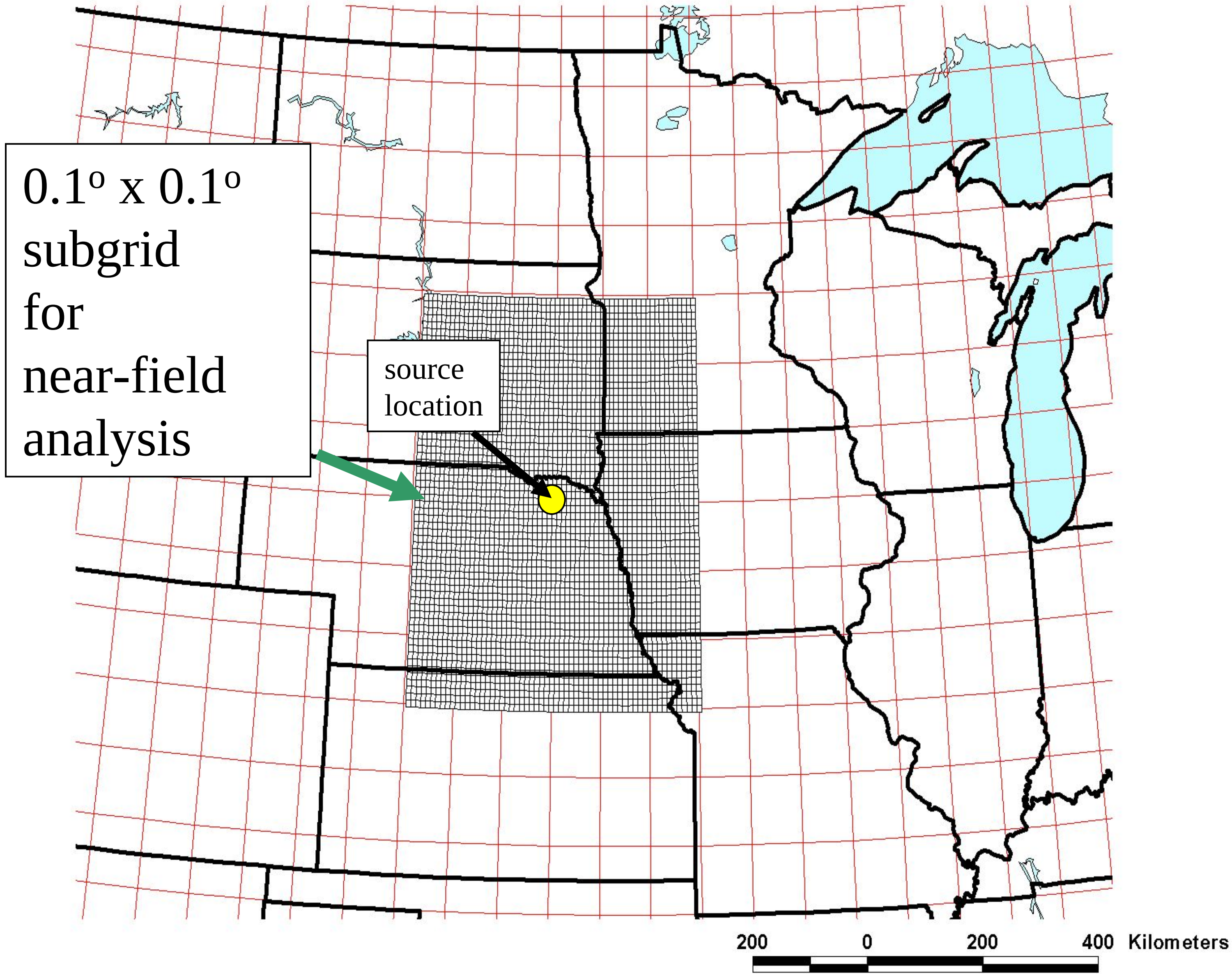
- **For each run, simulate fate and transport *everywhere*, but only keep track of impacts on each selected receptor (e.g., Great Lakes, Chesapeake Bay, etc.)**

- **Only run model for a limited number (~100) of hypothetical, individual unit-emissions sources throughout the domain**

- **Use spatial interpolation to estimate impacts from sources at locations not explicitly modeled**

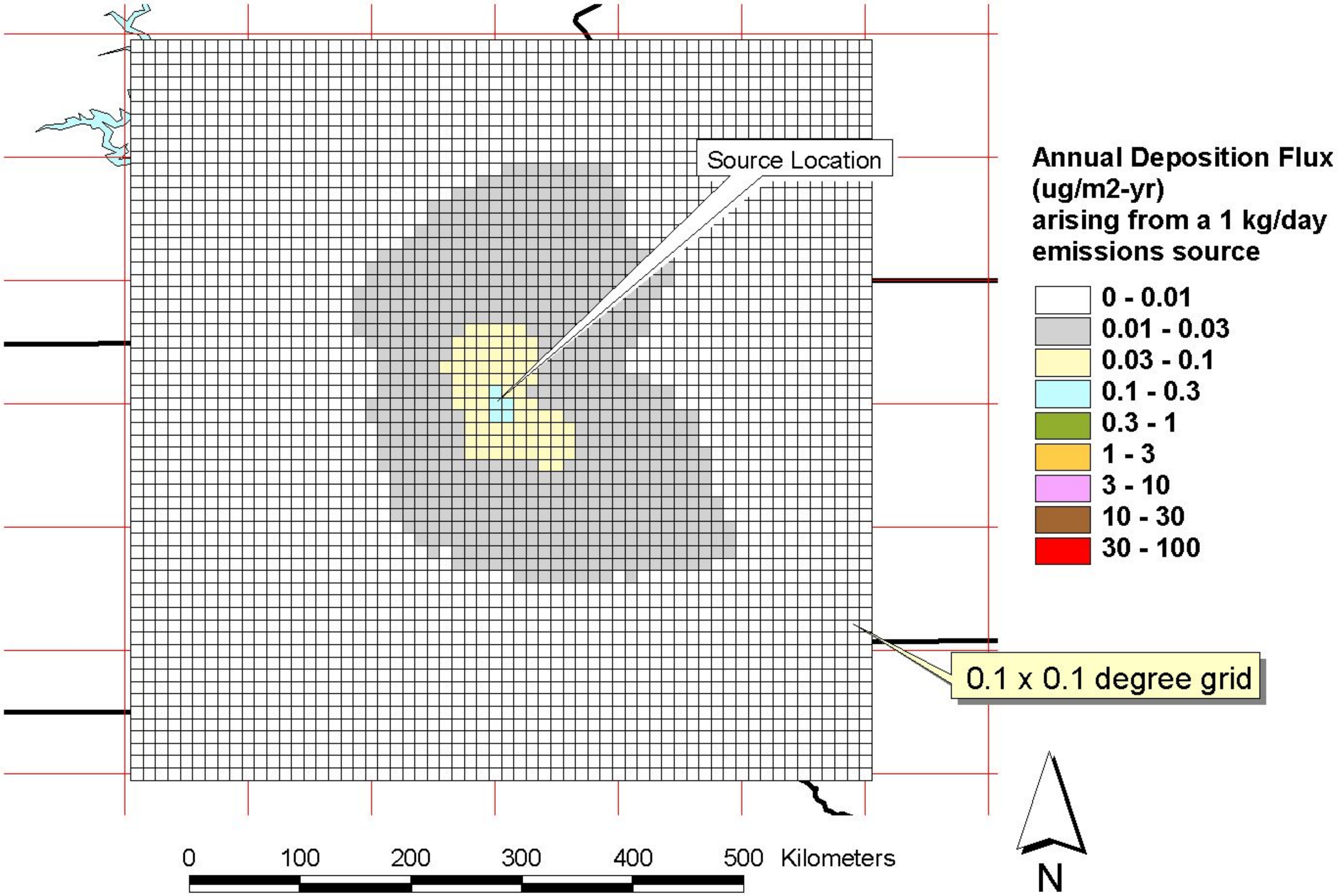
$0.1^o \times 0.1^o$ subgrid for near-field analysis





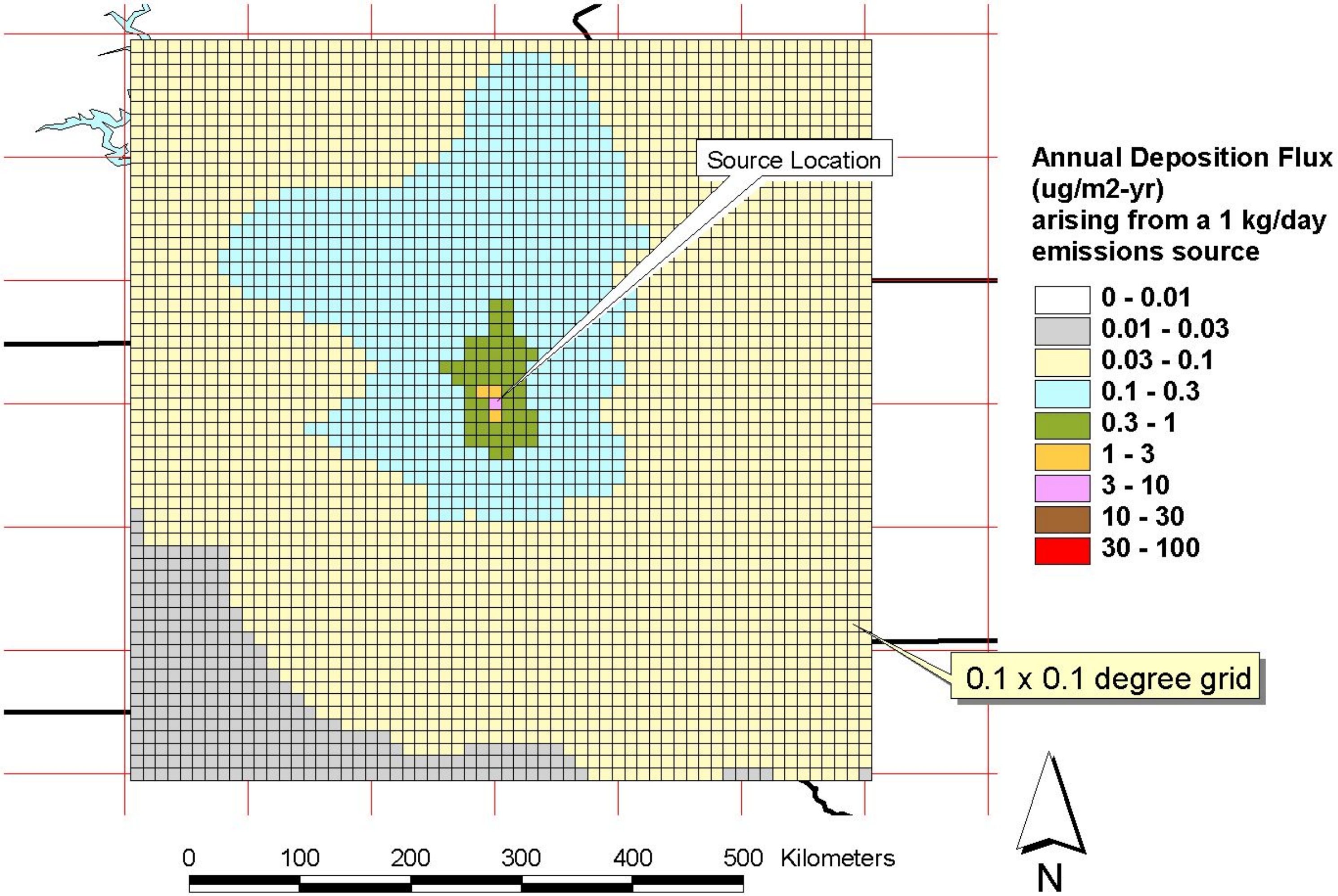
0.1° x 0.1° subgrid for near-field analysis
source location
200 0 200 400 Kilometers

Annual deposition summary for emissions of elemental Hg from a 250 meter high source



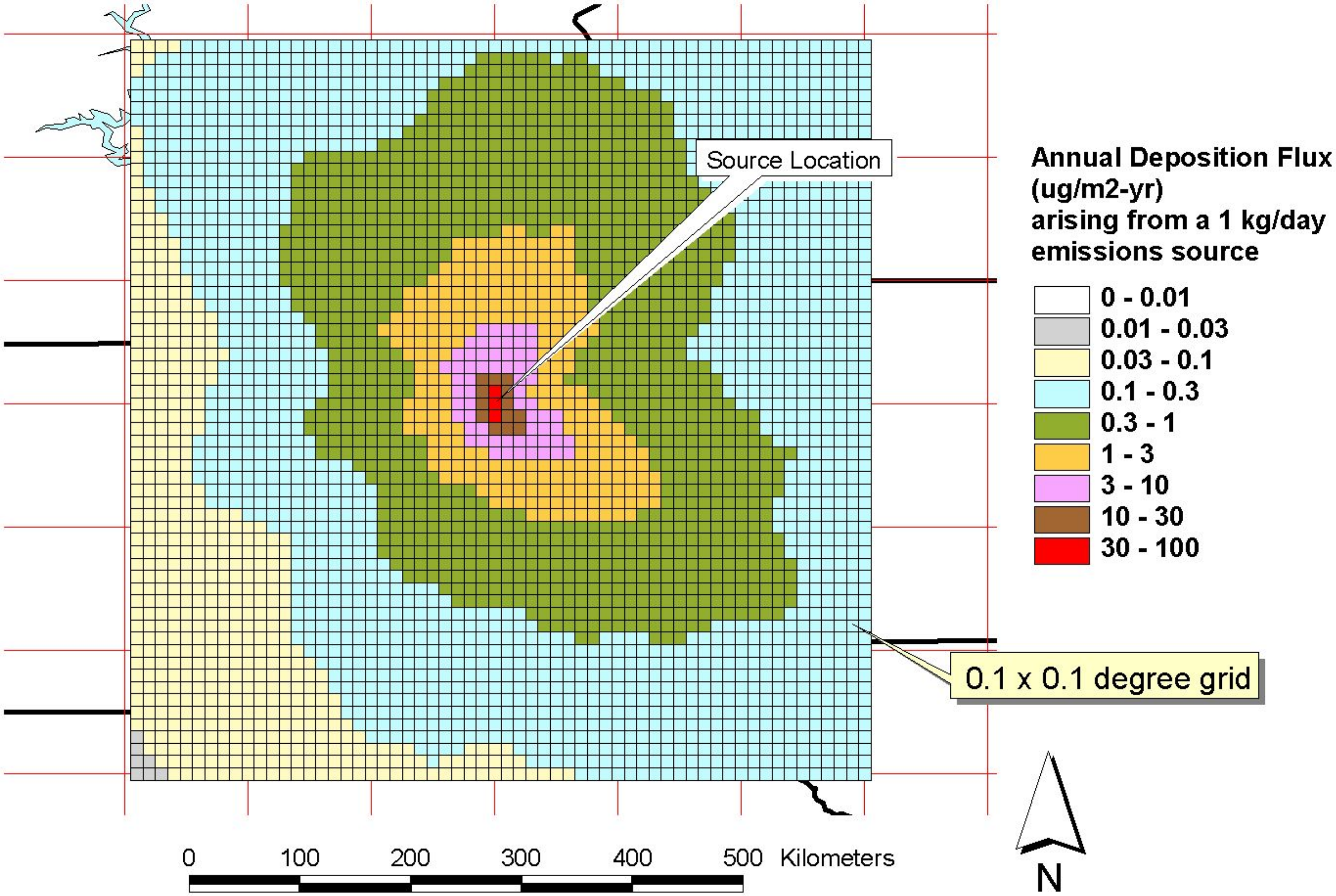
Hypothetical emissions source at lat = 42.5, long = -97.5;
simulation for entire year 1996 using archived NGM meteorology (180 km resolution)

Annual deposition summary for emissions of particulate Hg from a 250 meter high source



Hypothetical emissions source at lat = 42.5, long = -97.5;
simulation for entire year 1996 using archived NGM meteorology (180 km resolution)

Annual deposition summary for emissions of ionic Hg from a 250 meter high source



Hypothetical emissions source at lat = 42.5, long = -97.5;
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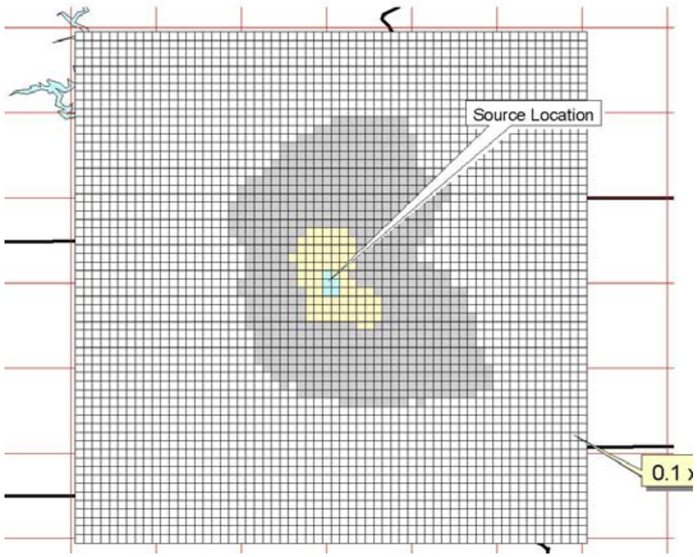
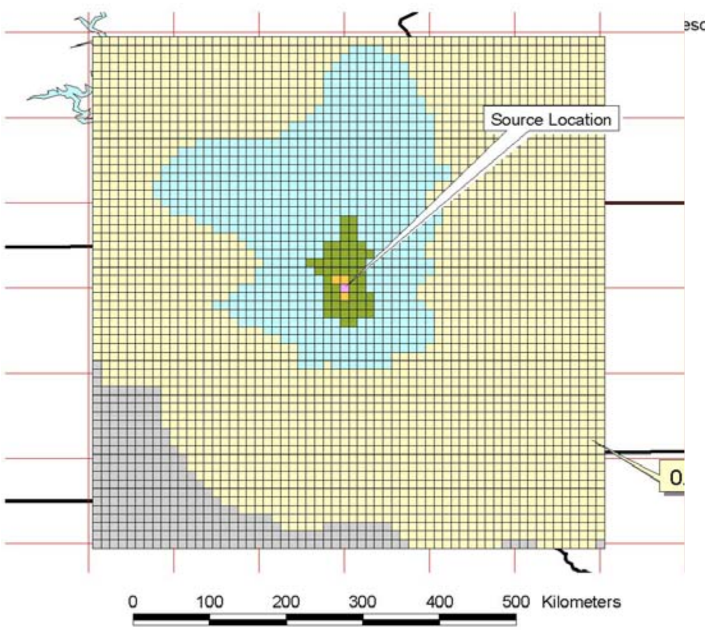
Annual deposition summary for emissions of elemental Hg from a 250 meter high source



Annual Deposition Flux (ug/m2-yr) arising from a 1 kg/day emissions source

- 0 - 0.01
- 0.01 - 0.03
- 0.03 - 0.1
- 0.1 - 0.3
- 0.3 - 1
- 1 - 3
- 3 - 10
- 10 - 30
- 30 - 100

Annual deposition summary for emissions of particulate Hg from a 250 meter high source



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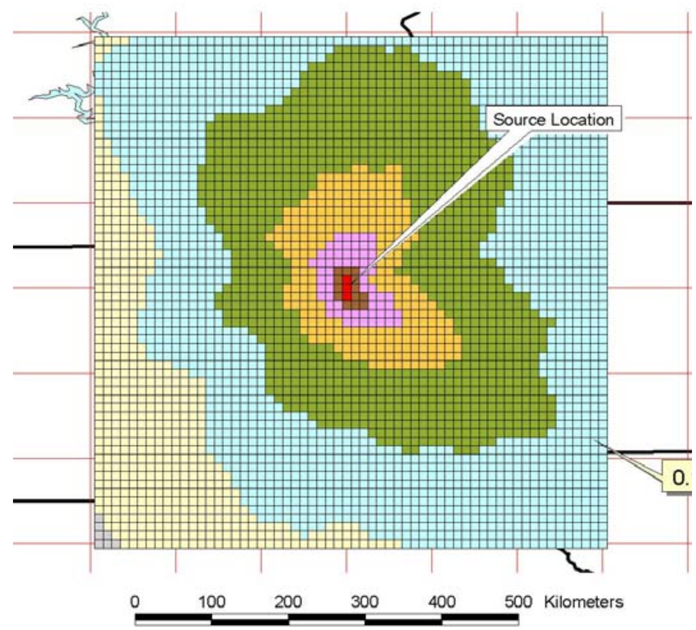
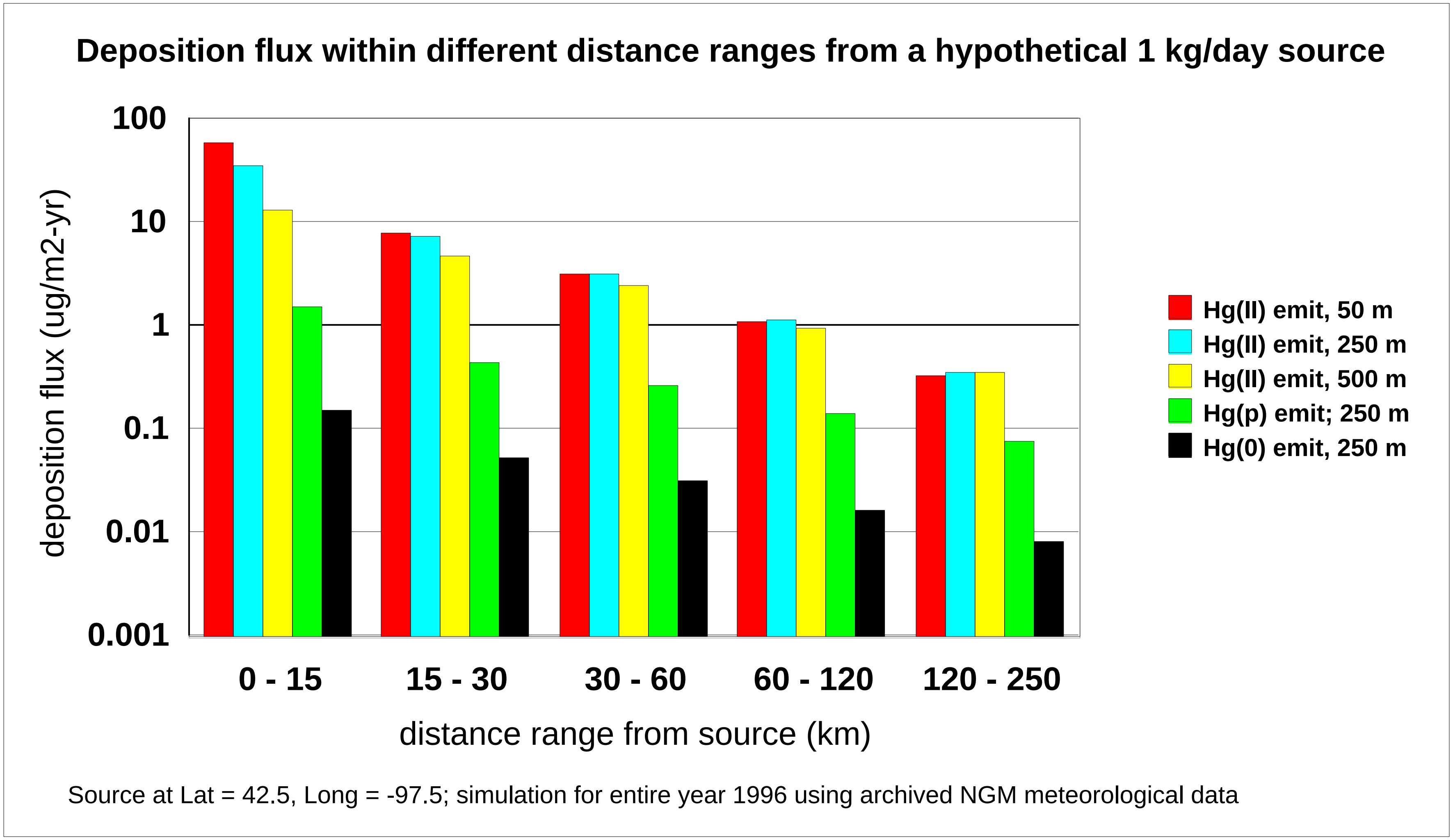
Annual deposition summary for emissions of ionic Hg from a 250 meter high source

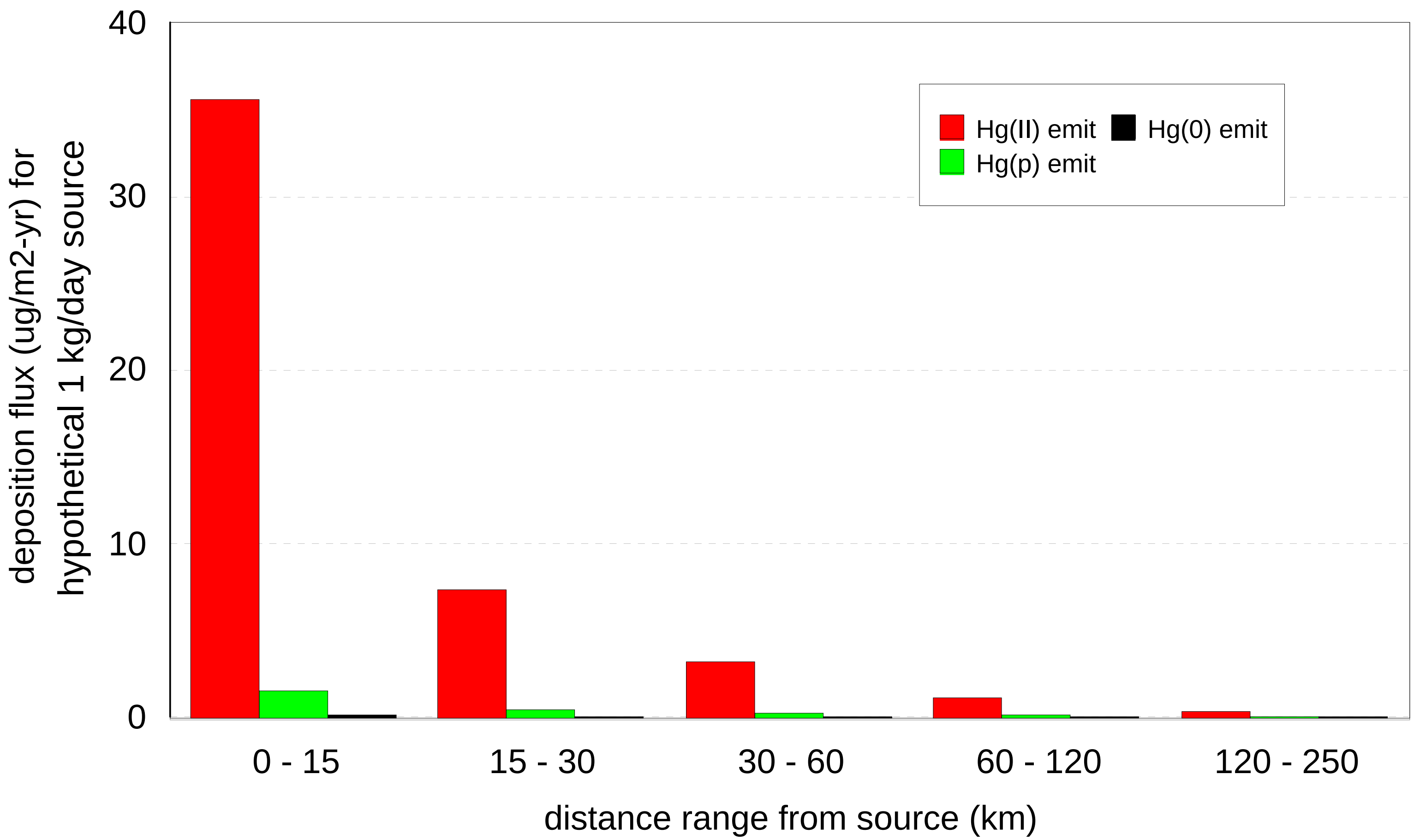


Hypothetical emissions source at lat = 42.5, long = -97.5;
simulation for entire year 1996 using archived NGM meteorology (180 km



Hypothesized rapid reduction of Hg(II) in plumes?
If true, then dramatic impact on modeling results…

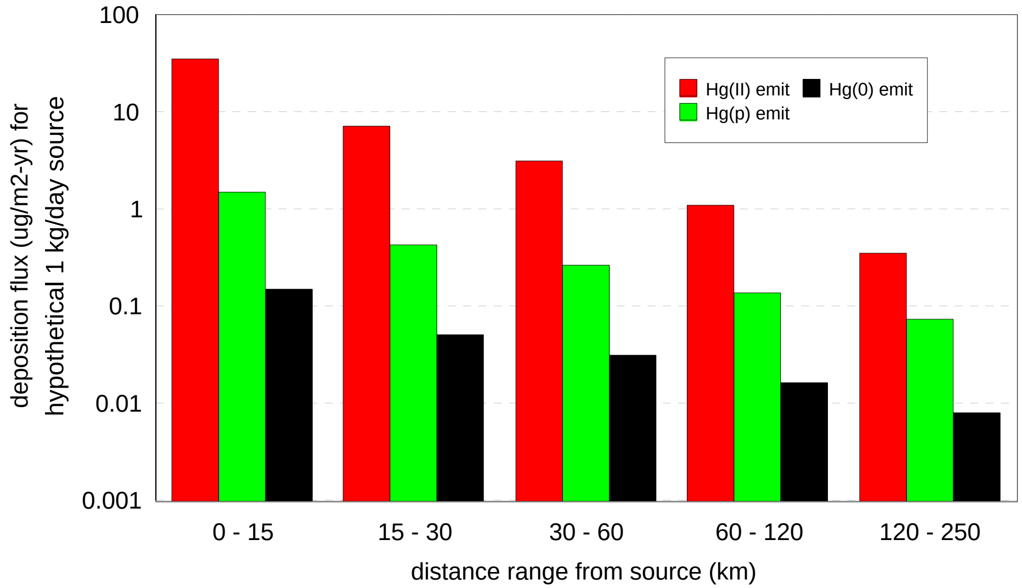
Why is emissions speciation information critical?



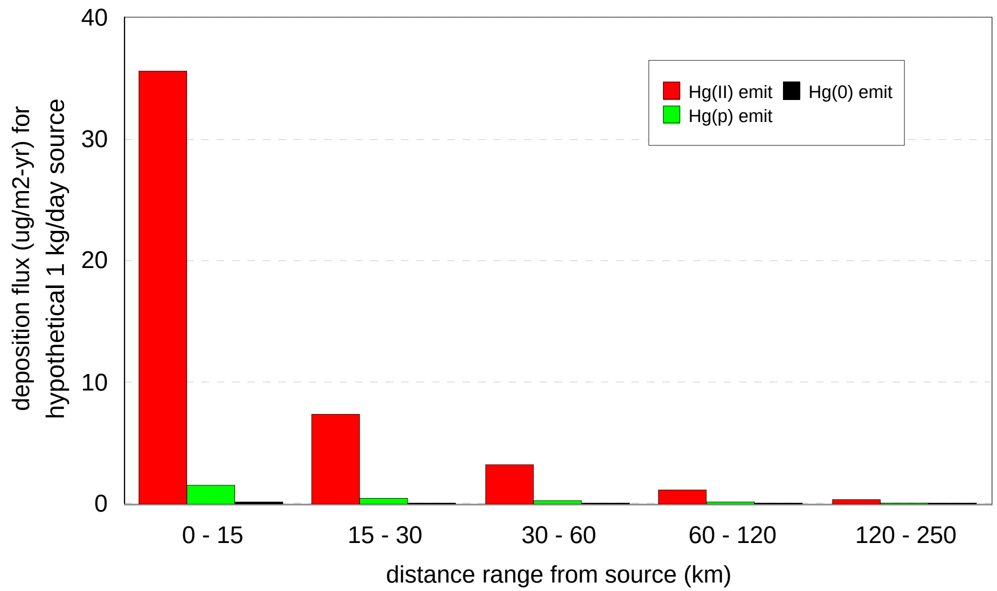
Linear

Why is emissions speciation information critical?

Logarithmic



Linear

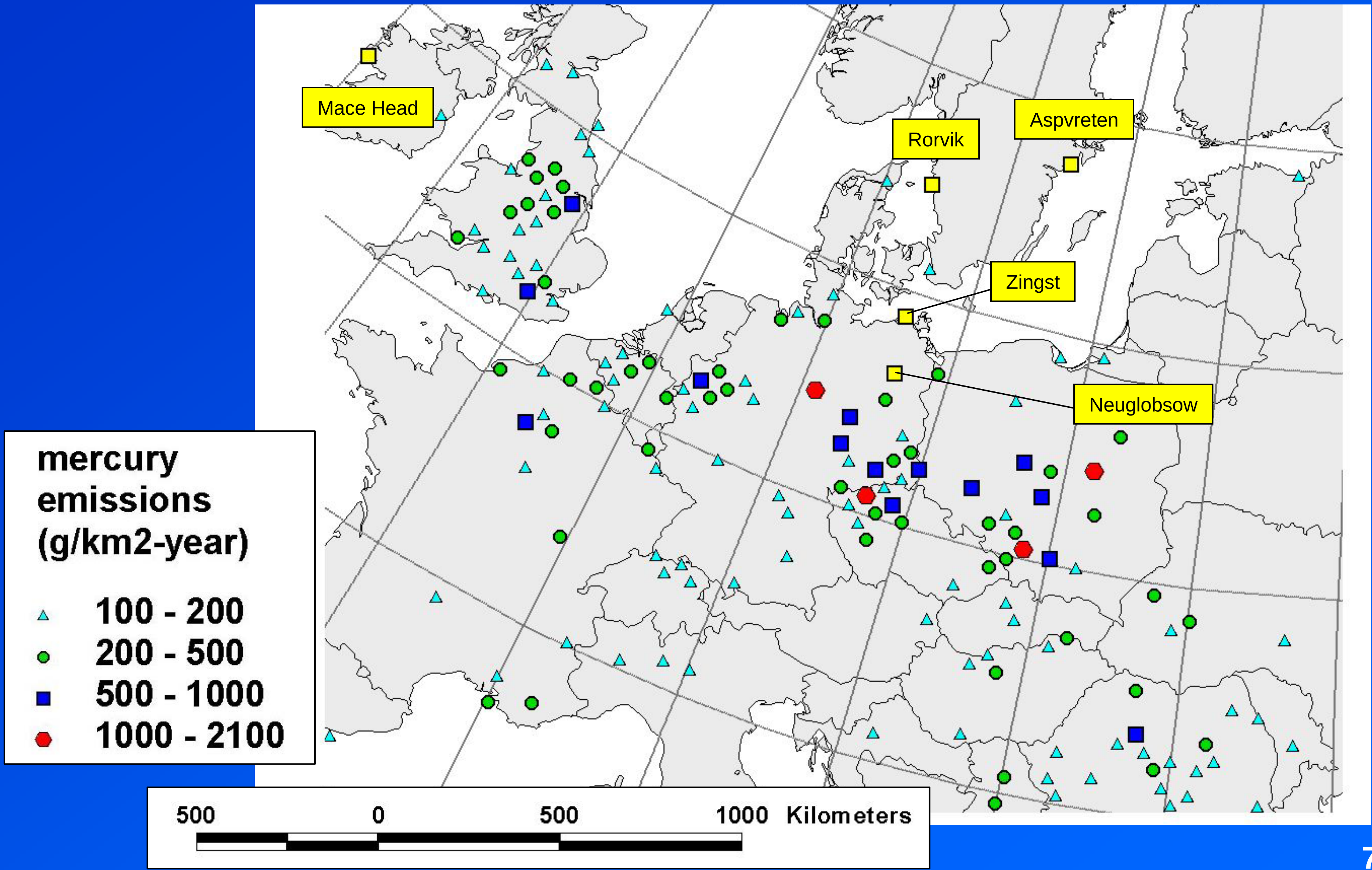


Emissions and Chemistry

- *The form of mercury emissions* (elemental, ionic, particulate) is often very poorly known, but *is a dominant factor in estimating deposition* (and associated source-receptor relationships)

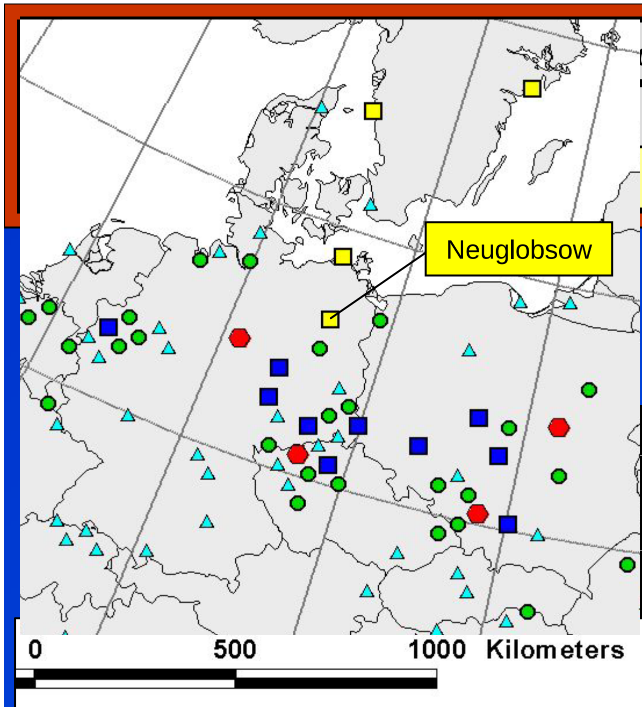
- Questions regarding atmospheric chemistry of mercury may also be very significant

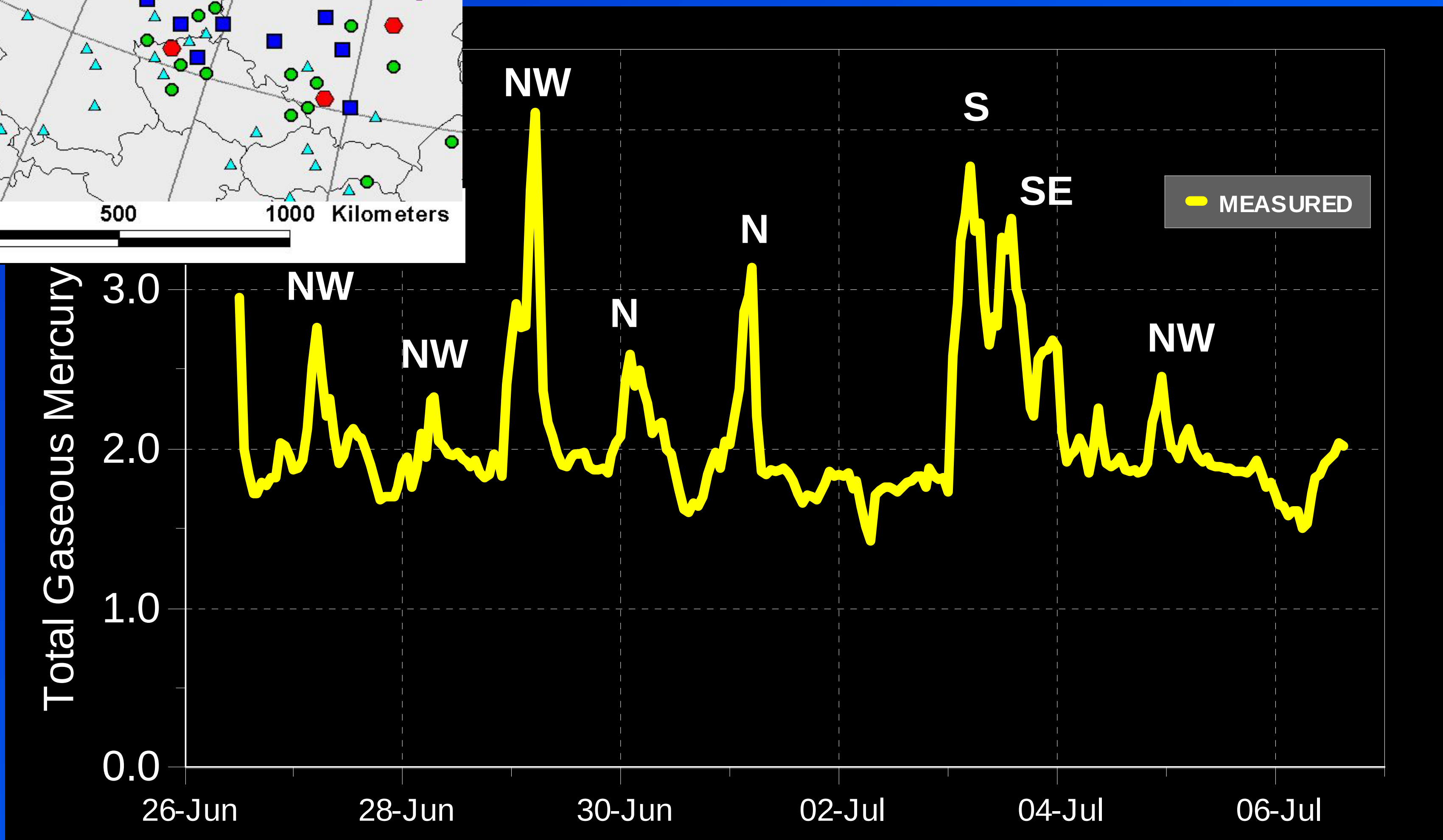
- *The above may contribute more to the overall uncertainties in atmospheric mercury models* than uncertainties in dry and wet deposition algorithms



Mace Head
Rorvik
Aspvreten
Zingst
Neuglobsow
mercury emissions (g/km2-year)
100 - 200
200 - 500
500 - 1000
1000 - 2100
500
0
500
1000
Kilometers

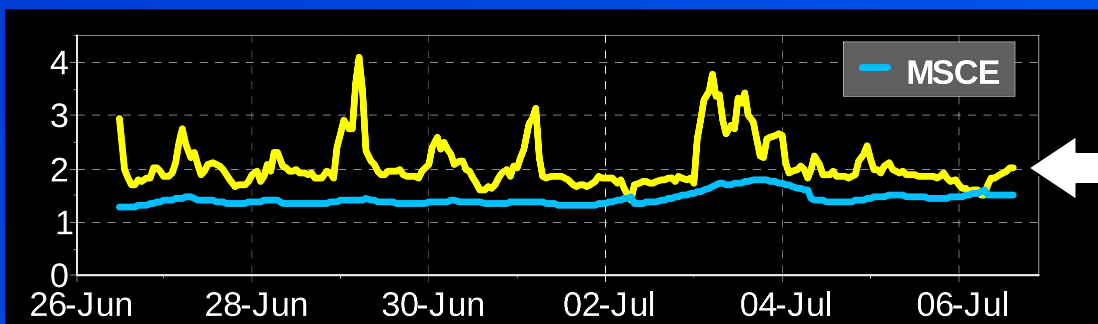
rcury at Neuglobsow: June 26 – July 6, 1995



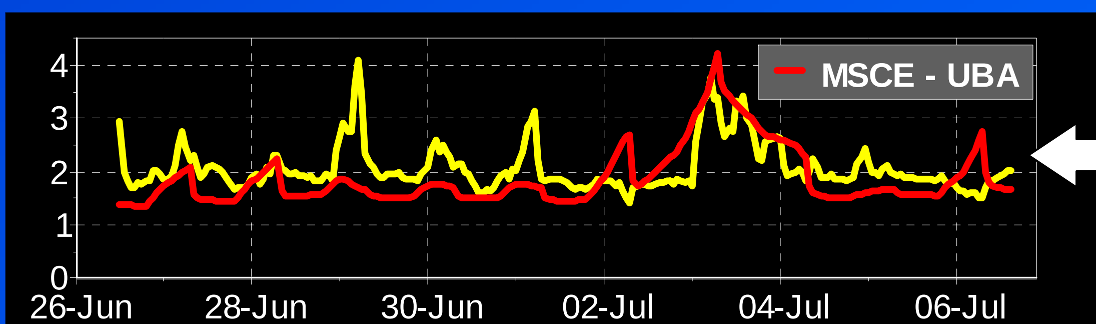


Total Gaseous Mercury (ng/m³) at Neuglobsow: June 26 – July 6, 1995

The emissions inventory is a critical input to the models…



Using default emissions inventory



Using alternative emissions inventory

Some Additional Measurement Issues (from a modeler's perspective)

- **Data availability**
- **Simple vs. Complex Measurements**
- **Process Information**

Process Information:
1. Dry Deposition - Resistance Formulation

$$V_d = \frac{1}{R_a + R_b + R_c + R_a R_b V_g} + V_g$$

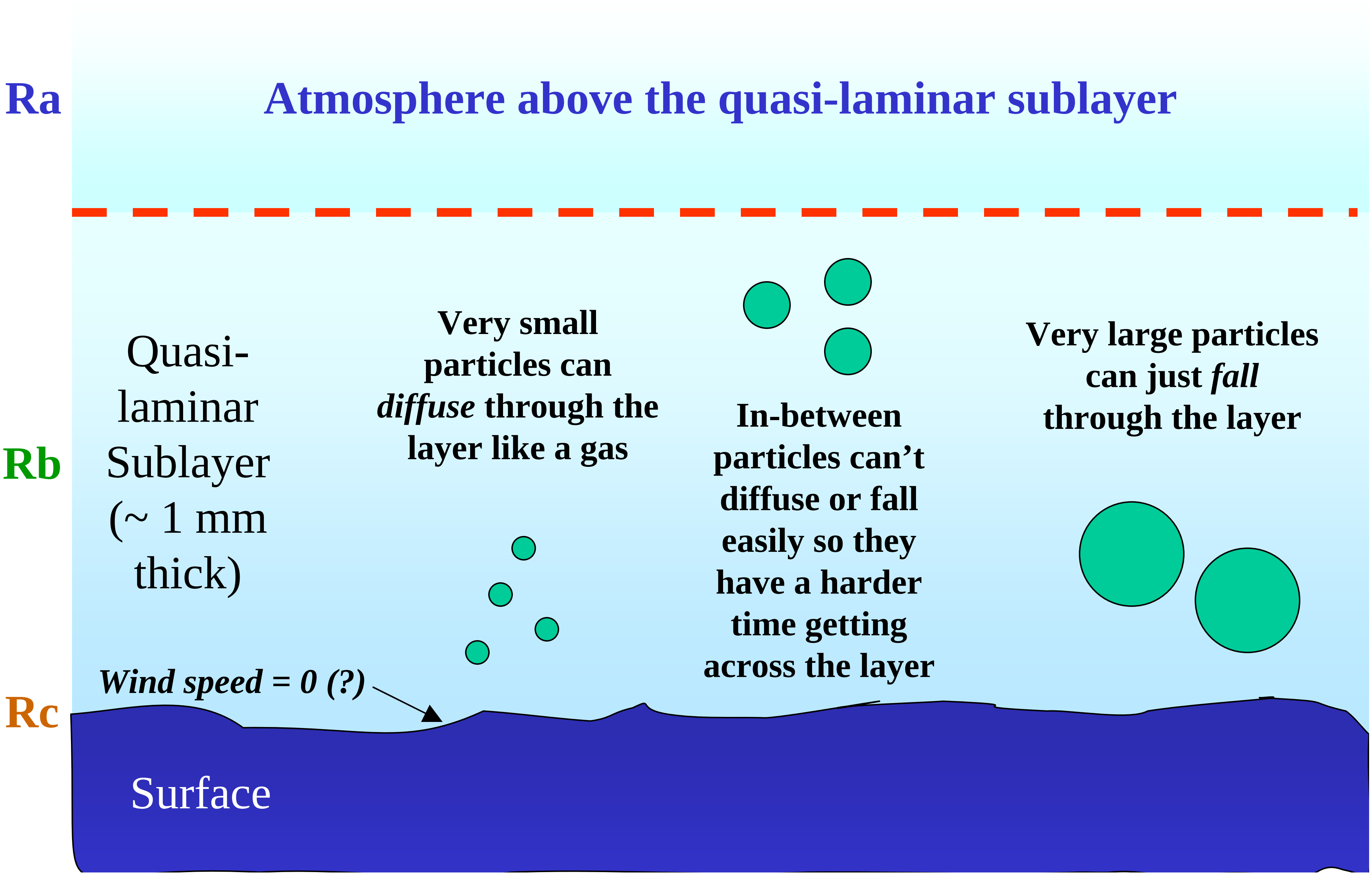
in which

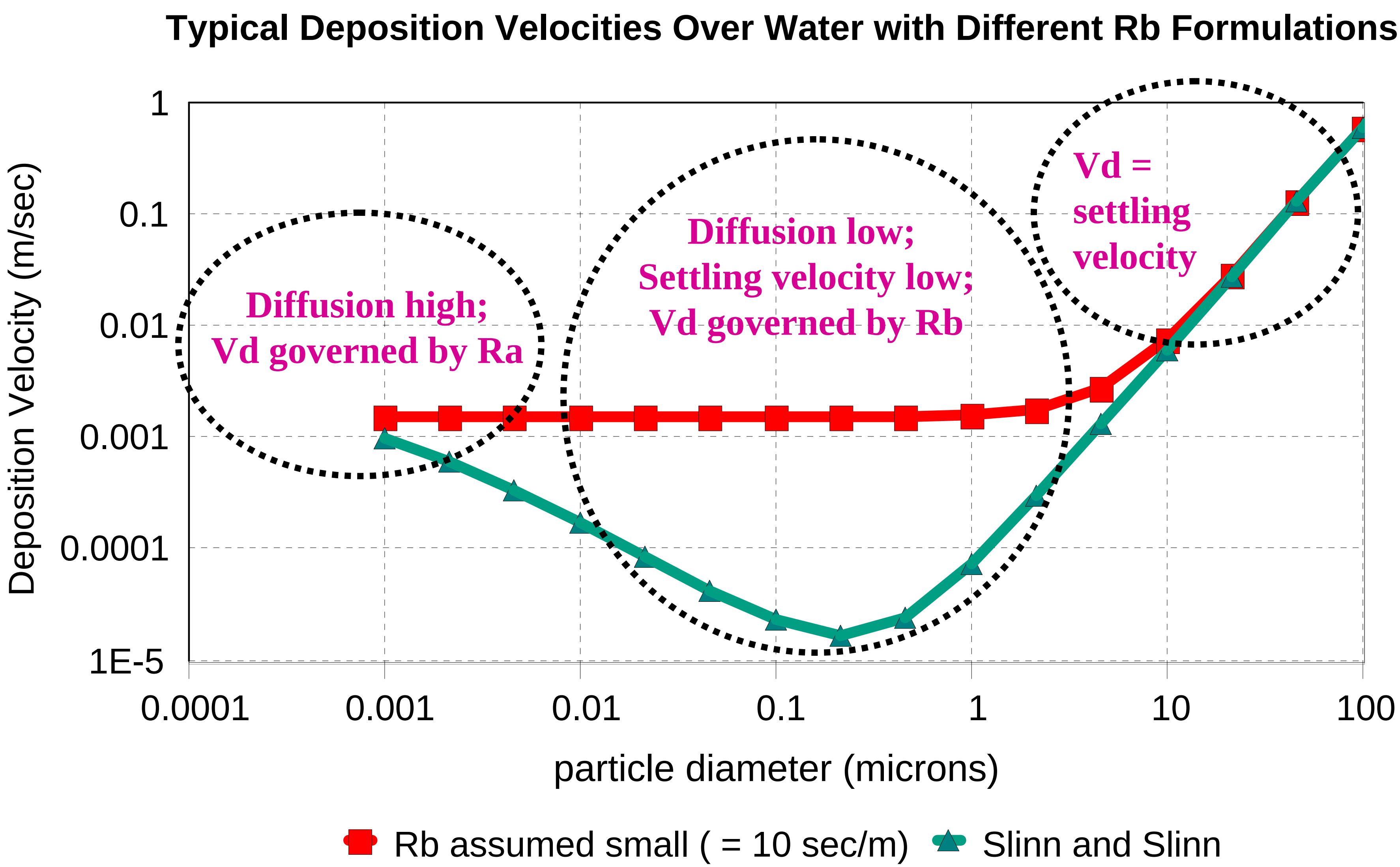
- R_a = aerodynamic resistance to mass transfer;
- R_b = resistance of the quasi-laminar sublayer;
- R_c = overall resistance of the canopy/surface (zero for particles)
- V_g = the gravitational settling velocity (zero for gases).

Dry Deposition

- depends intimately on vapor/particle partitioning and particle size distribution information
- resistance formulation [R_a, R_b, R_c...]
- for gases, key uncertainty often R_c (e.g., "reactivity factor" f_0)
- for particles, key uncertainty often R_b
- How to evaluate algorithms when phenomena hard to measure?

Particle dry deposition phenomena

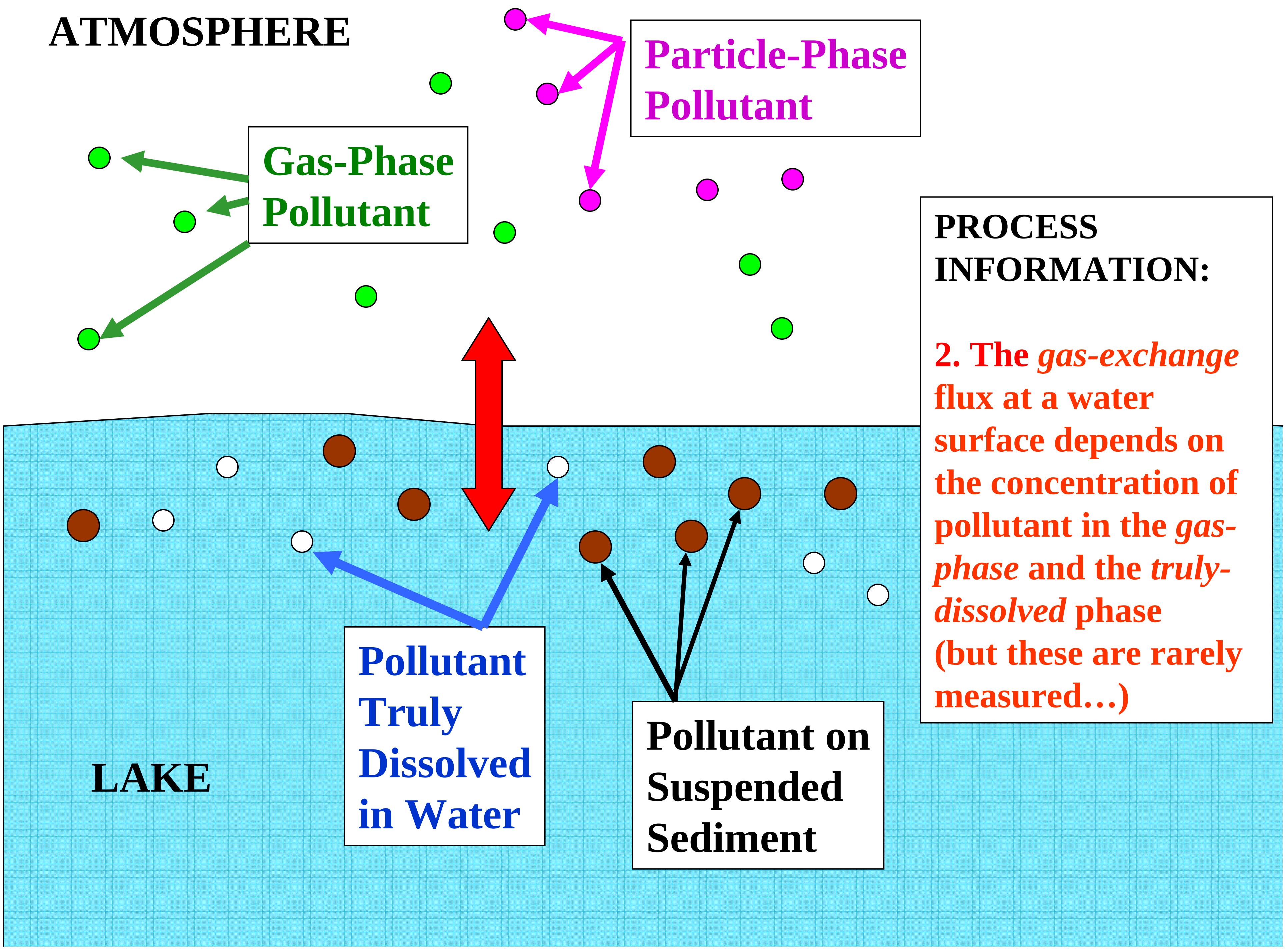




Typical Deposition Velocities Over Water with Different Rb Formulations
Deposition Velocity (m/sec)
1
0.1
0.01
0.001
0.0001
1E-5
0.0001
0.001
0.01
0.1
1
10
100
particle diameter (microns)
Diffusion high;
Vd governed by Ra
Diffusion low;
Settling velocity low;
Vd governed by Rb
Vd =
settling
velocity
Rb assumed small (= 10 sec/m)
Slinn and Slinn

Process information needed:

1. For particle dry deposition, must have particle size distributions!


ATMOSPHERE
Particle-Phase Pollutant
Gas-Phase Pollutant
PROCESS INFORMATION:
2. The *gas-exchange* flux at a water surface depends on the concentration of pollutant in the *gas-phase* and the *truly-dissolved* phase (but these are rarely measured…)
Pollutant Truly Dissolved in Water
Pollutant on Suspended Sediment
LAKE