The Atmospheric Transport and Deposition of Mercury

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Materials assembled for a discussion with the
Baltimore City Department of Law
City Hall, August 25, 2005

The Mercury Problem

- EPA has estimated that 1 out of every 6 children born in the U.S. may have already been exposed *in-utero* to levels of mercury that might cause problems with neurological development

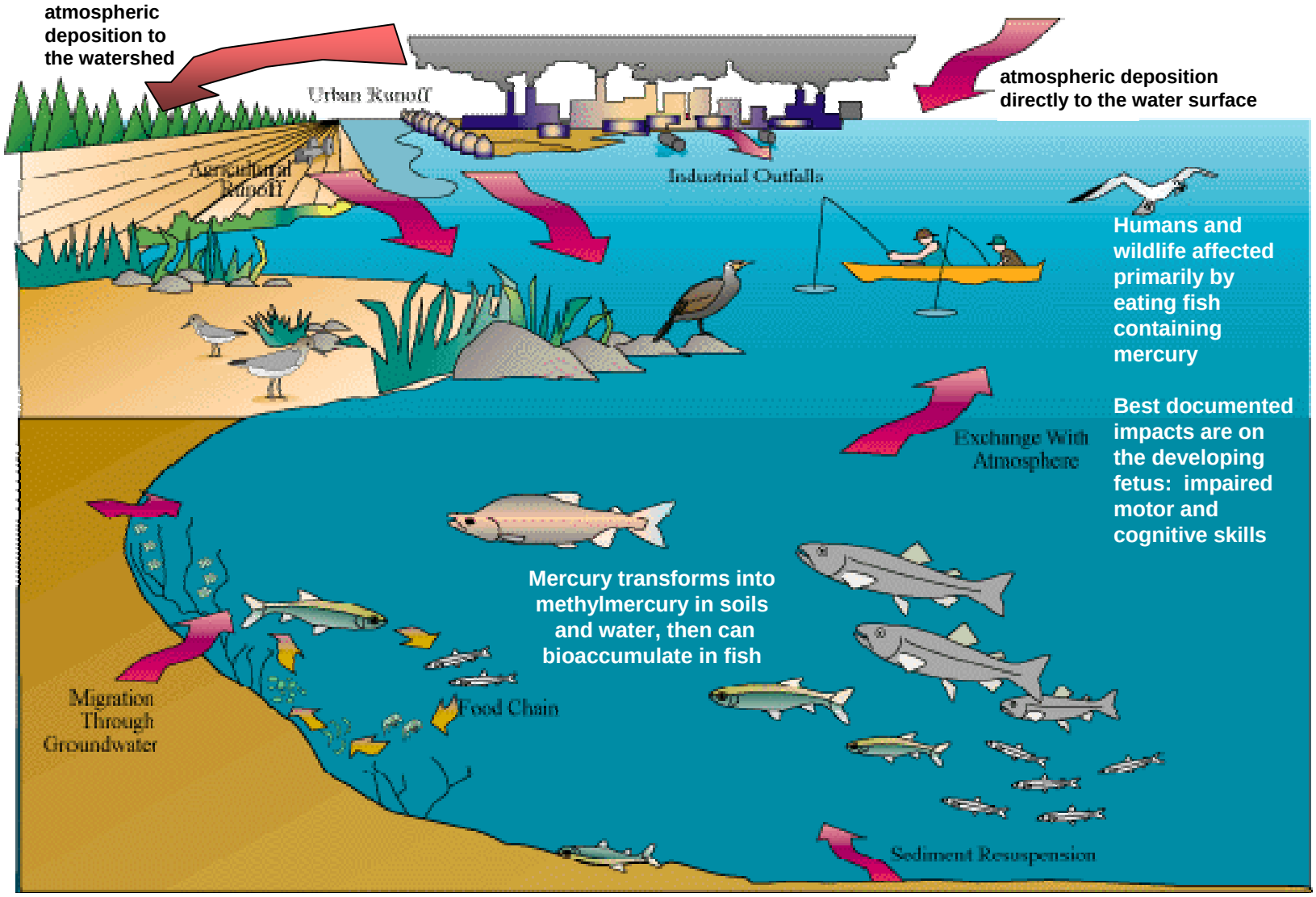
- There are additional potential mercury-related health hazards to children, adults, and to wildlife

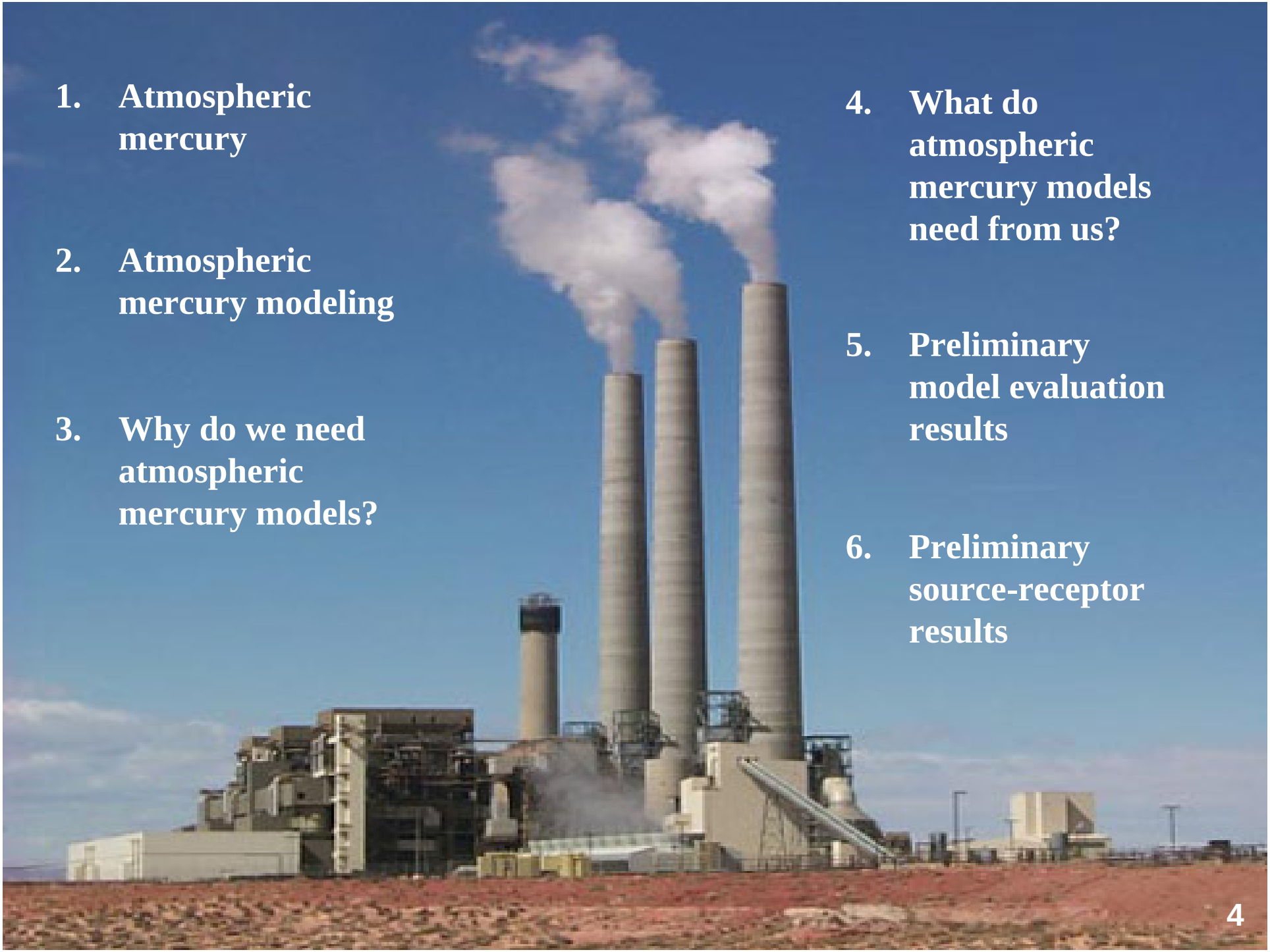
- Fish-consumption advisories due to mercury contamination are widespread throughout U.S. rivers, lakes, and coastal areas

- The primary exposure route is through fish consumption

- Atmospheric deposition is a significant – often the most significant – pathway for mercury loading to aquatic ecosystems

There are many ways in which mercury is introduced into a given aquatic ecosystem... atmospheric deposition can be a very significant pathway





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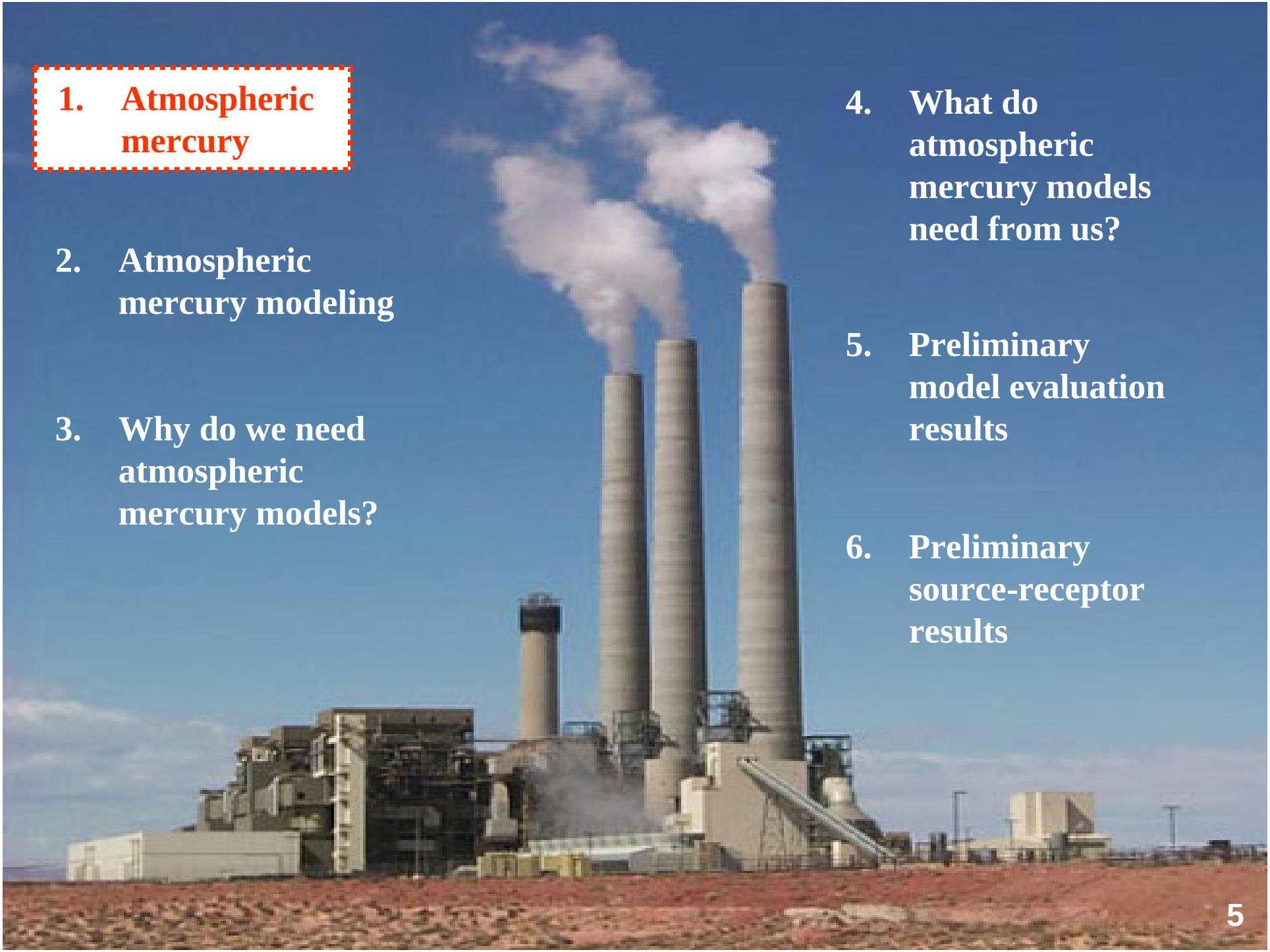
2. Atmospheric mercury modeling
3. Why do we need atmospheric mercury models?
4. What do atmospheric mercury models need from us?
5. Preliminary model evaluation results
6. Preliminary source-receptor results



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Three “forms” of atmospheric mercury

Elemental Mercury: Hg(0)

- **~ 95% of total Hg in atmosphere**
- ***not* very water soluble**
- **long atmospheric lifetime (~ 0.5 - 1 yr); globally distributed**

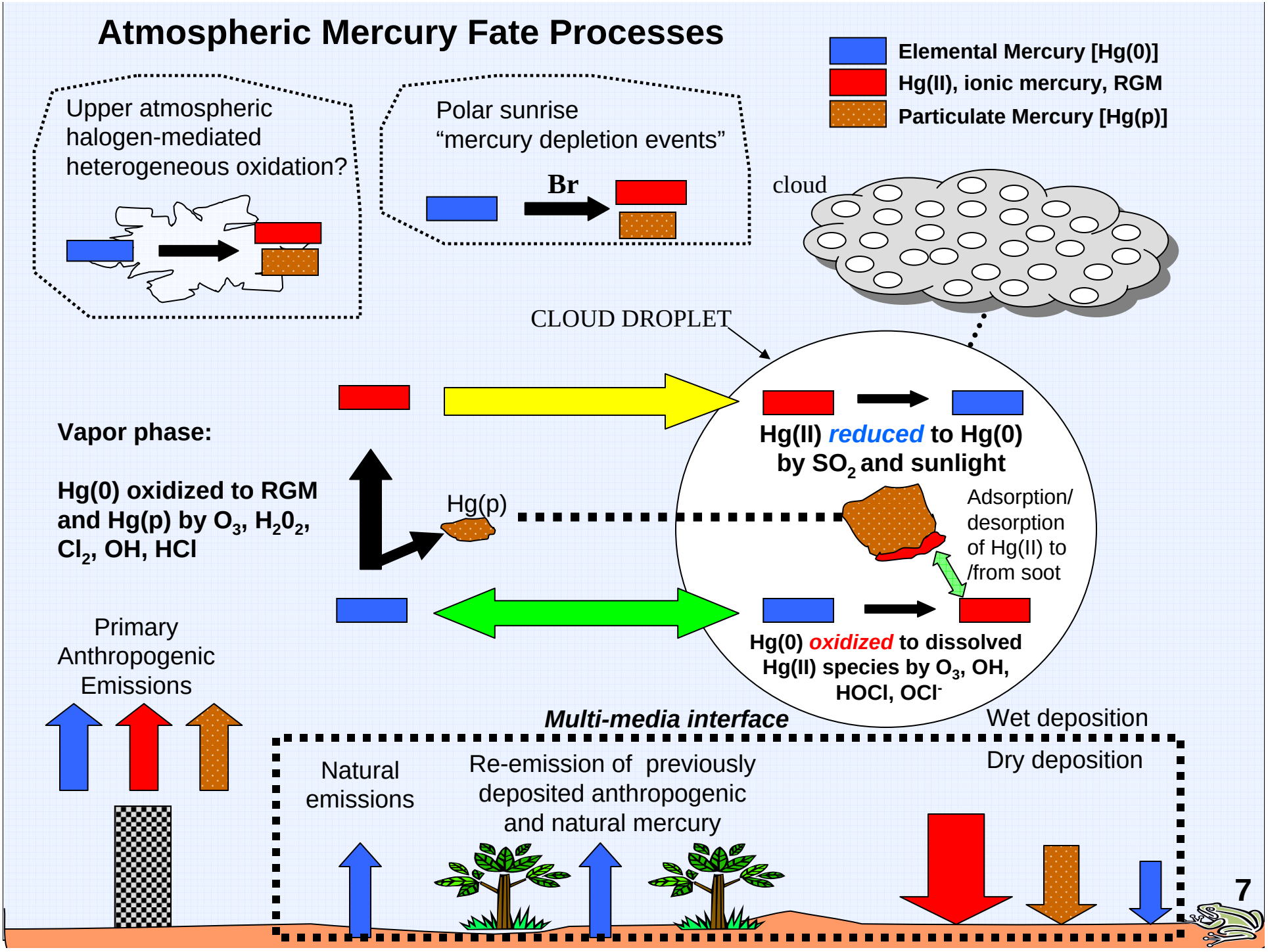
Reactive Gaseous Mercury (“RGM”)

- **a few percent of total Hg in atmosphere**
- **oxidized mercury: Hg(II)**
- **HgCl2, others species?**
- **somewhat operationally defined by measurement method**
- ***very* water soluble**
- **short atmospheric lifetime (~ 1 week or less);**
- **more local and regional effects**

Particulate Mercury (Hg(p)

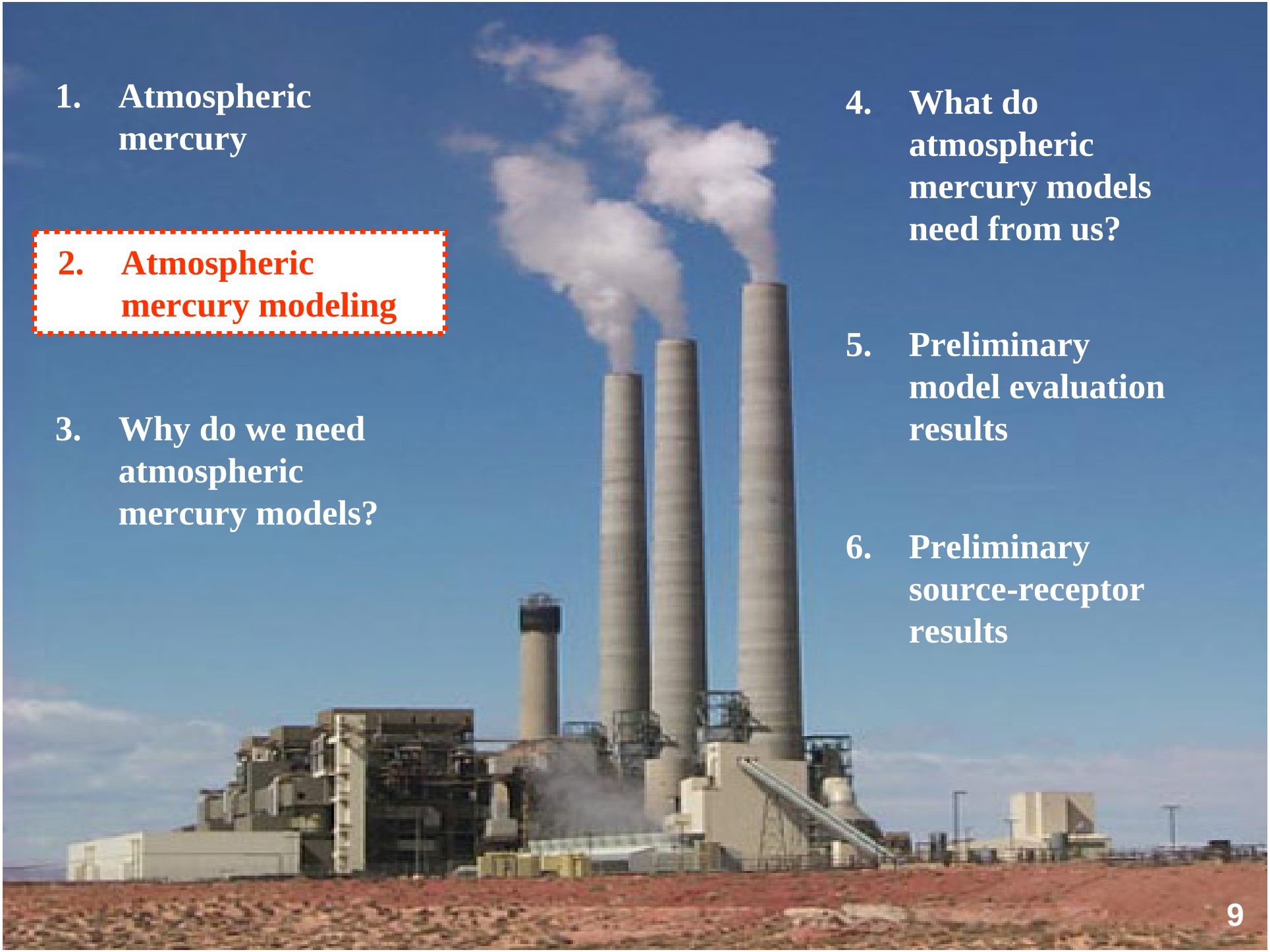
- **a few percent of total Hg in atmosphere**
- **not pure particles of mercury…**
  (Hg compounds associated with atmospheric particulate)
- **species largely unknown (in some cases, may be HgO?)**
- **moderate atmospheric lifetime (perhaps 1~ 2 weeks)**
- **local and regional effects**
- **bioavailability?**

Atmospheric Mercury Fate Processes



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	$(molar\text{-}sec)^{-1}$	Munthe (1992)
$Hg^0 + OHC \rightarrow Hg^{+2}$	2.0E+9	$(molar\text{-}sec)^{-1}$	Lin and Pehkonen(1997)
$HgSO_3 \rightarrow Hg^0$	$T*e^{((31.971*T)-12595.0)/T)}$ sec^{-1} [T = temperature (K)]		Van Loon et al. (2002)
$Hg(II) + HO_2C \rightarrow Hg^0$	~ 0	$(molar\text{-}sec)^{-1}$	Gardfeldt & Jonnson (2003)
$Hg^0 + HOCl \rightarrow Hg^{+2}$	2.1E+6	$(molar\text{-}sec)^{-1}$	Lin and Pehkonen(1998)
$Hg^0 + OCl^{-1} \rightarrow Hg^{+2}$	2.0E+6	$(molar\text{-}sec)^{-1}$	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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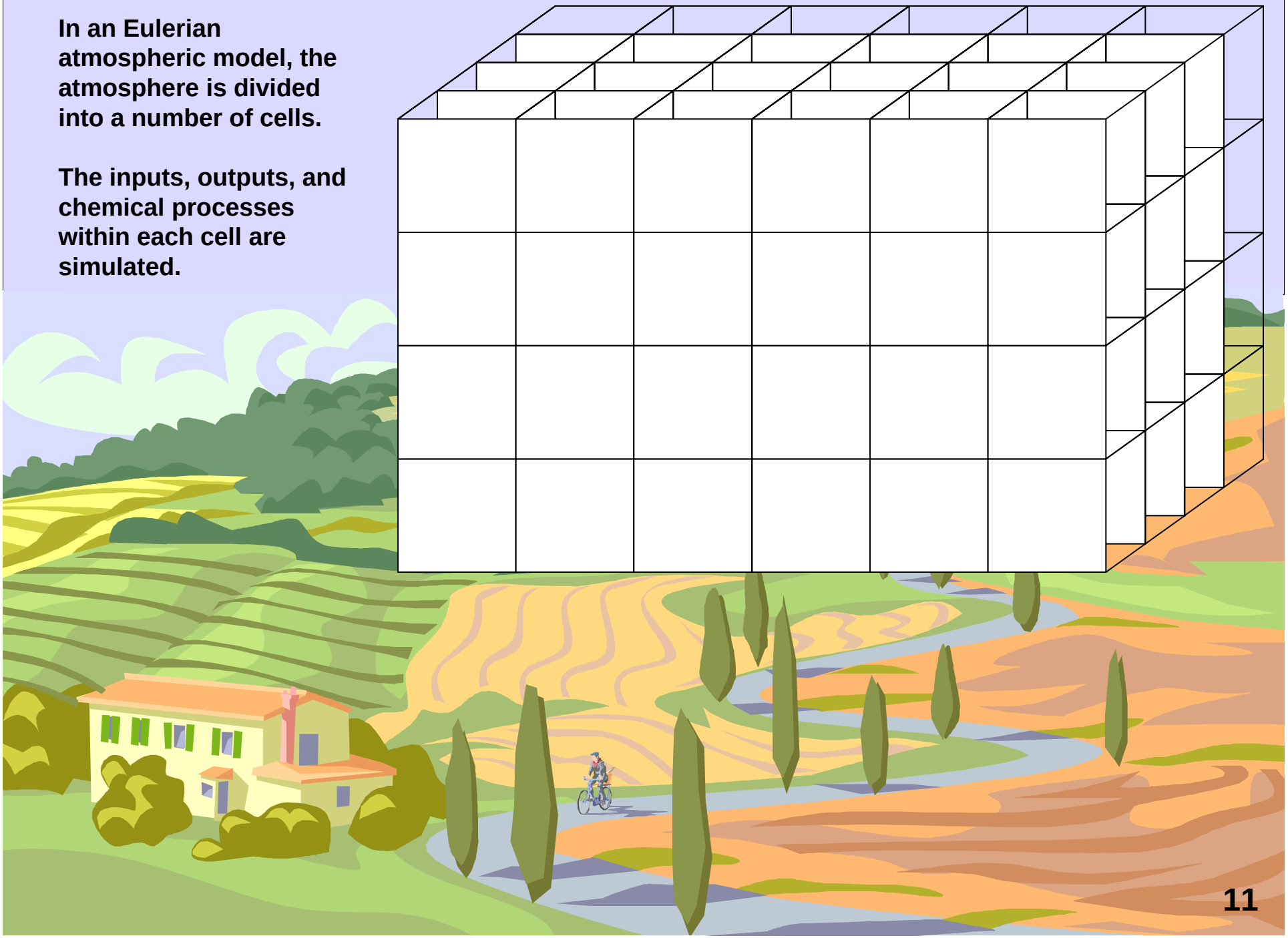
What is an atmospheric model?

- **a computer simulation of the fate and transport of emitted pollutants**

- **two different types of models**
  - **Eulerian**
  - **Lagrangian**

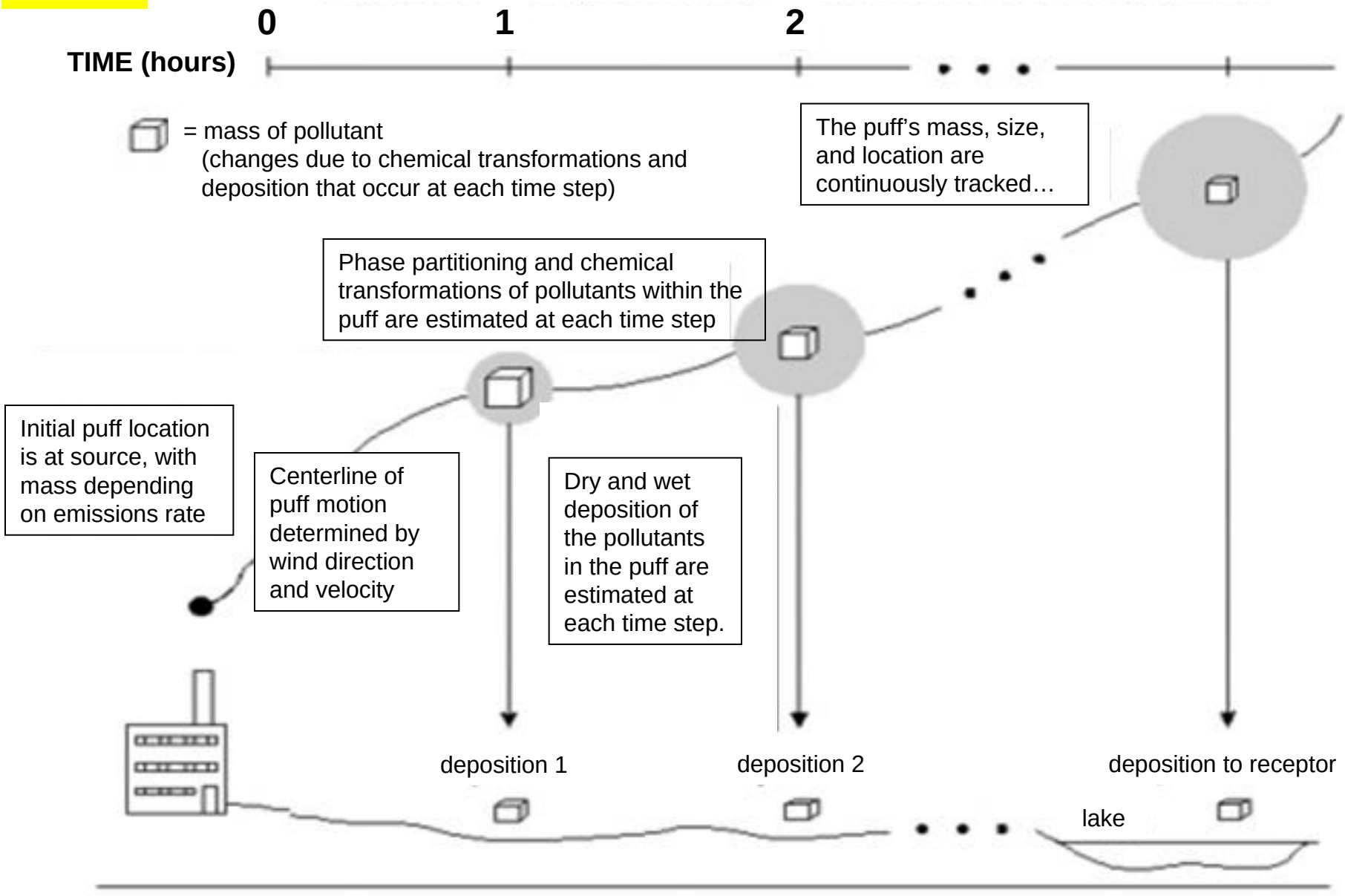
In an Eulerian atmospheric model, the atmosphere is divided into a number of cells.

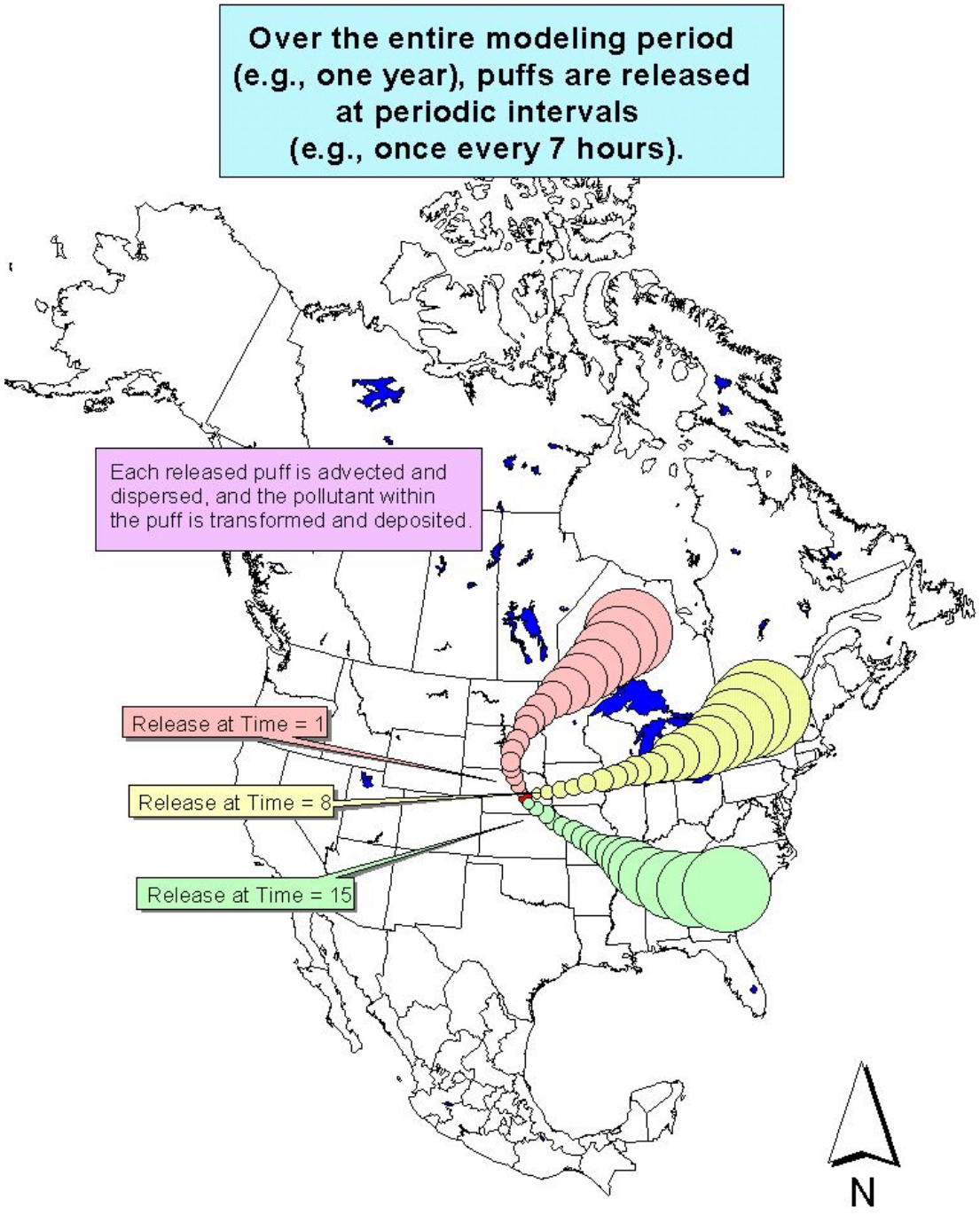
The inputs, outputs, and chemical processes within each cell are simulated.



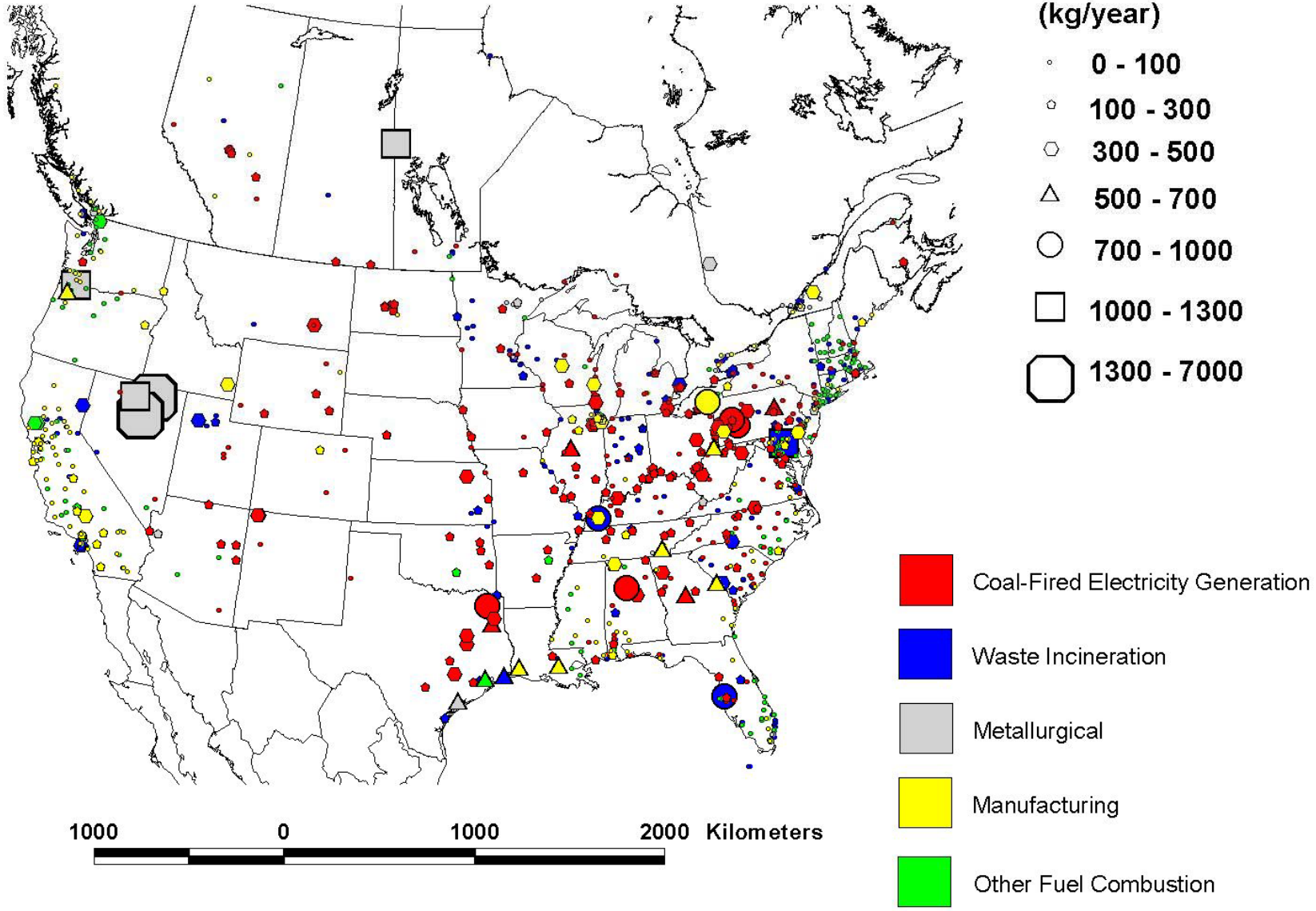
NOAA HYSPLIT MODEL

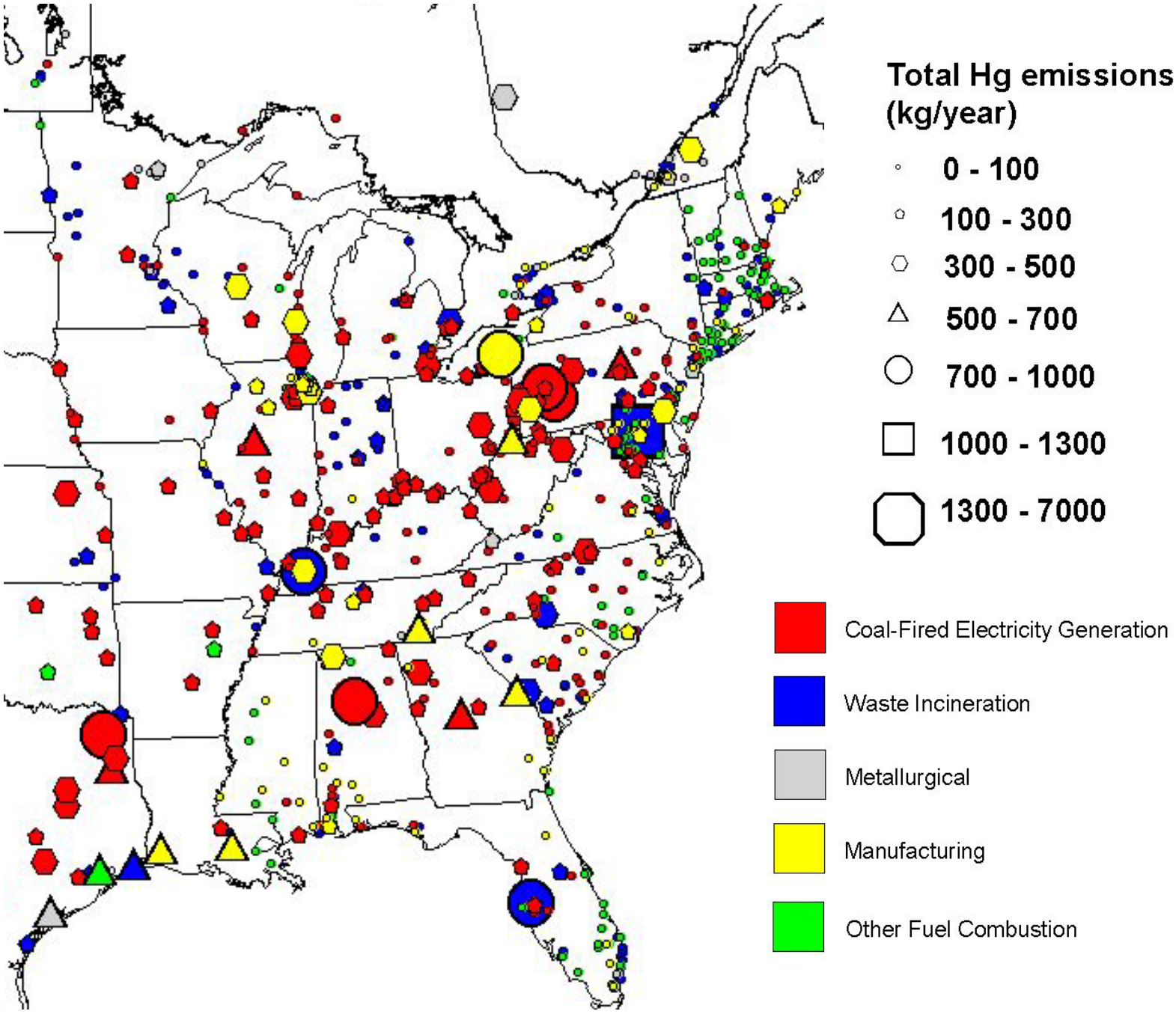
Lagrangian Puff Atmospheric Fate and Transport Model




Over the entire modeling period (e.g., one year), puffs are released at periodic intervals (e.g., once every 7 hours).
Each released puff is advected and dispersed, and the pollutant within the puff is transformed and deposited.
Release at Time = 1
Release at Time = 8
Release at Time = 15
N

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

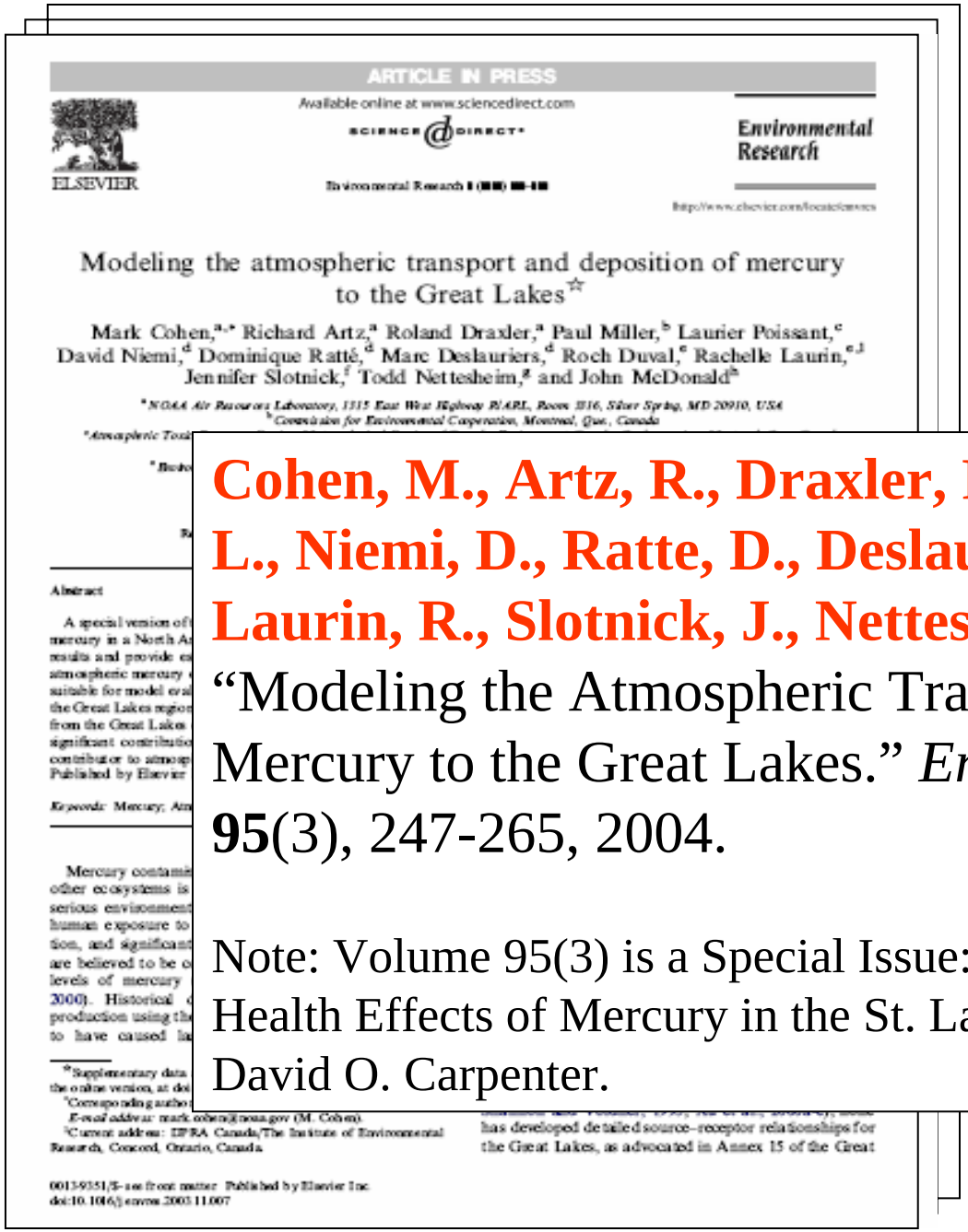



Total Hg emissions
(kg/year)
0 - 100
100 - 300
300 - 500
500 - 700
700 - 1000
1000 - 1300
1300 - 7000
Coal-Fired Electricity Generation
Waste Incineration
Metallurgical
Manufacturing
Other Fuel Combustion

- **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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Modeling the atmospheric transport and deposition of mercury to the Great Lakes☆

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Abstract

Keywords: Mercury; Atm

Mercury contami
other ecosystems is
serious environment
human exposure to
tion, and significant
are believed to be c
levels of mercury
2000). Historical
production using th
to have caused la

*Supplementary data
the online version, at do
*Corresponding autho
E-mail address: mark.cohen@noaa.gov (M. Cohen).
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has developed detailed source–receptor relationships for the Great Lakes, as advocated in Annex 15 of the Great

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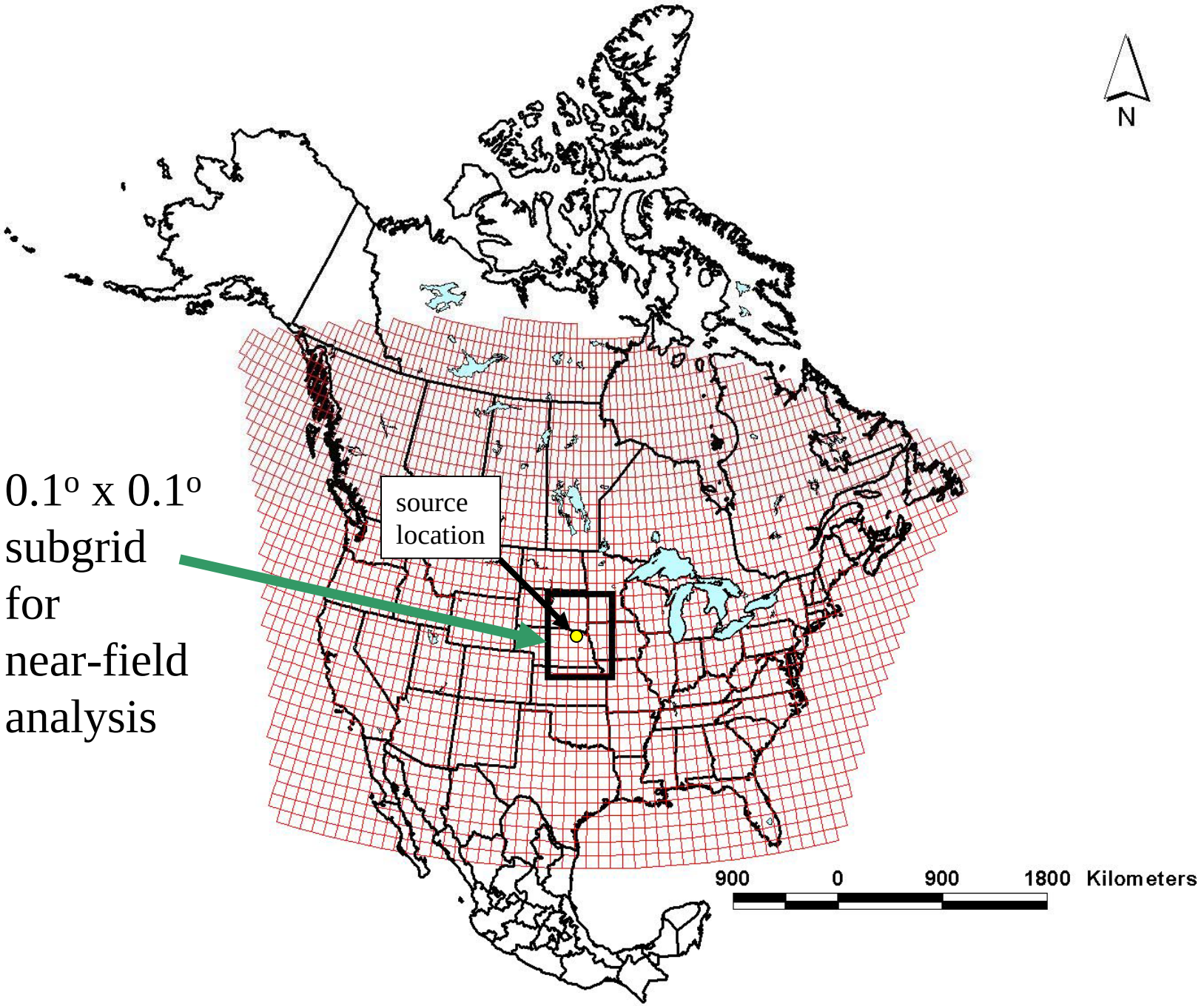
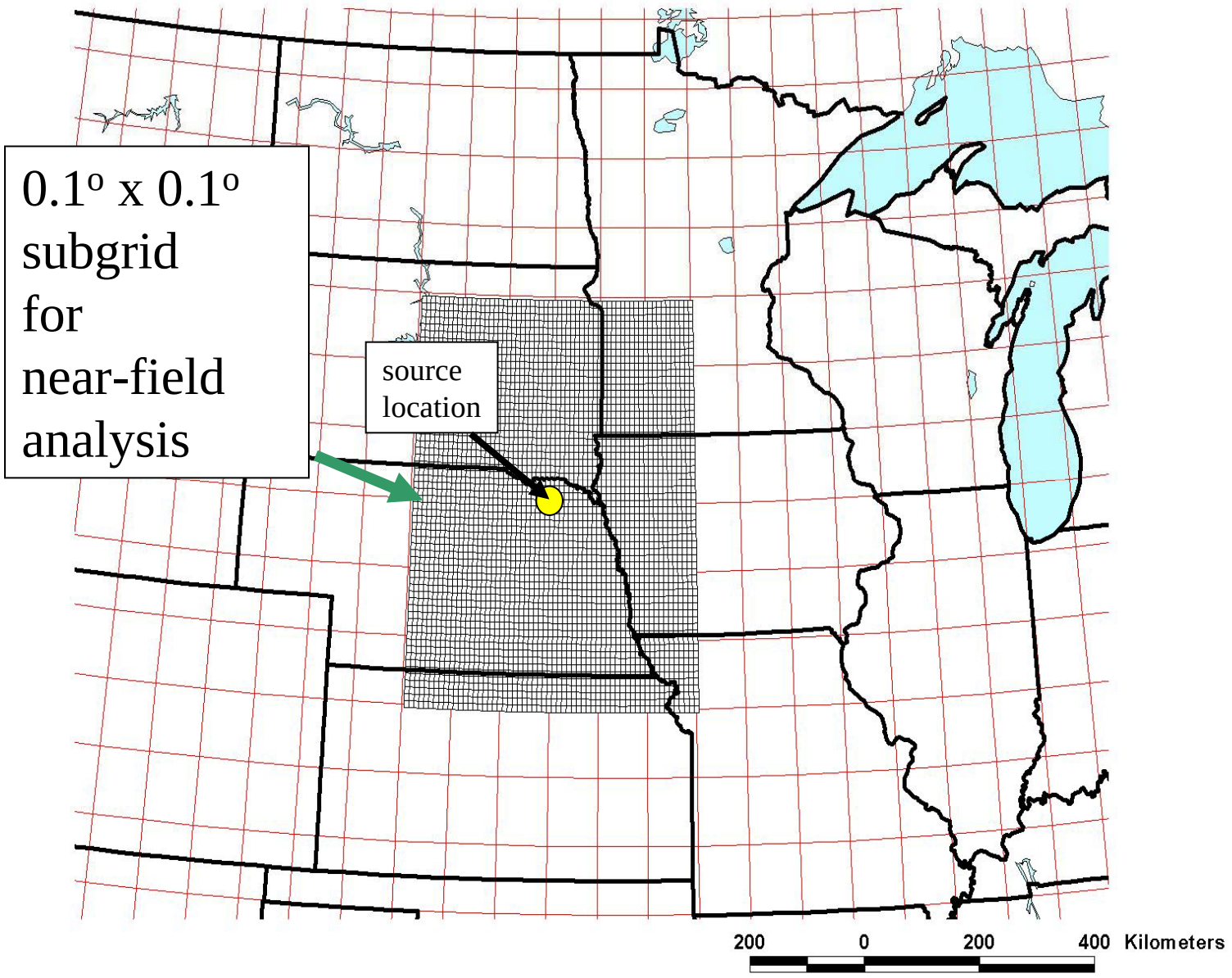
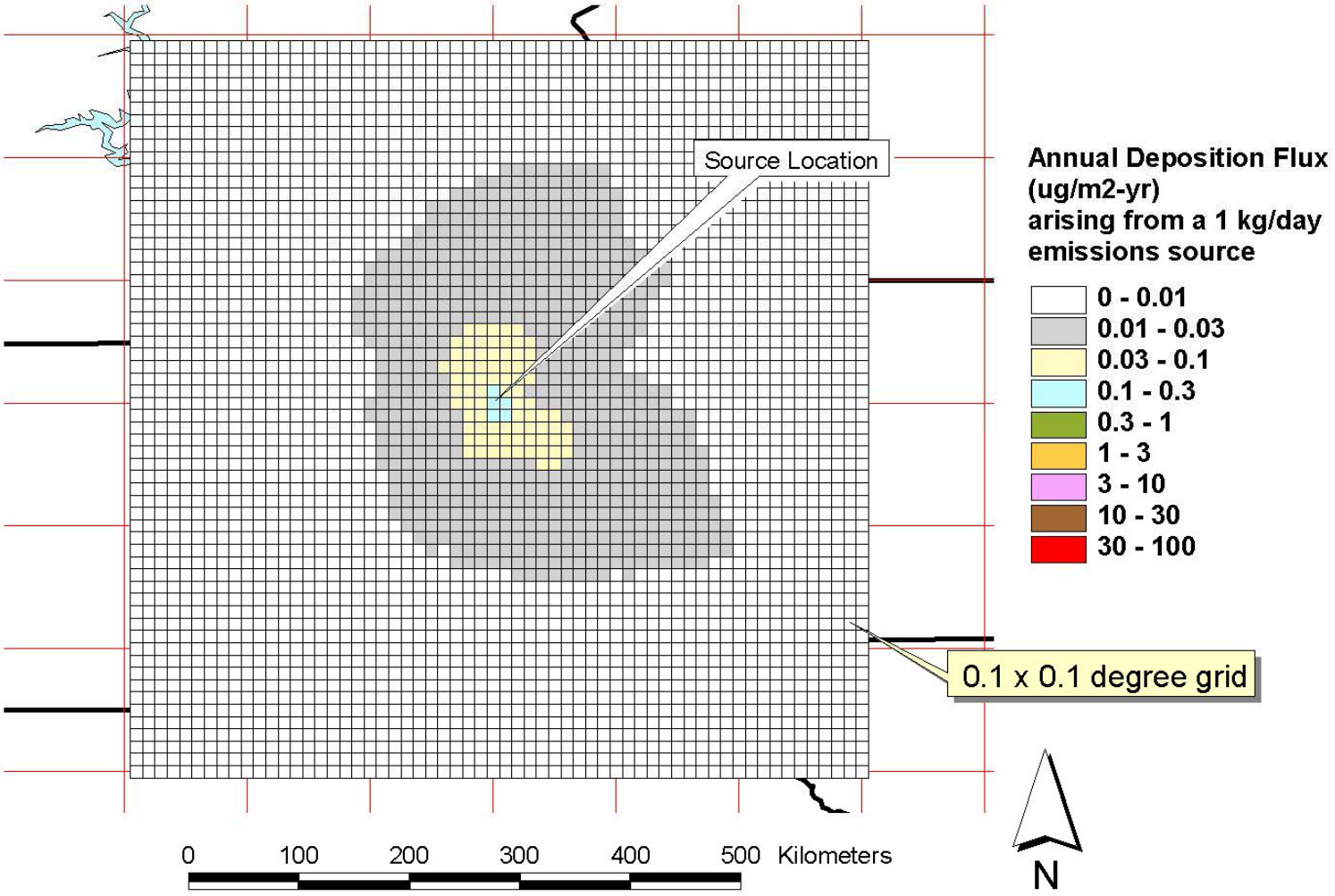
Note: Volume 95(3) is a Special Issue: "An Ecosystem Approach to Health Effects of Mercury in the St. Lawrence Great Lakes", edited by David O. Carpenter.



0.1^{o} x 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



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simulation for entire year 1996 using archived NGM meteorology (180 km resolution)

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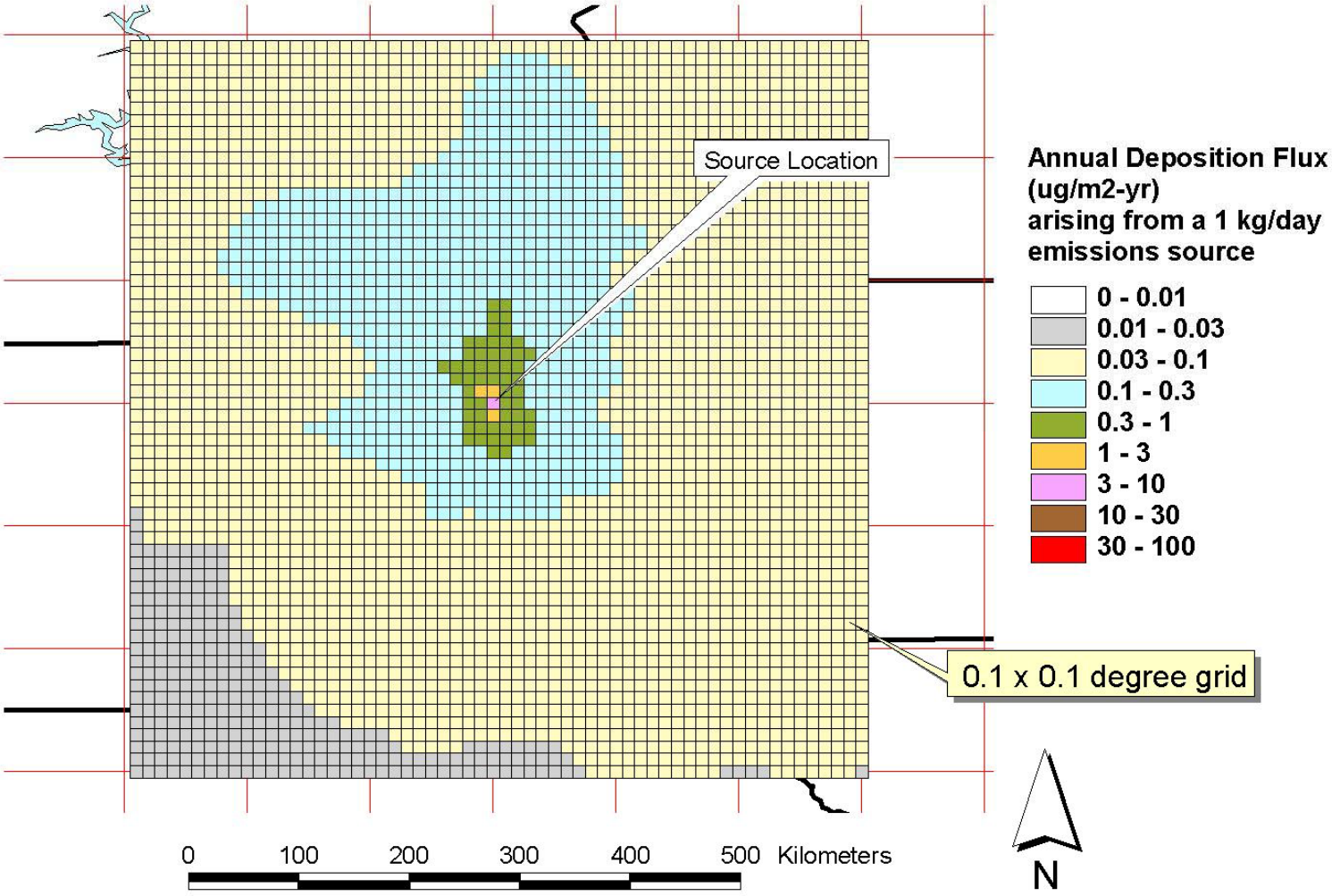

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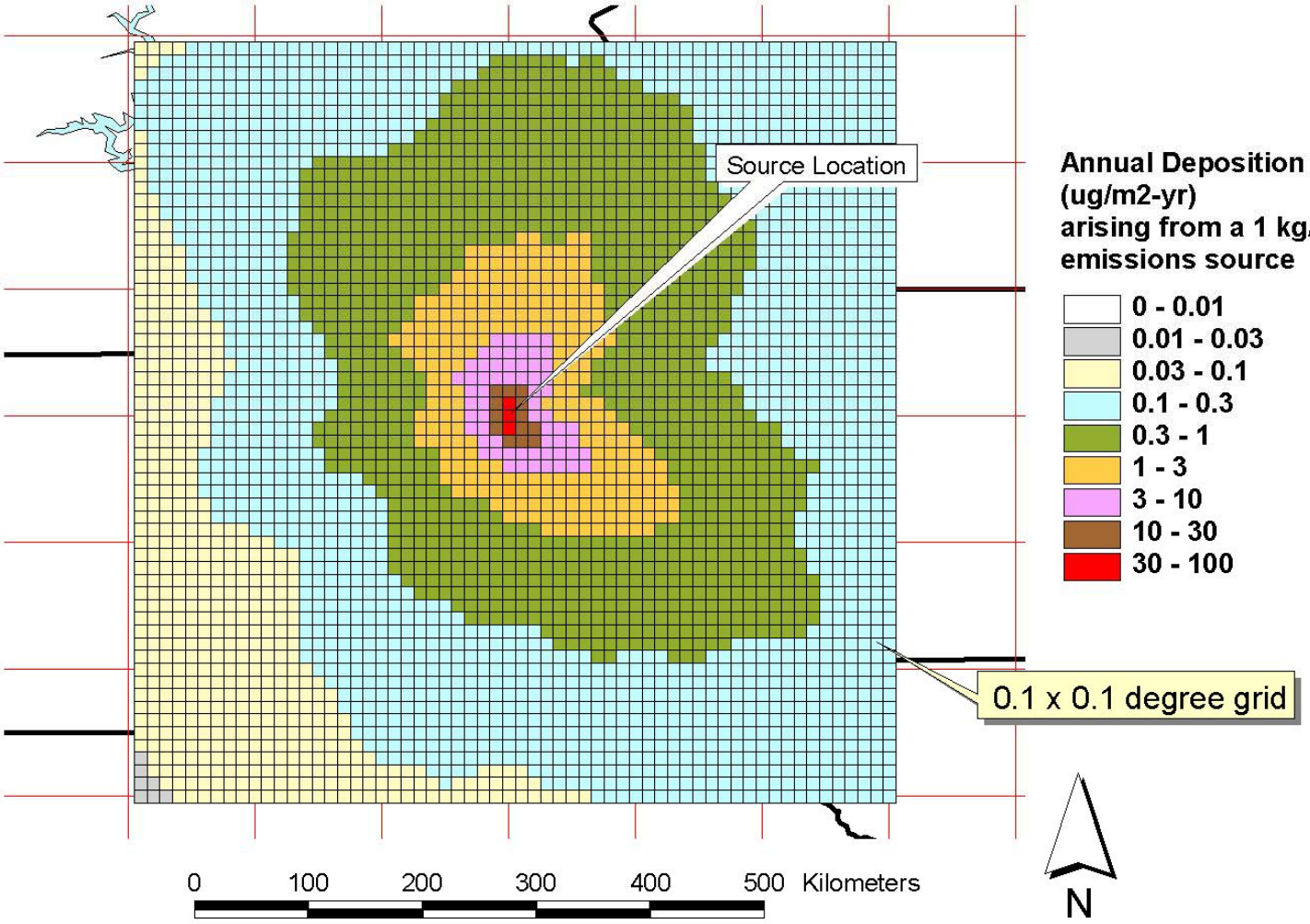

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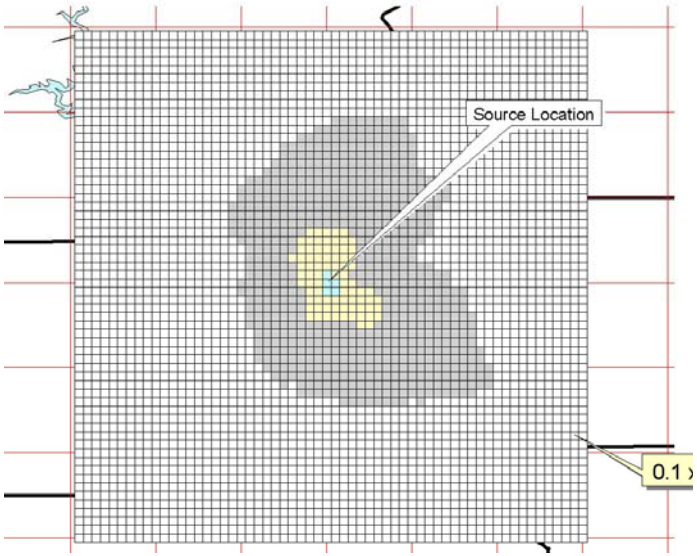

Annual Deposition Flux (ug/m2-yr) arising from a 1 kg/day emissions source

- 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

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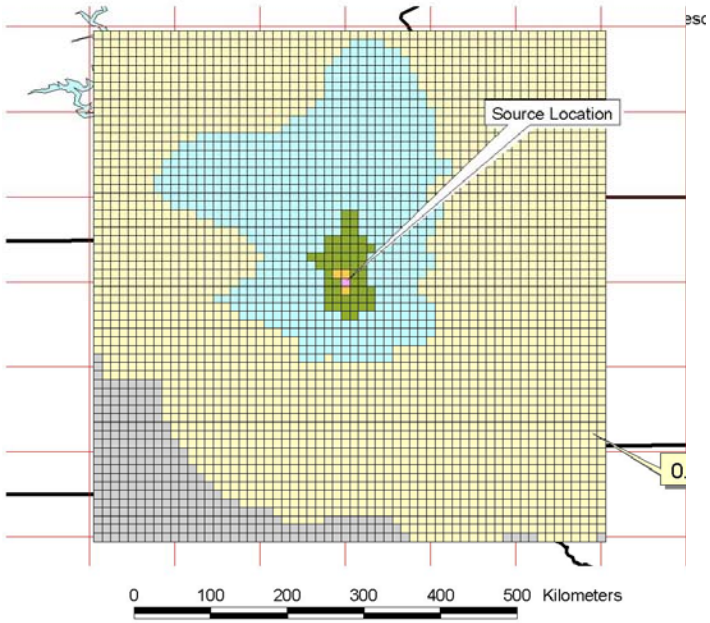

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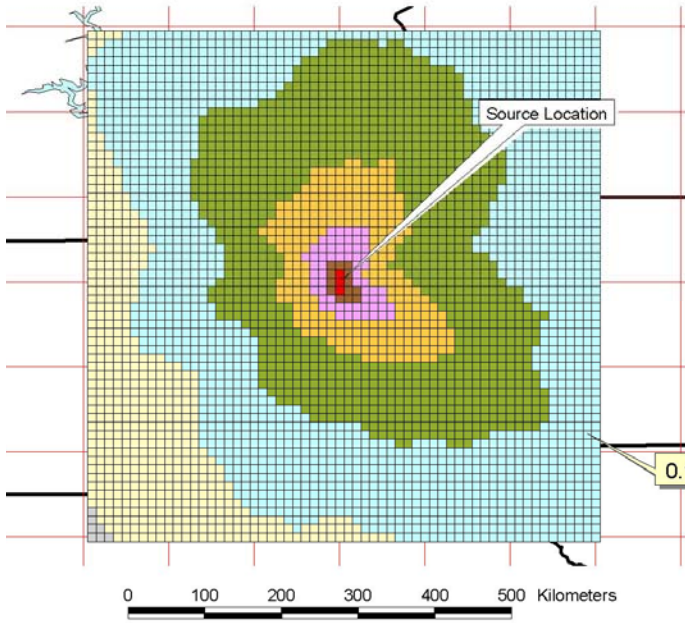

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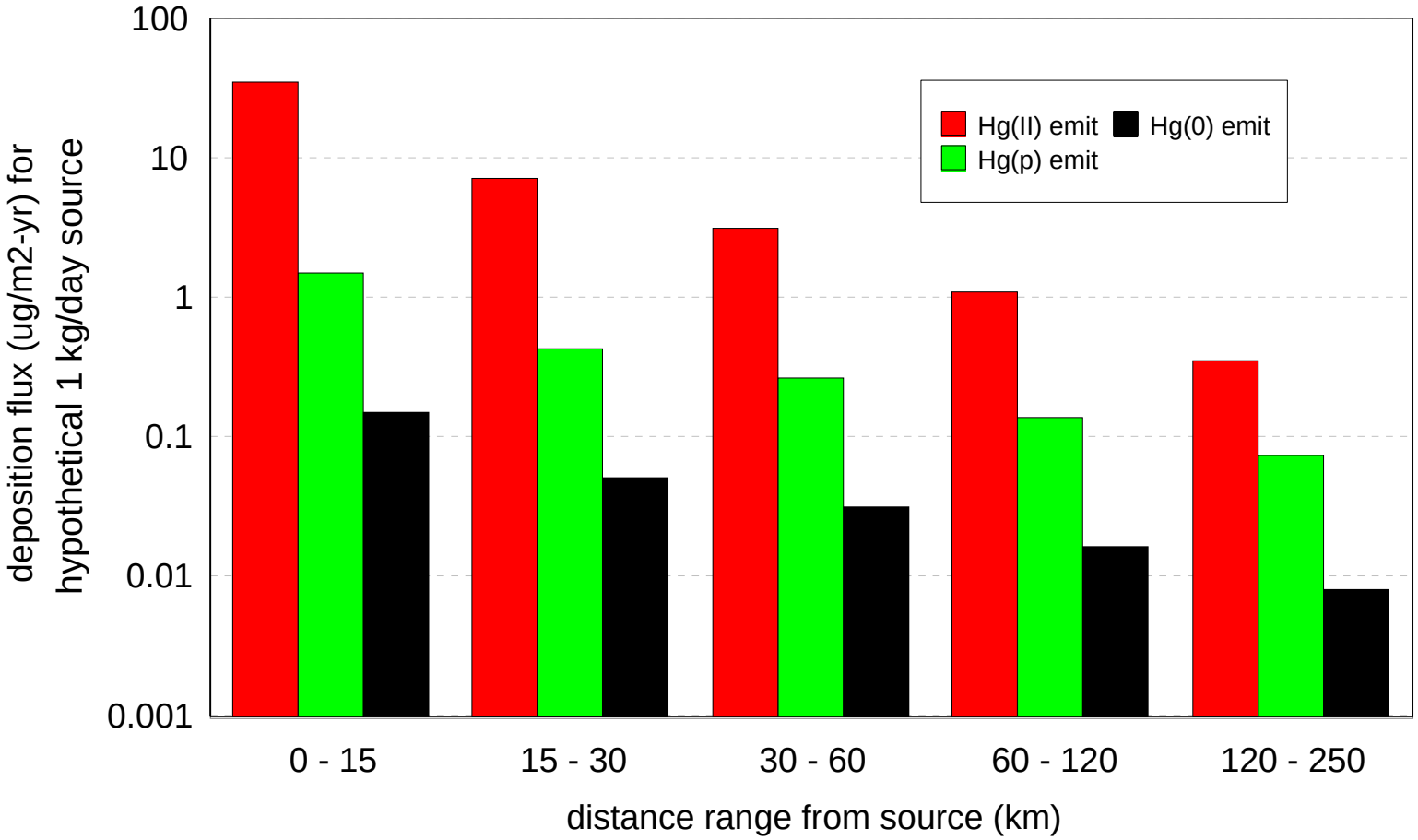
Why is emissions speciation information critical?



Logarithmic

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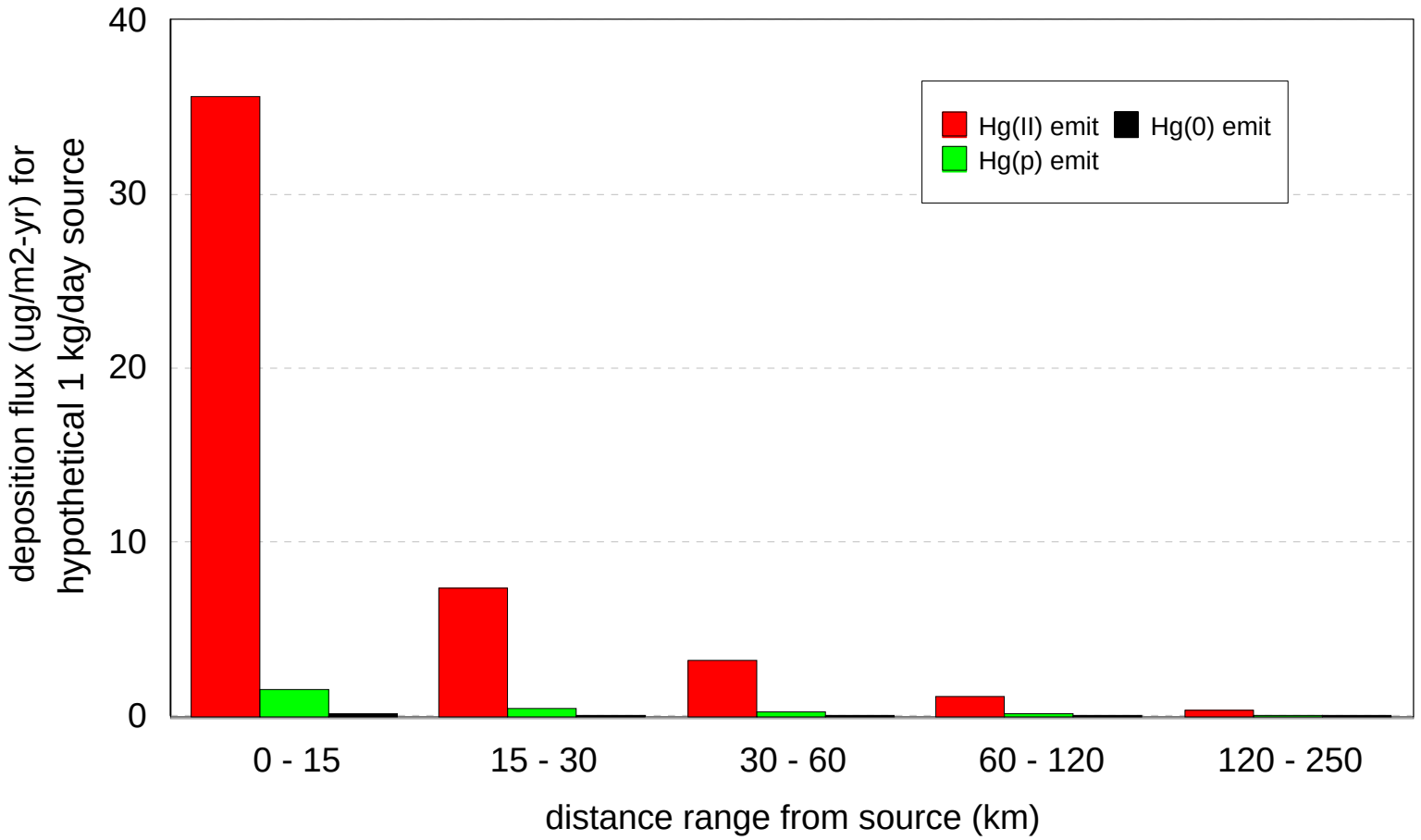

Linear

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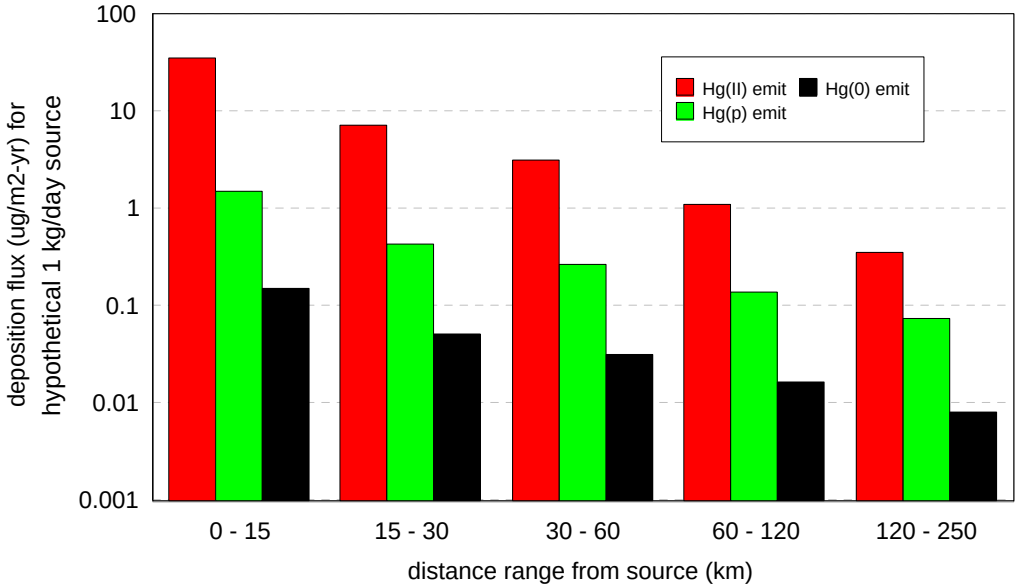
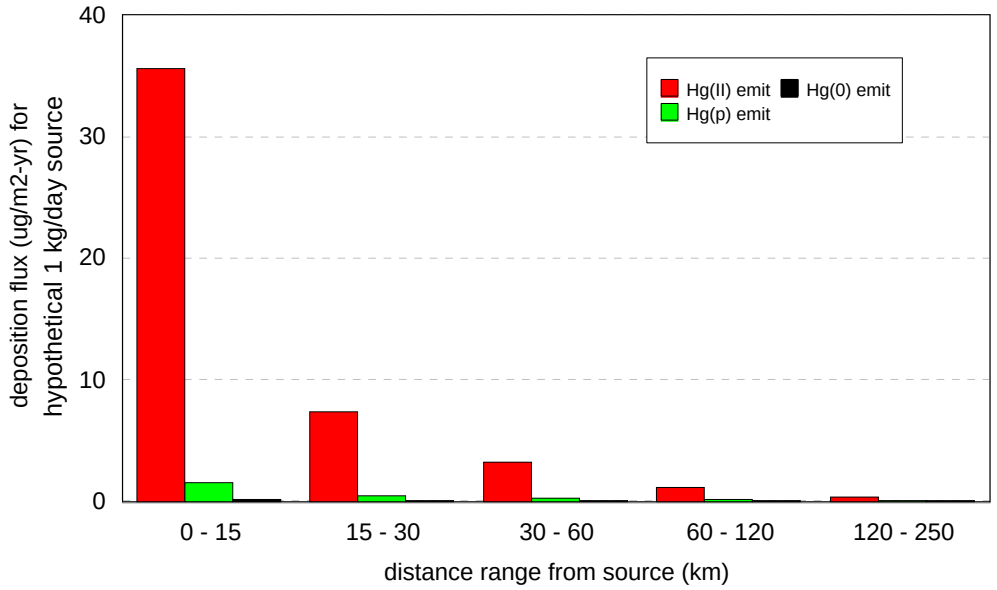
Logarithmic



Linear



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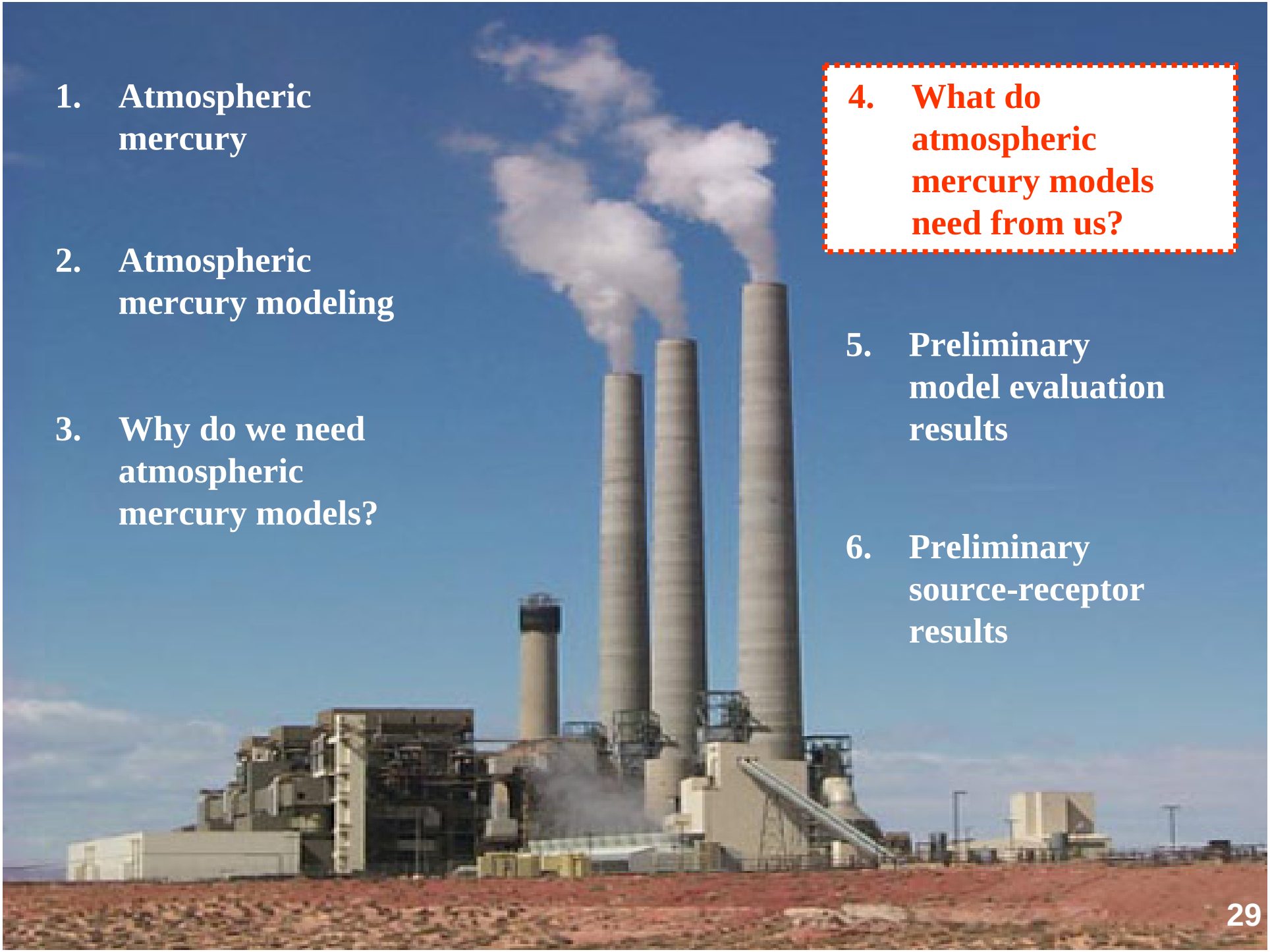
Why do we need atmospheric mercury models?

- 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



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What do atmospheric mercury models need?

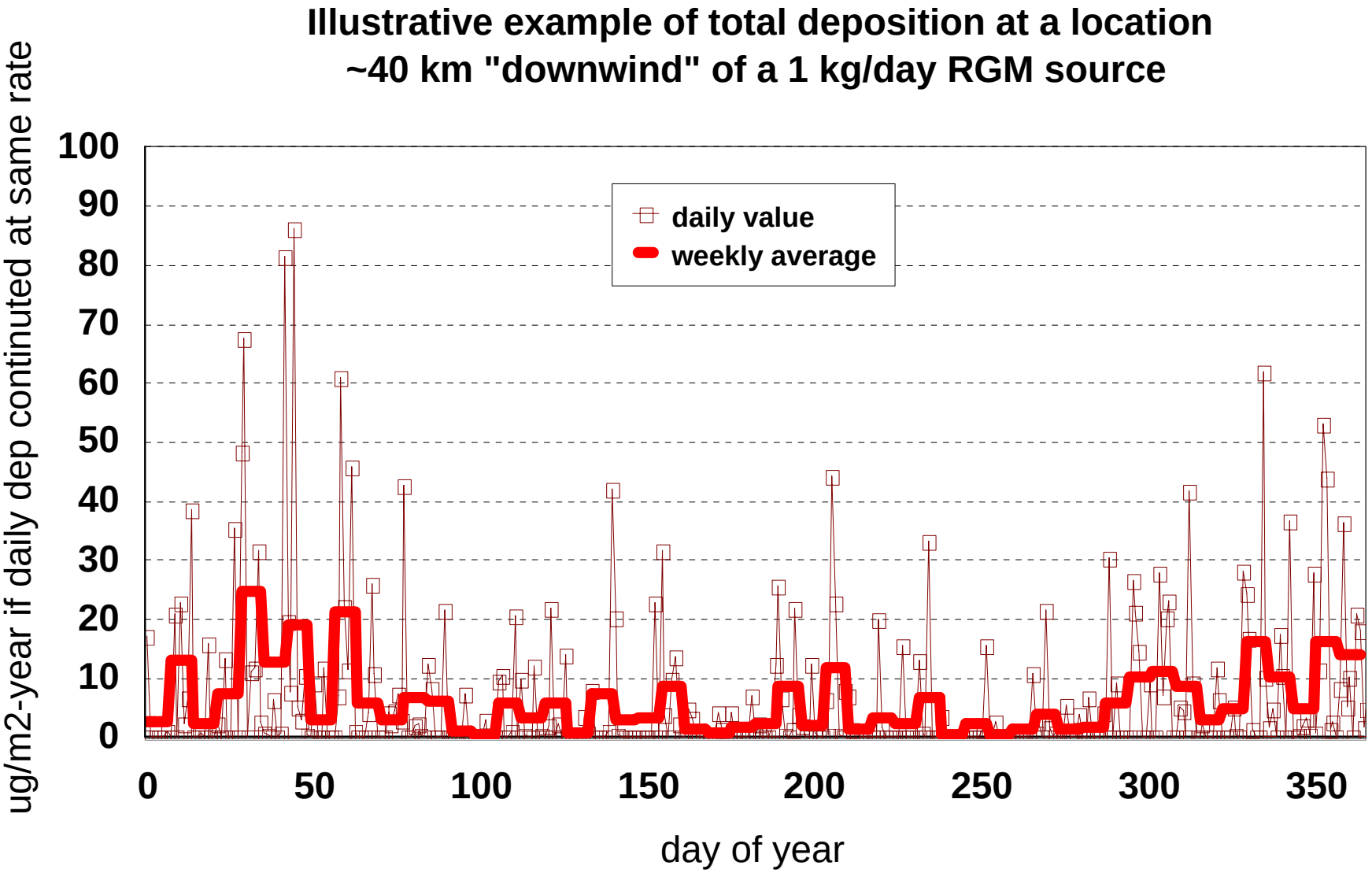
Emissions Inventories

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)


Illustrative example of total deposition at a location
~40 km "downwind" of a 1 kg/day RGM source
ug/m2-year if daily dep continuted at same rate
100
90
80
70
60
50
40
30
20
10
0
daily value
weekly average
0
50
100
150
200
250
300
350
day of year

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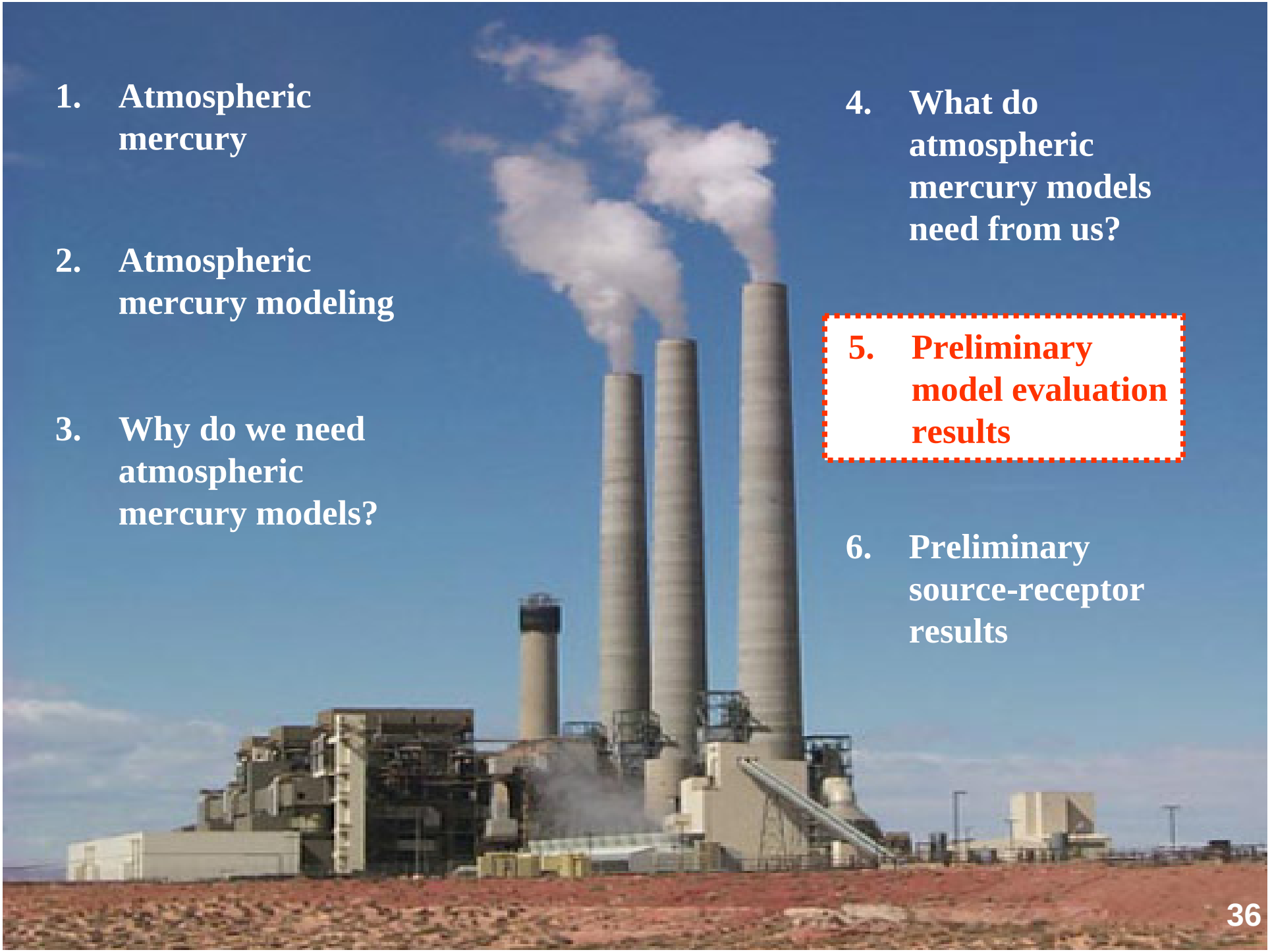

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- EMEP Model Intercomparison
  - Phase II – ambient concentrations
  - Phase III – wet and dry deposition
- Chesapeake Bay region

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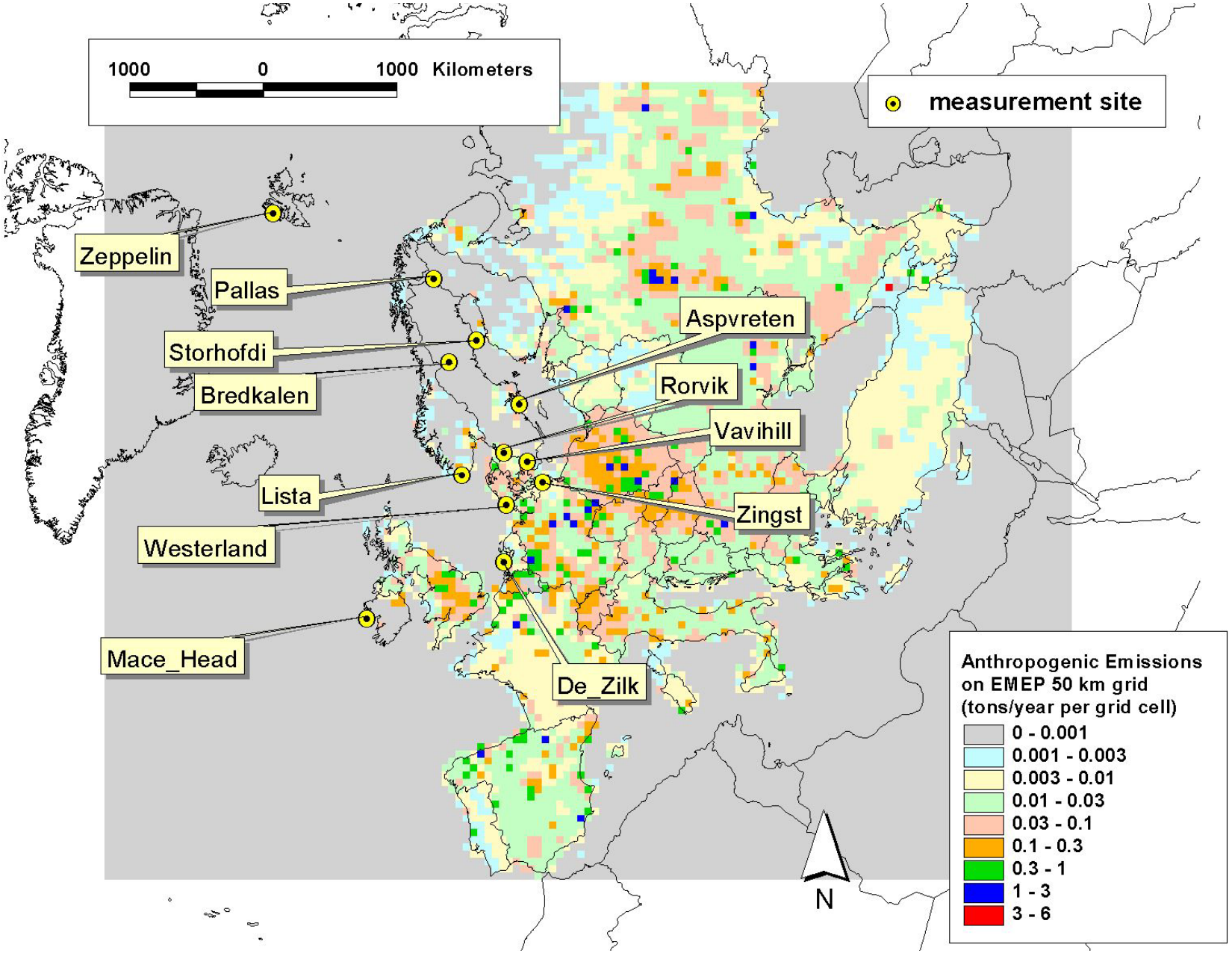
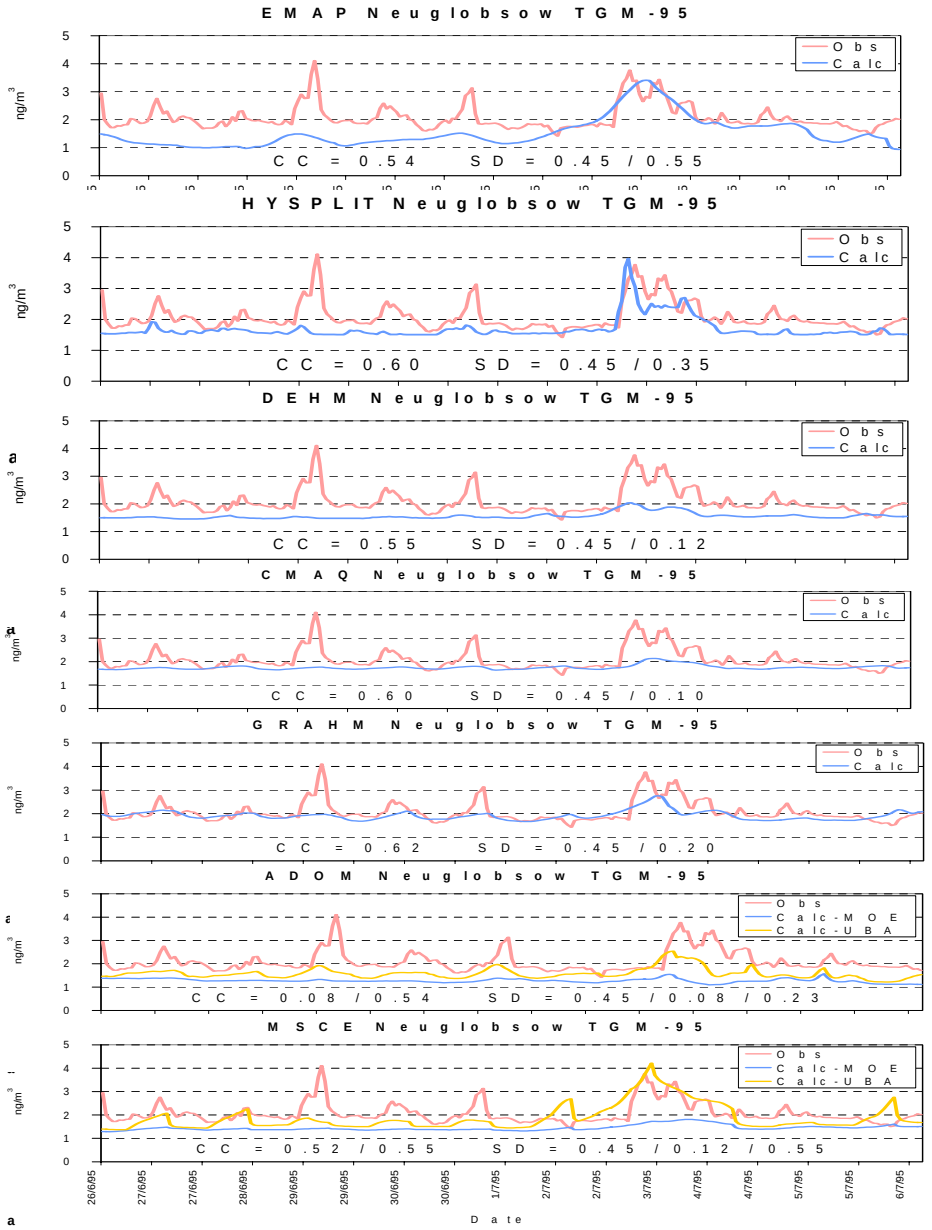
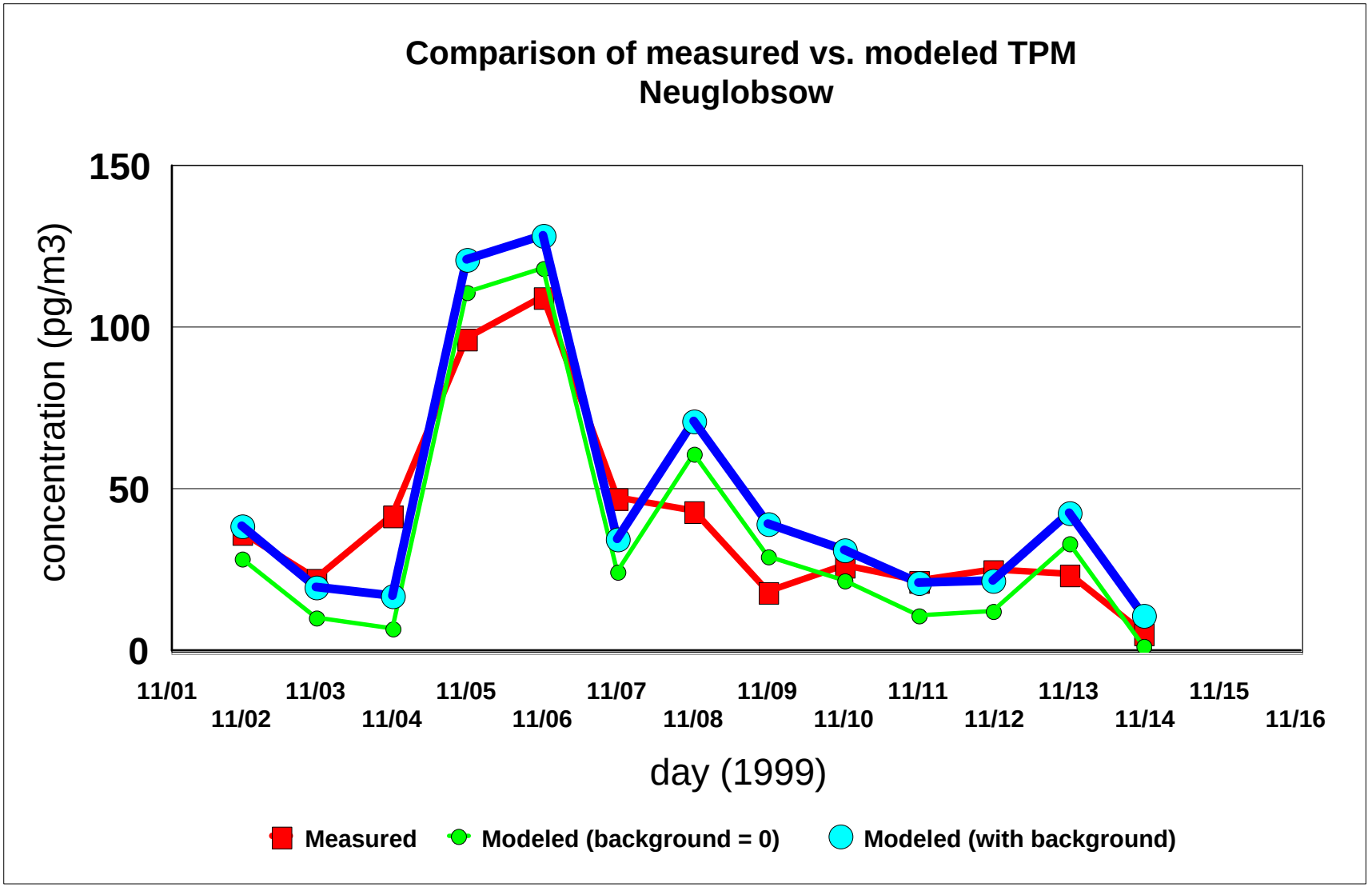
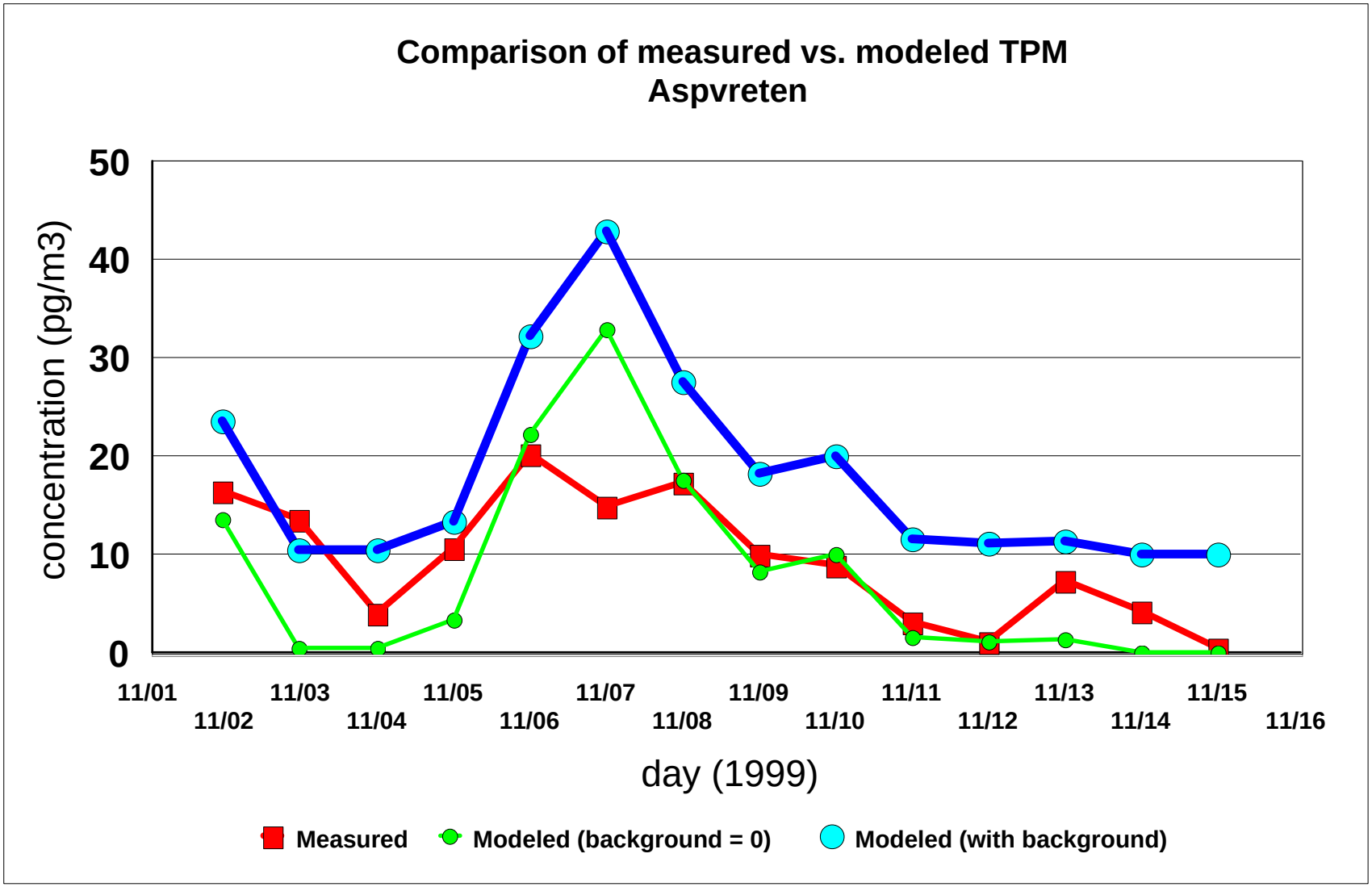
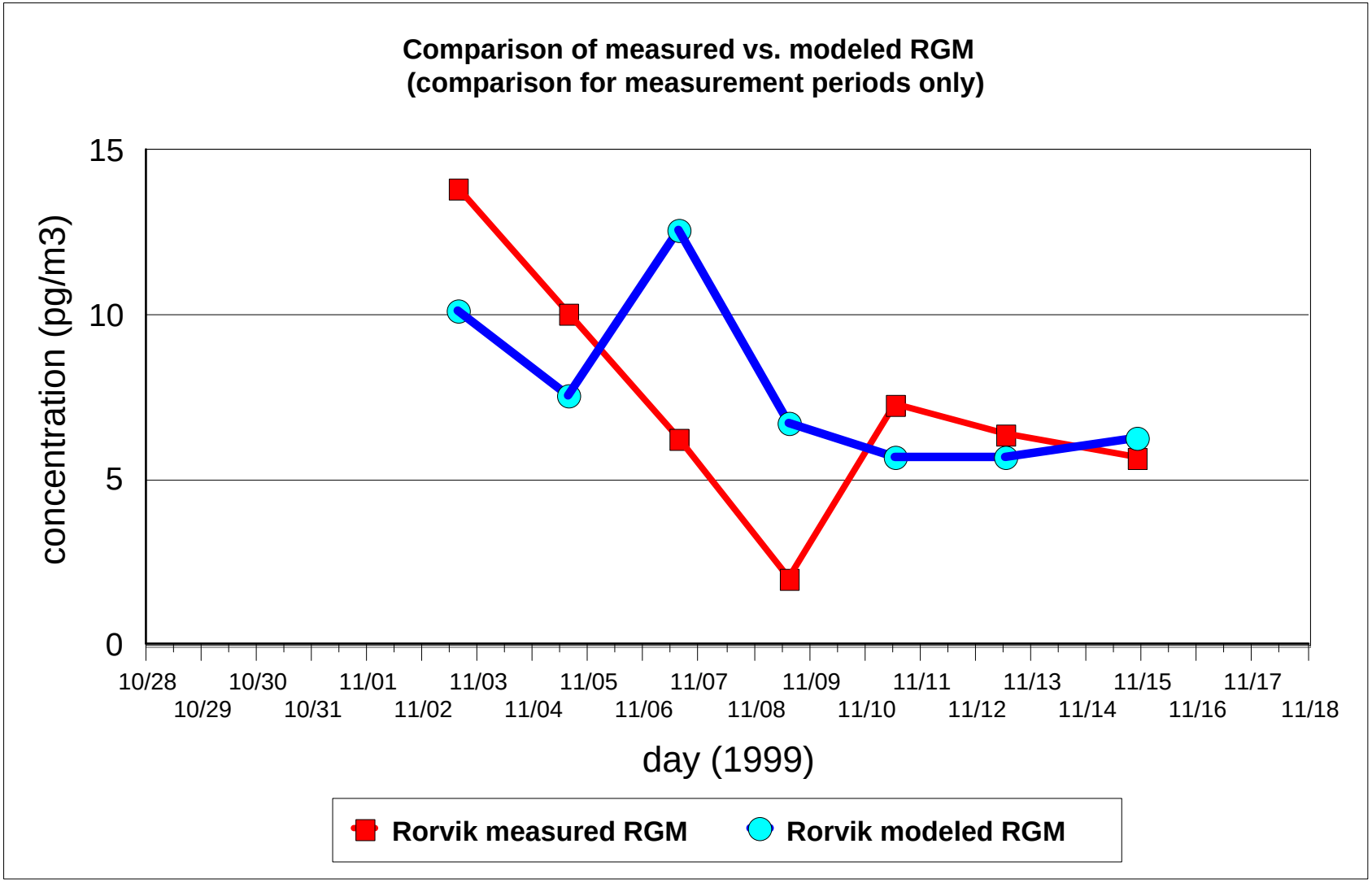

1000 0 1000 Kilometers
measurement site
Zeppelin
Pallas
Storhofdi
Bredkalen
Aspvreten
Rorvik
Vavihill
Lista
Zingst
Westerland
Mace_Head
De_Zilk
N
Anthropogenic Emissions on EMEP 50 km grid (tons/year per grid cell)
0 - 0.001
0.001 - 0.003
0.003 - 0.01
0.01 - 0.03
0.03 - 0.1
0.1 - 0.3
0.3 - 1
1 - 3
3 - 6

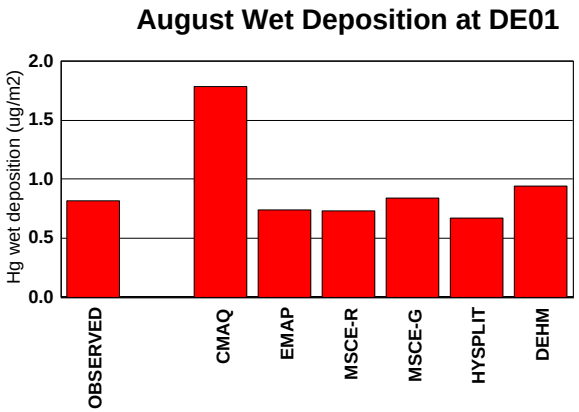
Measured and Simulated Total Gaseous Mercury at Neuglobsow during the 1995 episode

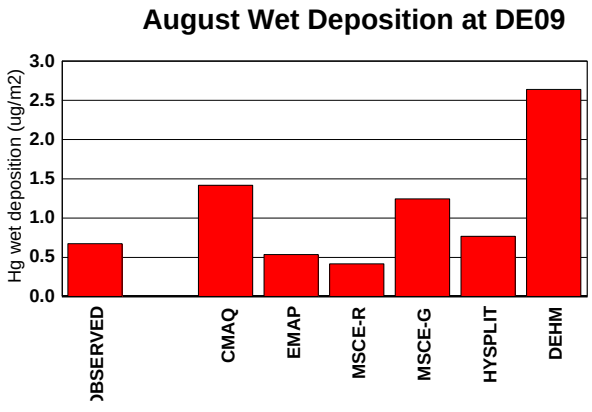


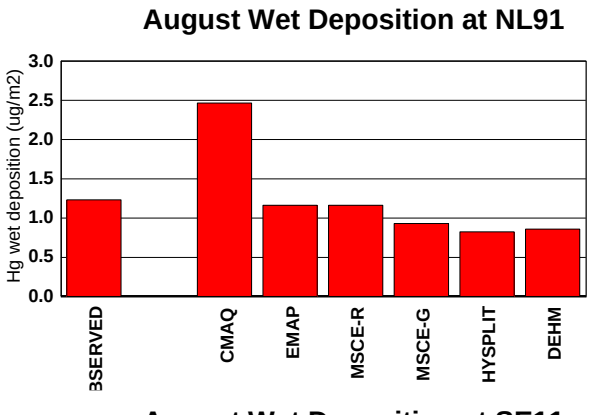

Comparison of measured vs. modeled TPM
Neuglobsow
150
100
50
0
concentration (pg/m3)
11/01
11/02
11/03
11/04
11/05
11/06
11/07
11/08
11/09
11/10
11/11
11/12
11/13
11/14
11/15
11/16
day (1999)
Measured
Modeled (background = 0)
Modeled (with background)

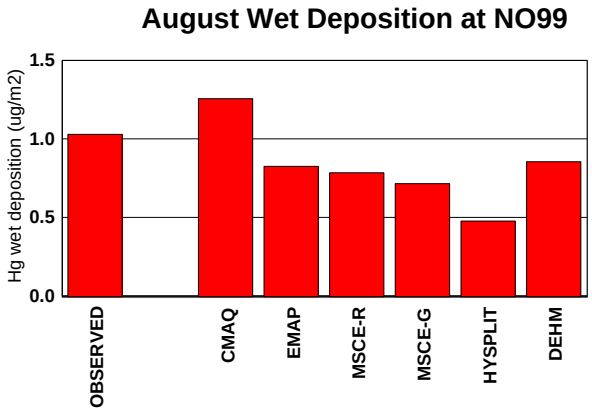

Comparison of measured vs. modeled TPM
Aspvreten
concentration (pg/m3)
50
40
30
20
10
0
11/01
11/02
11/03
11/04
11/05
11/06
11/07
11/08
11/09
11/10
11/11
11/12
11/13
11/14
11/15
11/16
day (1999)
Measured
Modeled (background = 0)
Modeled (with background)

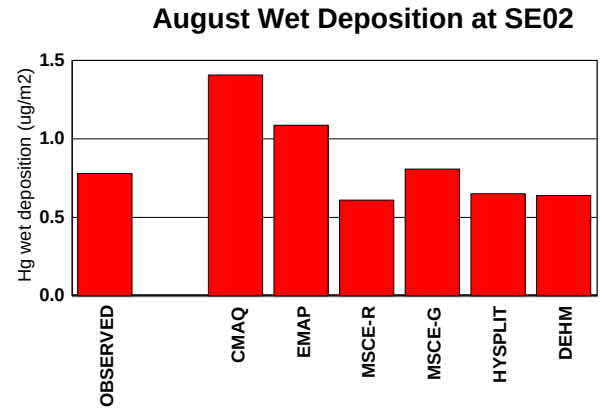

Comparison of measured vs. modeled RGM
(comparison for measurement periods only)
concentration (pg/m3)
15
10
5
0
10/28
10/29
10/30
10/31
11/01
11/02
11/03
11/04
11/05
11/06
11/07
11/08
11/09
11/10
11/11
11/12
11/13
11/14
11/15
11/16
11/17
11/18
day (1999)
Rorvik measured RGM
Rorvik modeled RGM

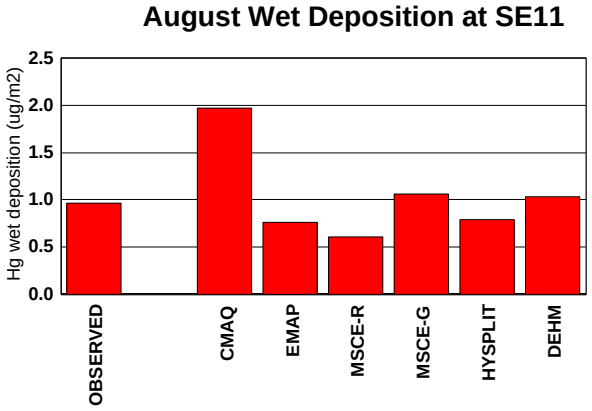

August Wet Deposition at DE01
Hg wet deposition (ug/m2)
2.0
1.5
1.0
0.5
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM

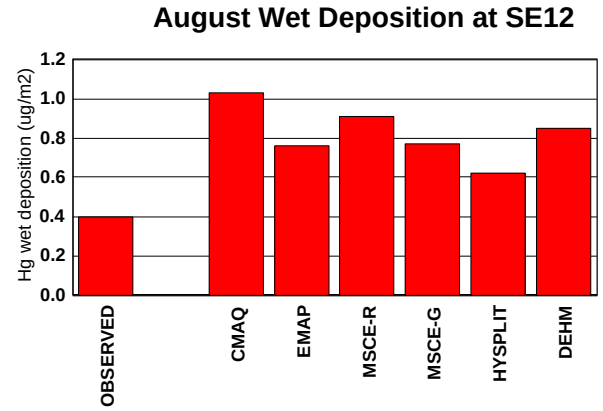

August Wet Deposition at DE09
Hg wet deposition (ug/m2)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM

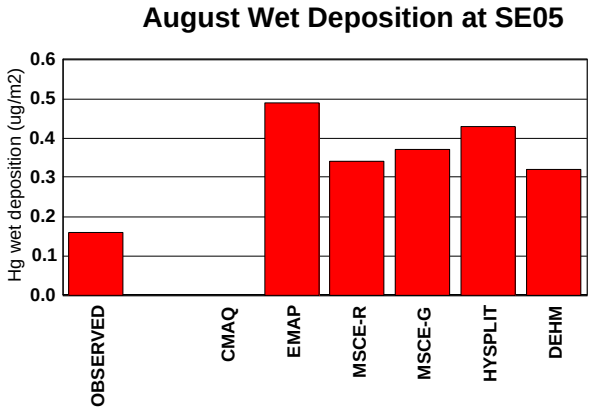

August Wet Deposition at NL91
Hg wet deposition (ug/m2)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM

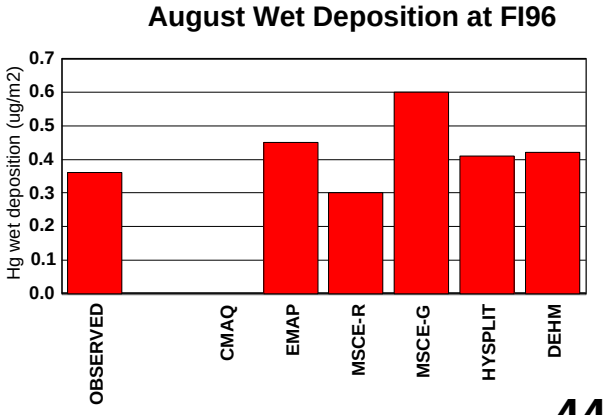

August Wet Deposition at NO99
Hg wet deposition (ug/m2)
1.5
1.0
0.5
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM


August Wet Deposition at SE02
Hg wet deposition (ug/m2)
1.5
1.0
0.5
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM


August Wet Deposition at SE11
Hg wet deposition (ug/m2)
2.5
2.0
1.5
1.0
0.5
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM

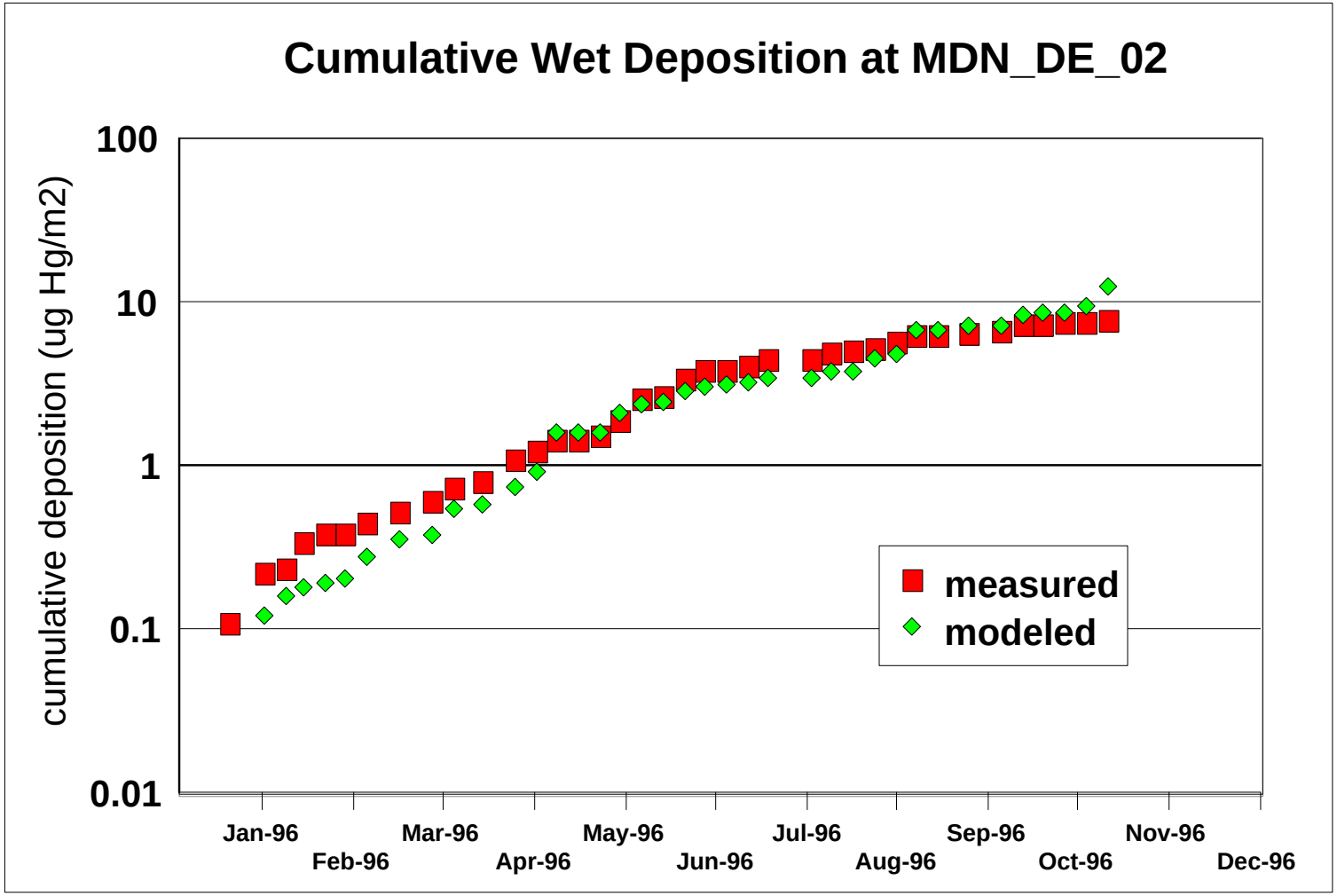

August Wet Deposition at SE12
Hg wet deposition (ug/m2)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM


August Wet Deposition at SE05
Hg wet deposition (ug/m2)
0.6
0.5
0.4
0.3
0.2
0.1
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM

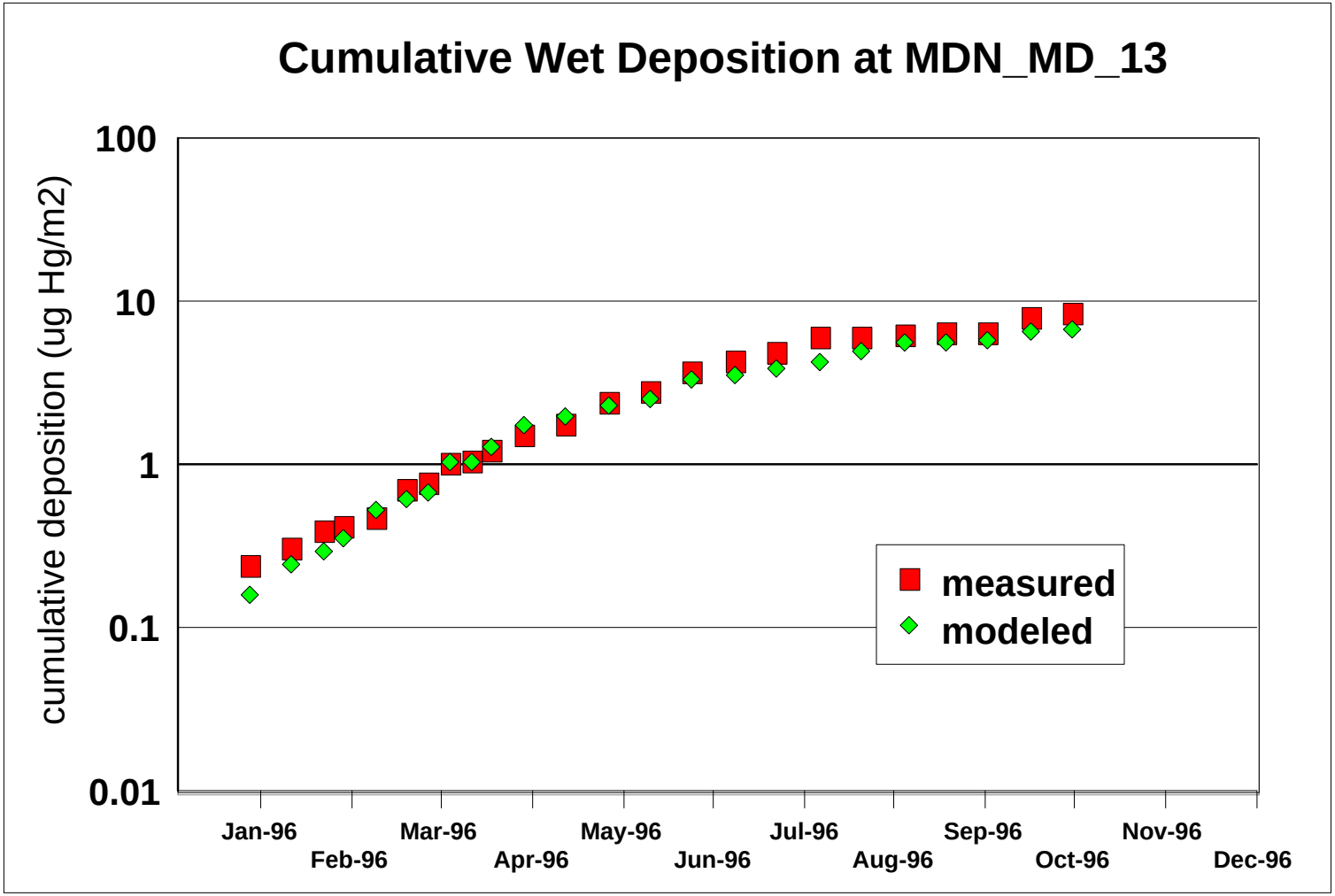

August Wet Deposition at FI96
Hg wet deposition (ug/m2)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
OBSERVED
CMAQ
EMAP
MSCE-R
MSCE-G
HYSPLIT
DEHM

- EMEP Model Intercomparison
  - Phase II – ambient concentrations
  - Phase III – wet and dry deposition
- Chesapeake Bay region

Modeled vs. Measured Wet Deposition at Mercury Deposition Network Site DE_02 during 1996



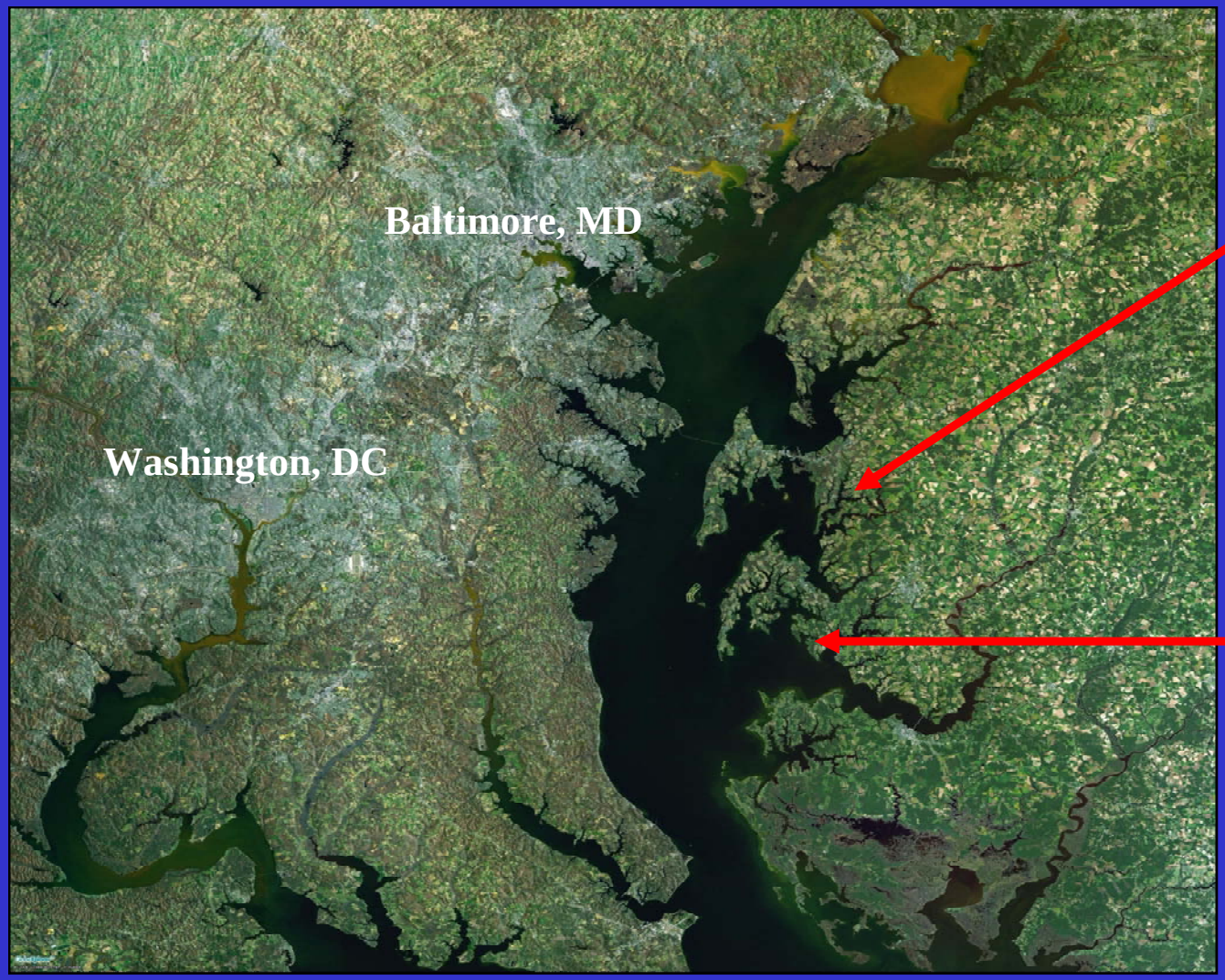
Modeled vs. Measured Wet Deposition at Mercury Deposition Network Site MD_13 during 1996

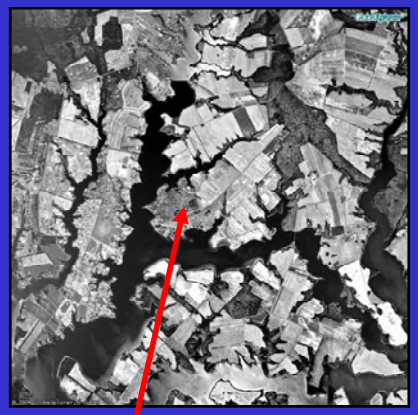


Summer 2004 Chesapeake Bay Atmospheric Hg Study (June – August 2004)

- NOAA Cooperative Oxford Lab: ***Bob Wood***
- NOAA Air Resources Lab Atmospheric Turbulence and Diffusion Division (ATDD): ***Steve Brooks***
- NOAA Air Resources Lab HQ Division: ***Winston Luke, Paul Kelley, Mark Cohen, Richard Artz***
- NOAA Chesapeake Bay Office: ***Maggie Kerchner***
- Frontier GeoSciences: ***Bob Brunette, Gerard van der Jagt, Eric Prestbo***
- Univ. of MD Wye Res. and Educ. Center: ***Mike Newall***

Summer 2004 Measurement Sites

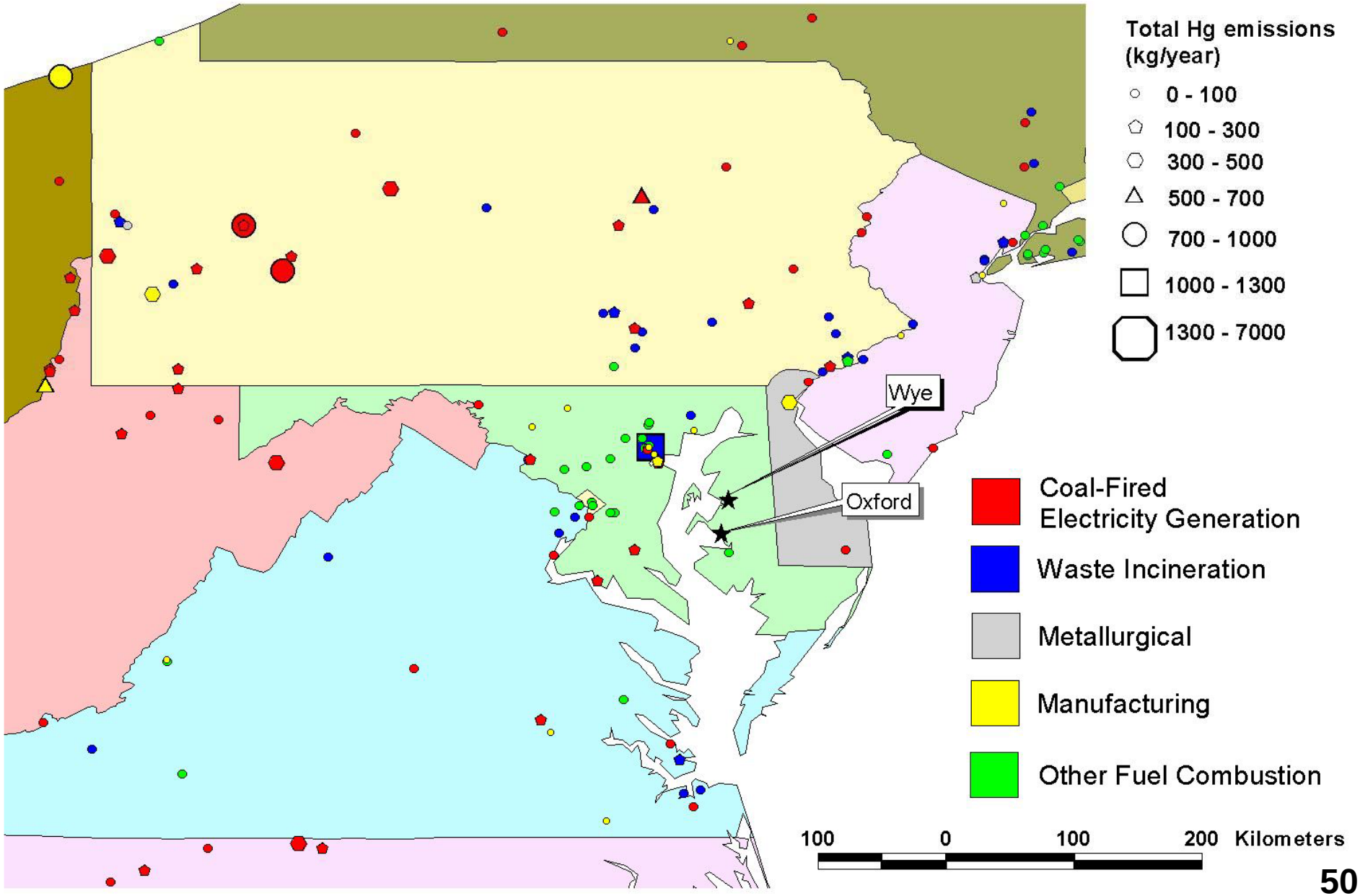




Wye Research and
Education Center
(38.9131EN, 76.1525EW)

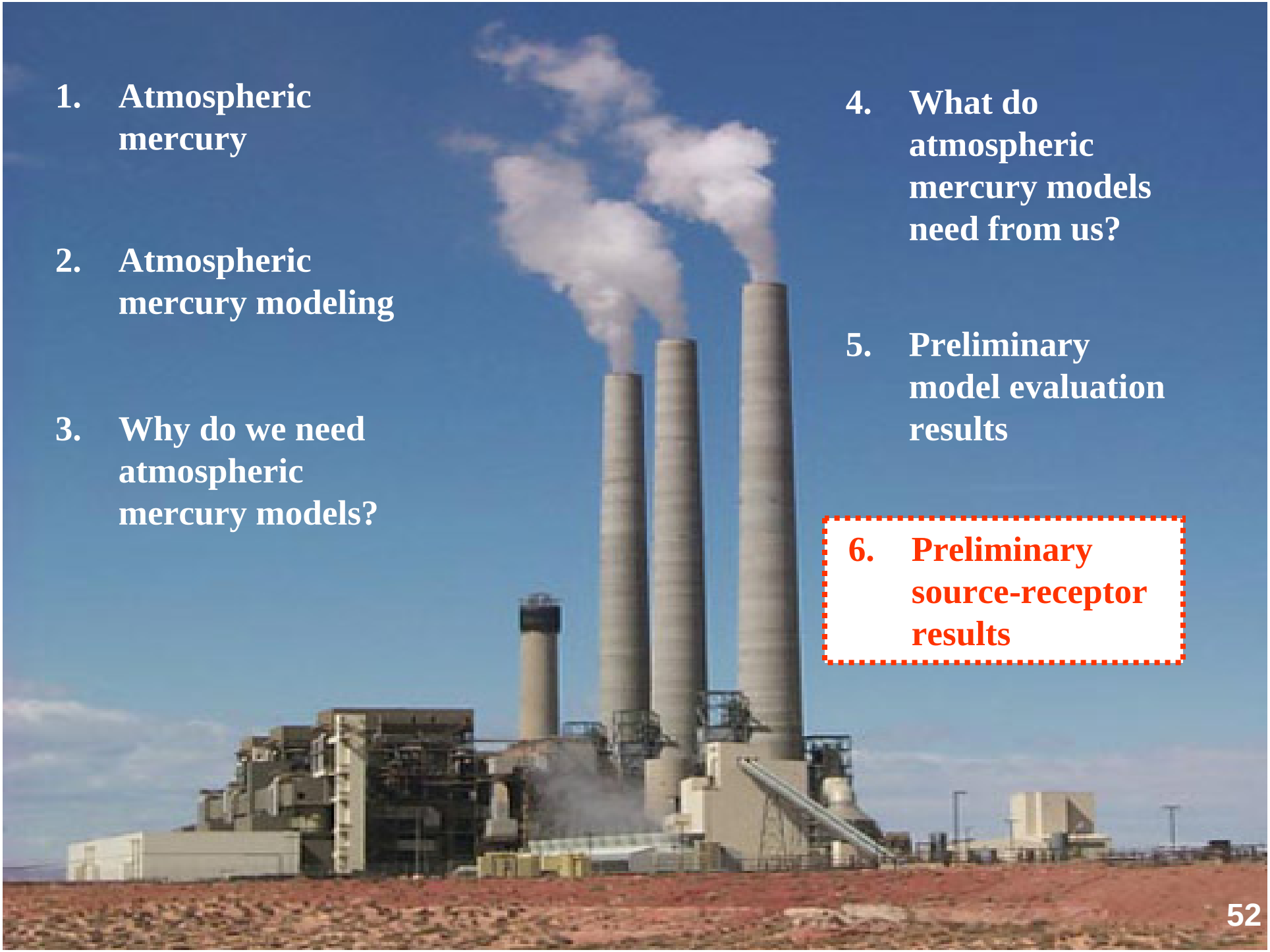
Cooperative Oxford Lab
(38.678EN, 76.173EW)

regional emissions (1999) and sampling sites for summer 2004 Ches Bay Hg study



Summer 2004 Chesapeake Bay Atmospheric Hg Study (June – August 2004)

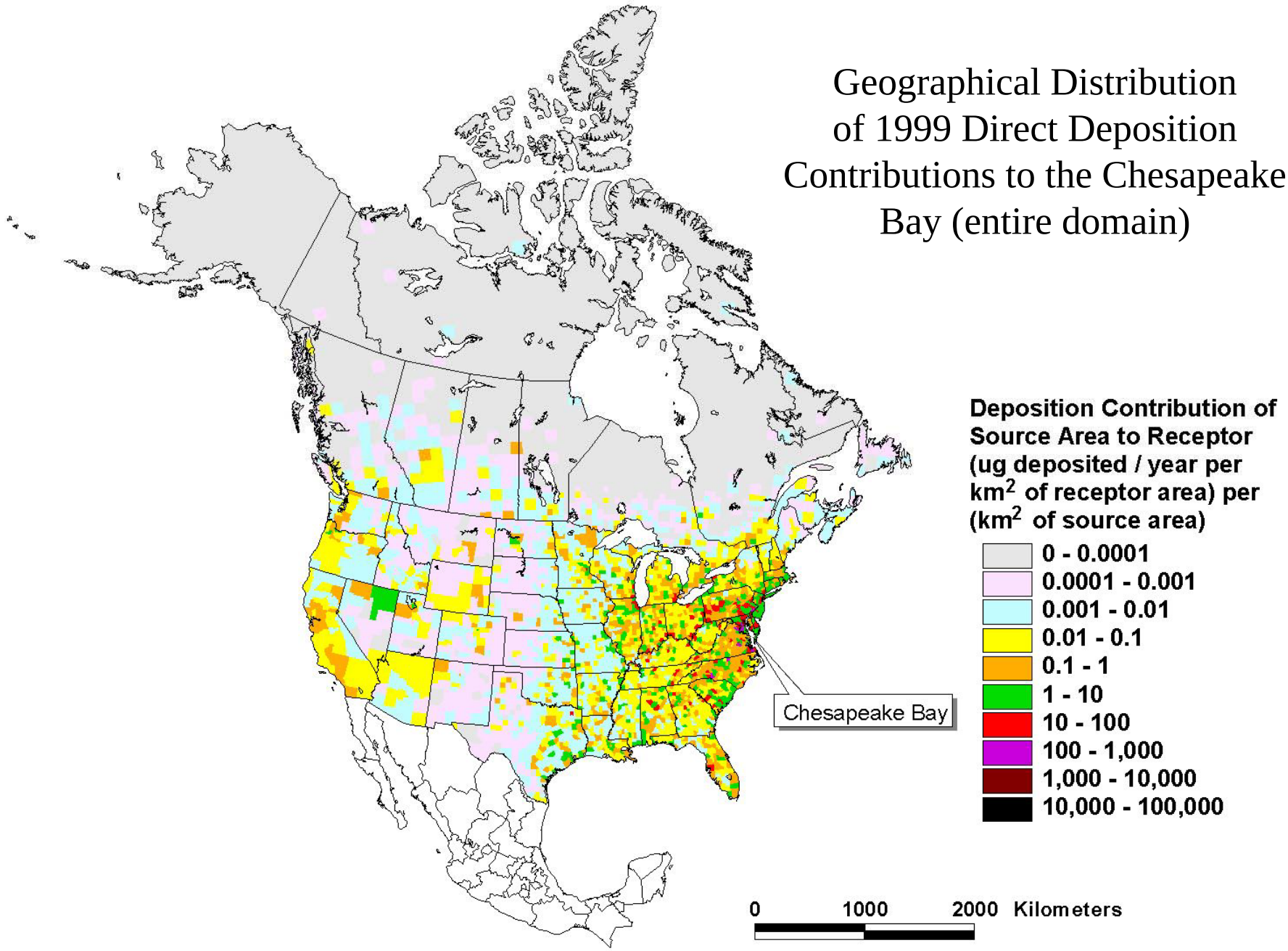
	Oxford	Wye
Event-based precipitation samples analyzed for Hg	✓	✓
Speciated Hg concentrations in ambient air (RGM, Hg(p), Hg^0)	✓	✓
Ambient concentration of ozone and sulfur dioxide	✓ (continuous)	✓ (weekly via AirMON Dry)
Ambient concentration of carbon monoxide	✓	
Meteorology	✓	✓ (via NADP/NTN site)
Major ions in precipitation		✓ (via NADP/NTN site)



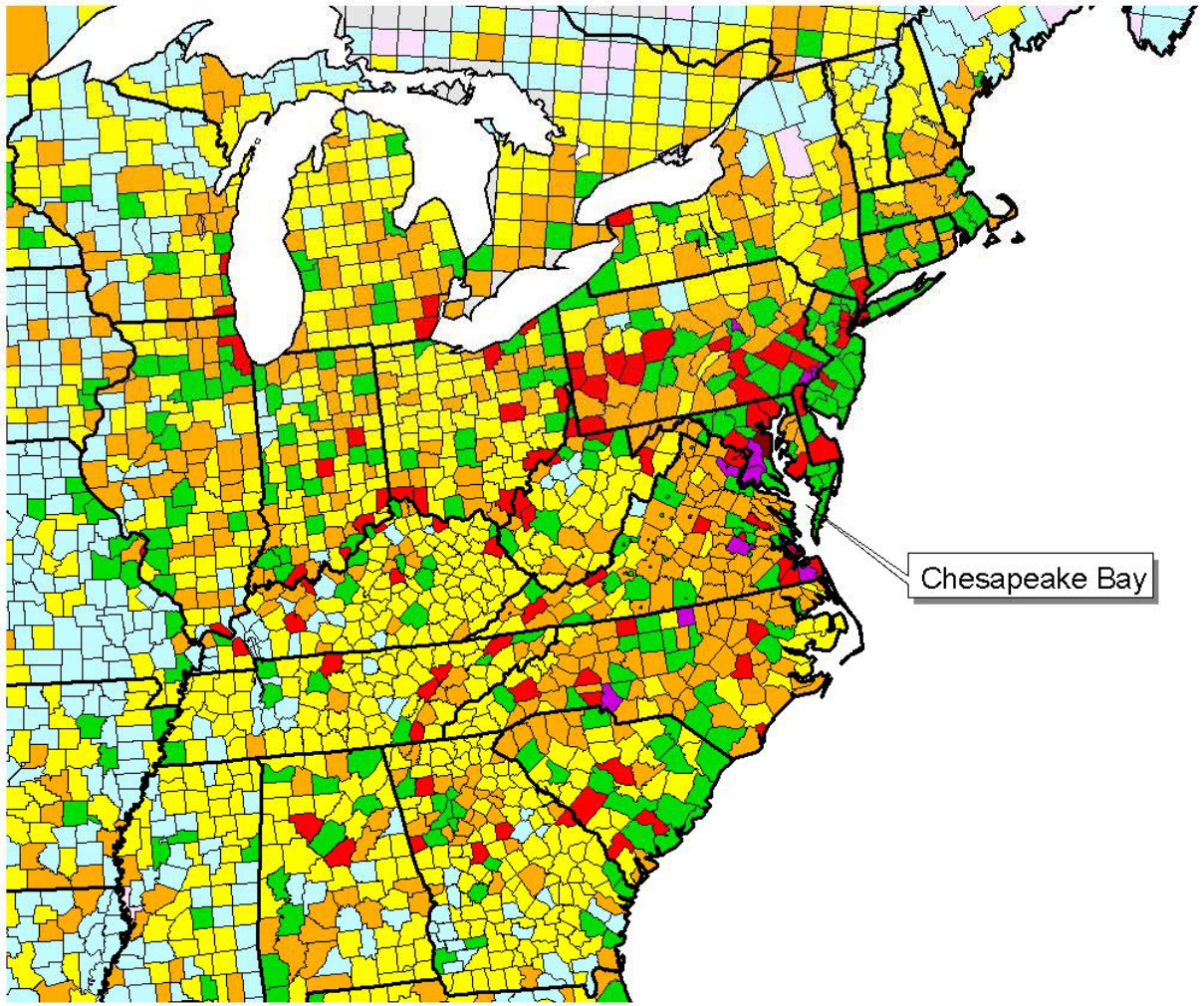
1. Atmospheric mercury
2. Atmospheric mercury modeling
3. Why do we need atmospheric mercury models?
4. What do atmospheric mercury models need from us?
5. Preliminary model evaluation results
6. Preliminary source-receptor results

Example of Detailed Results: 1999 Results for Chesapeake Bay

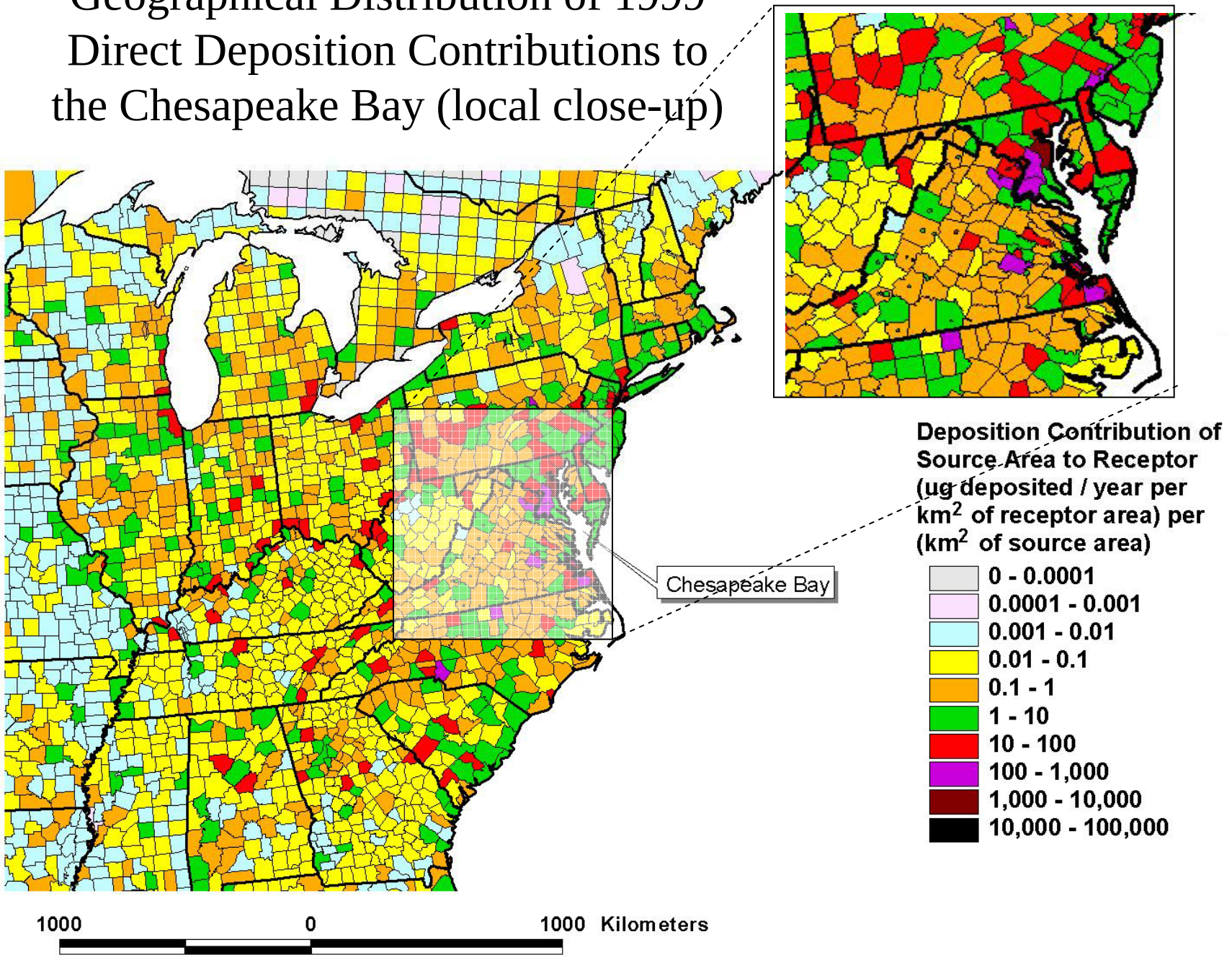
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (entire domain)



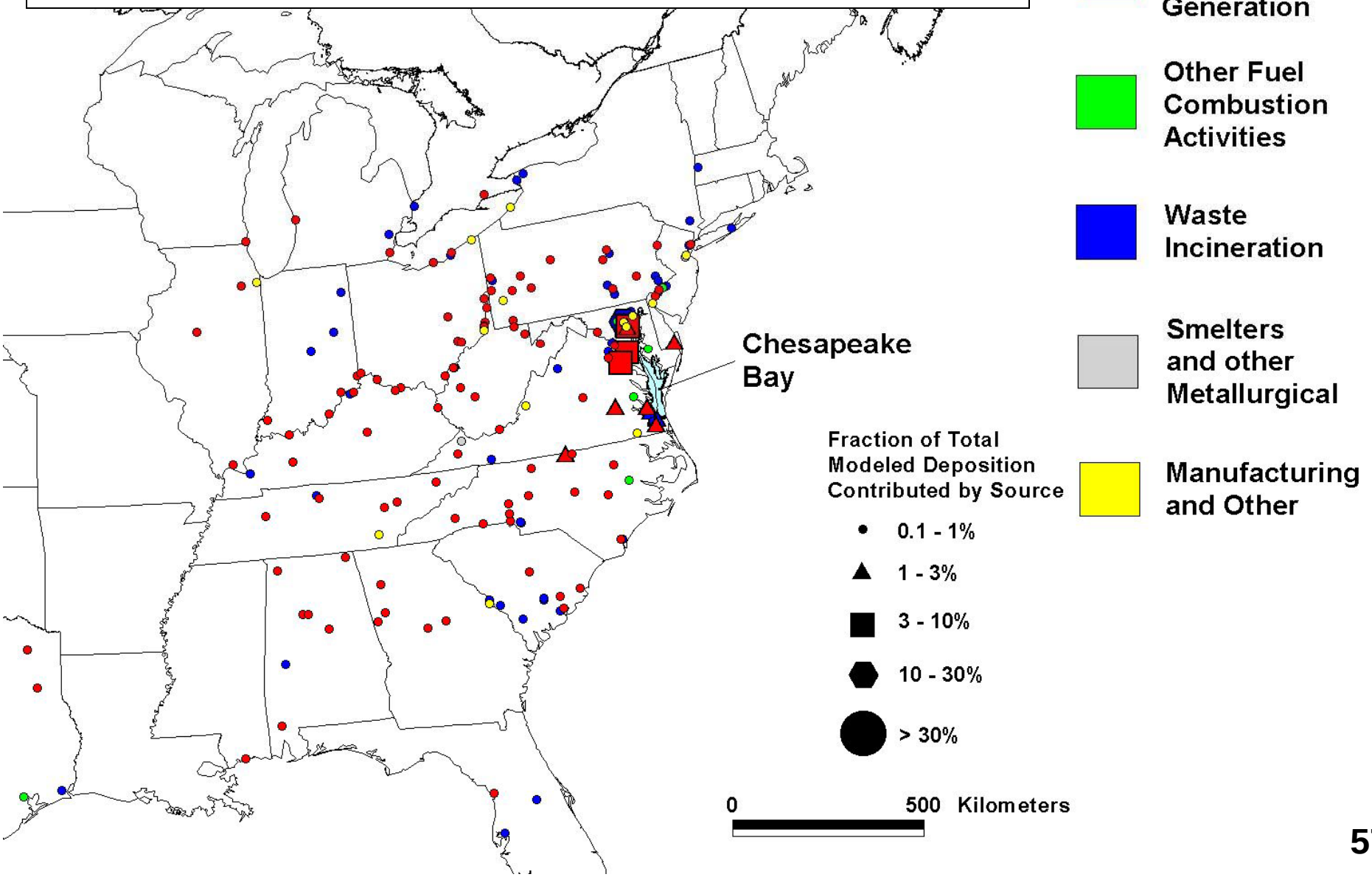
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (regional close-up)



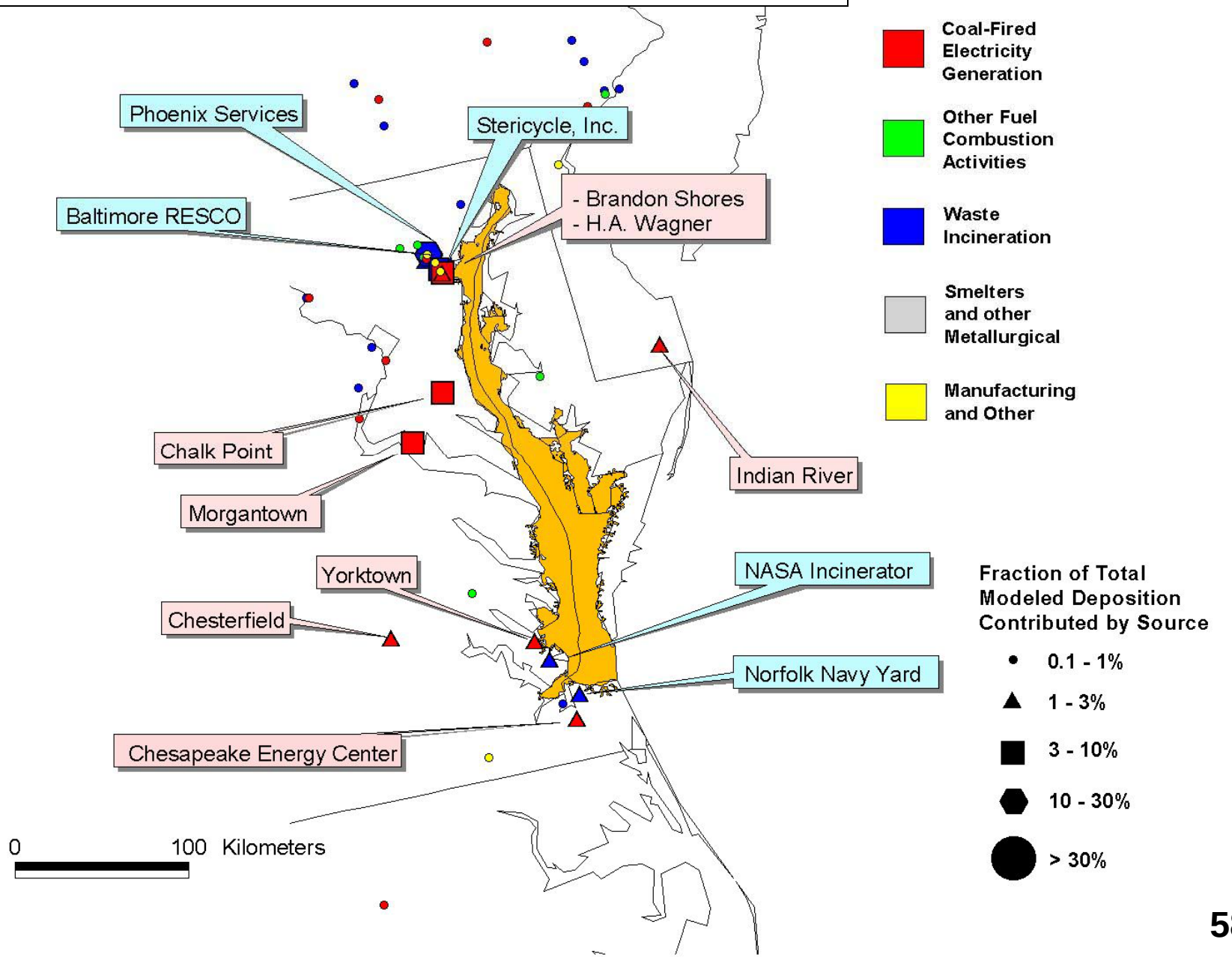
Geographical Distribution of 1999 Direct Deposition Contributions to the Chesapeake Bay (local close-up)



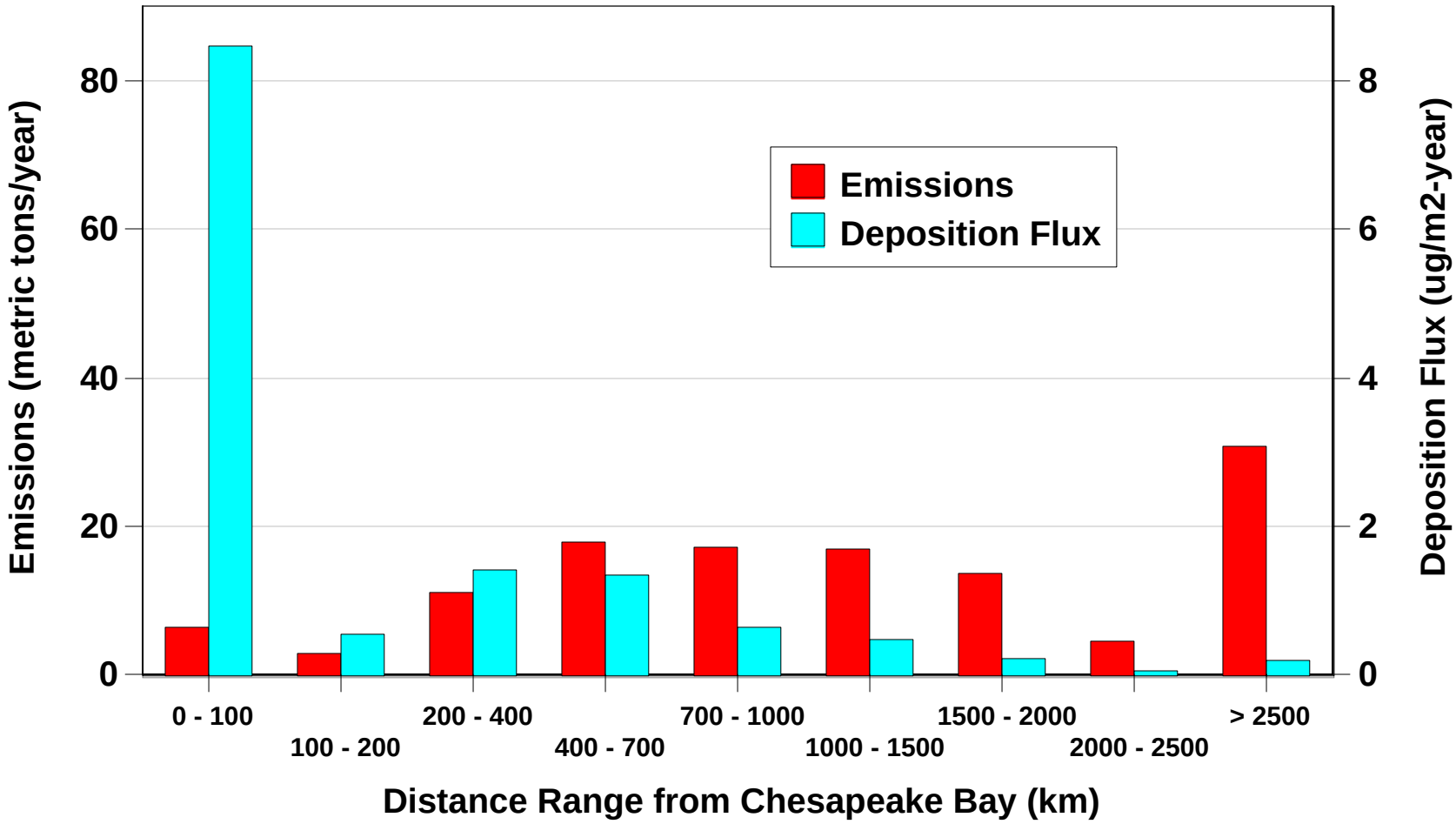
Largest Regional Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay



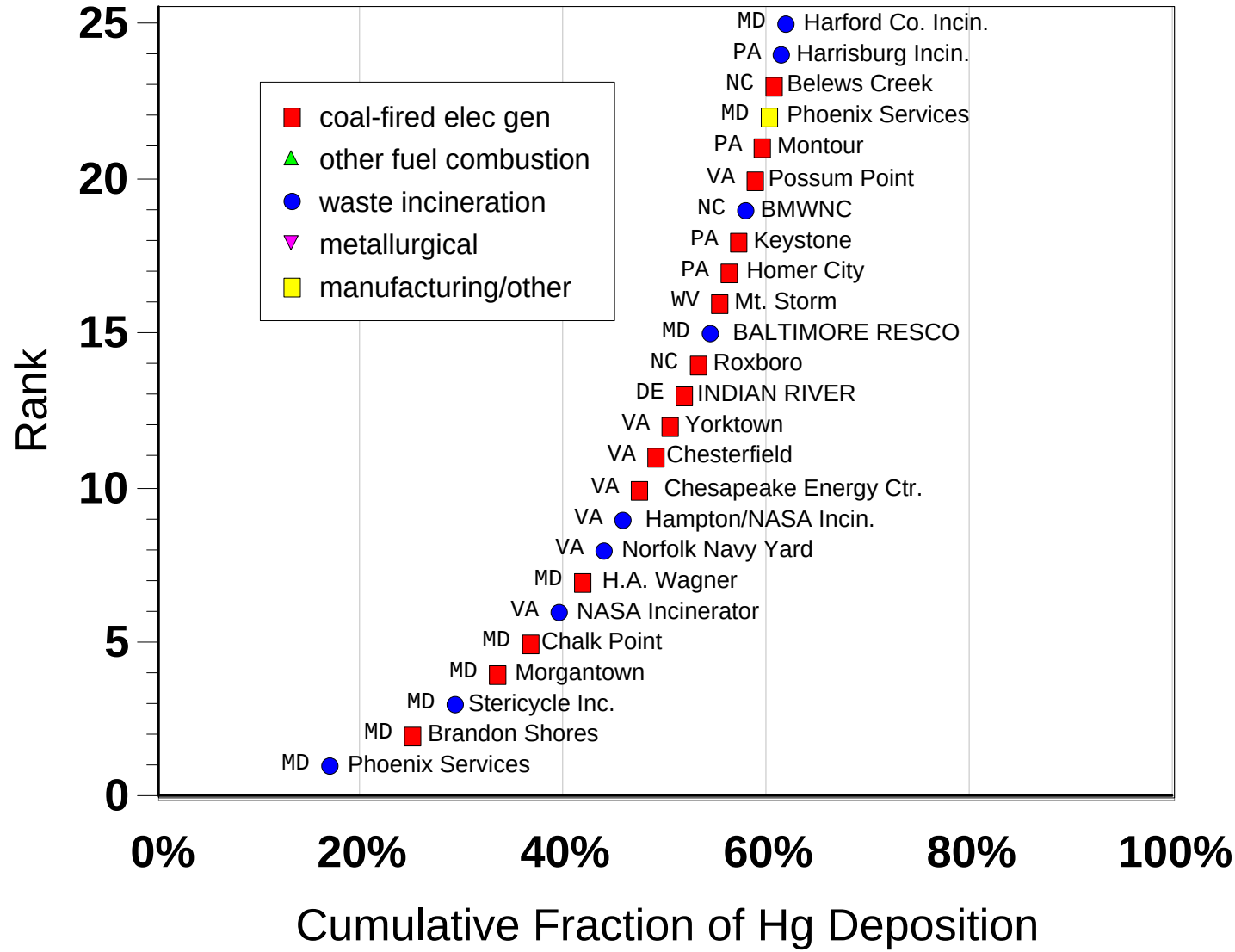
Largest Local Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay



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

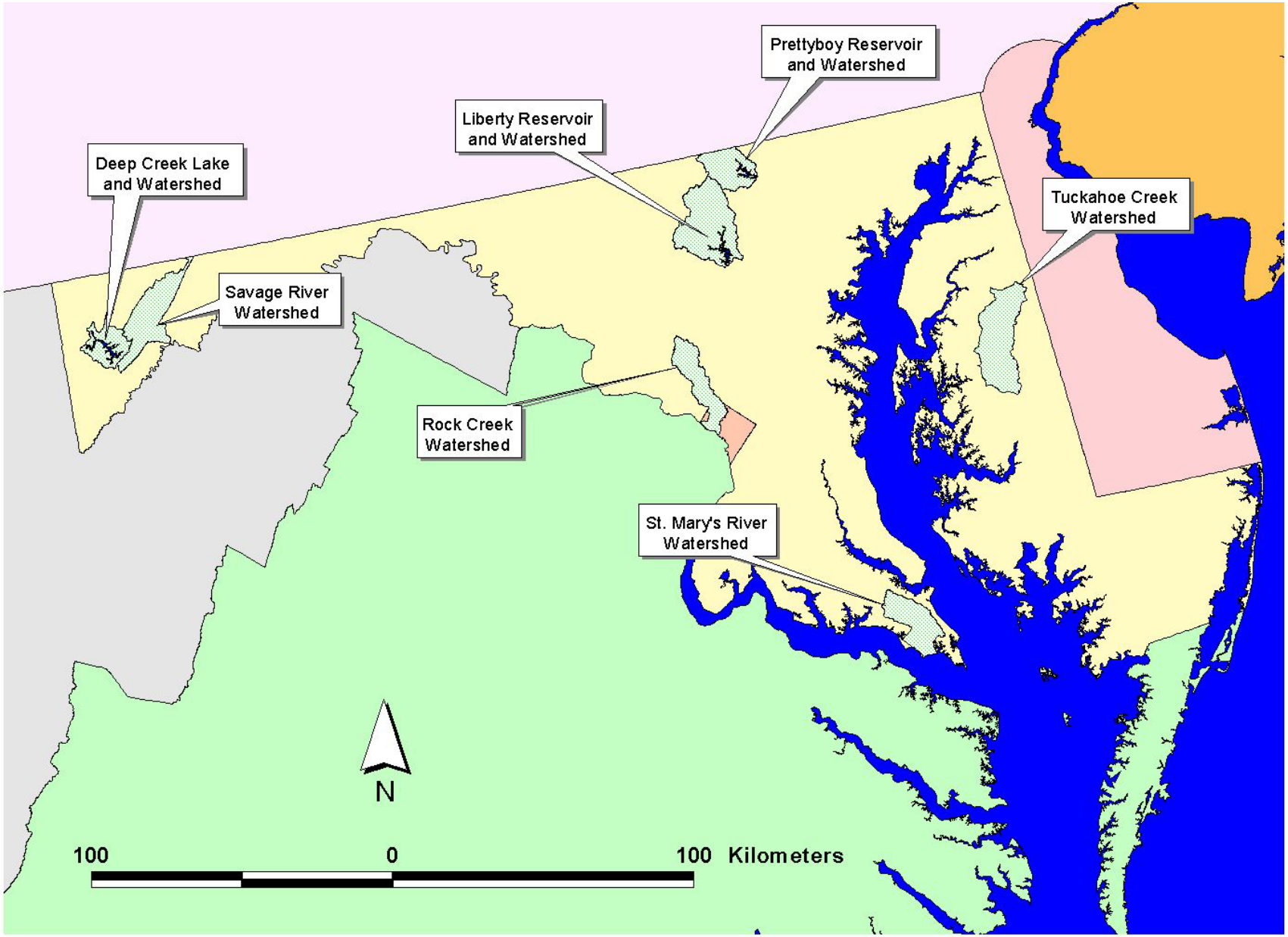


Top 25 Contributors to 1999 Hg Deposition Directly to the Chesapeake Bay

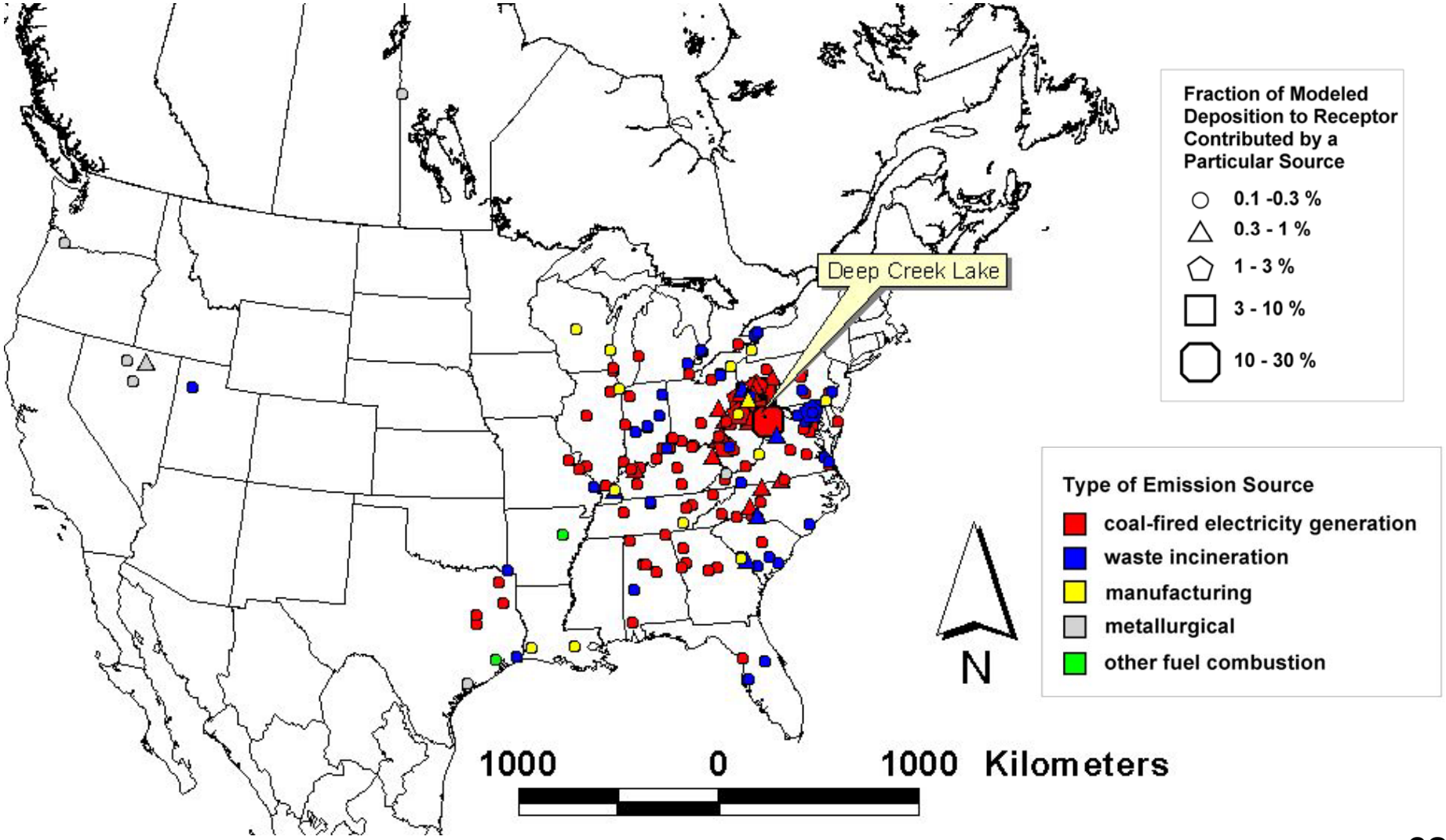


Preliminary Results for other Maryland Receptors

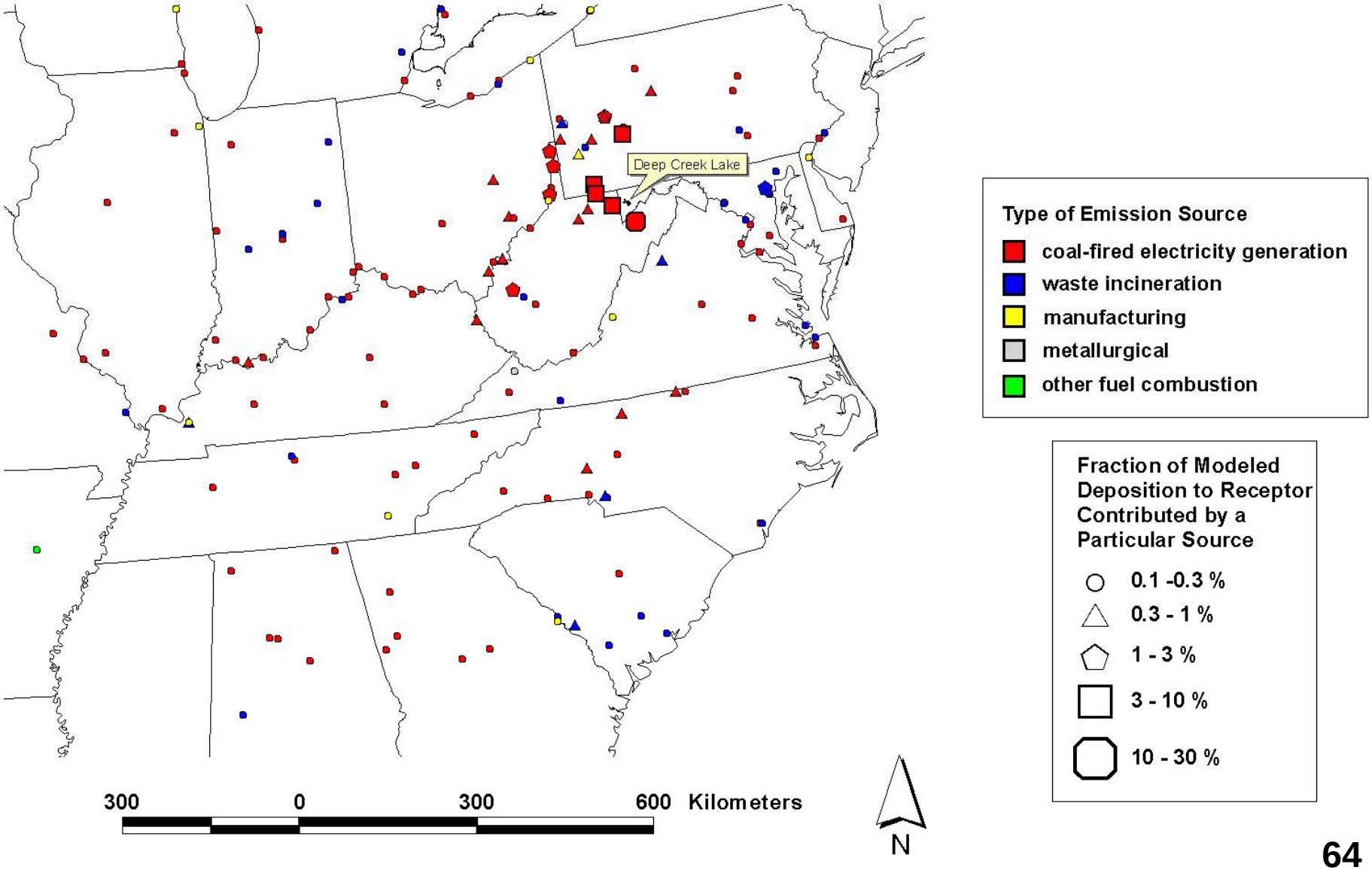
Maryland Receptors Included in Recent Preliminary HYSPLIT-Hg modeling *(but modeling was not optimized for these receptors!)*



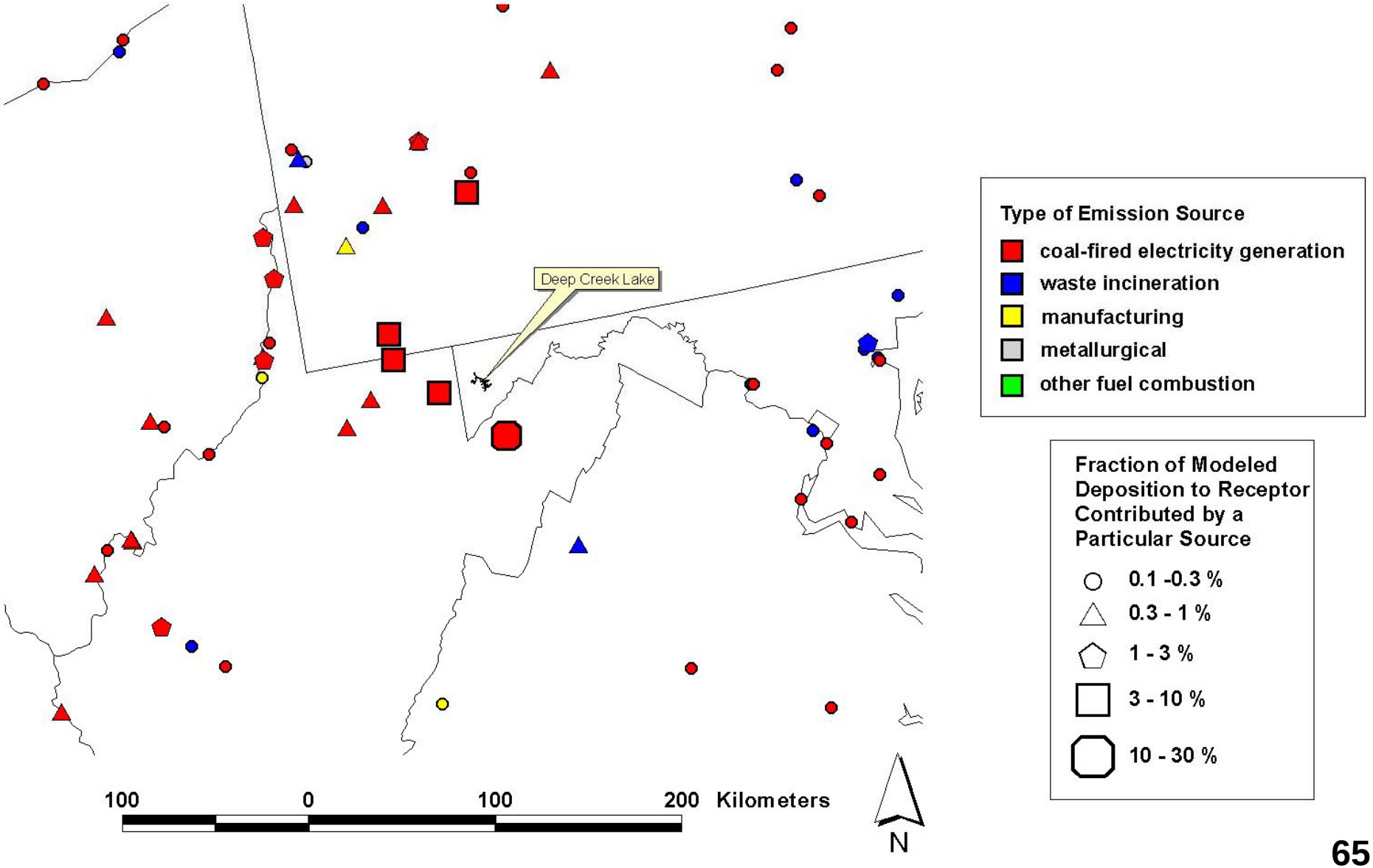
Largest Modeled Atmospheric Deposition Contributors Directly to Deep Creek Lake based on 1999 USEPA Emissions Inventory (national view)



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



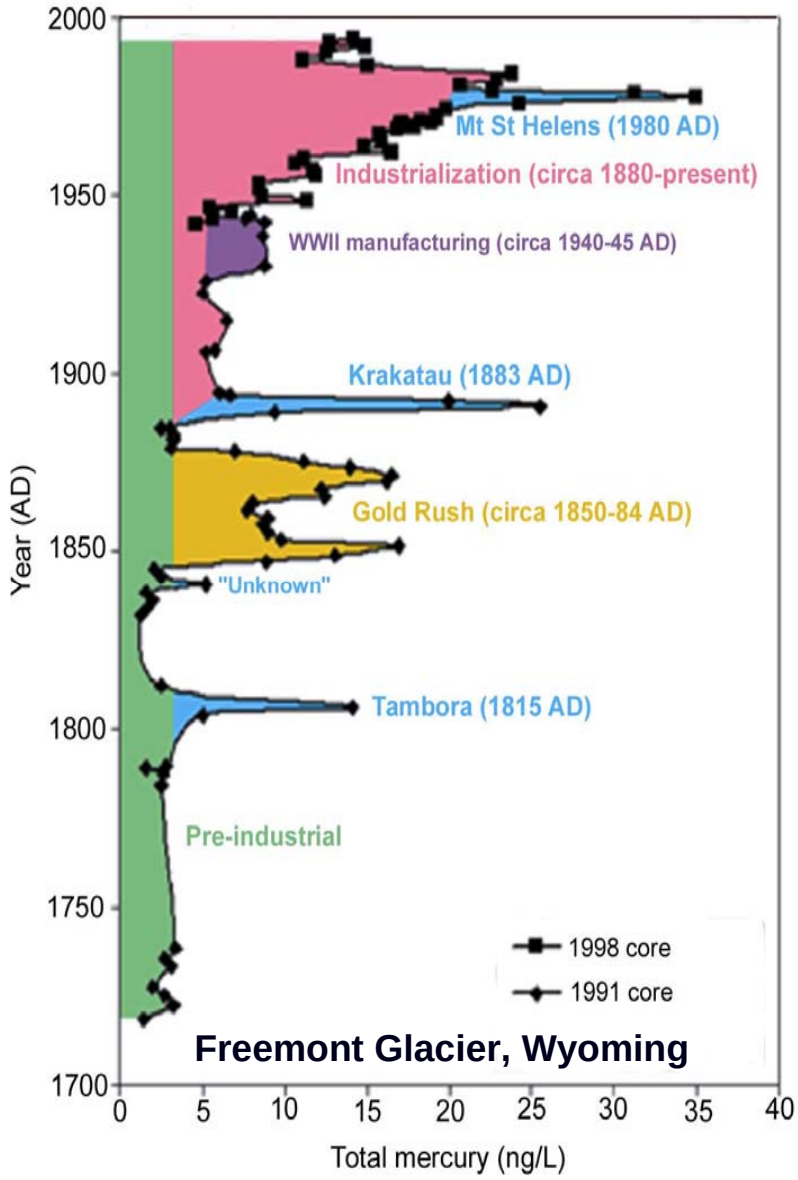
Largest Modeled Atmospheric Deposition Contributors Directly to Deep Creek Lake based on 1999 USEPA Emissions Inventory (close-up view)



source-attribution – the “big picture”

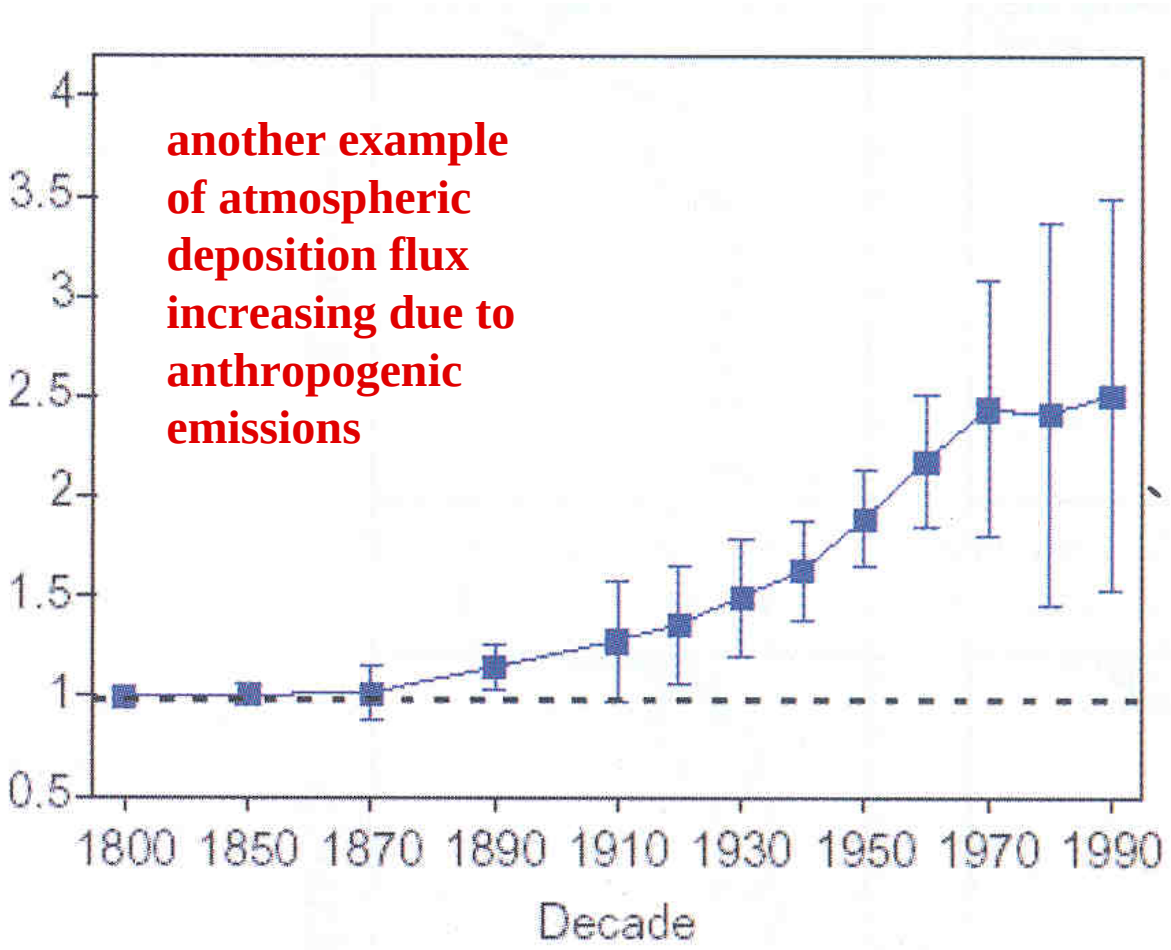
Natural vs. anthropogenic mercury?

Studies show that anthropogenic activities have typically increased bioavailable Hg concentrations in ecosystems by a factor of 2 – 10



source: USGS, Shuster et al., 2002

Hg flux / pre-industrial Hg flux



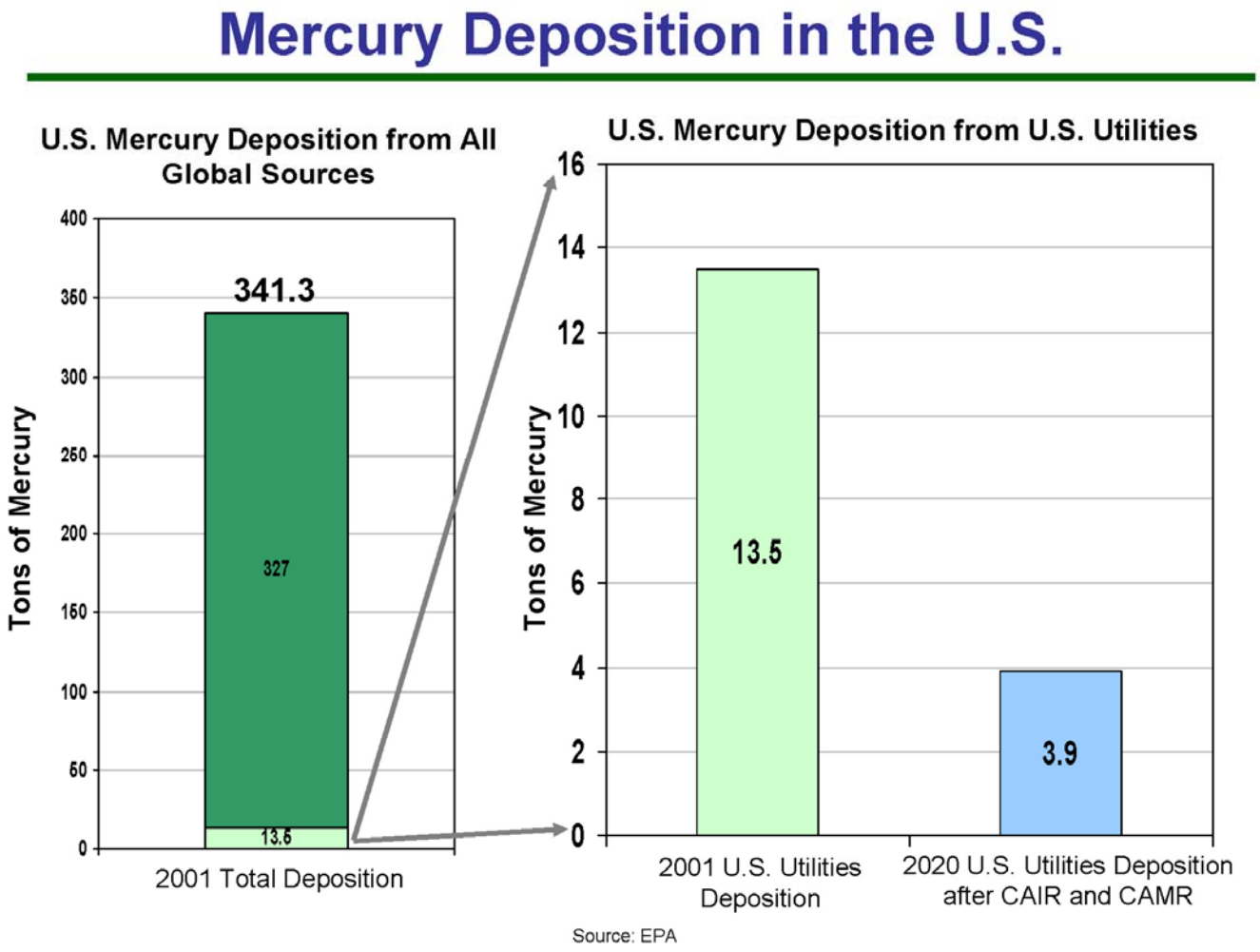
Average mercury accumulation rate relative to pre-industrial (1800-1850) accumulation rate in five lakes in Northern Alaska (based on sediment cores)

from Fitzgerald et al. (2005), "Modern and Historic Atmospheric Mercury Fluxes in Northern Alaska: Global Sources and Arctic Depletion" *Environ Sci Tech* **39**, 557-568

What is the relative importance of global, national, regional, and local sources?

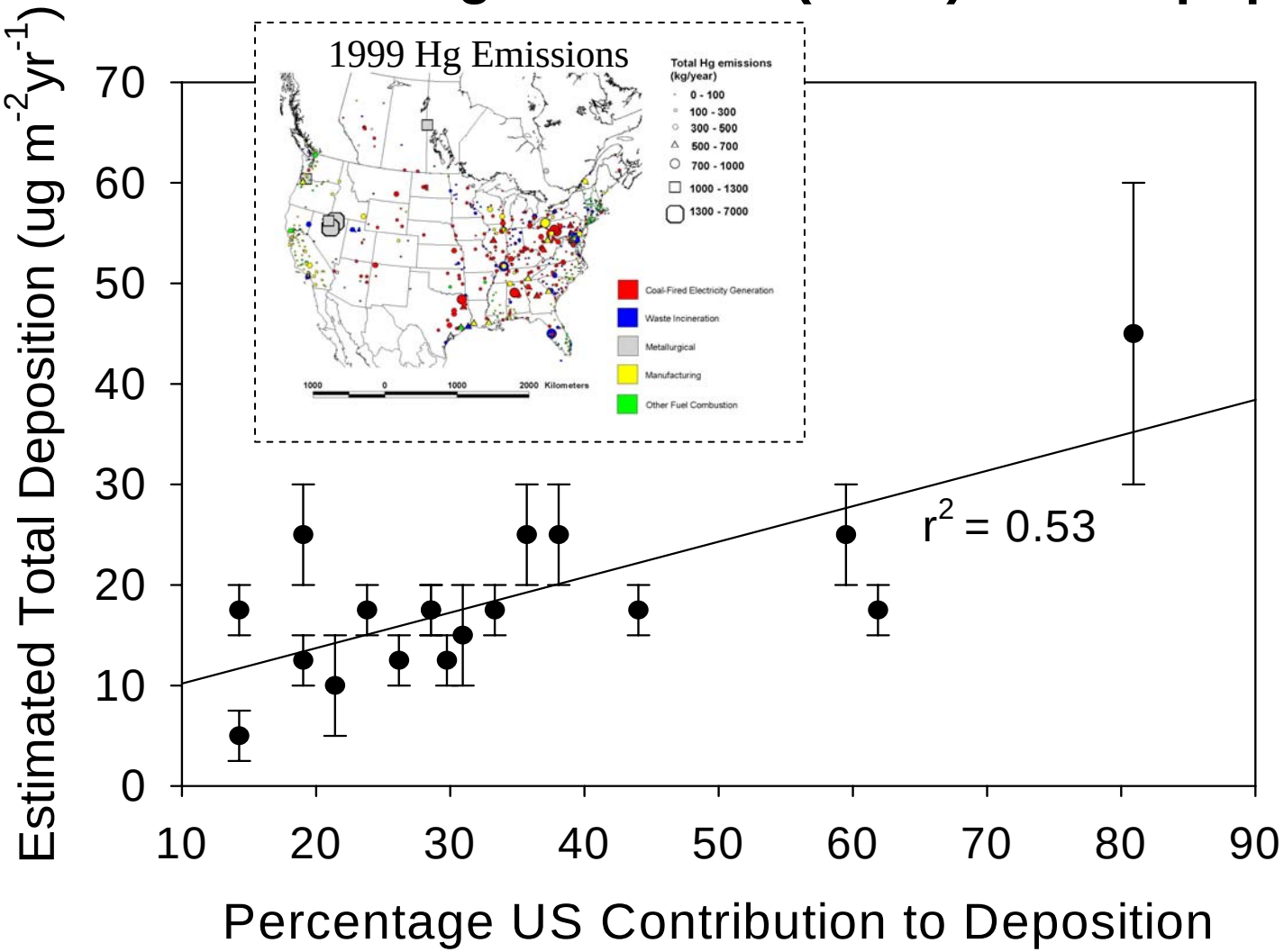
Data used by EPA to support recent Clean Air Mercury Rule

?



(slide courtesy of Rob Mason, Univ. of CT)

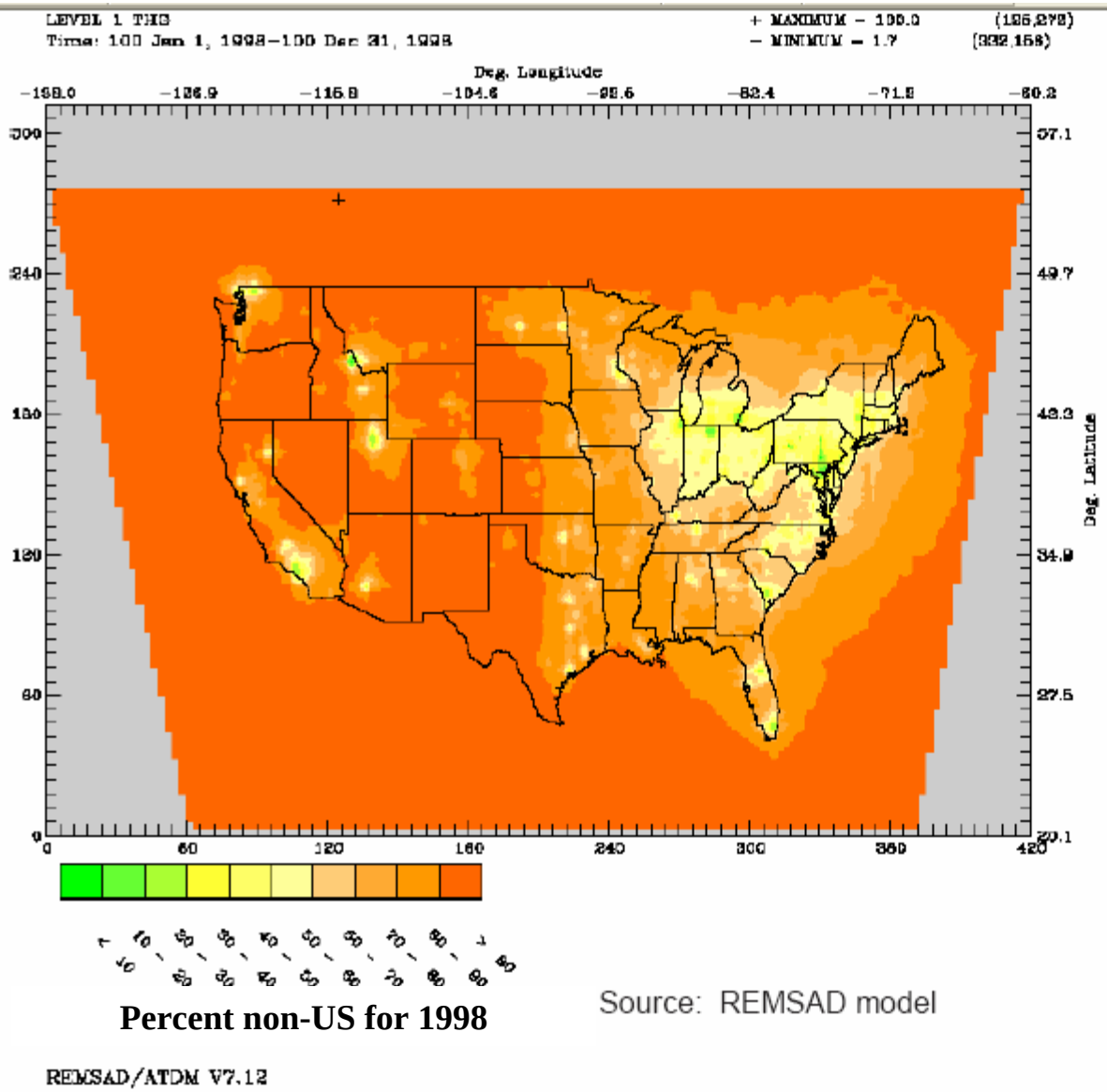
Data from Seigneur et al. (2004) model paper



the Seigneur et al. (2004) paper shows that, for regions where deposition is high, the impact of local/regional sources is the main cause for these elevated concentrations.

[Seigneur et al., (2004), "Global Source Attribution for Mercury Deposition in the United States", *ES&T* **38**, 555-569.]

Results from the EPA REMSAD Mercury model



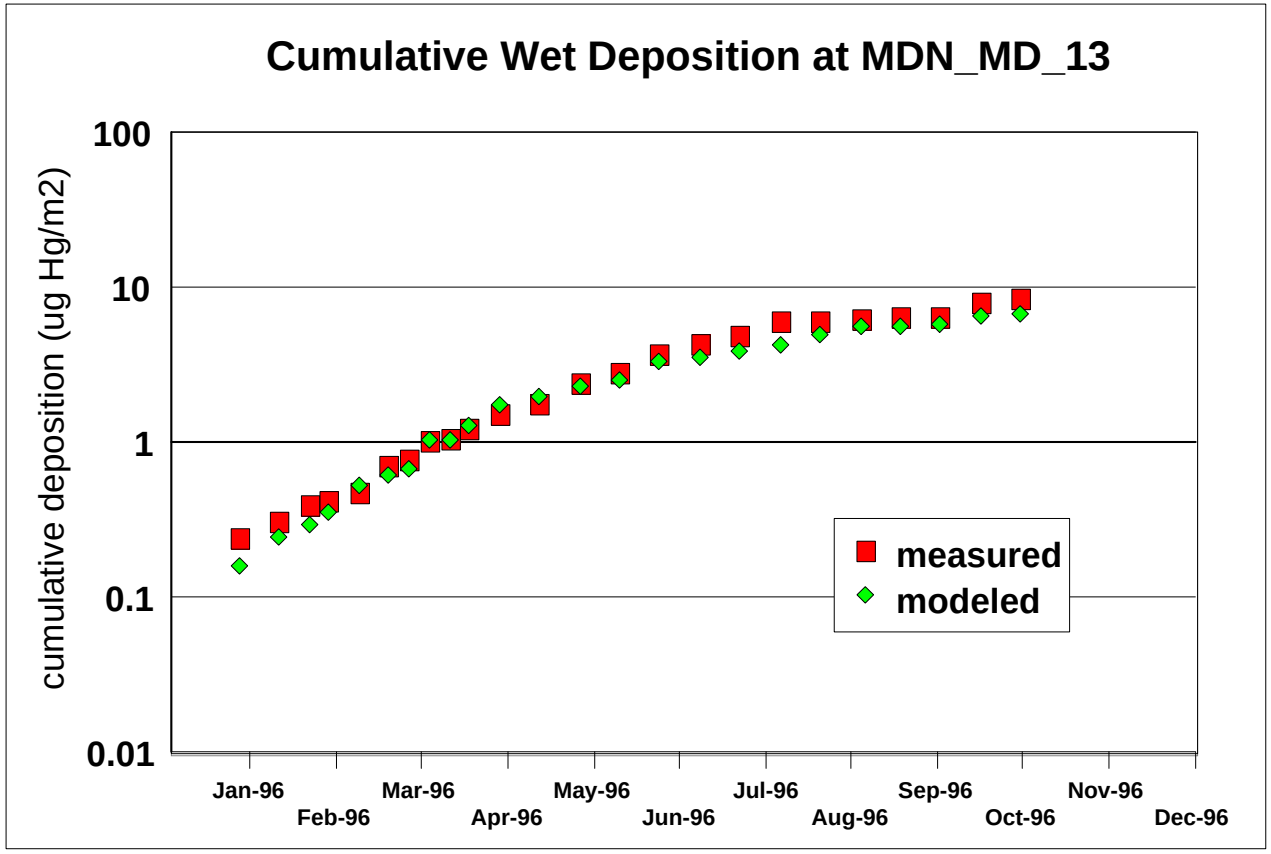
Percent non-US for 1998

Source: REMSAD model

- Based on this modeling approximately half of U.S. mercury deposition is from domestic anthropogenic sources and half is from other sources
- Domestic sources dominate deposition for large part of Eastern U.S.
- Global sources are dominant in the Western U.S.

Source: slide developed by Anne Pope for the Hg Roundtable conference call April 21, 2005

Modeled vs. Measured Wet Deposition at Mercury Deposition Network Site MD_13 during 1996



HYSPLIT modeling has shown that in areas of significant local and regional anthropogenic sources, ambient measurements can be explained reasonably well by considering only these local and regional anthropogenic sources.

Source-apportionment answers depend a lot on where you are

- **For areas *without large emissions sources***
  - **the deposition may be relatively low,**
  - **but what deposition there is may largely come from natural and global sources**
- **For areas *with large emissions sources***
  - **the deposition will be higher**
  - **and be more strongly influenced by these large emissions sources...**

What is the relative importance of global, national, regional, and local sources?

Possible answers are emerging as our understanding improves, but there is no scientific consensus yet...

Conclusions

- **Source-attribution information is important**
- **Impacts are episodic & depend on form of mercury emitted**
- **Modeling needed to get source-attribution information**
- **(more!) Monitoring needed for model evaluation & refinement**
- **Many uncertainties but useful model results are emerging –**
  these HYSPLIT model results are being extended to include global & natural emissions, and re-emissions
- **The question of the relative importance of global vs. national vs. regional vs. local sources is complex –**
  the answer depends on *location* and on *what model* one is using...

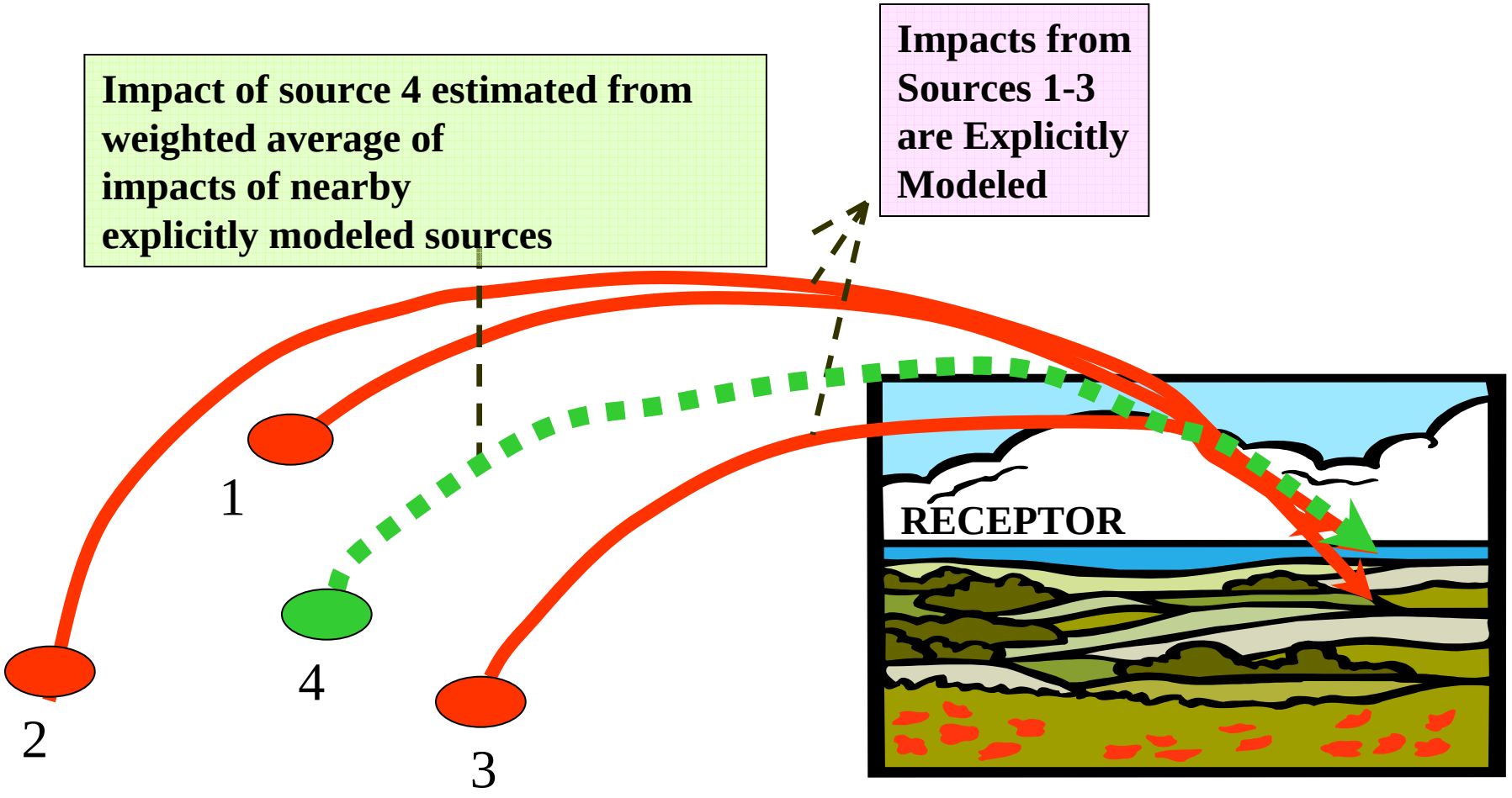
EXTRA SLIDES

- **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**

Spatial interpolation

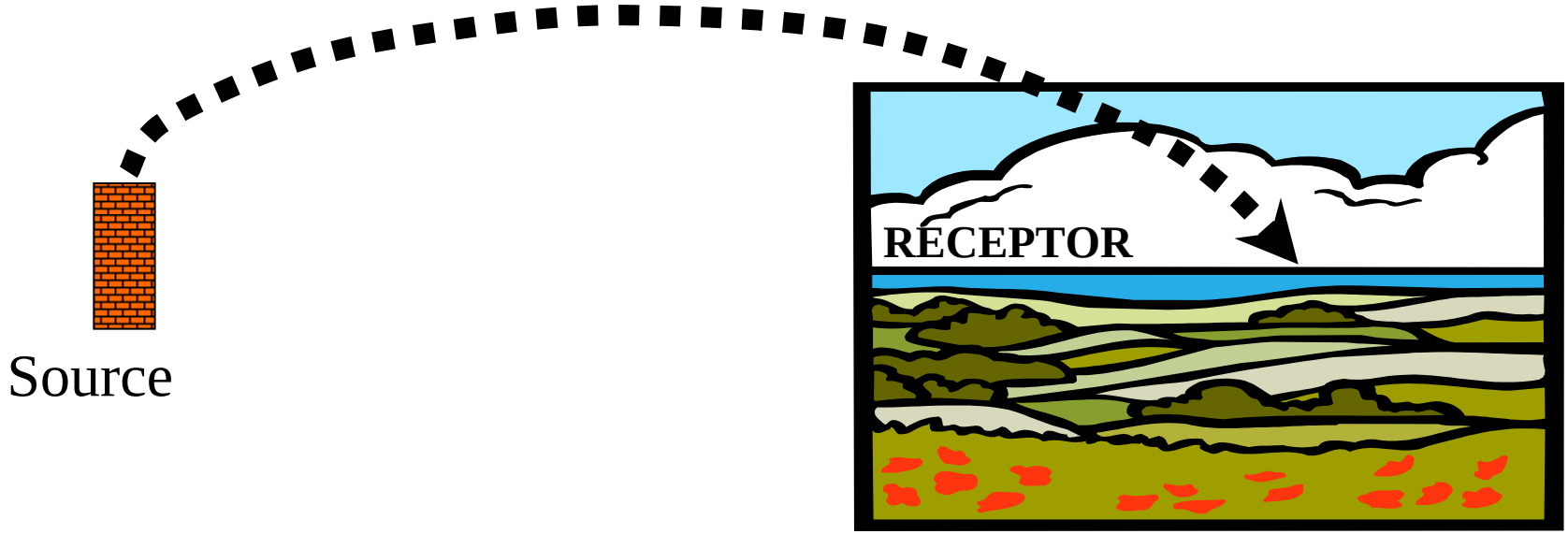


- **Perform separate simulations at each location for emissions of pure Hg(0), Hg(II) and Hg(p)**

  [*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)

=

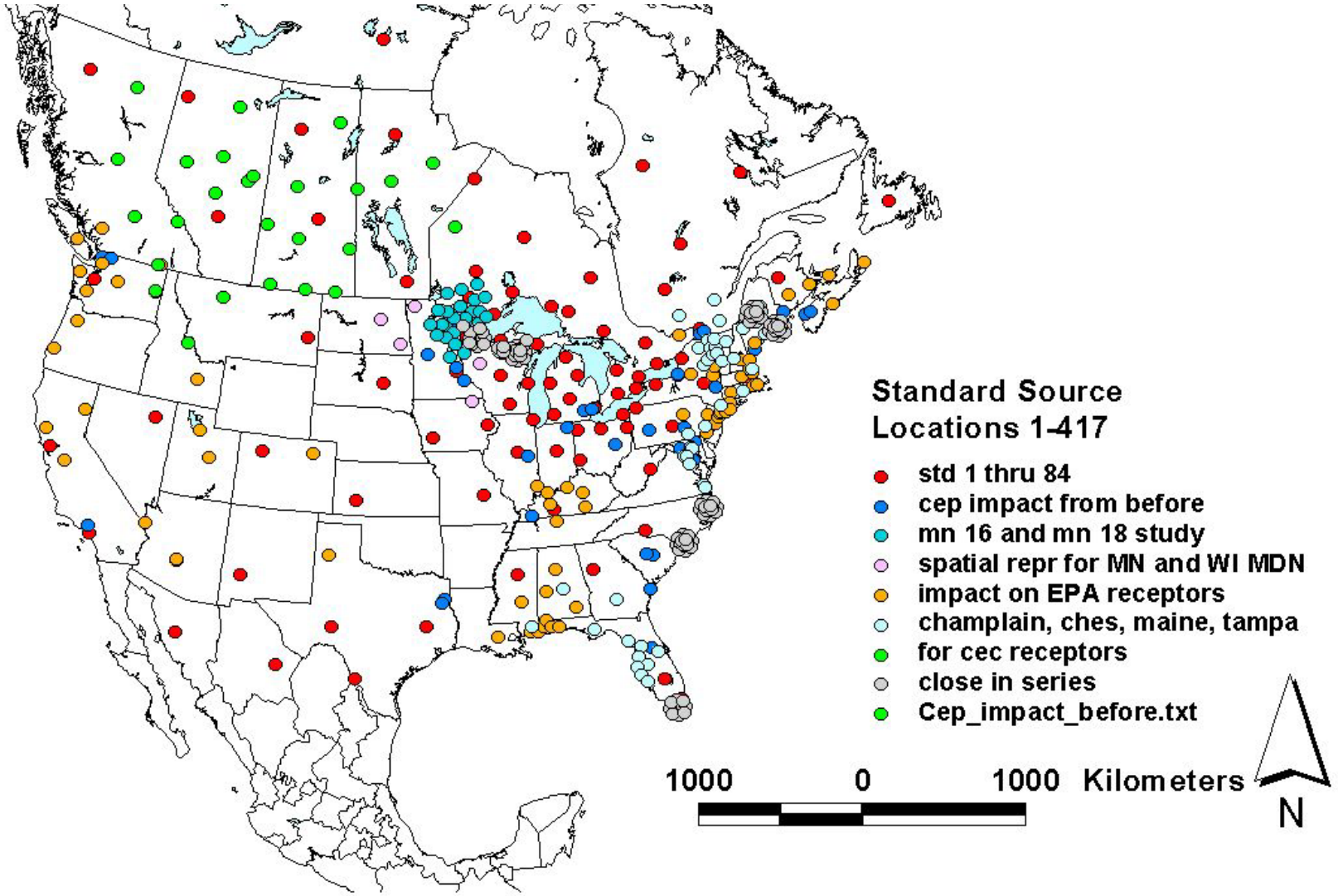
0.3 x **Impact of Source Emitting Pure Hg(0)**

+

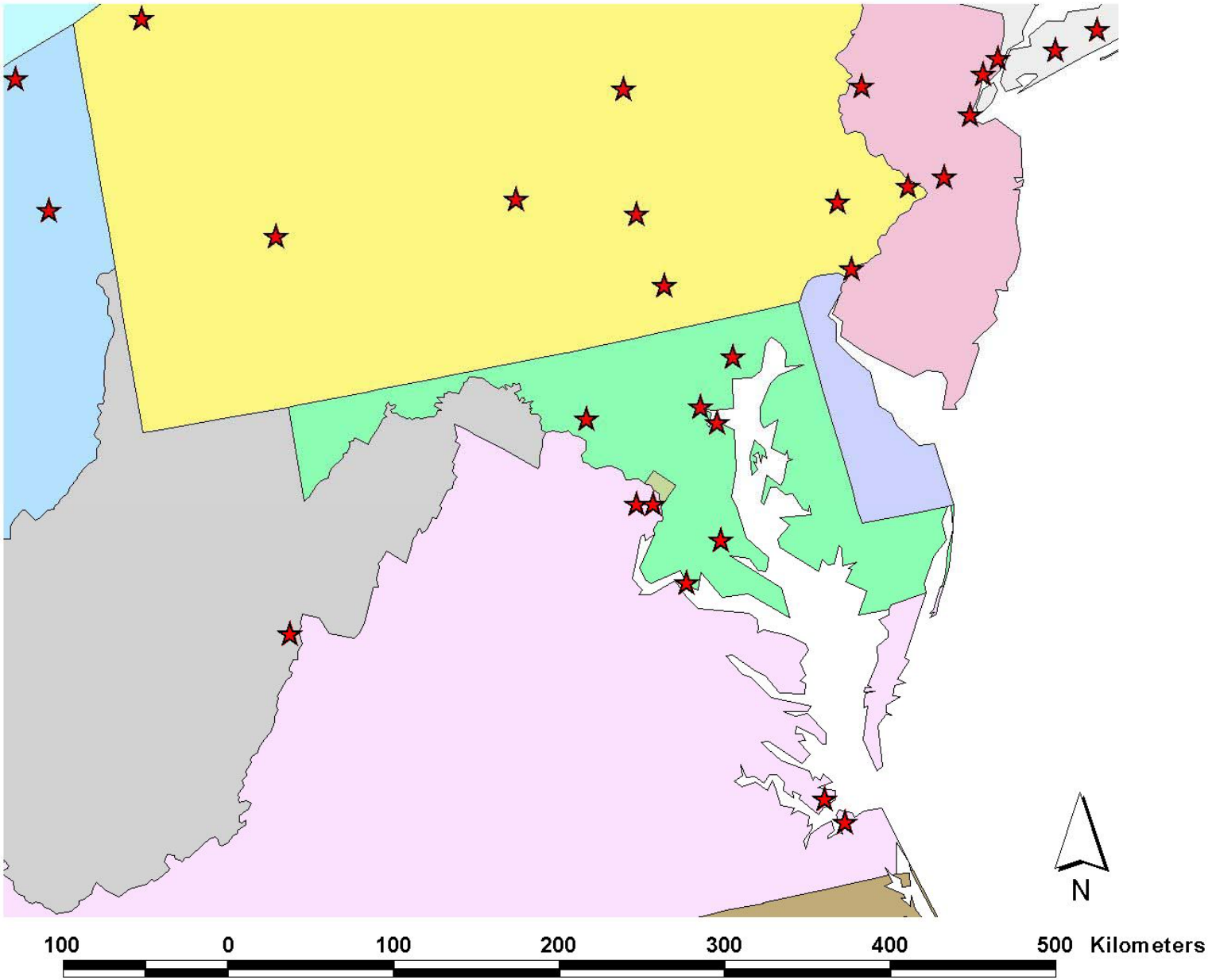
0.5 x **Impact of Source Emitting Pure Hg(II)**

+

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



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**