Source-apportionment for atmospheric mercury deposition: Where does the *mercury* in *mercury deposition* come from?

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Presentation at USGS, Eastern Region
2004 Mercury Workshop, August 17-18, 2004, Reston, VA
(revised version January 2005)

For many (but not all) aquatic ecosystems, much of the loading comes directly or indirectly through the atmospheric pathway...

For the atmospheric pathway:

- **How much of the mercury in atmospheric mercury deposition comes from local, regional, national, continental, and global sources?**
- **How important are different source types?**

1. The impact of any given mercury emissions source on any receptor is highly variable

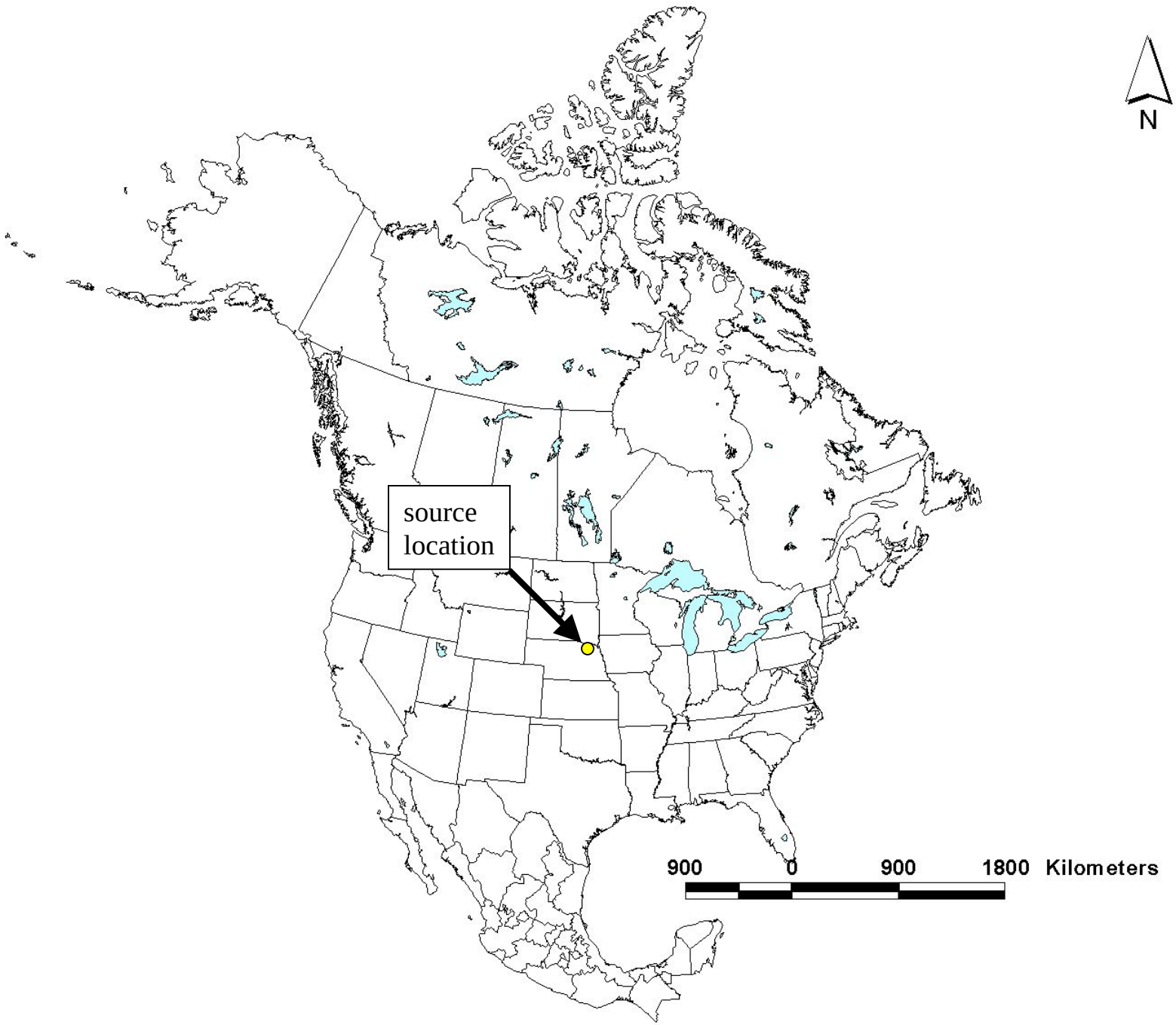
- **extreme spatial and temporal variations**
- **Think about the weather and then add all the chemistry and physics of mercury's interactions with the "weather"**

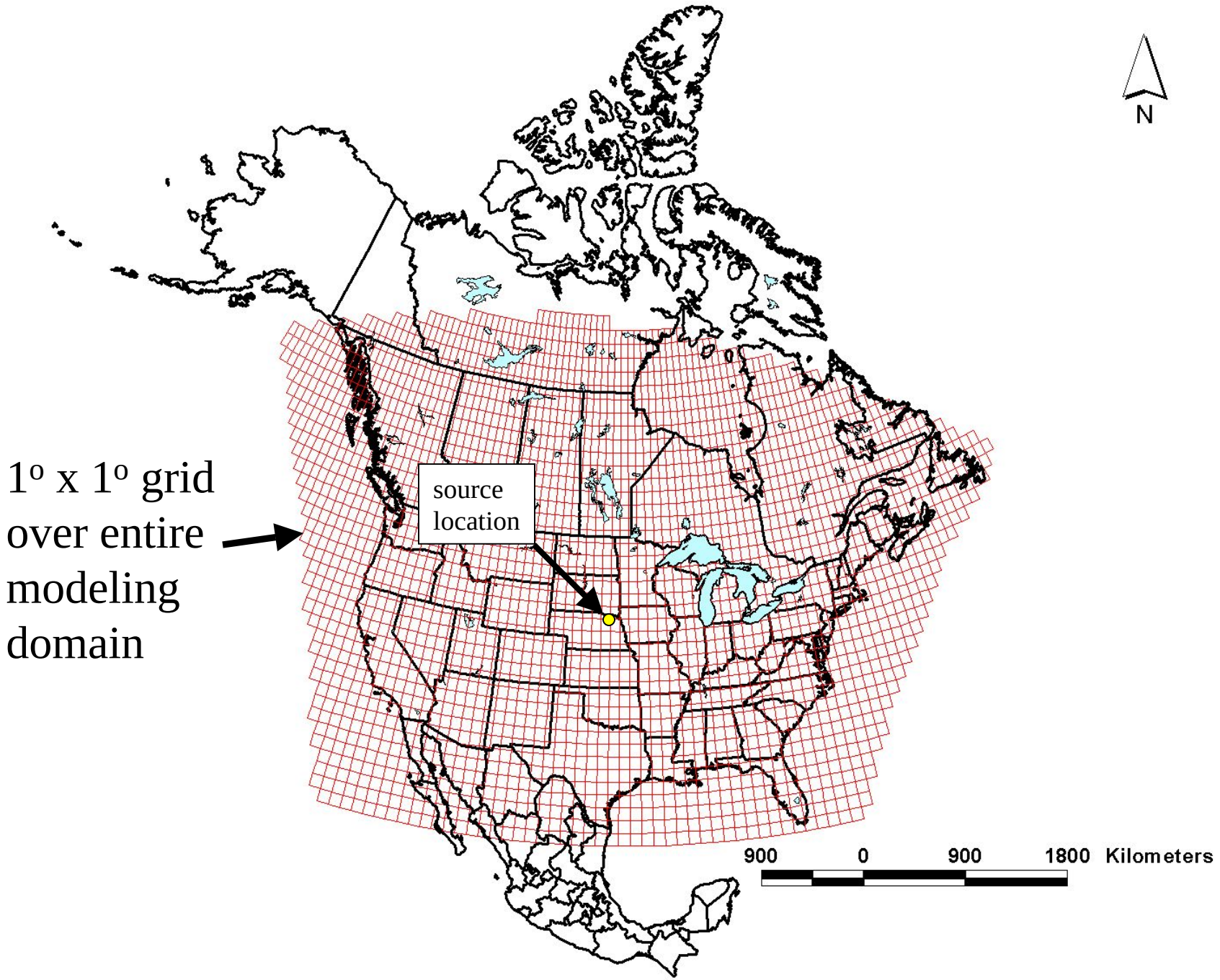
2. The impact of any given mercury emissions source on any receptor is highly dependent on the "type" of mercury emitted

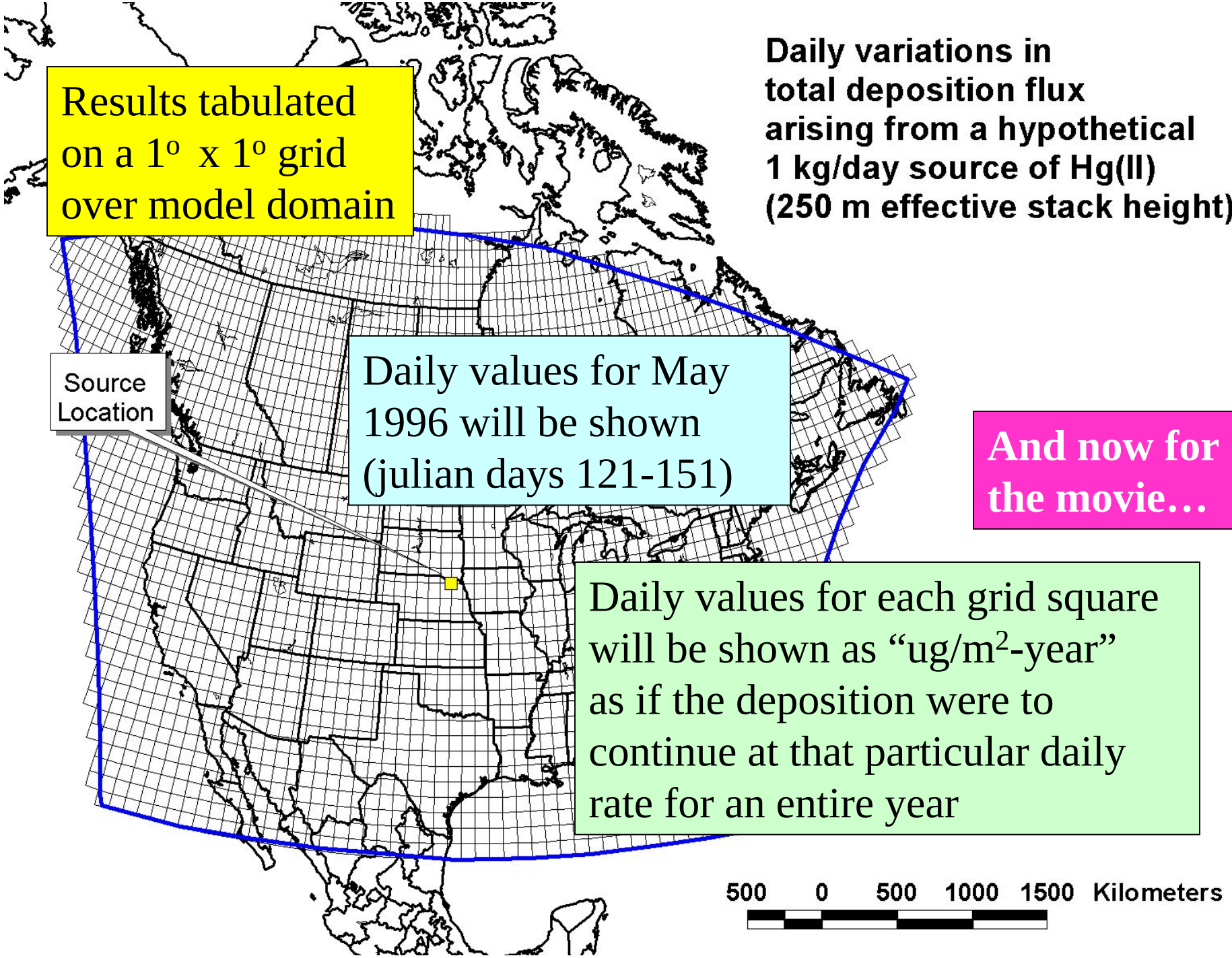
- **Elemental mercury - Hg^0 - is not readily dry or wet deposited, and its conversion to ionic Hg or Hg(p) is relatively slow**
- **Particulate mercury – Hg(p) - is moderately susceptible to dry and wet deposition**
- **Ionic mercury – also called Reactive Gaseous Mercury or RGM – is very easily dry and wet deposited**
  - ***Conversion of RGM to Hg^0 in plumes*?**

Example simulation of the atmospheric fate and transport of mercury emissions:

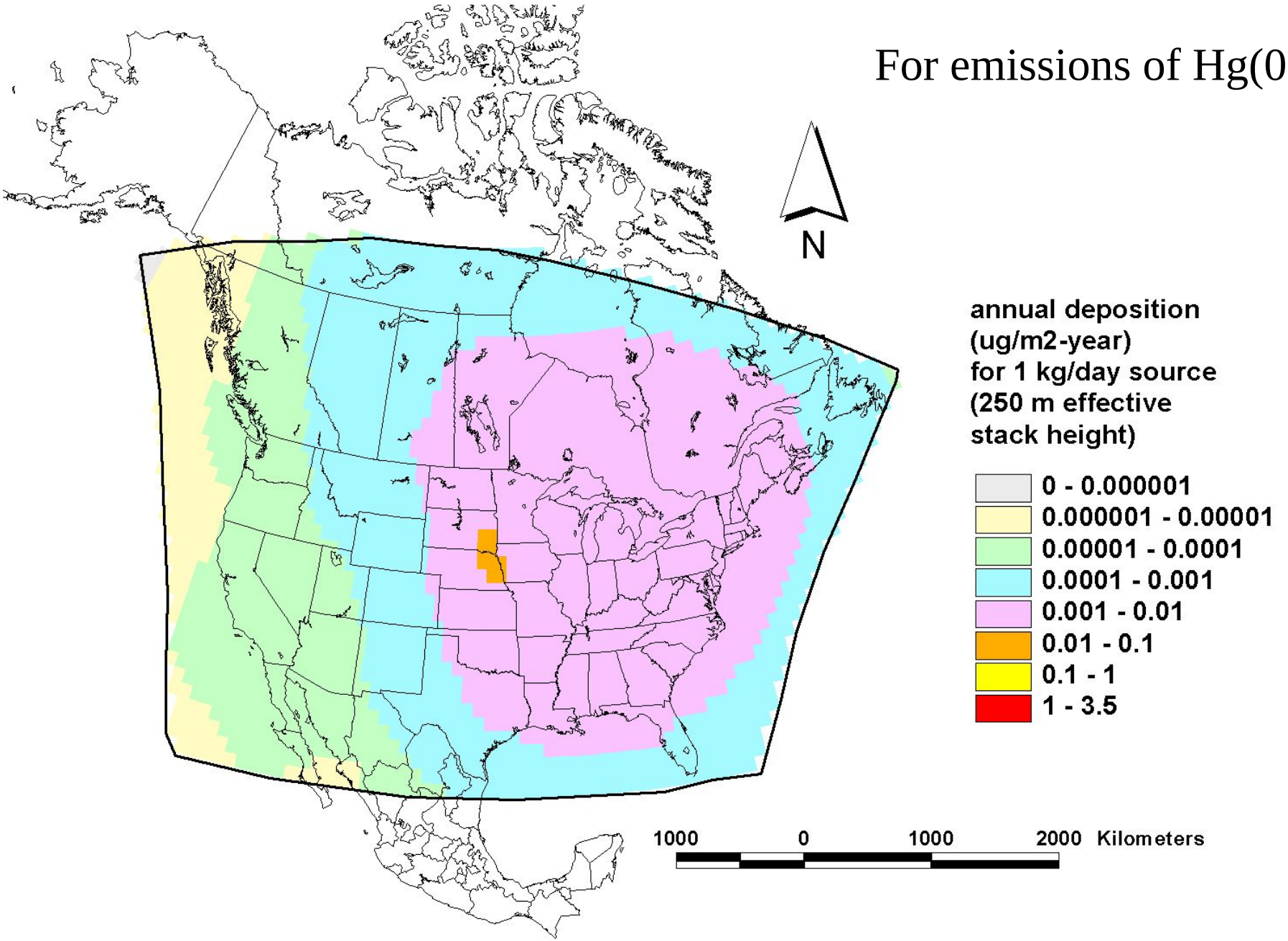
- **hypothetical 1 kg/day source of RGM, Hg(p) or Hg(0)**
- **source height 250 meters**
- **results tabulated on a 1^{o} x 1^{o} receptor grid**
- **annual results (1996)**


N
source
location
900 0 900 1800 Kilometers

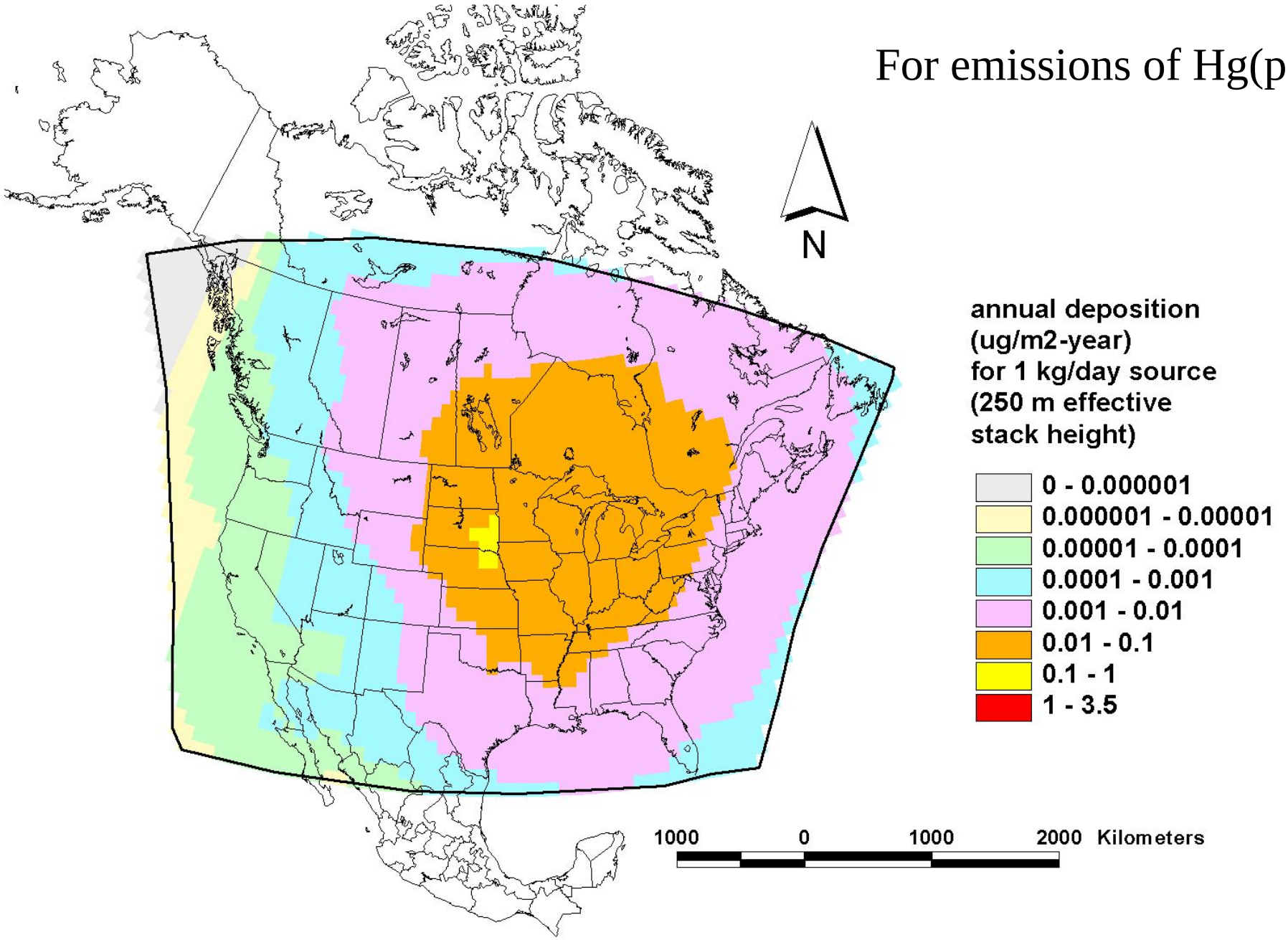

1º x 1º grid over entire modeling domain
source location
N
900 0 900 1800 Kilometers


Results tabulated on a 1° x 1° grid over model domain
Daily variations in total deposition flux arising from a hypothetical 1 kg/day source of Hg(II) (250 m effective stack height)
Source Location
Daily values for May 1996 will be shown (julian days 121-151)
And now for the movie…
Daily values for each grid square will be shown as "ug/m2-year" as if the deposition were to continue at that particular daily rate for an entire year
500 0 500 1000 1500 Kilometers

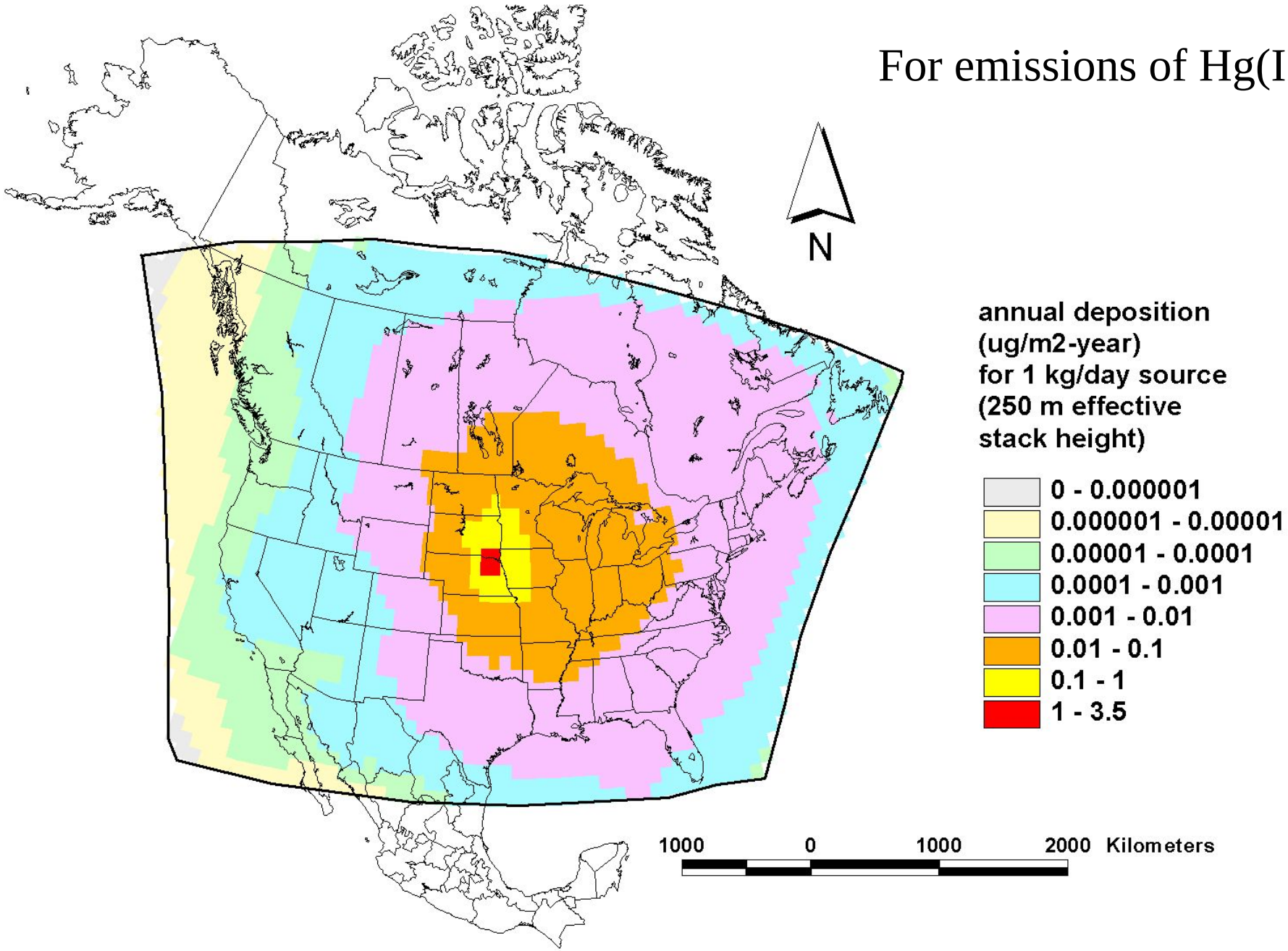
For emissions of Hg(0)



For emissions of Hg(p)

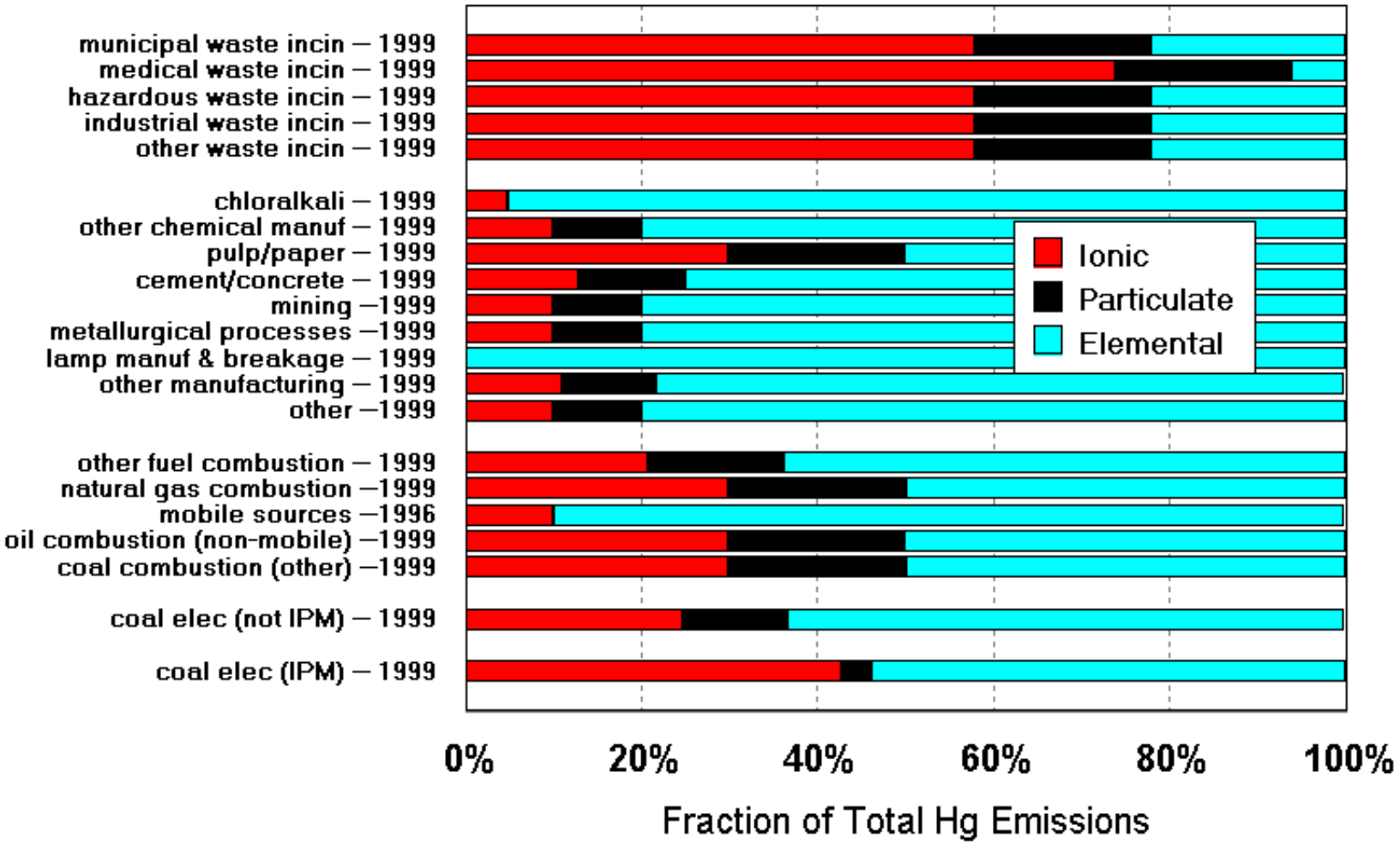


For emissions of Hg(II)

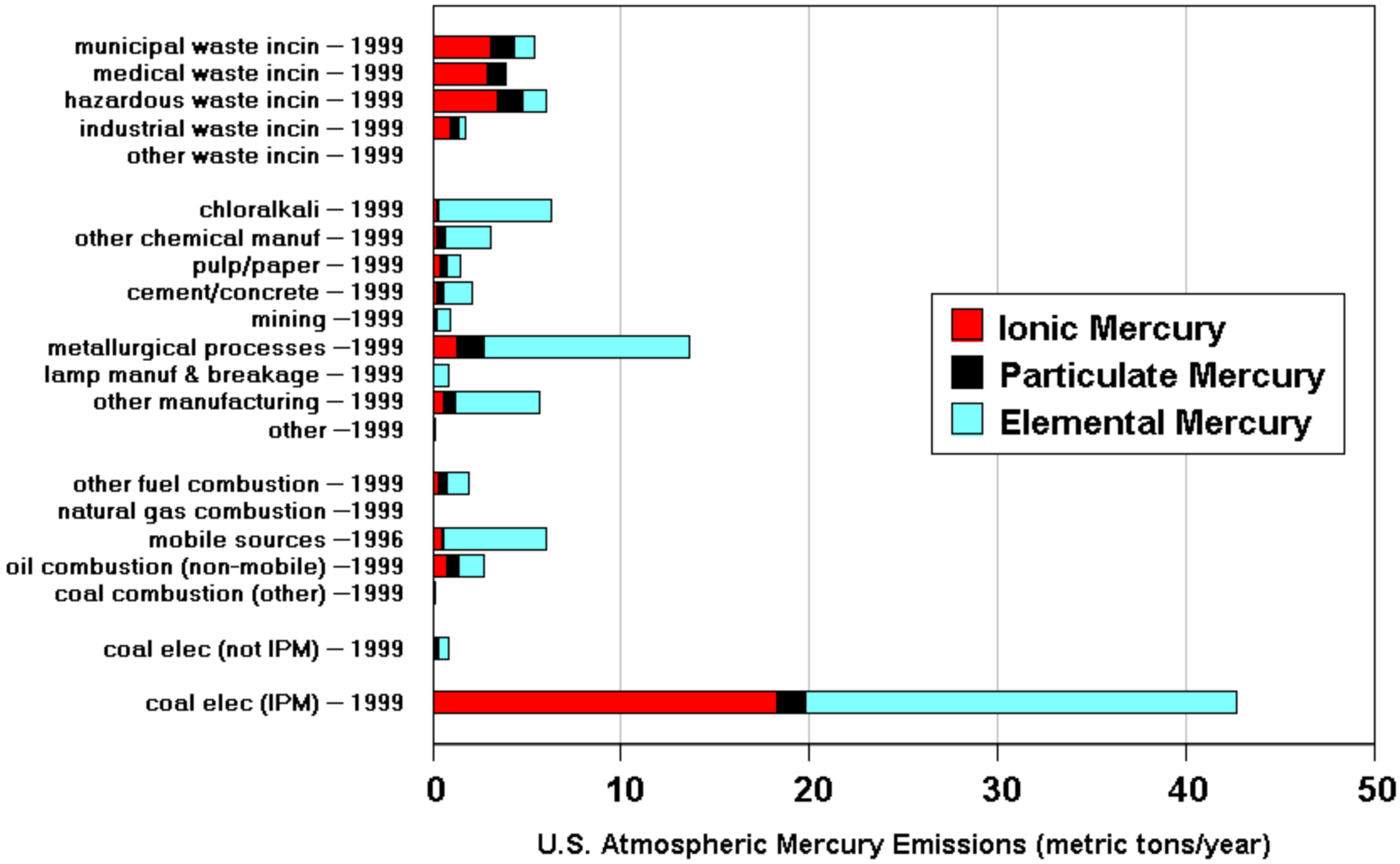


Estimated Speciation Profile for 1999 U.S. Atmospheric Anthropogenic Mercury Emissions

Very uncertain for most sources



Estimated 1999 U.S. Atmospheric Anthropogenic Mercury Emissions

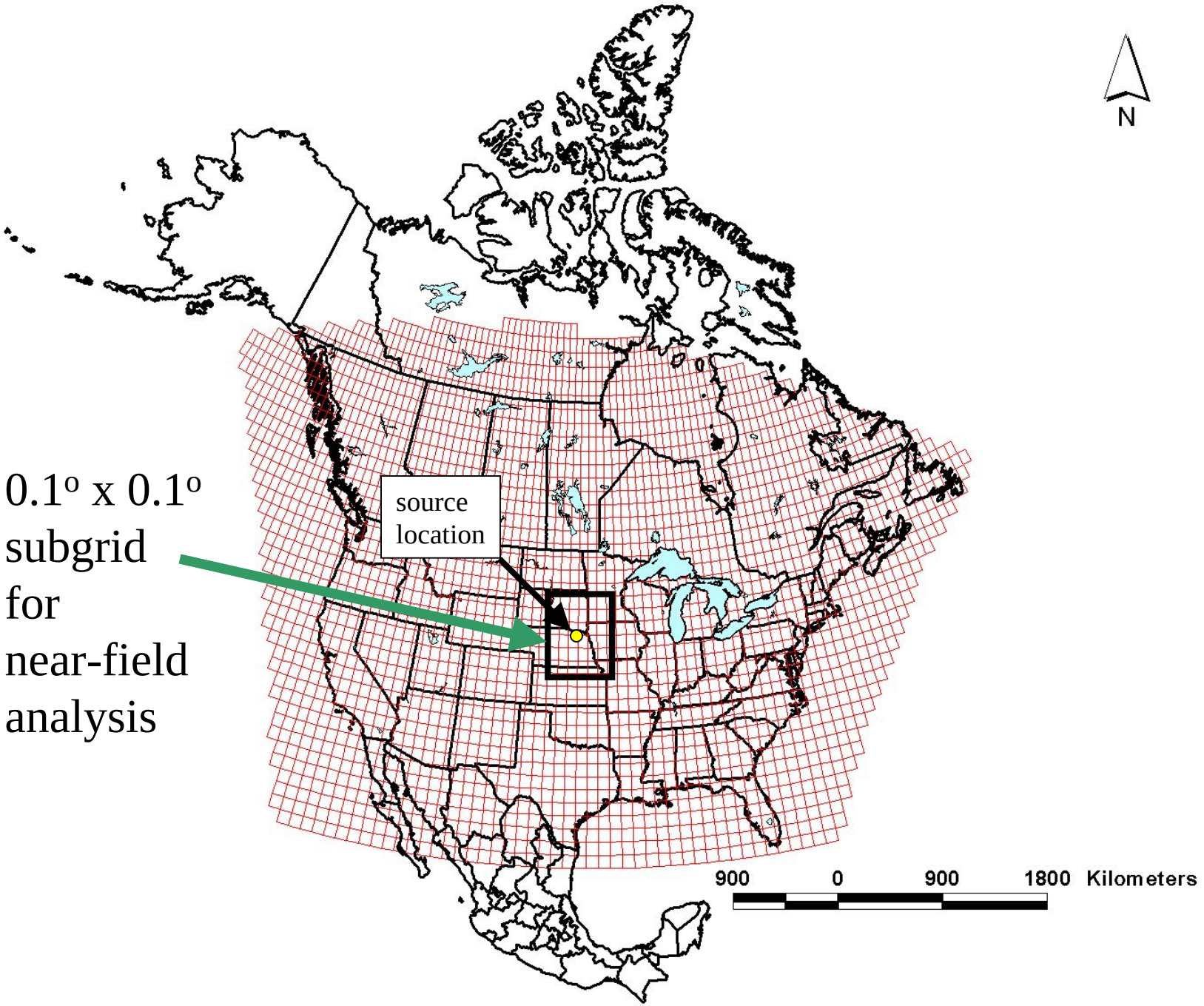


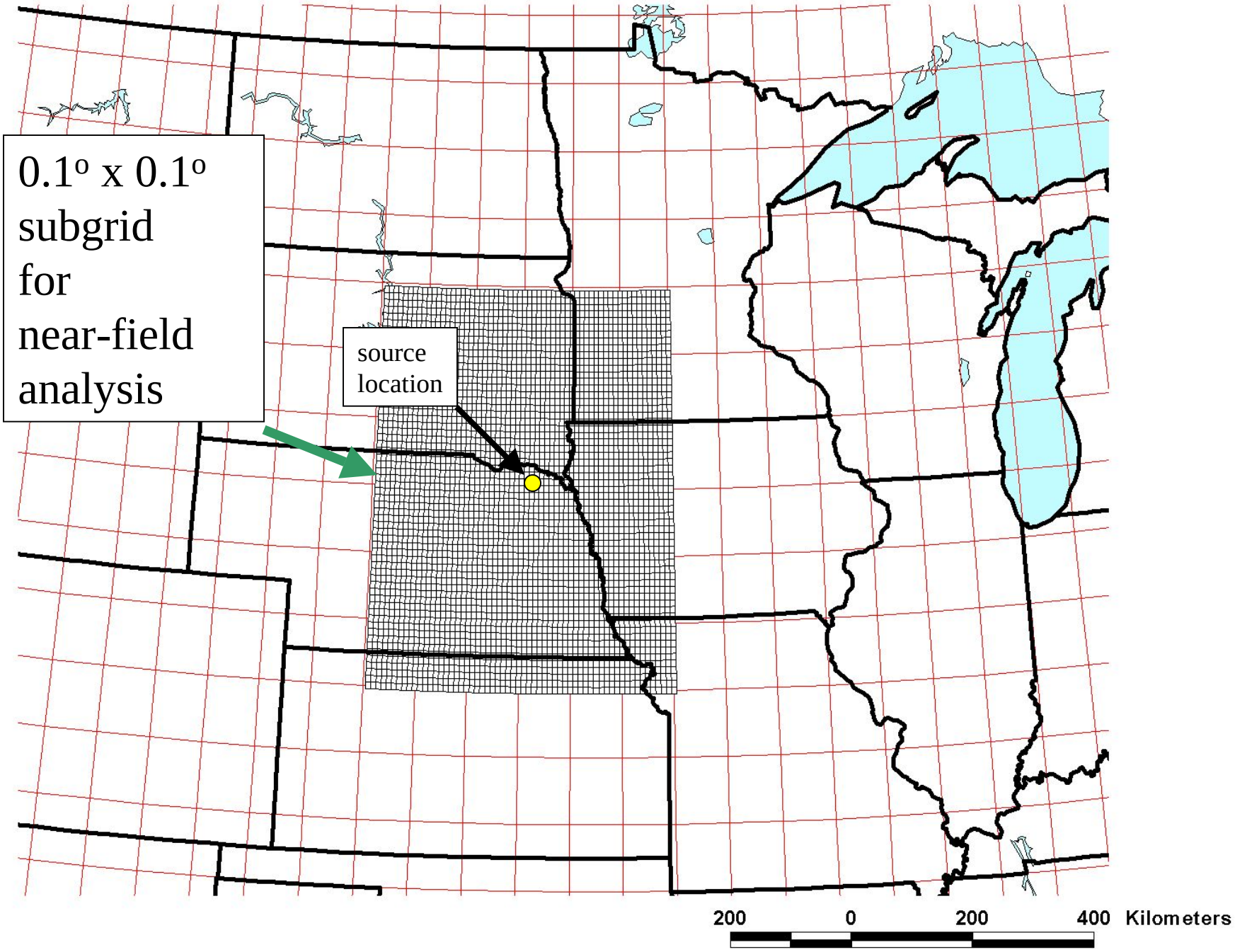
- **Each type of source has a very different emissions speciation profile**

- **Even within a given source type, there can be big differences – depending on process type, fuels and raw materials, pollution control equipment, etc.**

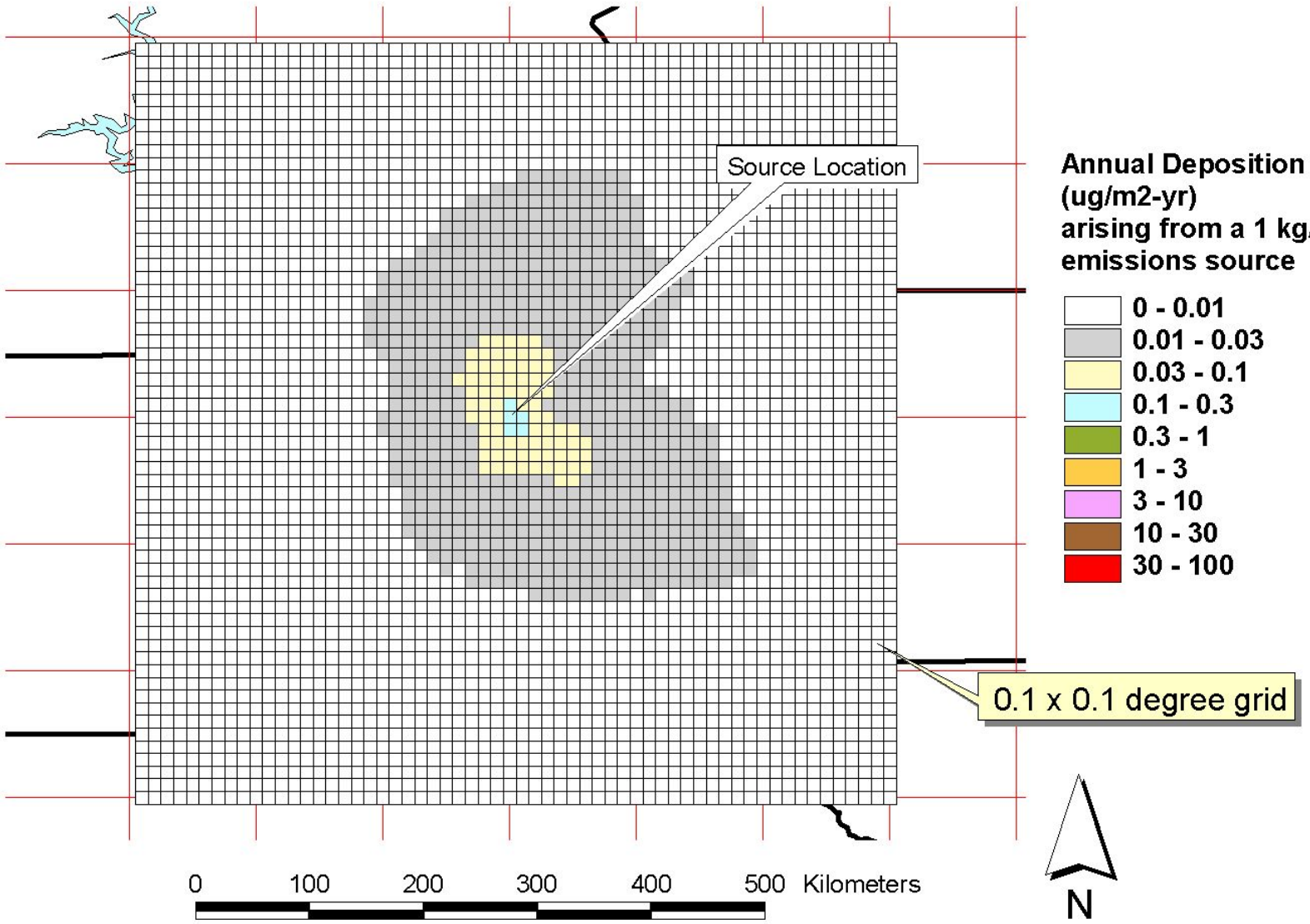
3. There can be large local and regional impacts from any given source

- **same hypothetical 1 kg/day source of RGM**
- **source height 250 meters**
- **exactly the same simulation, but results tabulated on a 0.1^{o} x 0.1^{o} receptor grid**
- **overall results for an entire year (1996)**


0.1° x 0.1°
subgrid
for
near-field
analysis
source
location
N
900
0
900
1800
Kilometers

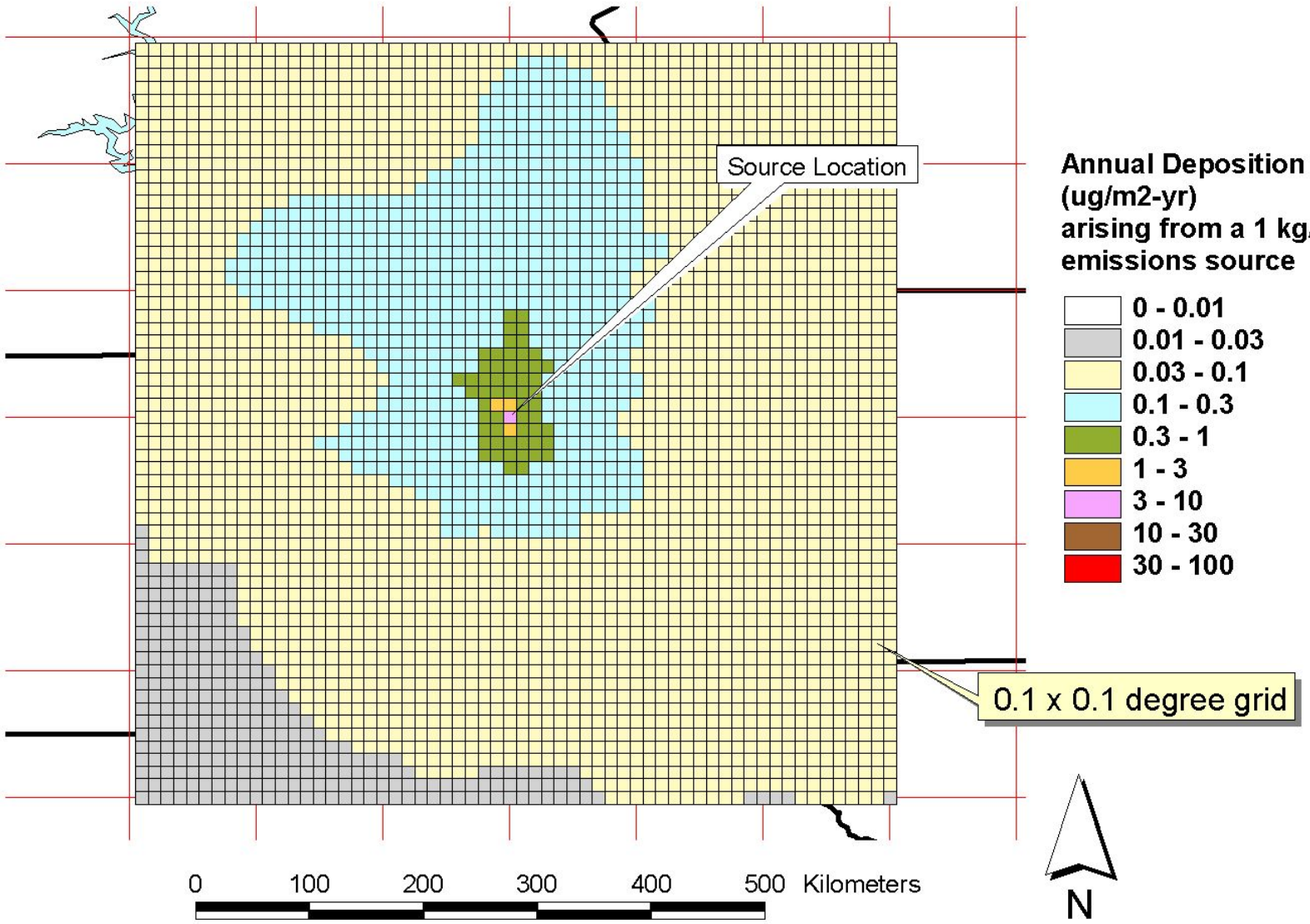

0.1° x 0.1°
subgrid
for
near-field
analysis
source
location
200
0
200
400
Kilometers

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



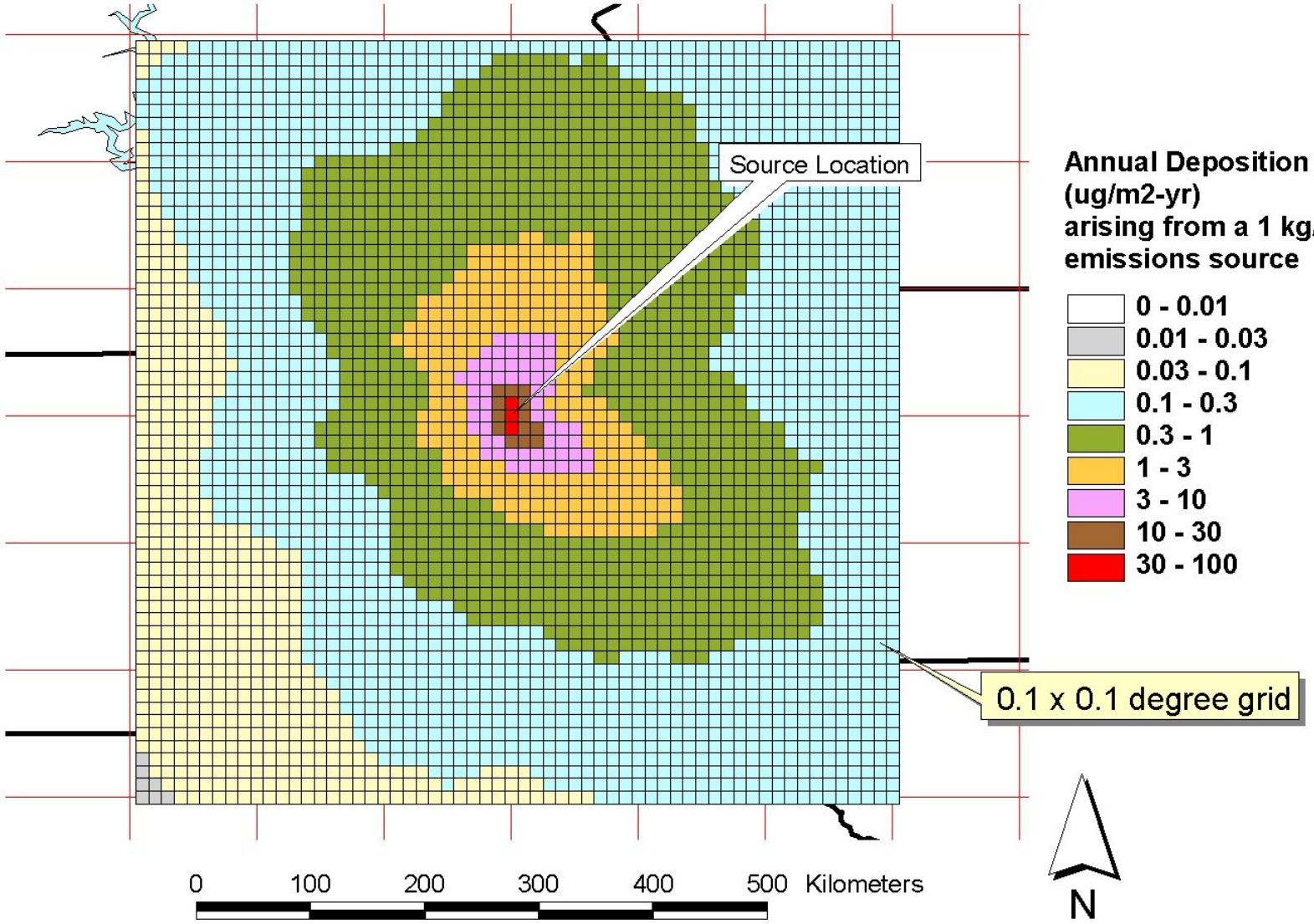
Hypothetical emissions source at lat = 42.5, long = -97.5;
simulation for entire year 1996 using archived NGM meteorology (180 km resolution)

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



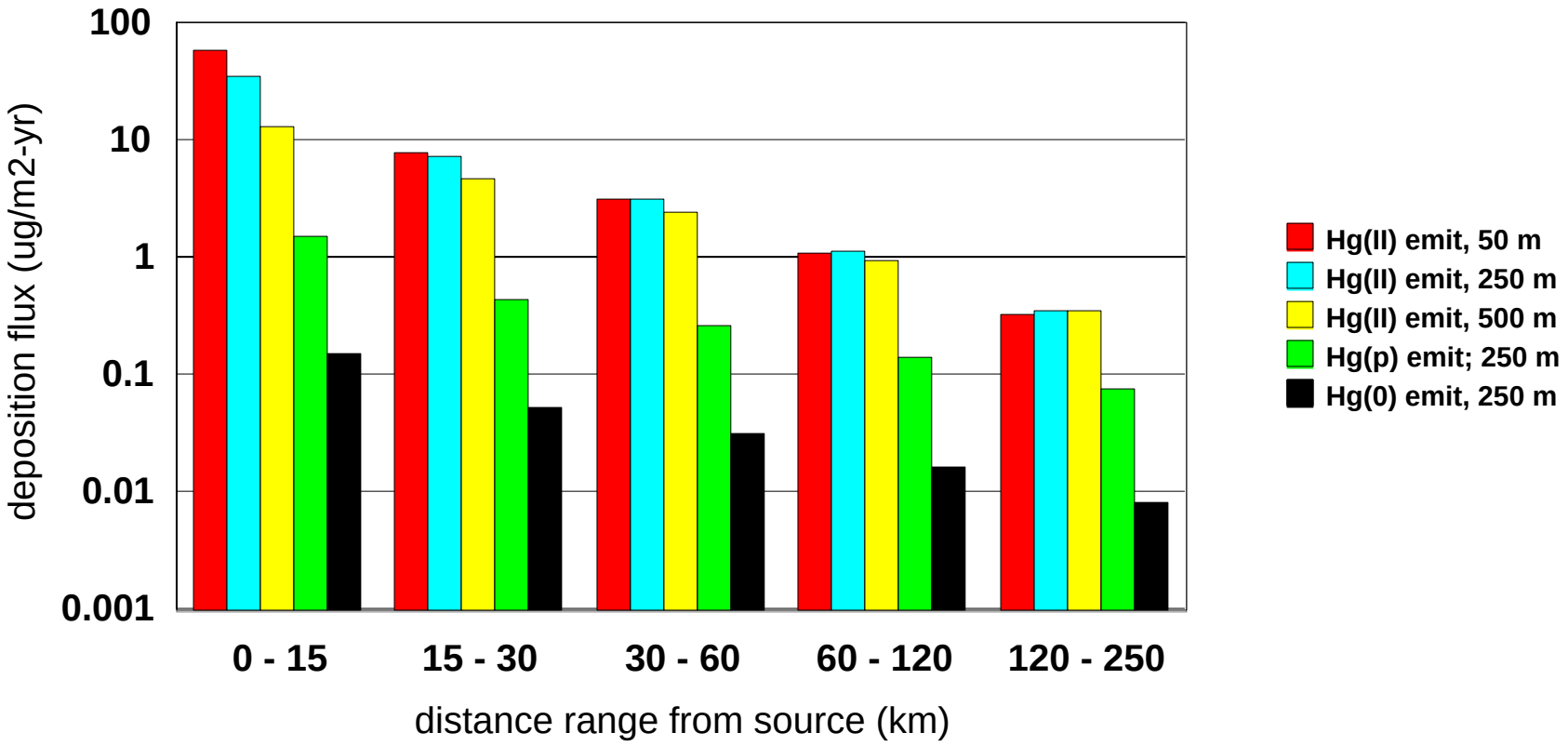
Hypothetical emissions source at lat = 42.5, long = -97.5;
simulation for entire year 1996 using archived NGM meteorology (180 km resolution)

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



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

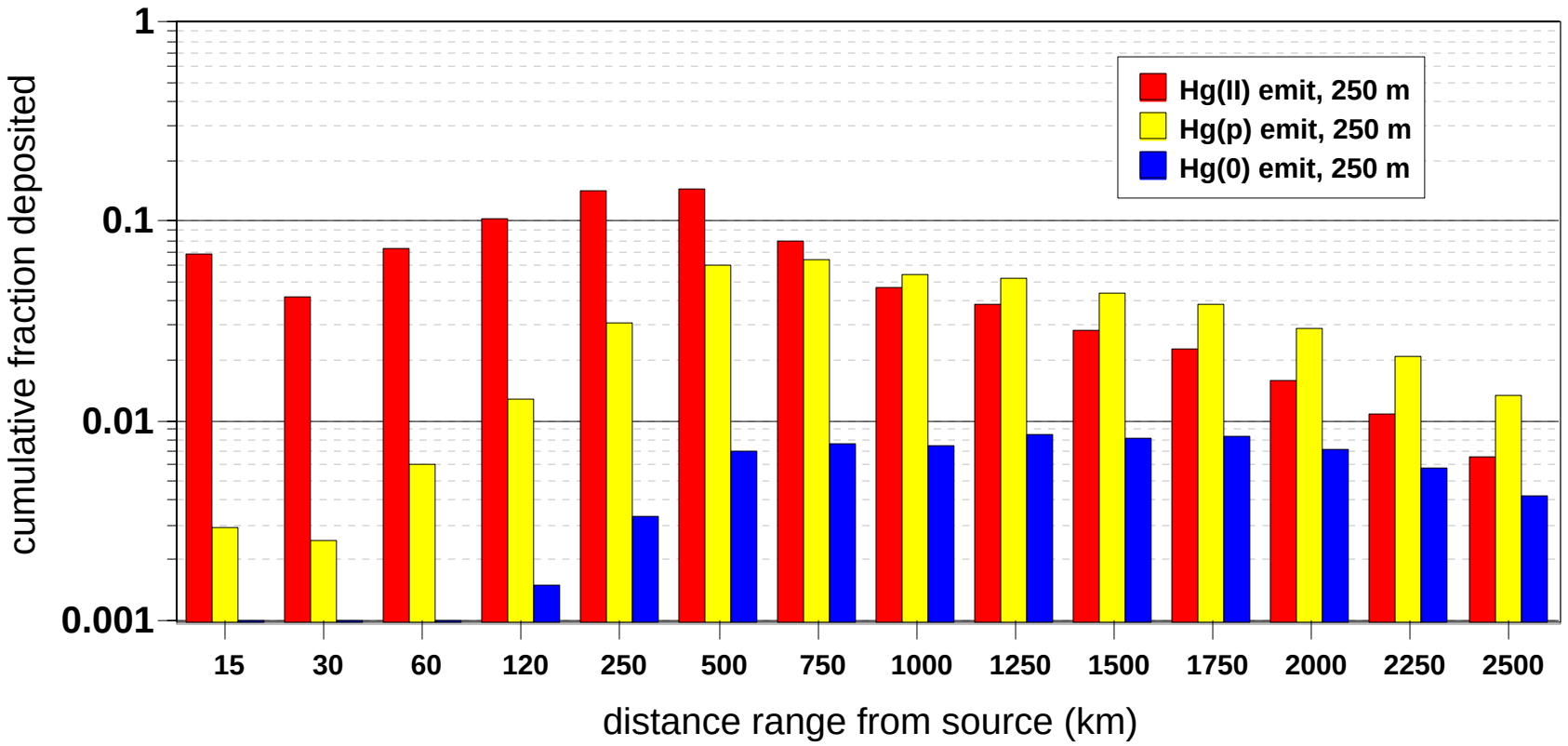
Deposition flux within different distance ranges from a hypothetical 1 kg/day source



Source at Lat = 42.5, Long = -97.5; simulation for entire year 1996 using archived NGM meteorological data

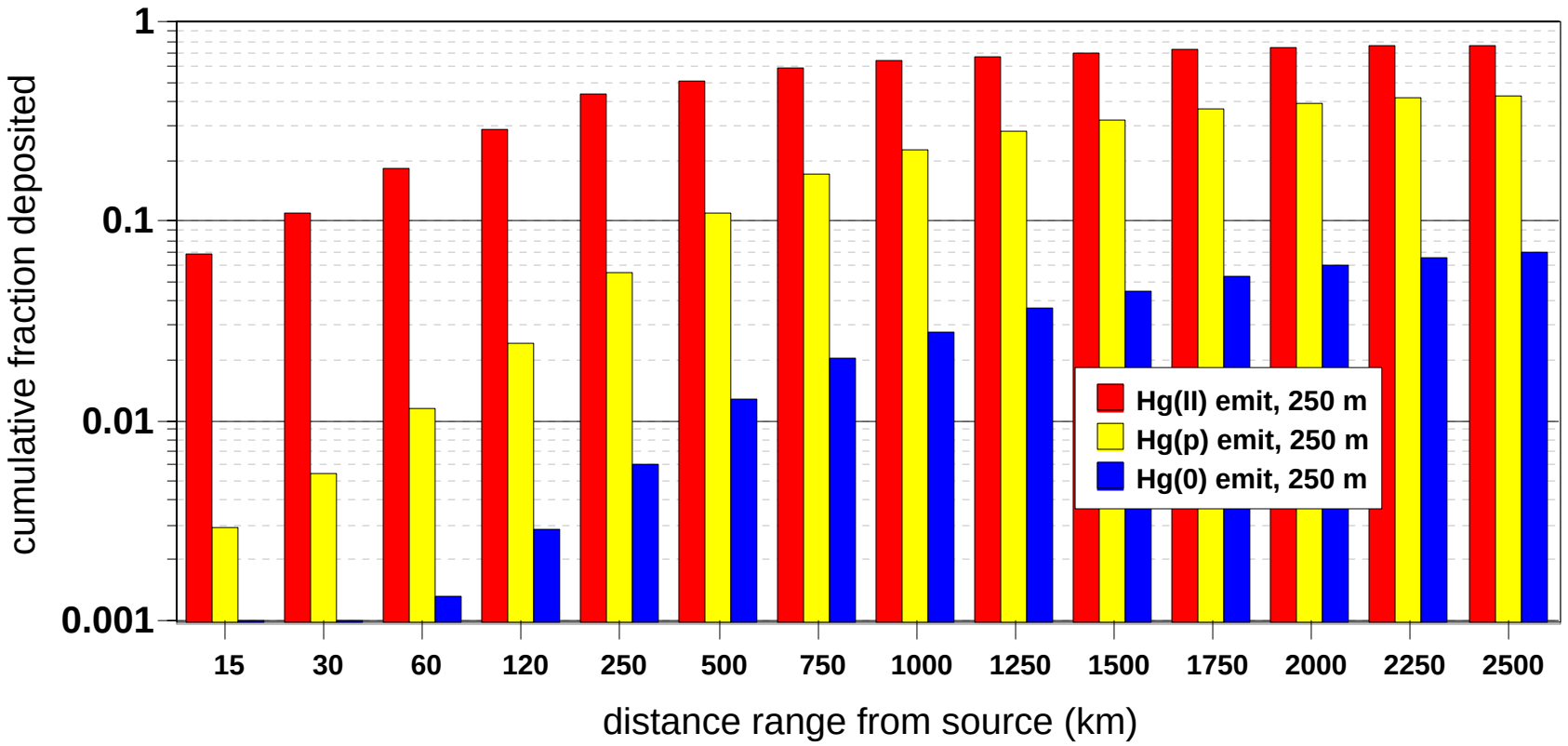
4. At the same time, medium to long range transport can't be ignored

Fraction deposited within concentric regions away from a hypothetical source



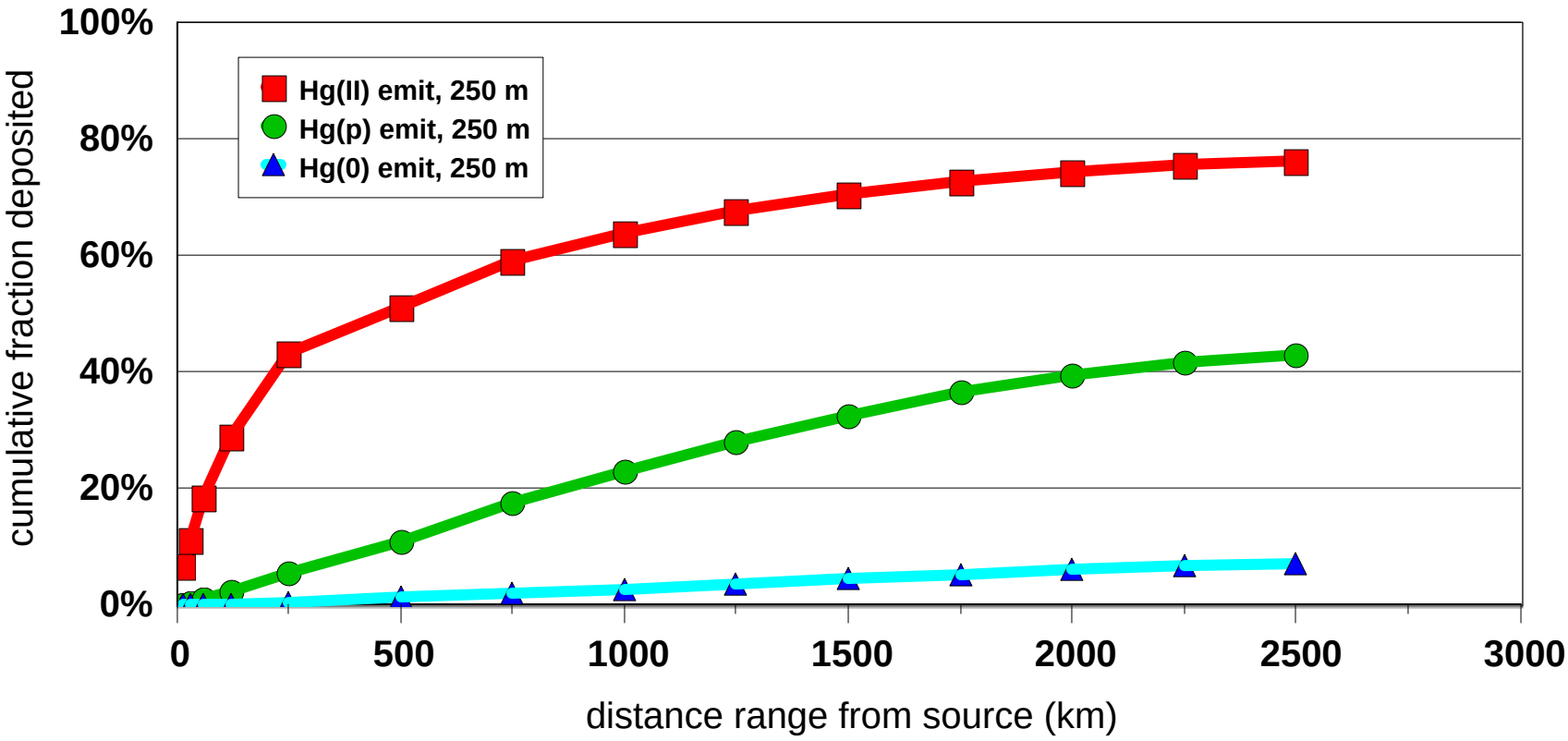
Source at Lat = 42.5, Long = -97.5; simulation for entire year 1996 using archived NGM meteorological data

Cumulative fraction deposited within different distances from a hypothetical source



Source at Lat = 42.5, Long = -97.5; simulation for entire year 1996 using archived NGM meteorological data

Cumulative fraction deposited out to different distance ranges from a hypothetical source

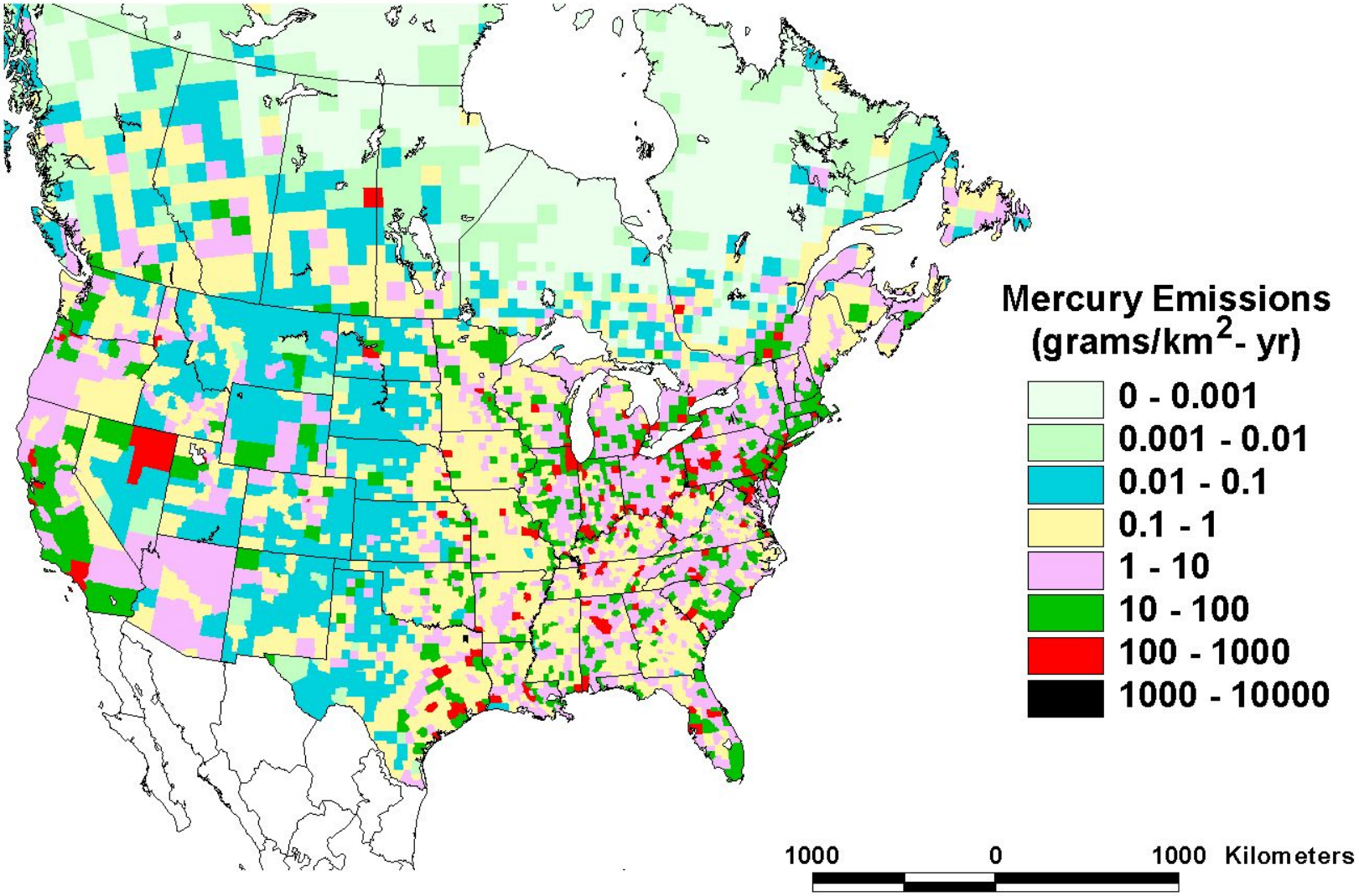


Source at Lat = 42.5, Long = -97.5; simulation for entire year 1996 using archived NGM meteorological data

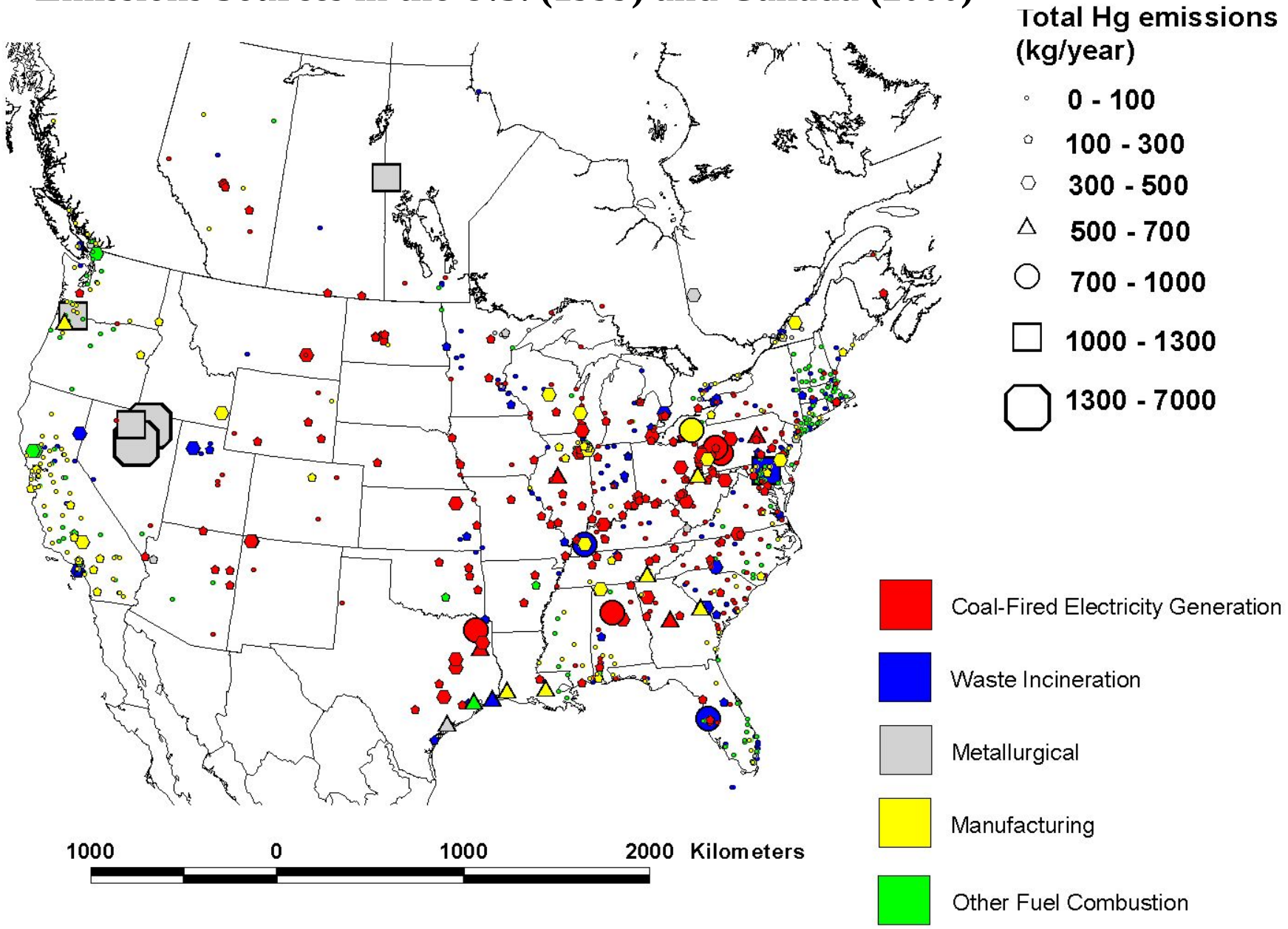
5. There are a lot of sources…

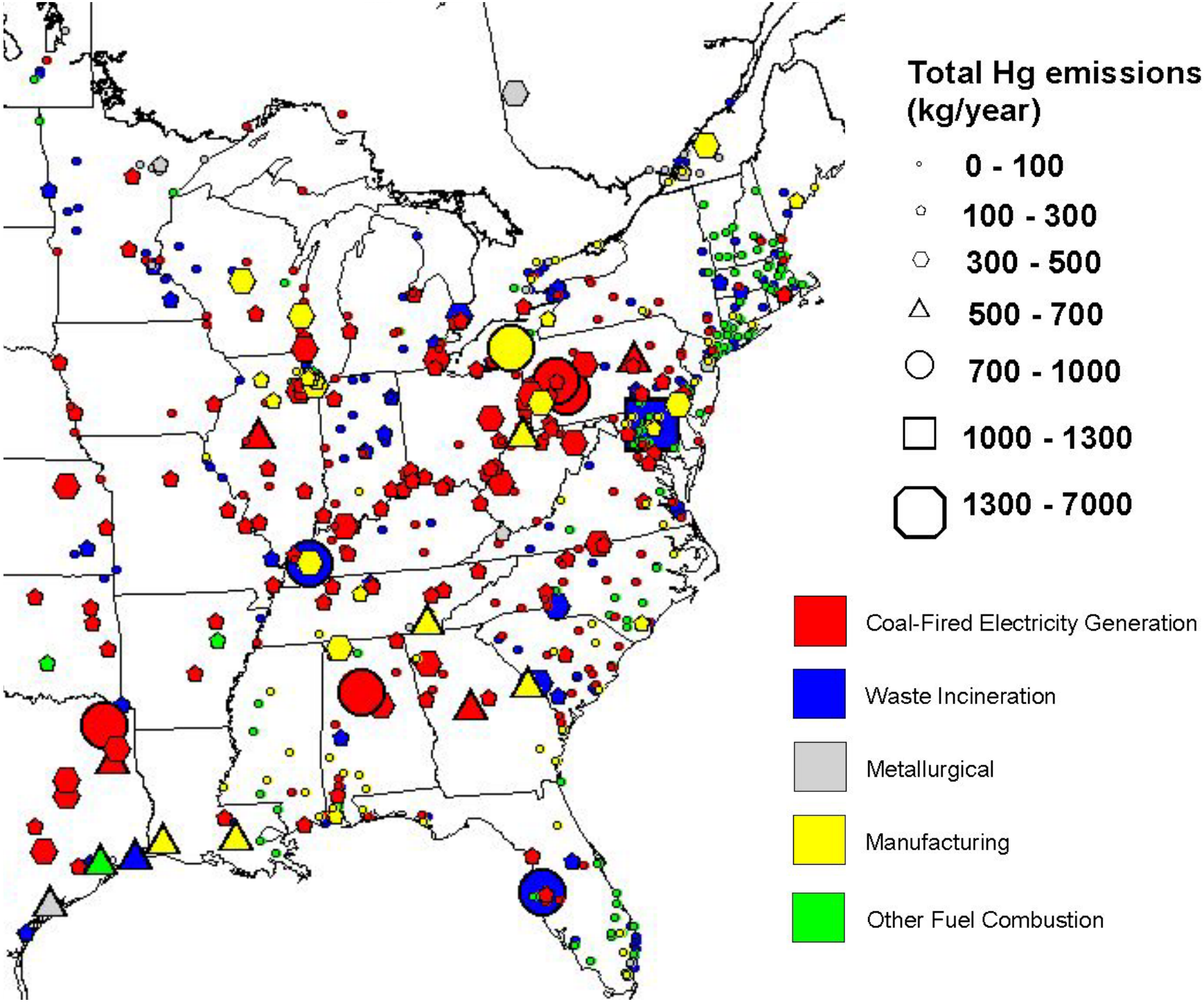
- **Large spatial and temporal variations**
- **Each source emits mercury forms in different proportions**
- **A lot of different sources can contribute significant amounts of mercury through atmospheric deposition to any given receptor**

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



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

6. Getting the source-apportionment information we all want is difficult

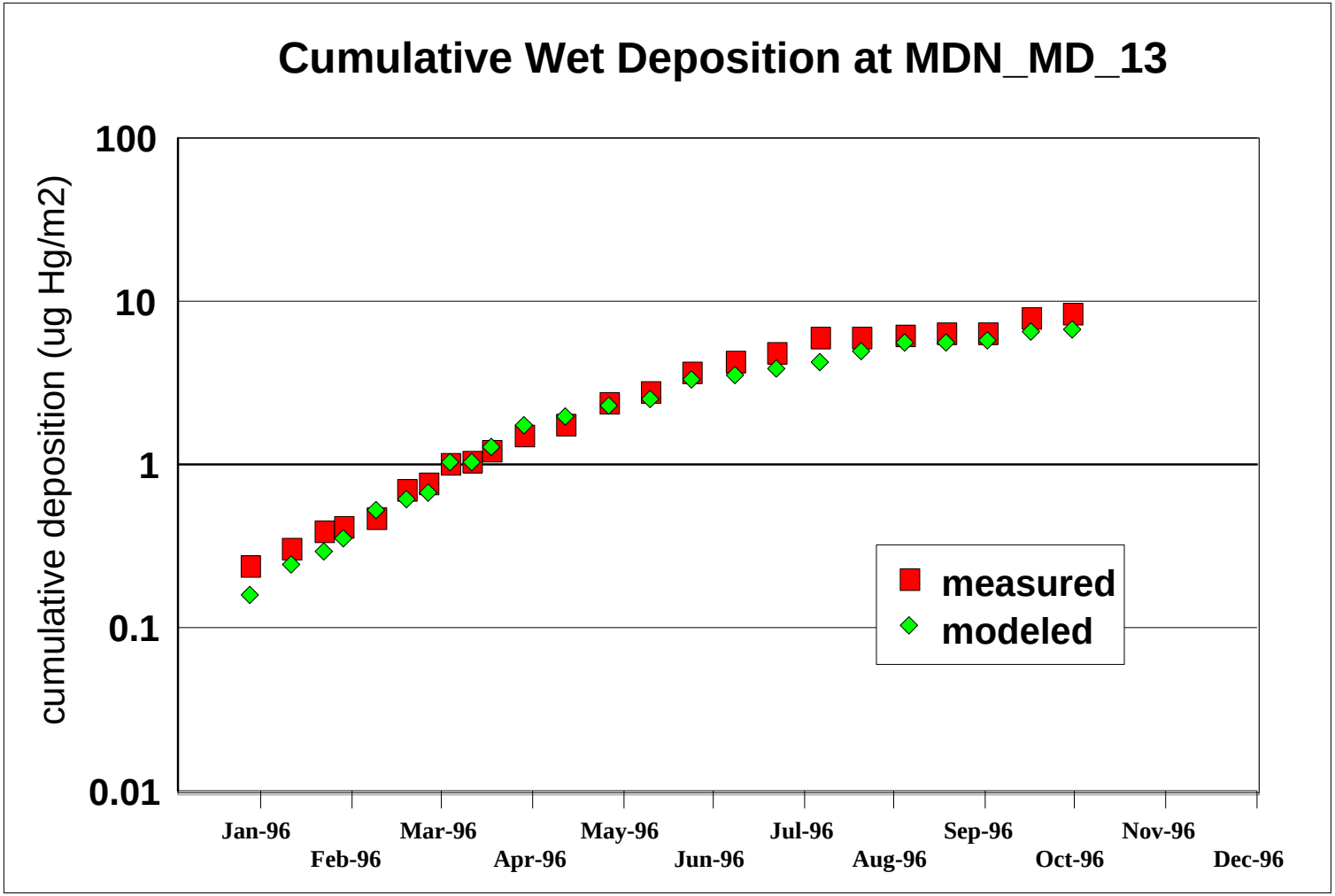
- **With measurements alone, generally impossible**
- **Coupling measurements with back-trajectory analyses yields only a little information**
- **Comprehensive fate and transport modeling – "forward" from emissions to deposition – holds the promise of generating detailed source-receptor information**

7. There are a lot of uncertainties in current comprehensive fate and transport models

- **atmospheric chemistry of mercury**
- **concentrations of key reactants**
- **mercury emissions (amounts & speciation profile)**
- **meteorological data (e.g., precipitation)**

8. Nevertheless, many models seem to be performing reasonably well, i.e., are able to explain a lot of what we see

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



Convention on Long-Range Transboundary Air Pollution

emep

Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe

TECHNICAL REPORT 1/2003 June 2003

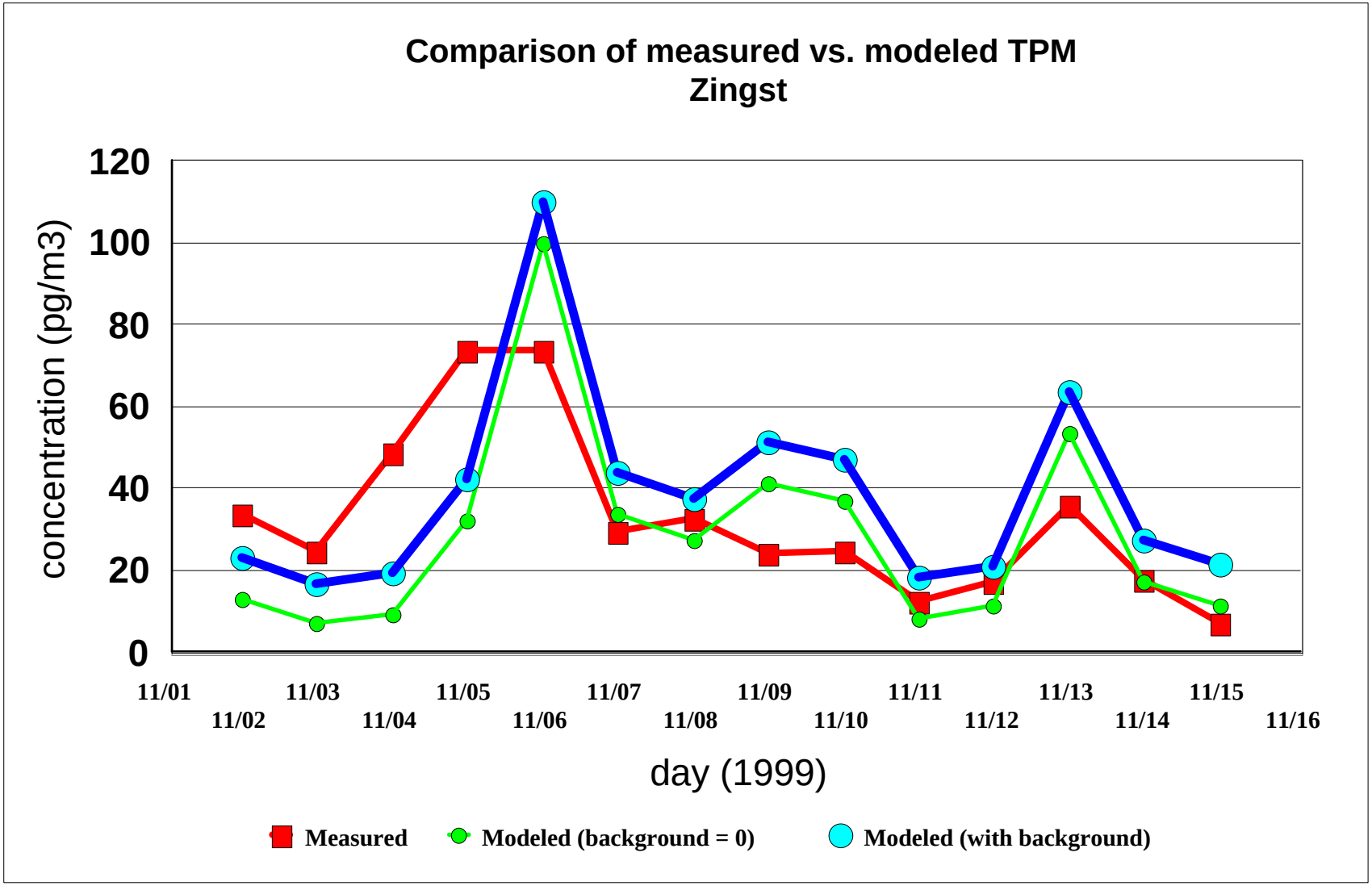
Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury

Stage II. Comparison of modeling results with observations obtained during short-term measuring campaigns

Technical Report 1/2003

A. Ryaboshapko, R. Artz,
R. Bullock, J. Christensen,
M. Cohen, A. Dastoor,
D. Davignon, R. Draxler,
R. Ebinghaus, I. Ilyin,
J. Munthe, G. Petersen,
D. Syrakov

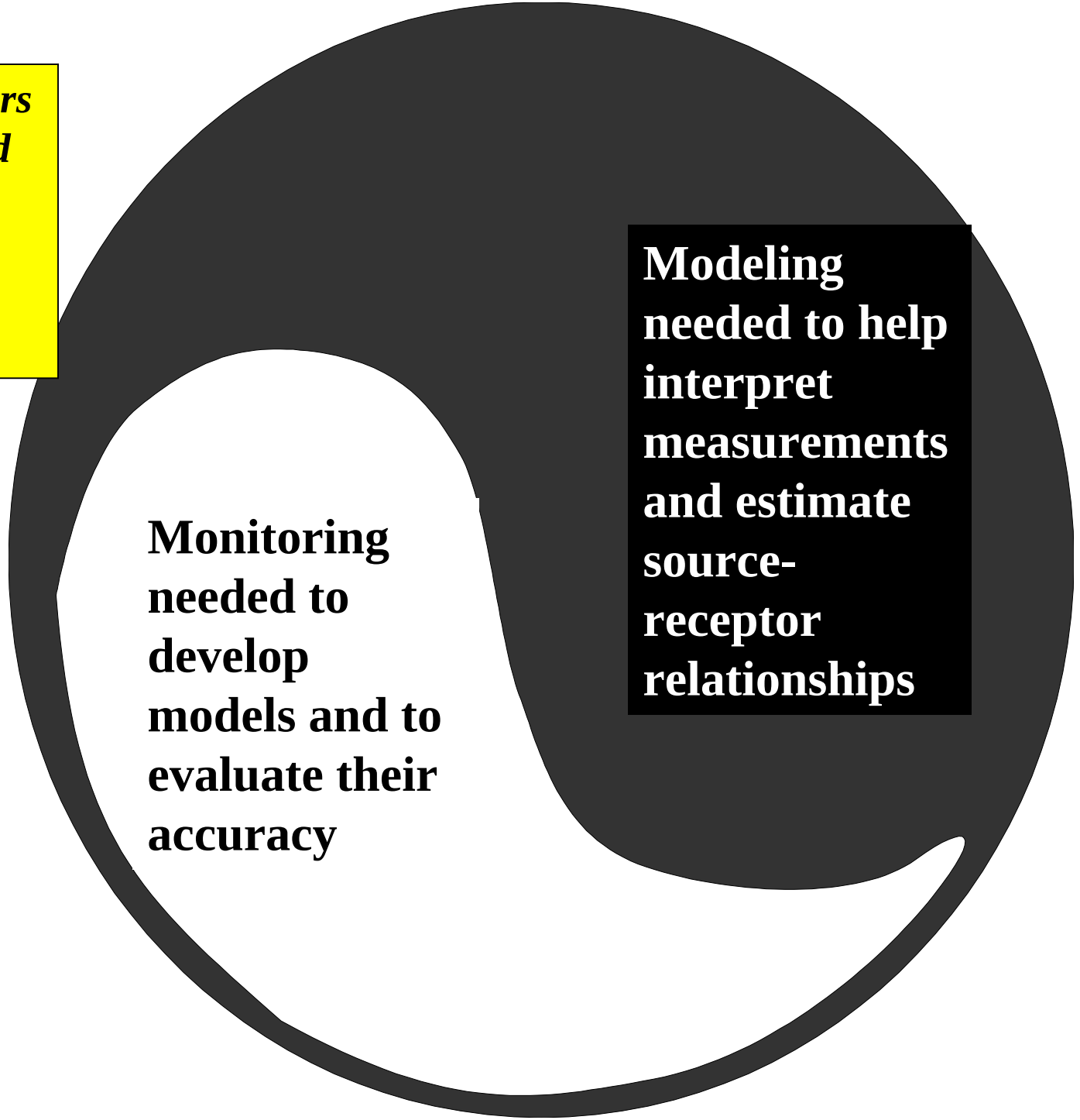
msc-e


Comparison of measured vs. modeled TPM
Zingst
concentration (pg/m3)
120
100
80
60
40
20
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)

9. A model does not have to be perfect in order to be useful

- **Often, most decisions just require qualitatively reasonable results**
- **And realistically, most if not all data and information used in decision-making has uncertainties (e.g., public health impacts, economic impacts)**
- **So, we shouldn't demand perfection of models**

10. To get the answers we need, we need to use both monitoring and modeling -- <u>together</u>



11. MDN is GREAT!…but there are some big gaps in atmospheric monitoring – making it very difficult to evaluate and improve models

- **We desperately need national MDN-like network to measure ambient air concentrations of Hg0, Hg(p), and RGM, *with readily available data***
- **What is RGM? What is Hg(p)?**
- **Both "background/regional" *and* near-source measurements needed…**
- **Measurements at different heights in the atmosphere**

Dry Deposition?

- ***Dry deposition is important, and difficult – if not impossible – to measure reliably with current techniques…***
- ***Essentially all dry deposition estimates made currently are made by applying models***
- ***National ambient network of speciated ambient measurements will help to evaluate and improve models of dry deposition***

Source-Apportionment

where does the *mercury* in *mercury deposition* come from?

Source-apportionment answers depend on

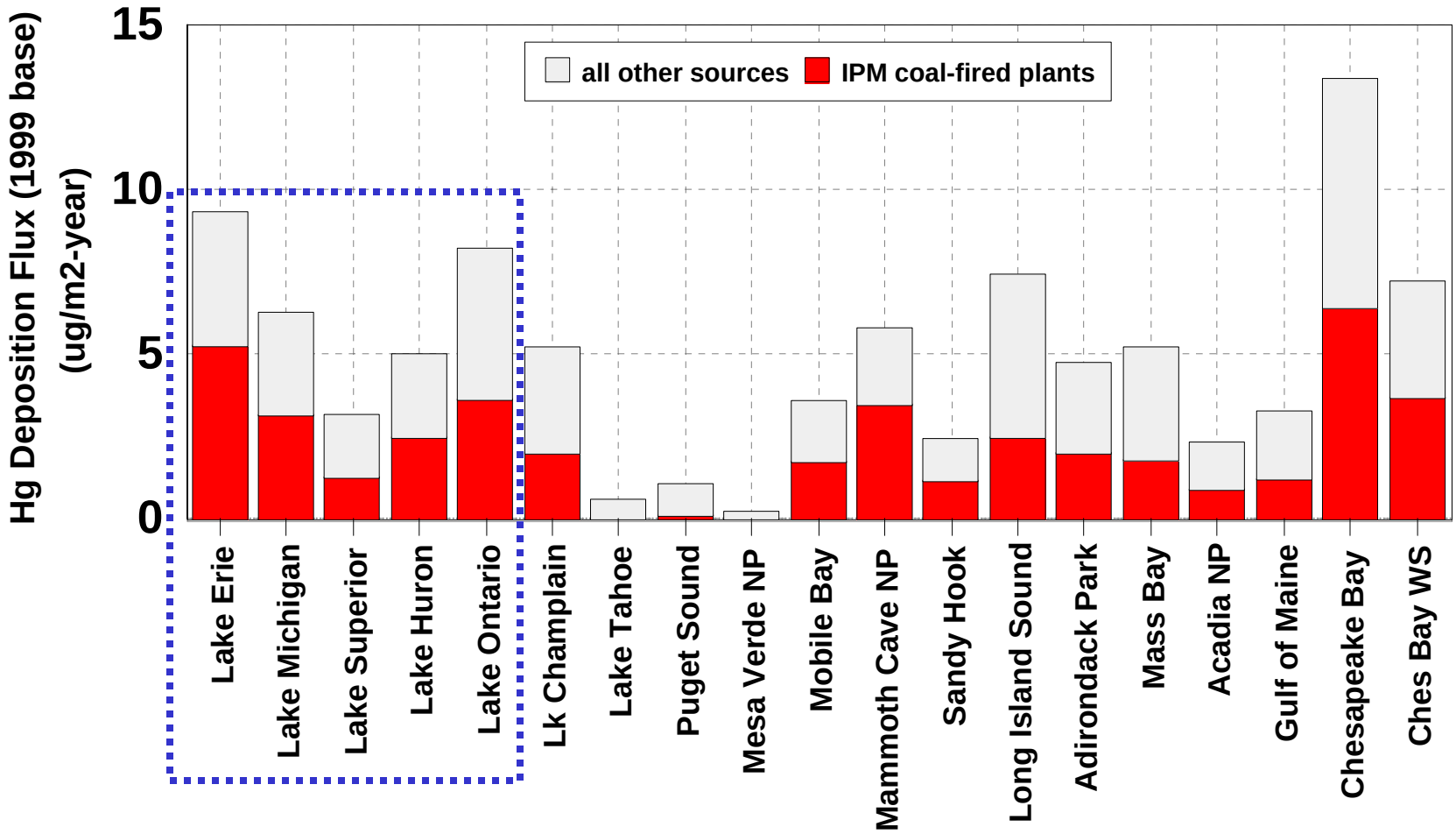
- ***where*** **you are, and**
- ***when*** **you are**

(and the effects of deposition will be different in each ecosystem)

- **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**

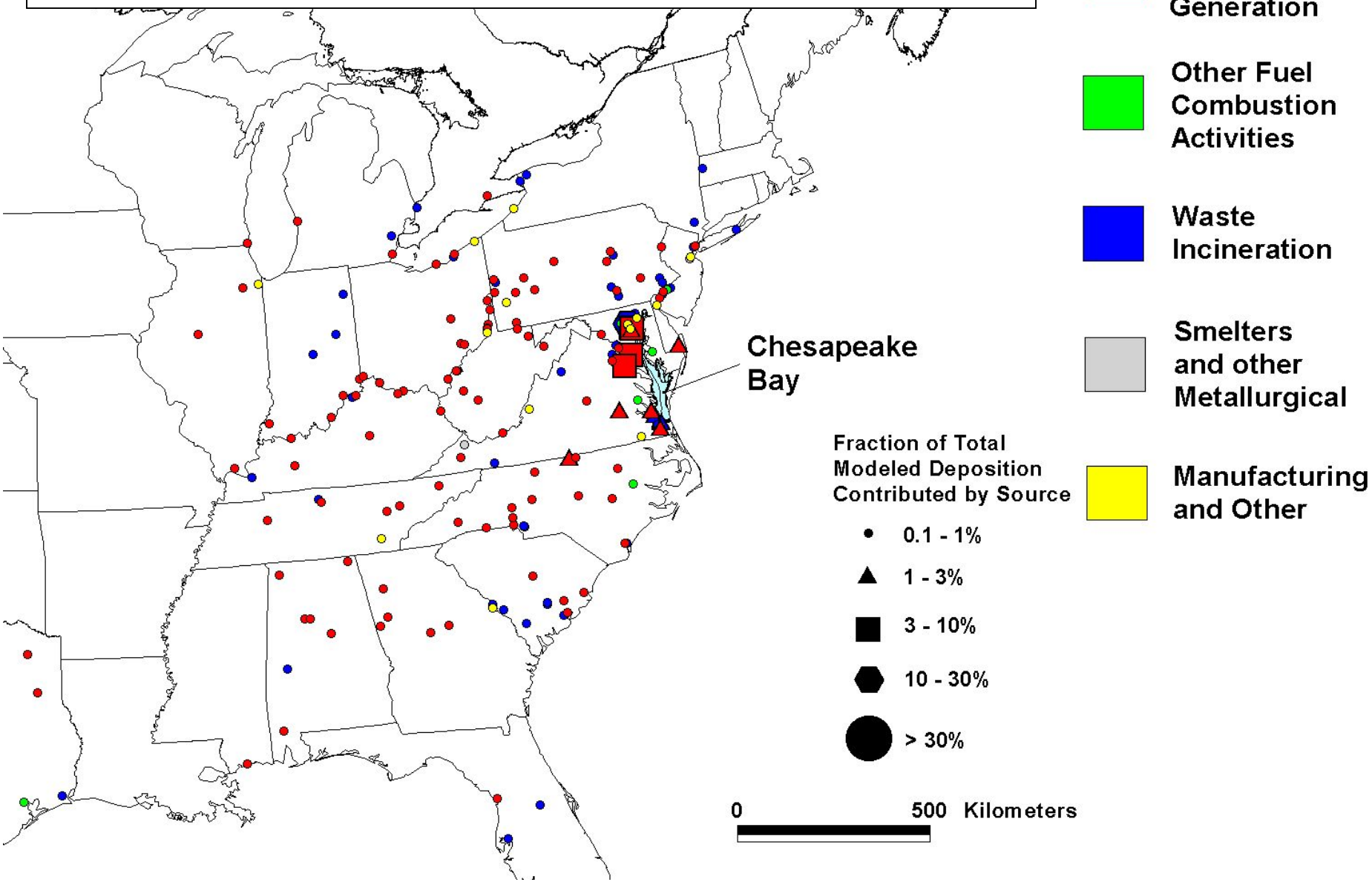
- **For areas *with large emissions sources***
  - **the deposition will be higher**
  - **and be more strongly influenced by these large emissions sources...**

Mercury deposition at selected receptors arising from 1999 base-case emissions from anthropogenic sources in the United States and Canada
(IPM coal fired plants are large coal-fired plants in the U.S. only)

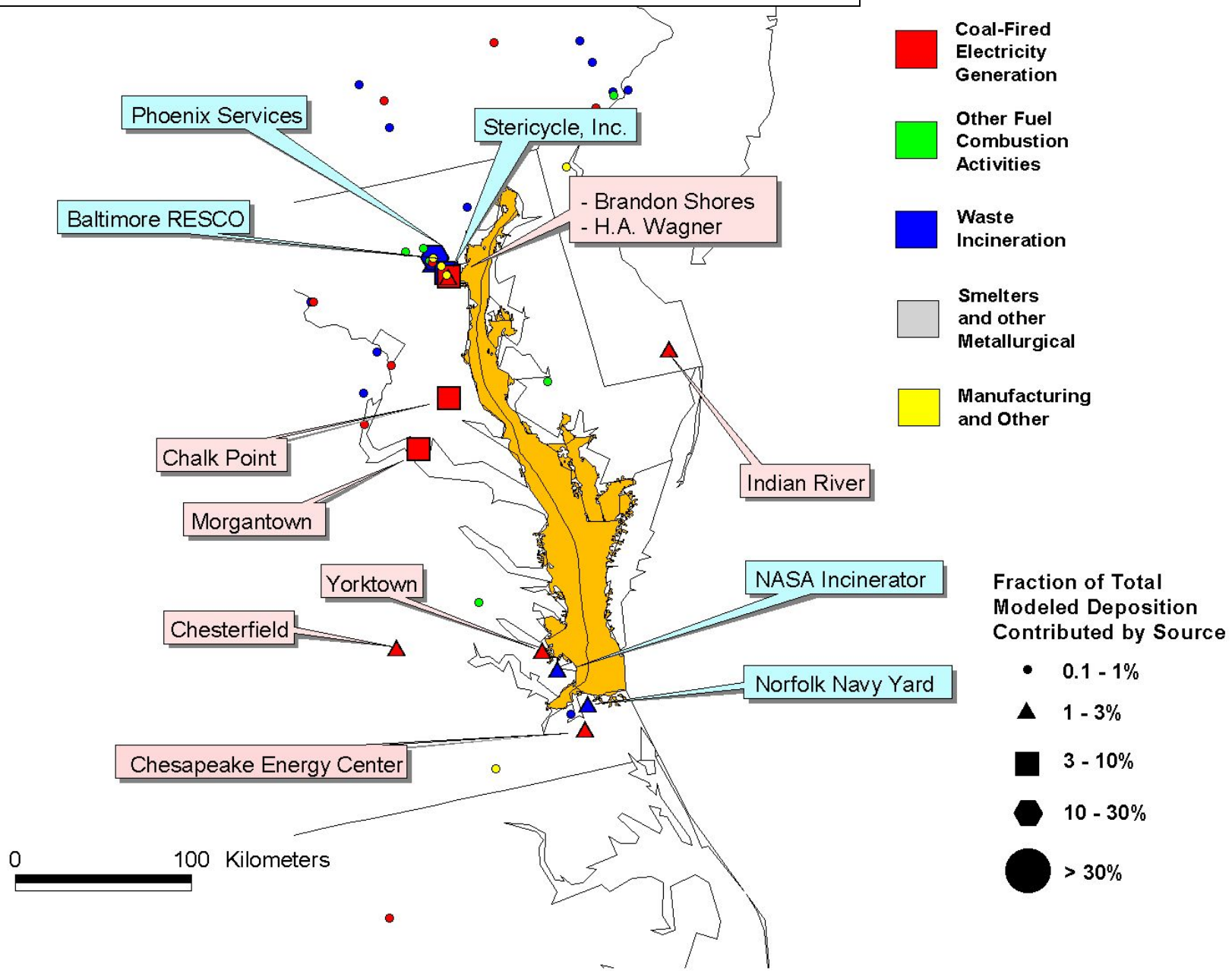


Example of modeling results: Chesapeake Bay

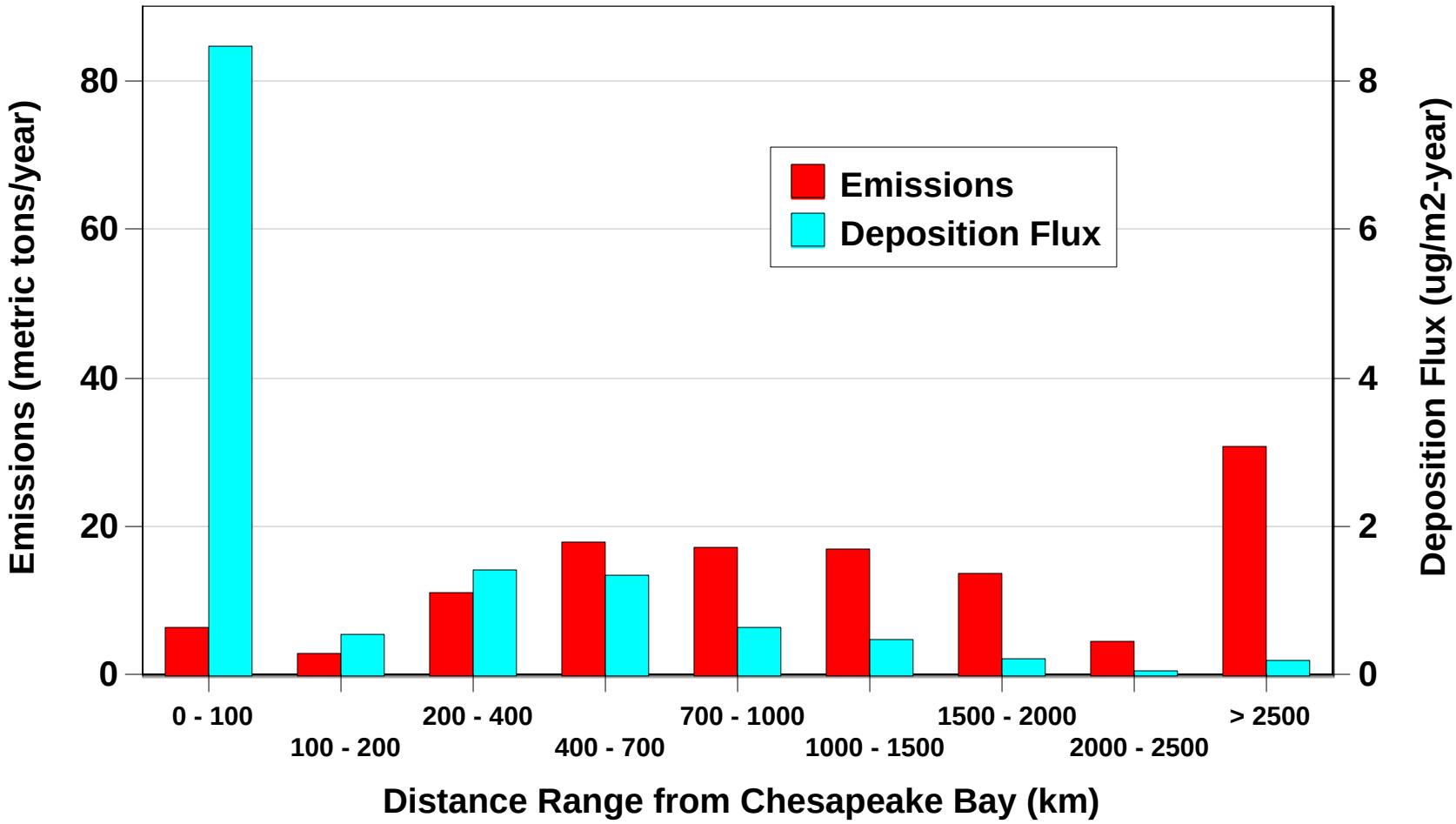
Largest Regional Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay

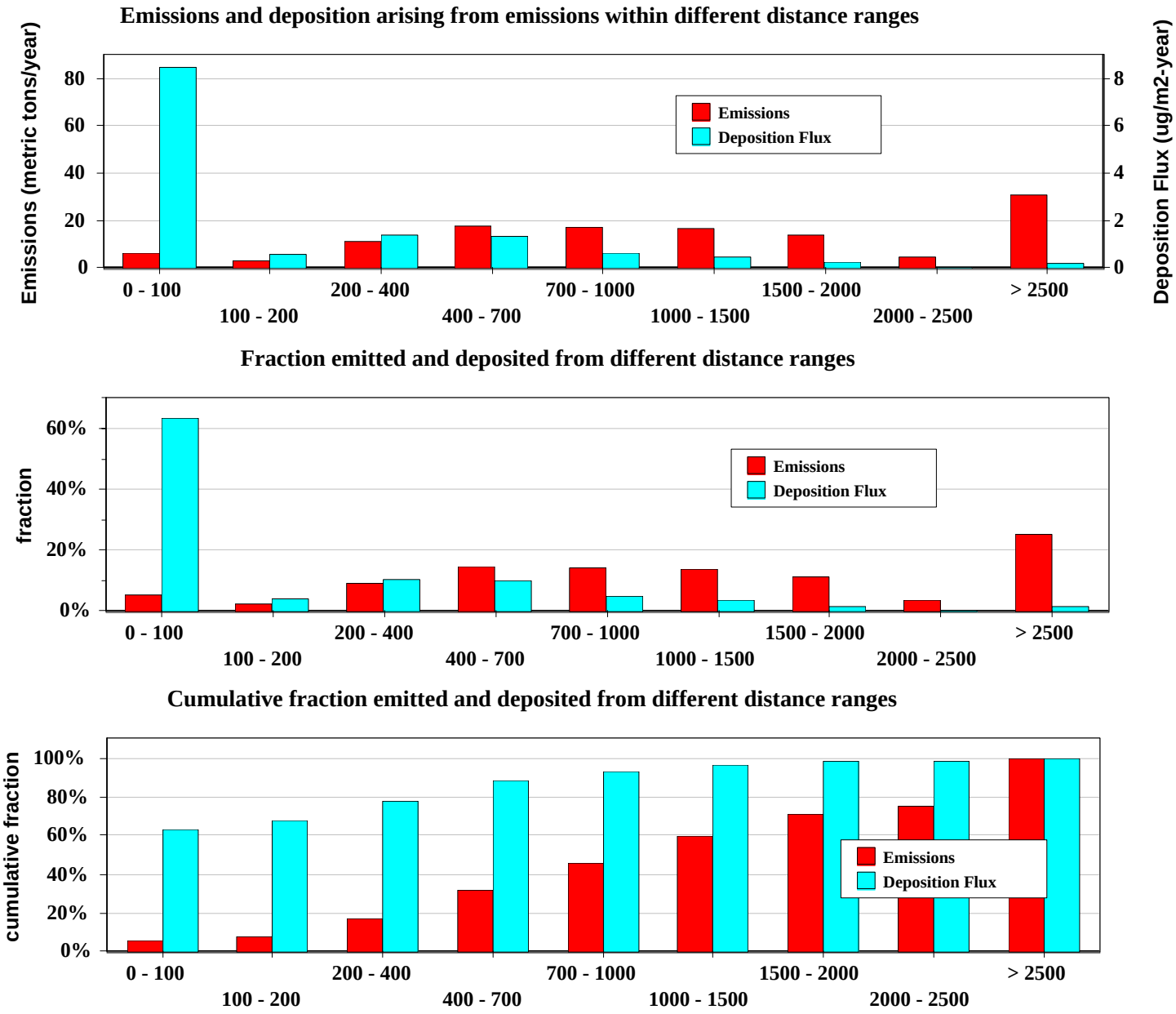


Largest Local Individual Sources Contributing to 1999 Mercury Deposition Directly to the Chesapeake Bay

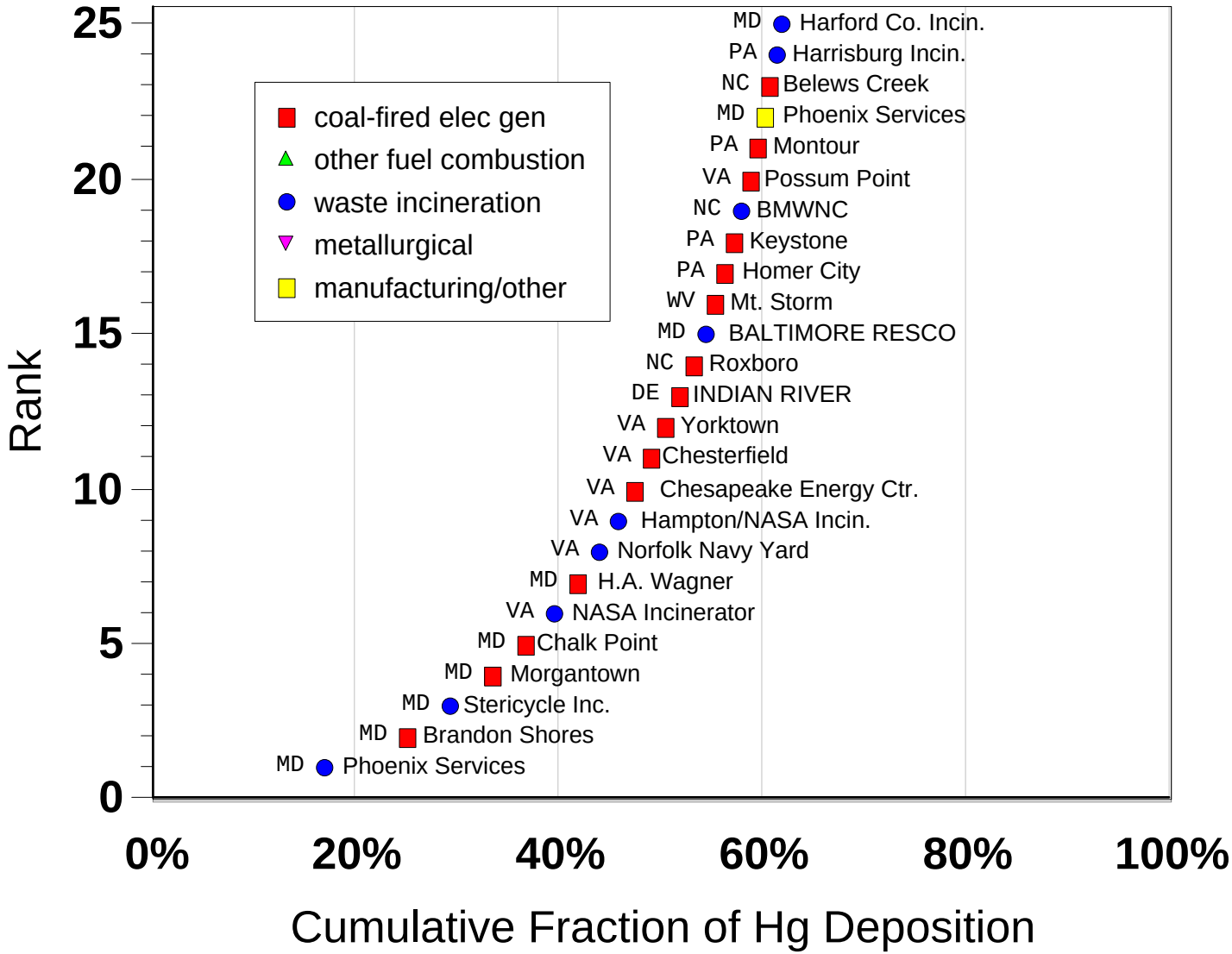


Emissions and Direct Deposition Contributions from Different Distance Ranges Away From the Chesapeake Bay




Emissions and deposition arising from emissions within different distance ranges
Emissions (metric tons/year)
Deposition Flux (ug/m2-year)
Emissions
Deposition Flux
0
20
40
60
80
0
2
4
6
8
0 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 1500
1500 - 2000
2000 - 2500
> 2500
Fraction emitted and deposited from different distance ranges
fraction
0%
20%
40%
60%
Emissions
Deposition Flux
0 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 1500
1500 - 2000
2000 - 2500
> 2500
Cumulative fraction emitted and deposited from different distance ranges
cumulative fraction
0%
20%
40%
60%
80%
100%
Emissions
Deposition Flux
0 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 1500
1500 - 2000
2000 - 2500
> 2500
Distance Range from the Chesapeake Bay (km)

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



Conclusions

- Source-attribution information is important
- Impacts are episodic & depend on form of mercury emitted
- Modeling needed to get source-attribution information
- (more!) Monitoring for model evaluation & refinement
- Models don't have to be perfect to give useful information
- Many uncertainties but useful model results are emerging
- Many opportunities exist for improvements in modeling/monitoring integrated approaches to develop source-attribution information