Modeling the Atmospheric Deposition of Mercury to Lake Champlain

(from Anthropogenic Sources in the U.S. and Canada)

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Coordination of Atmospheric Deposition Research in the Lake Champlain Basin
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Burlington, Vermont

Key questions regarding atmospheric deposition:

1. How much is being deposited in each Lake?

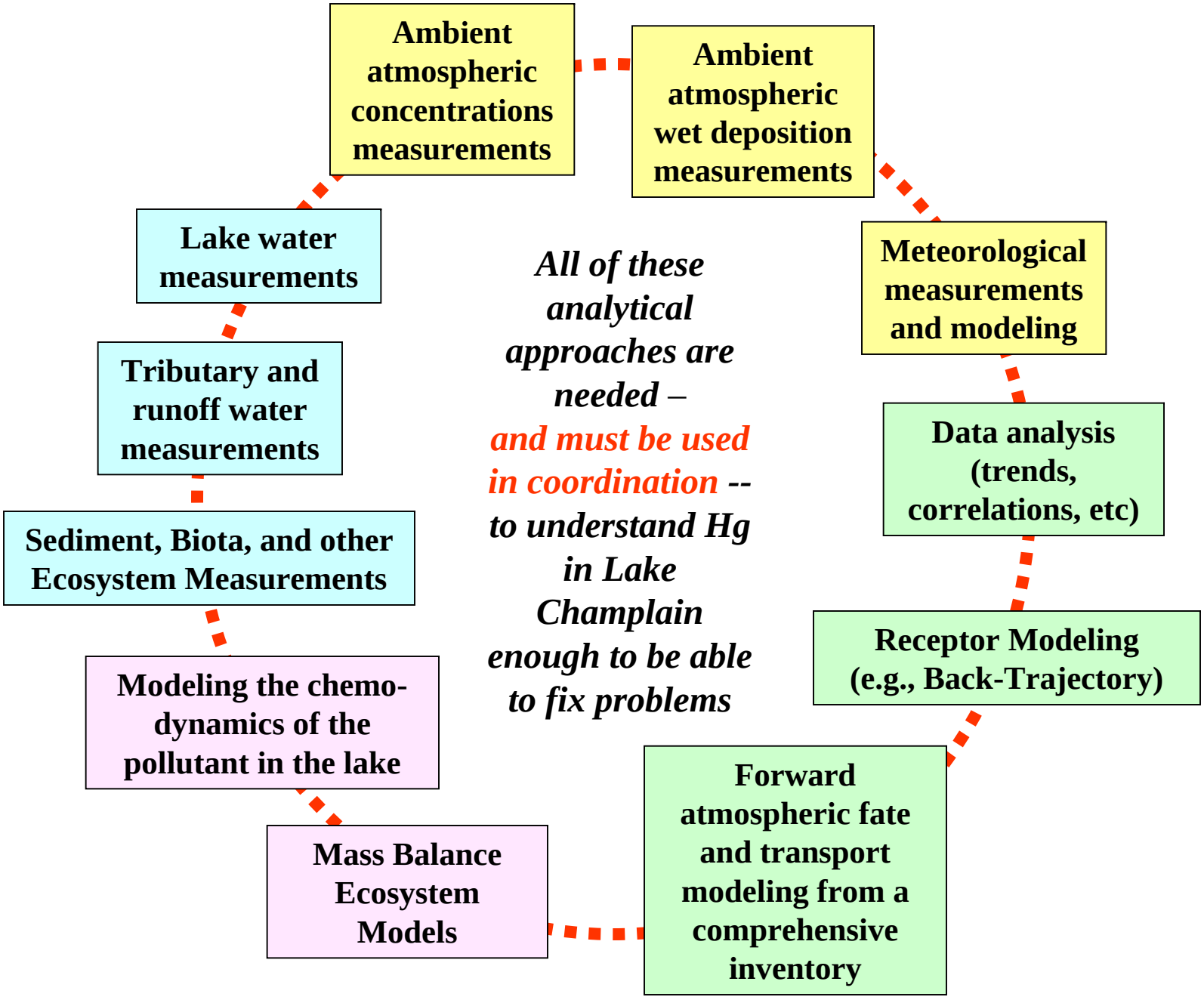
2. How important is direct deposition to a given lake relative to indirect loading via deposition to the lake's watershed?

3. How important is atmospheric deposition relative to other loading pathways (e.g., direct discharge to the Lake or its tributaries)

4. What is the relative importance of the contributions from local, regional, national, continental, and global sources?

5. What is the relative importance of contributions from different types of sources, e.g, coal fired utilities, incinerators, natural emissions, etc.?

We need to know all these things to efficiently direct action to reduce the contamination levels in a given lake.


Ambient atmospheric concentrations measurements
Ambient atmospheric wet deposition measurements
Lake water measurements
Meteorological measurements and modeling
Tributary and runoff water measurements
Data analysis (trends, correlations, etc)
Sediment, Biota, and other Ecosystem Measurements
Receptor Modeling (e.g., Back-Trajectory)
Modeling the chemo-dynamics of the pollutant in the lake
Forward atmospheric fate and transport modeling from a comprehensive inventory
Mass Balance Ecosystem Models
All of these analytical approaches are needed – and must be used in coordination -- to understand Hg in Lake Champlain enough to be able to fix problems

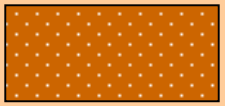
Three “forms” of atmospheric mercury

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”)

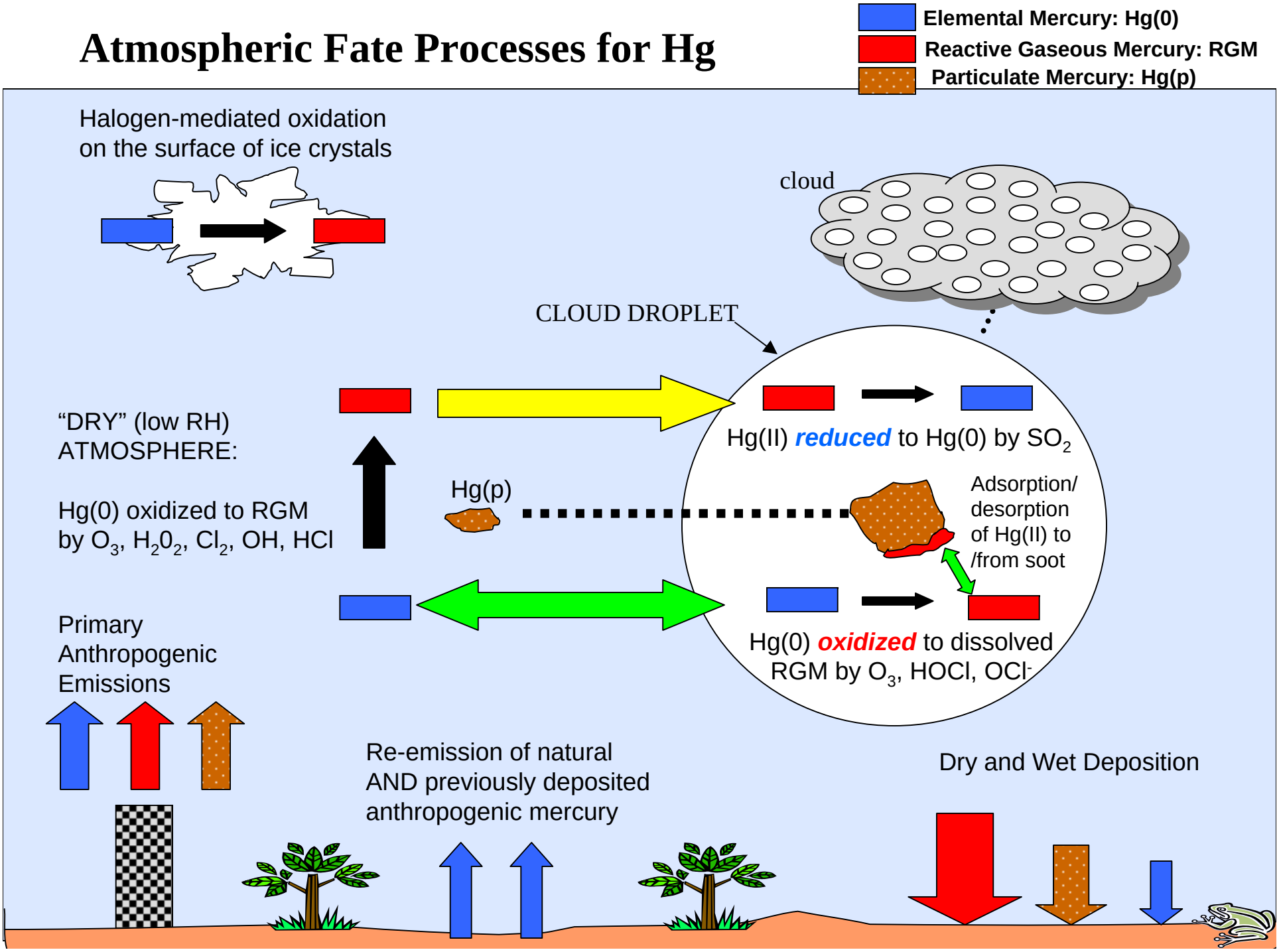
- **a few percent of total Hg in atmosphere**
- **oxidized mercury: Hg(II)**
- **HgCl2, others species?**
- **somewhat operationally defined by measurement method**
- ***very* water soluble**
- **short atmospheric lifetime (~ 1 week or less);**
- **more local and regional effects**



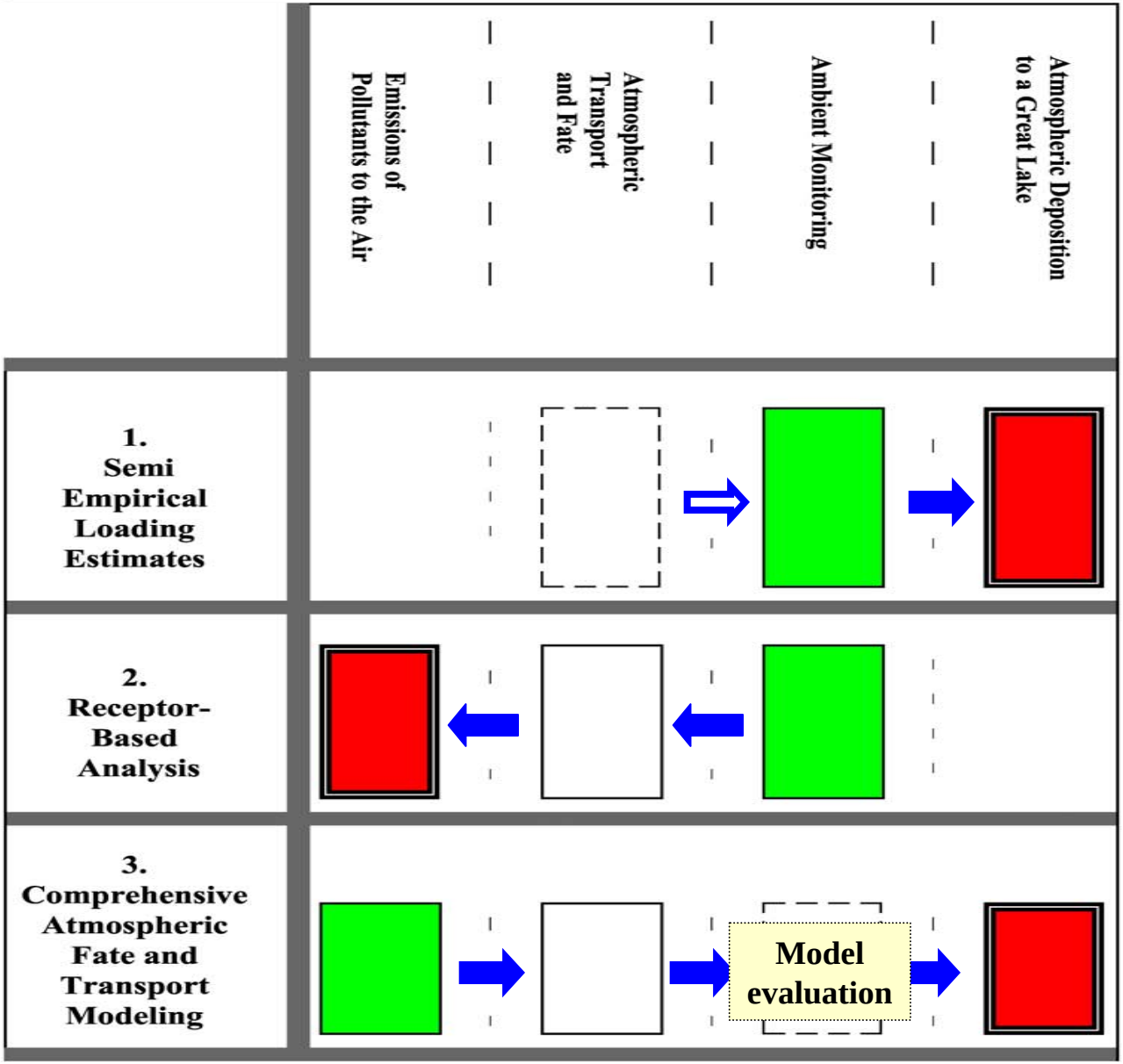
Particulate Mercury (Hg(p)

- **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 Fate Processes for Hg



Methodological Approaches for Analysis of the Atmospheric Deposition Pathway



Can't reliably estimate the *amount* of deposition or *source-receptor relationships* using monitoring alone…

Modeling can potentially give you these answers, but cannot be done credibly without using monitoring to ground-truth the results

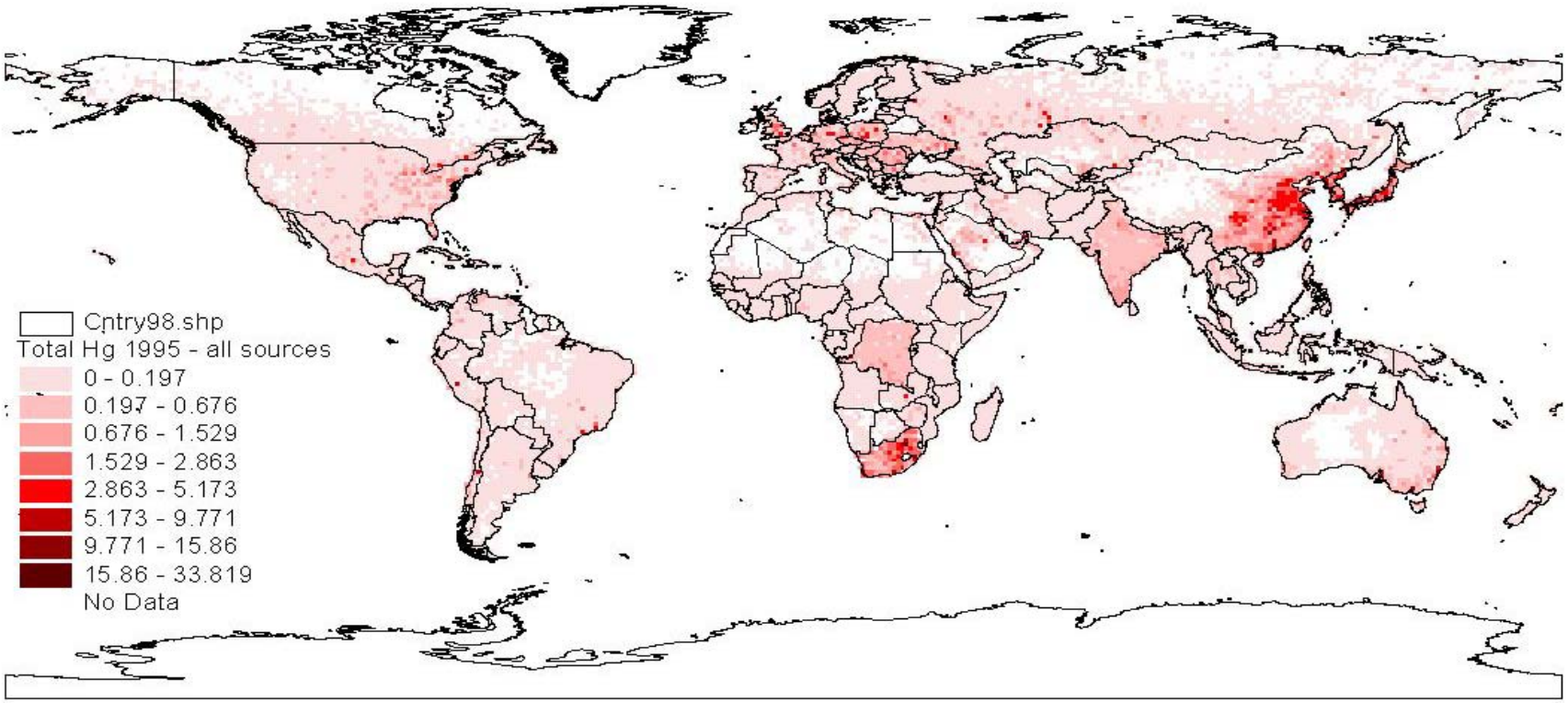
- **We are generally *not* actually interested in the concentration or deposition at a single monitoring site…**

- **We are interested in the deposition to an *entire* water body, or to a particular ecosystem**

- ***We are just using the few monitoring sites that we might have to give us a clue as to what the total impact might be…***

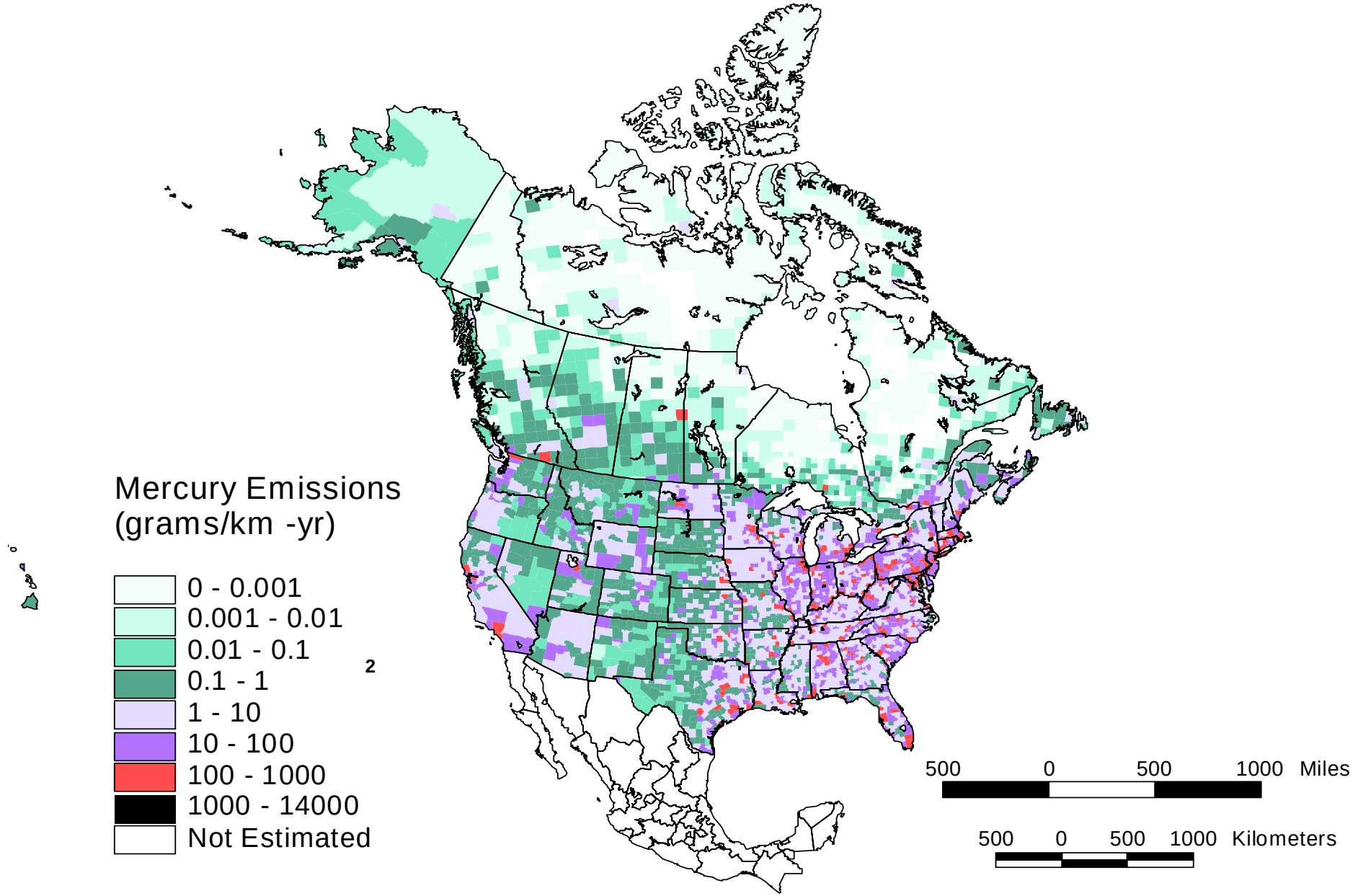
Overall Methodology

- **Start with atmospheric mercury *emissions inventory***
- **Perform *atmospheric fate and transport modeling* of these emissions (using a modified version of NOAA's HSYPLIT model)**
- **Keep track of *source-receptor information* during the modeling**
- **Evaluate the modeling by *comparison* of the predictions *against ambient monitoring data***
- ***If model is performing satisfactorily*, report source-receptor results from the simulations**
- **(Similar to earlier work with dioxin and atrazine)**

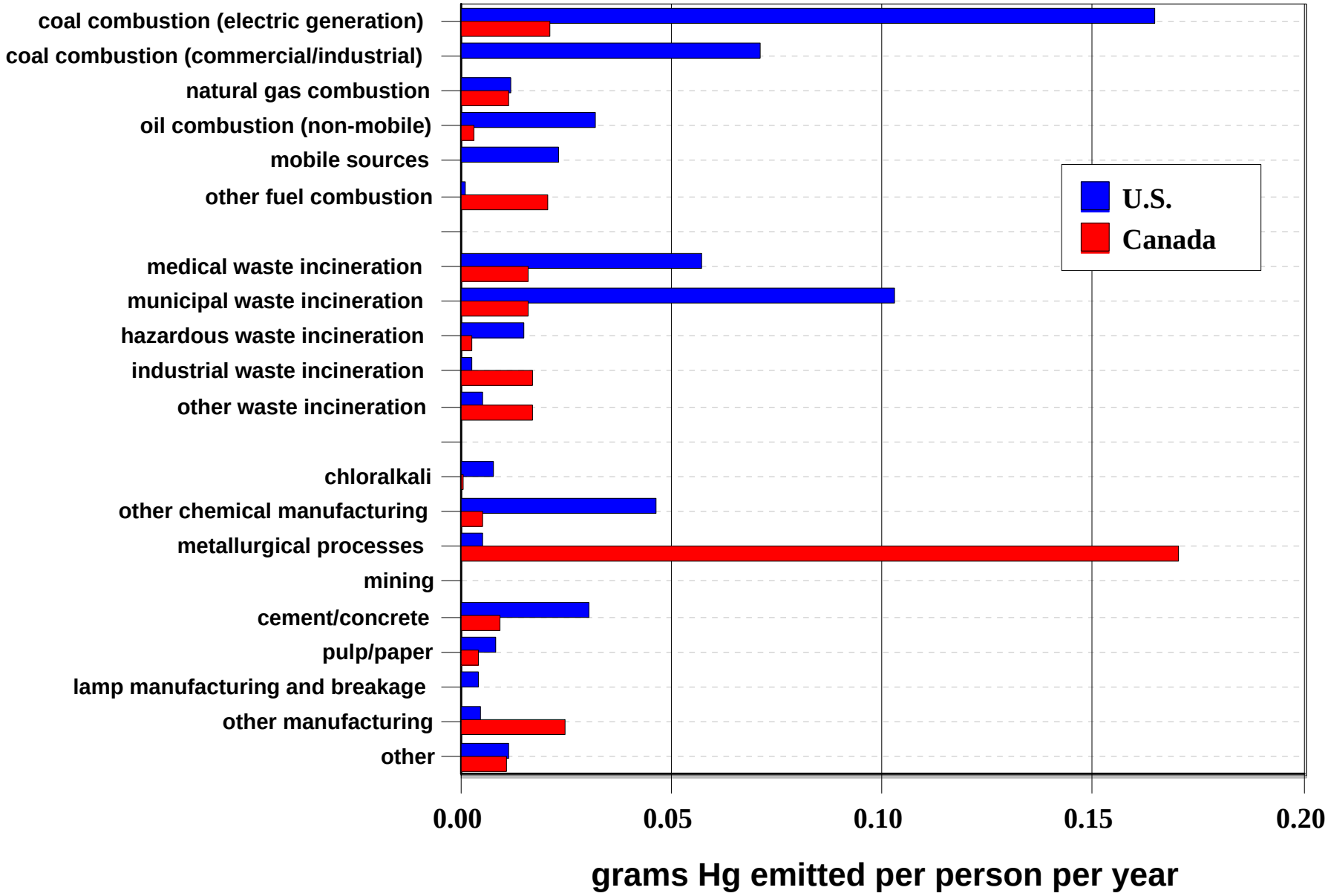


1995 Global Hg Emissions Inventory
Josef Pacyna,NILU, Norway (2001)

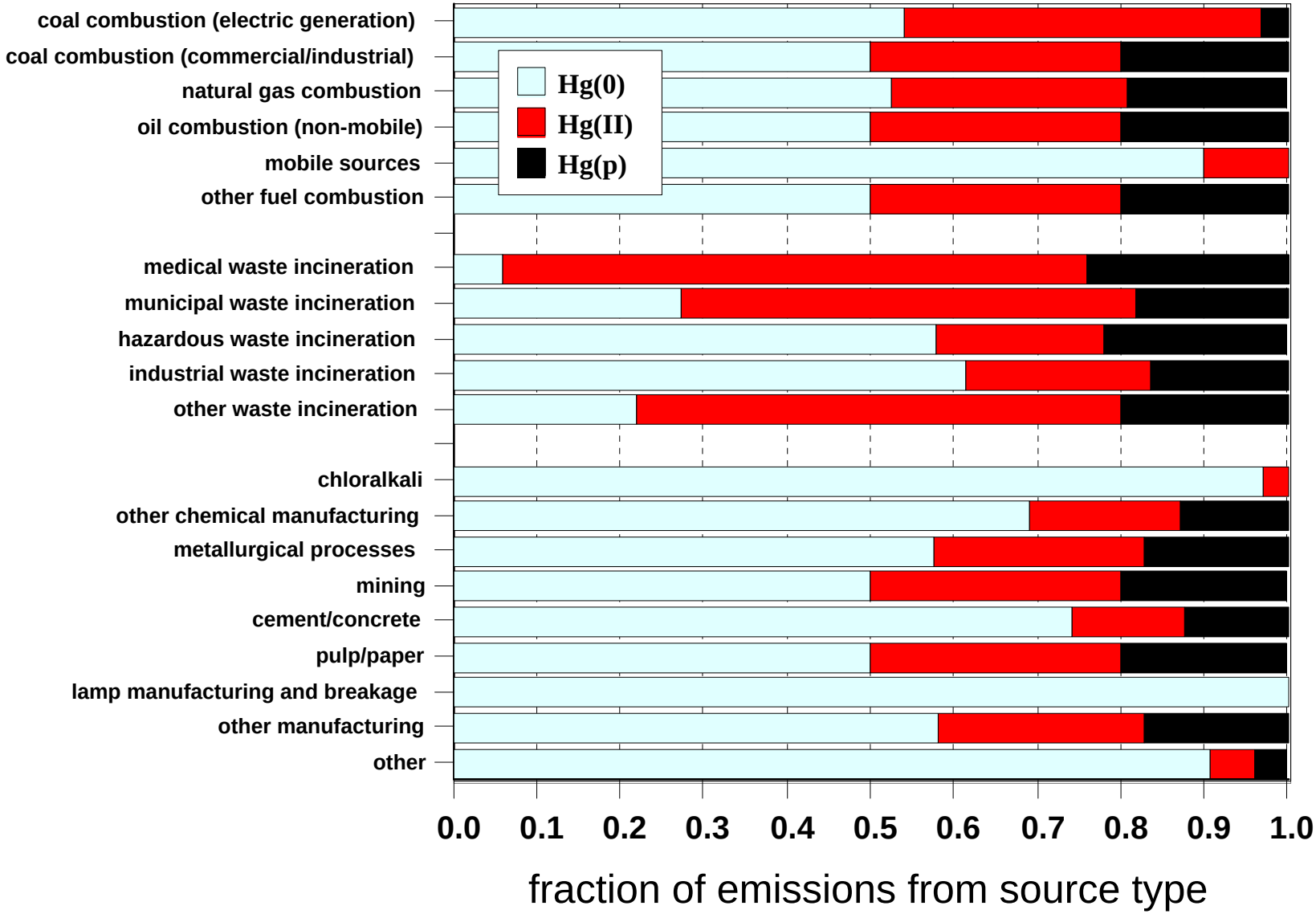
Geographic Distribution of Estimated Anthropogenic Mercury Emissions in the U.S. and Canada for 1995-1996



Annual *Per Capita* Mercury Emissions from U.S. and Canadian Anthropogenic Sources, 1995-1996



Speciation Profile of Mercury Emissions from U.S. and Canadian Anthropogenic Sources, 1995-1996



Speciated Annual Mercury Emissions from U.S. and Canadian Anthropogenic Sources During 1995-1996

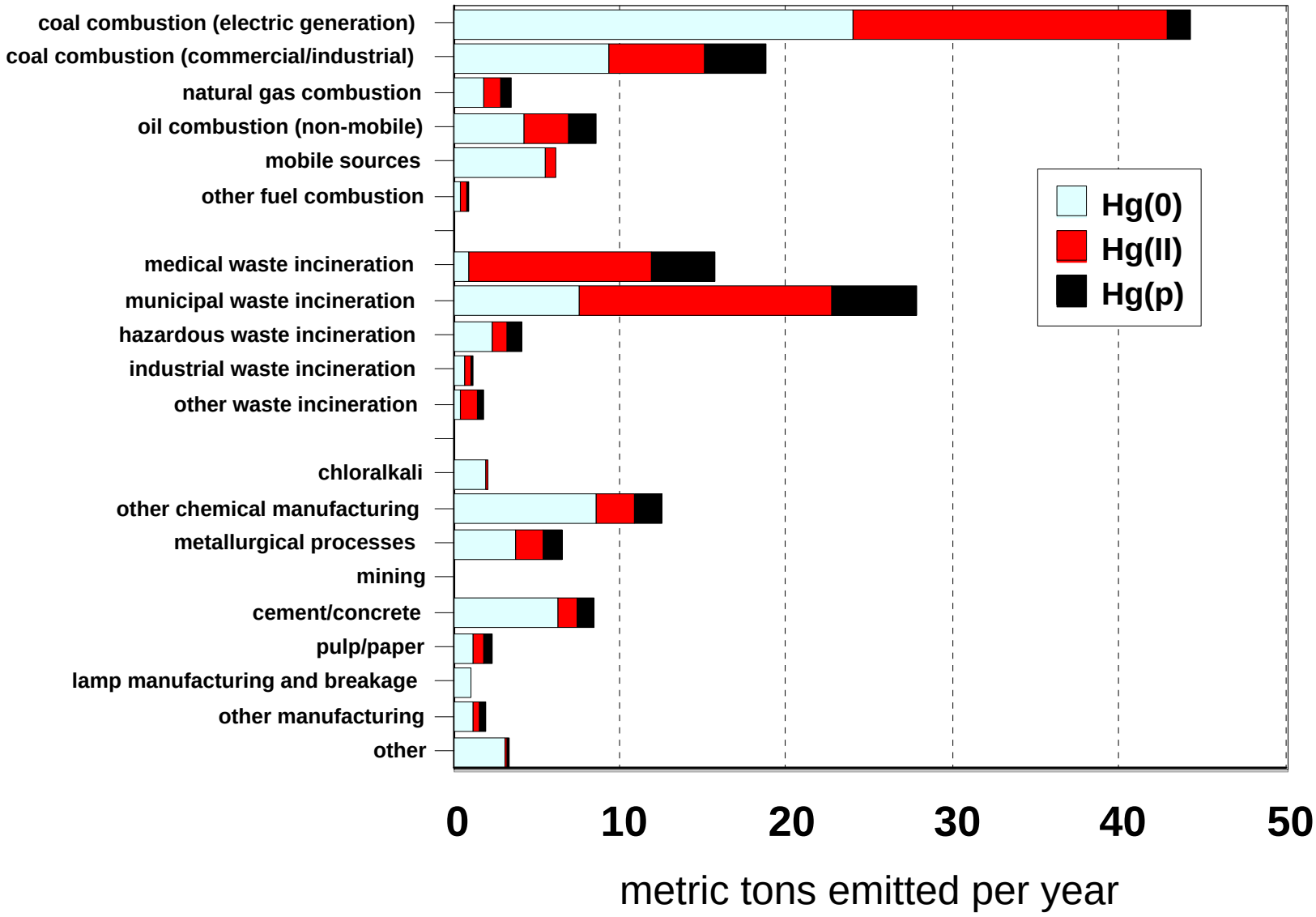
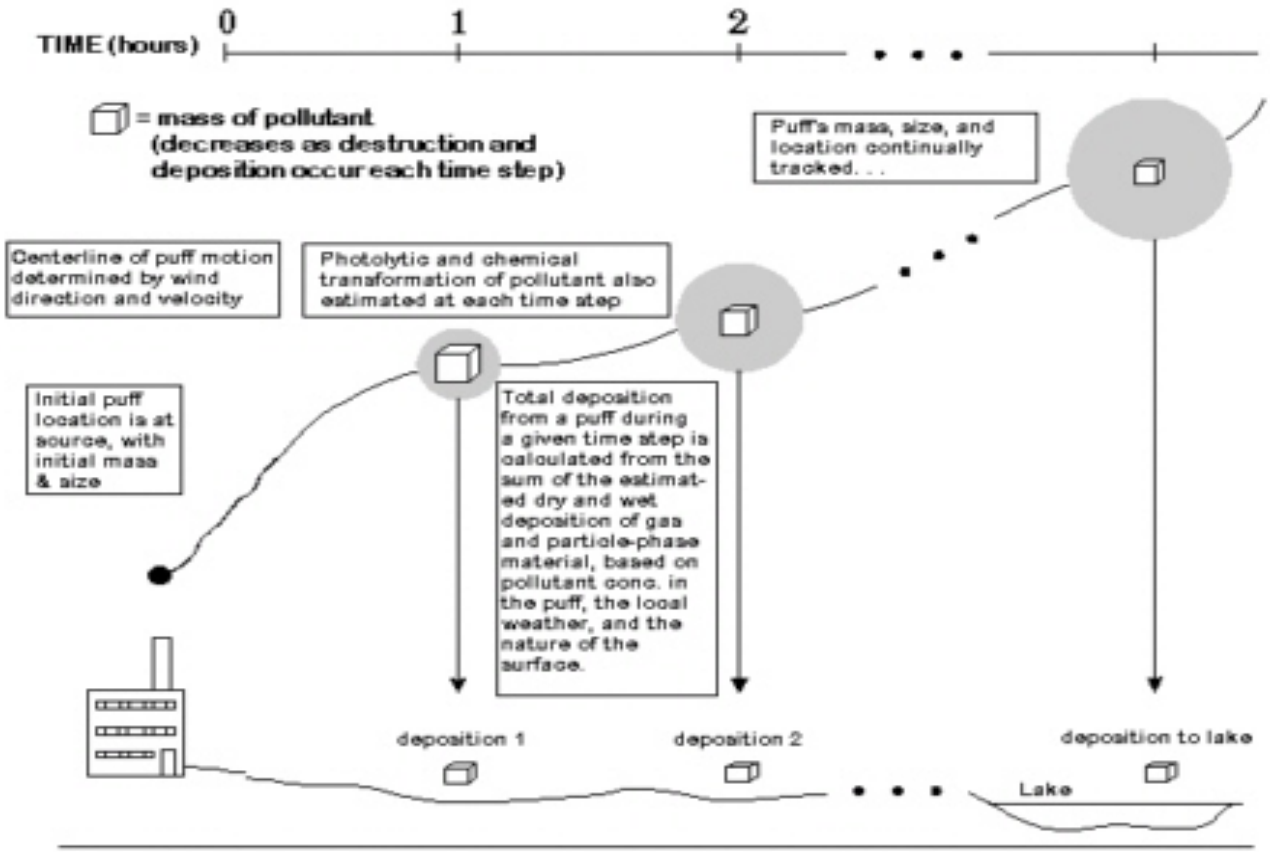
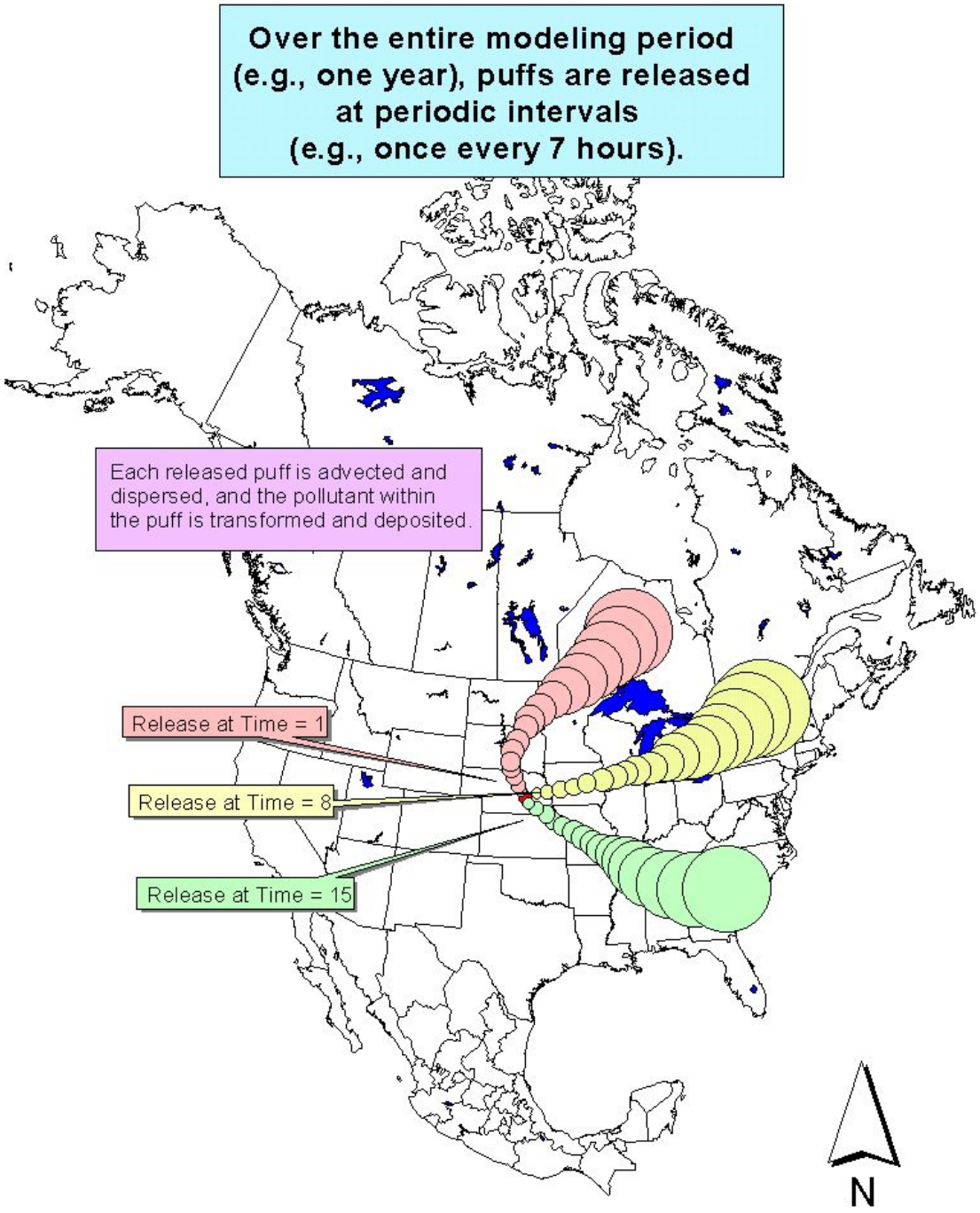
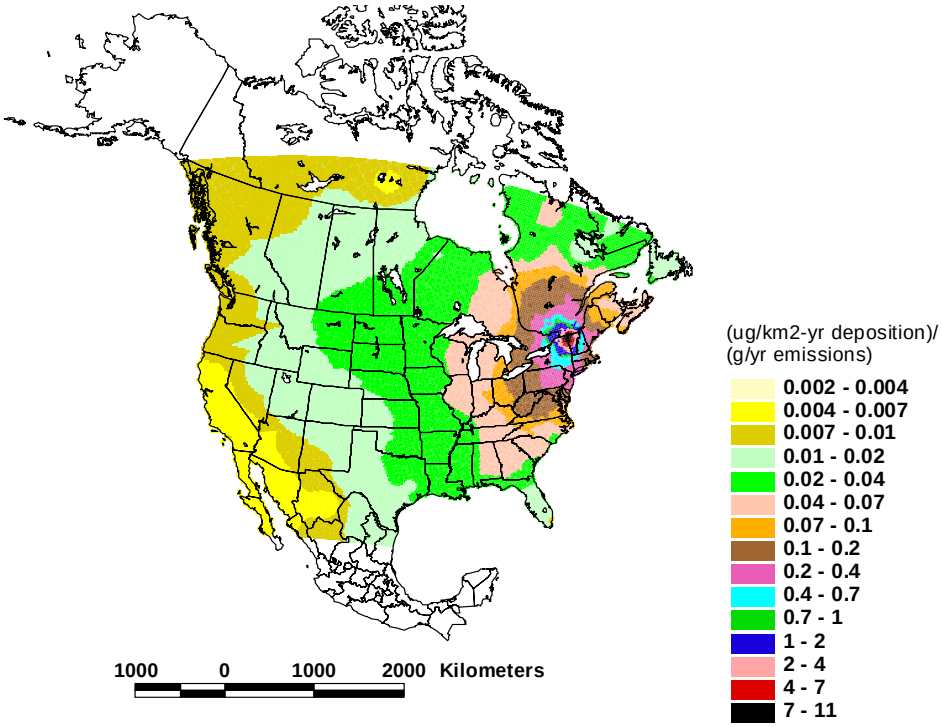


Figure 1. Lagrangian Puff Air Transport and Deposition Model




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

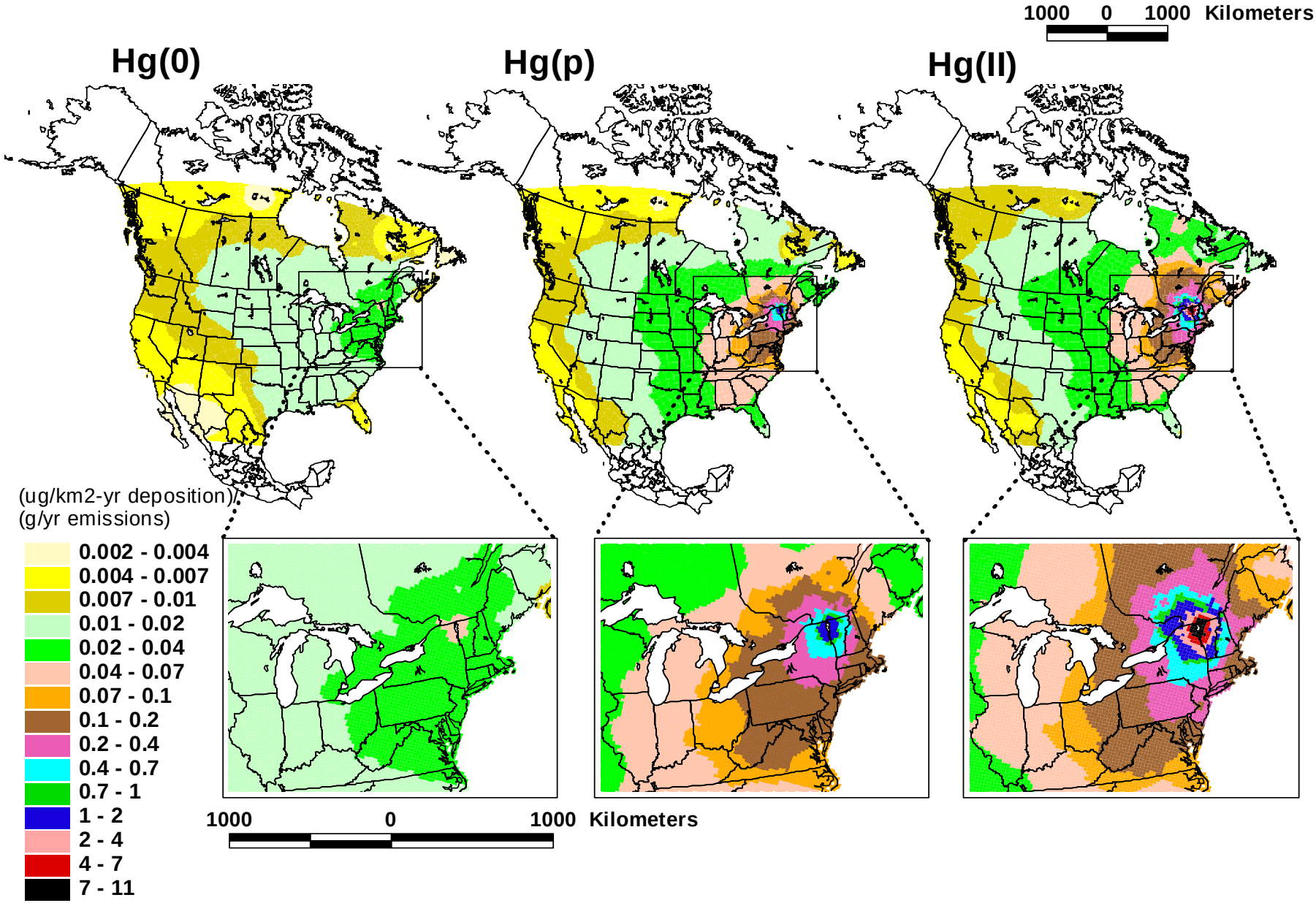
at any given location,
the ***transfer coefficient***
is defined as the amount
that would be deposited
in the given receptor
(in this case, Lake Champlain)
if there were emissions
at that location.



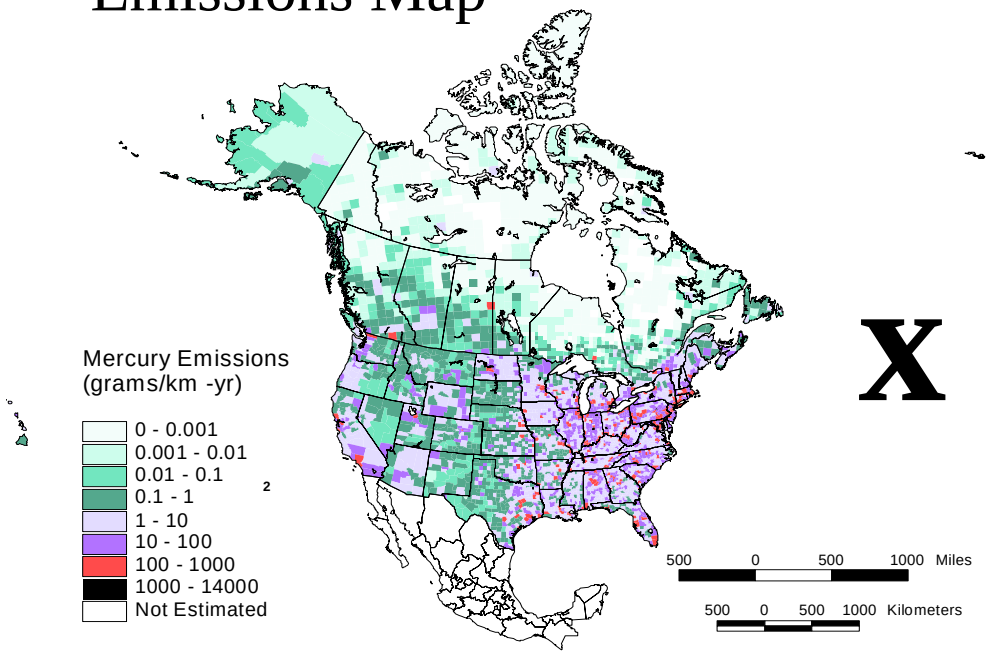
Transfer Coefficients

- **refer to hypothetical emissions;**
  are independent of actual emissions
- **can be formulated with different units**
  [total Hg deposition flux (ug/km2-yr) / emissions (g/yr)]
- **will depend on the pollutant *[Hg(II)]***
- **will depend on the receptor**
  [Lake Champlain]
- **and the time period being modeled**
  [entire year 1996]

Annual 1996 Transfer Coefficients for Different Forms of Mercury to Lake Champlain

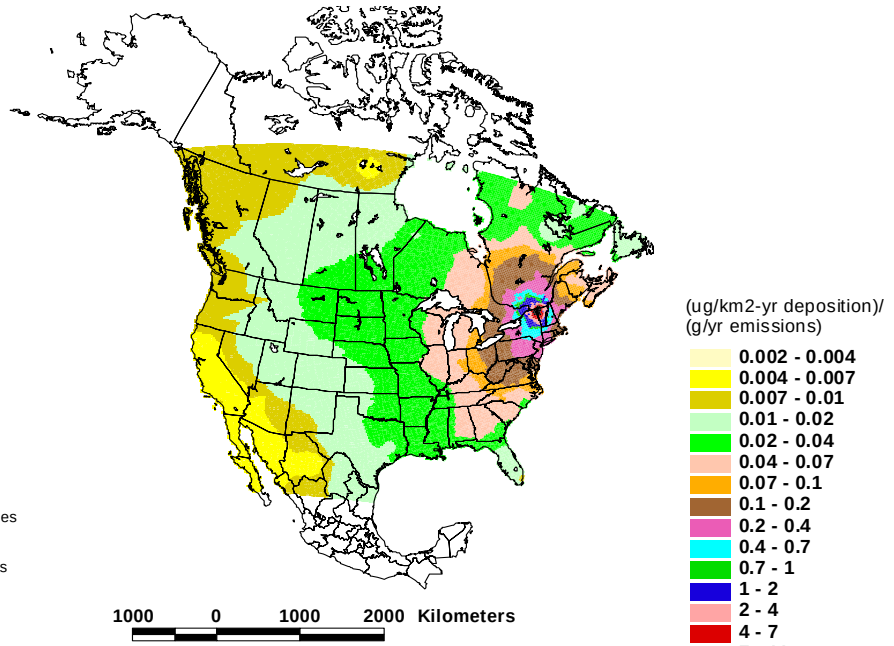


Emissions Map



X

Transfer Coefficient Map



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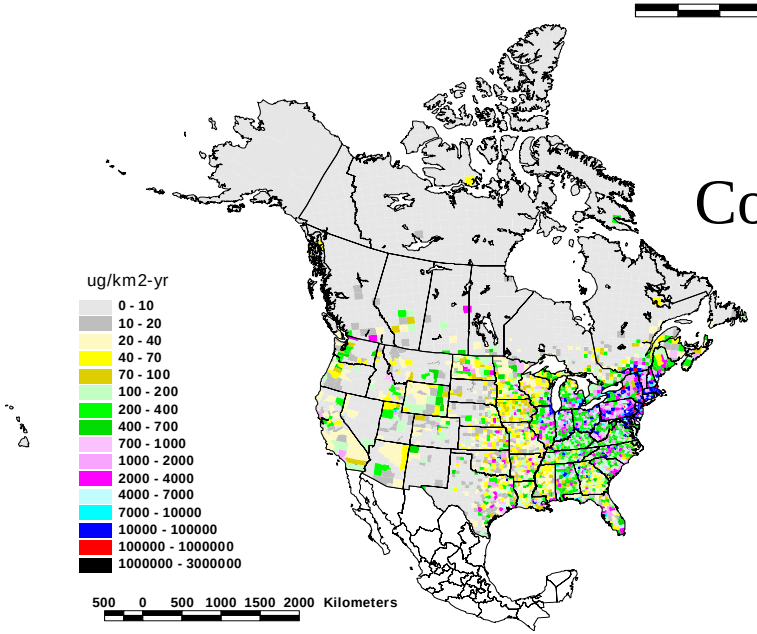
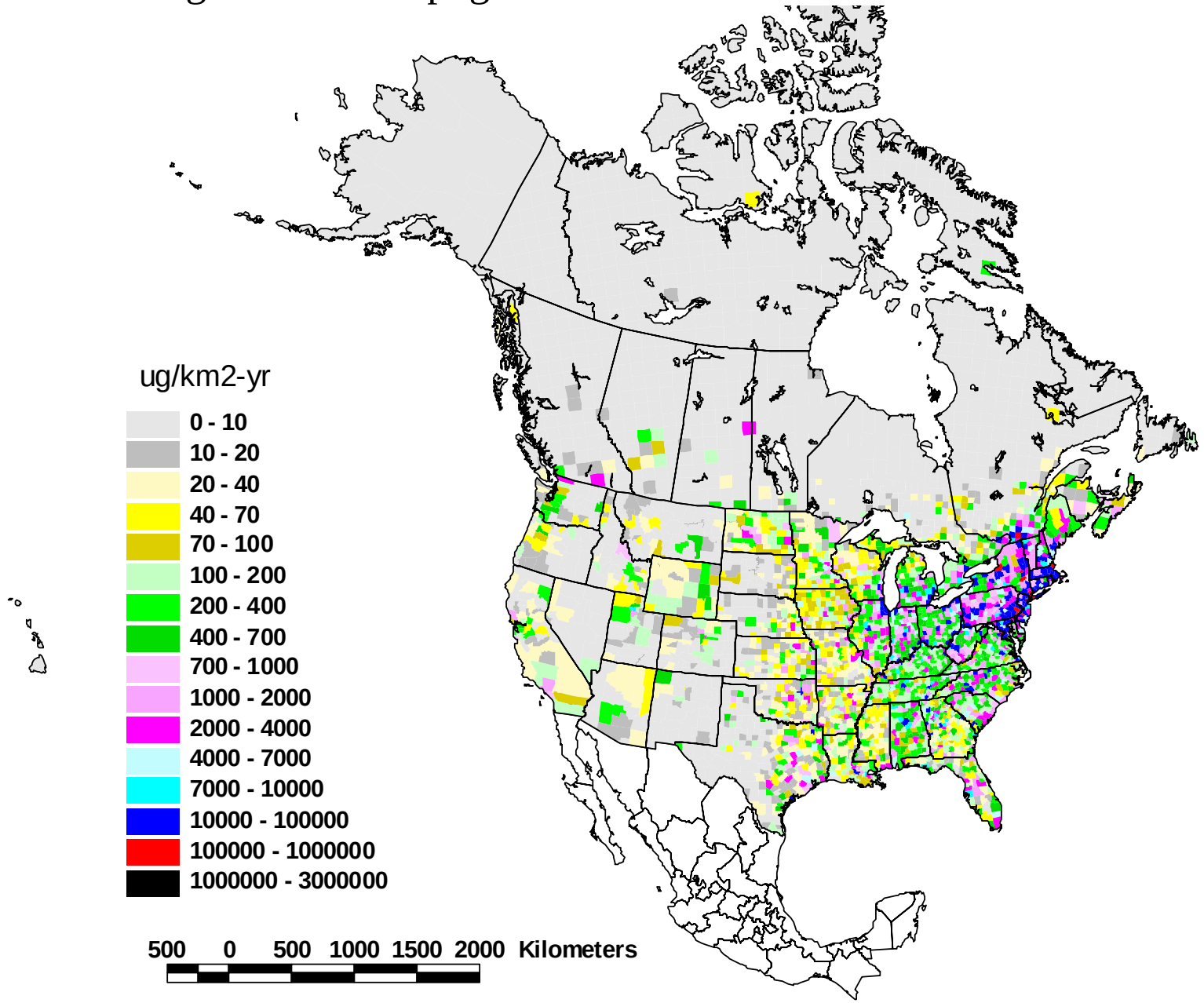
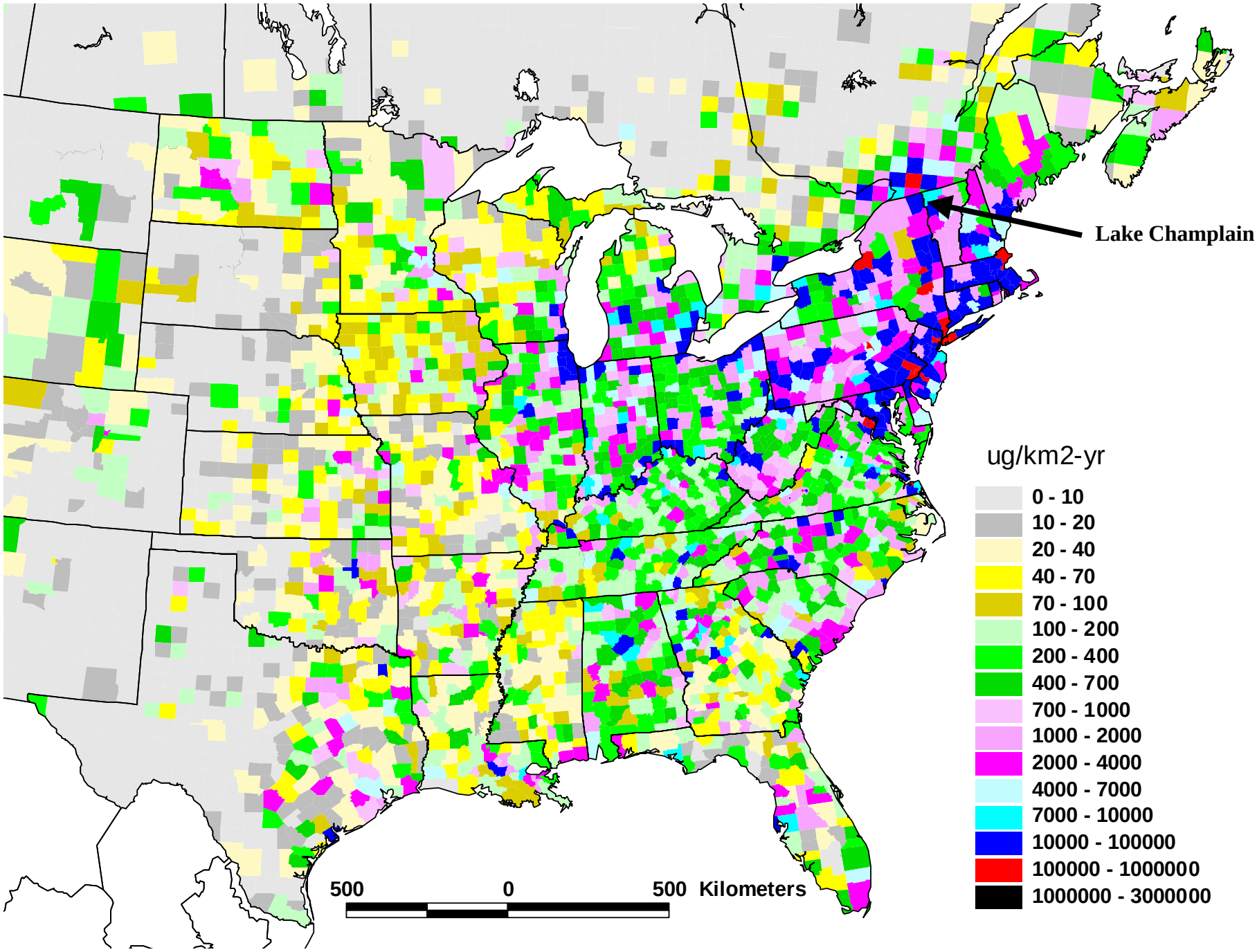
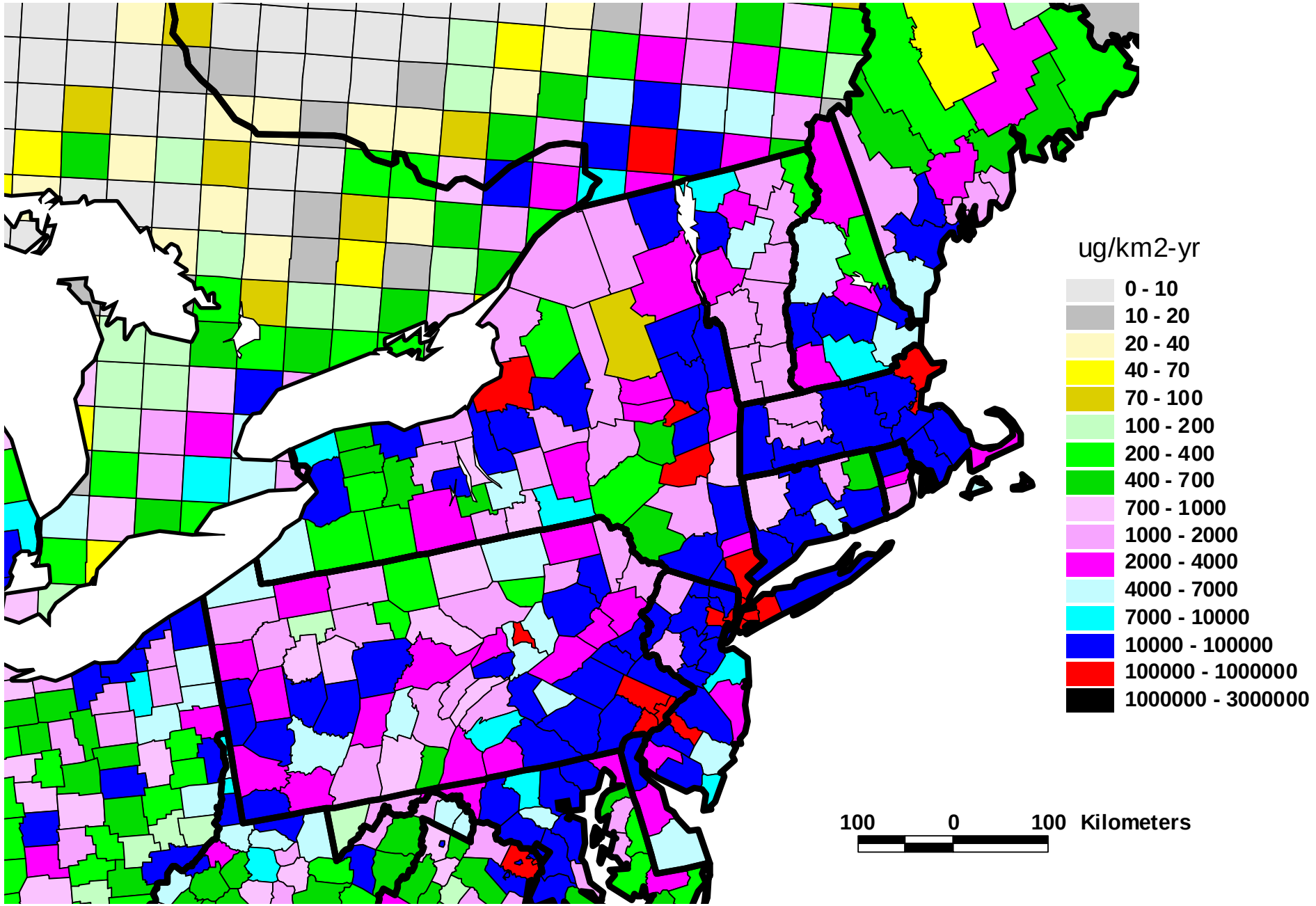
Contribution Map



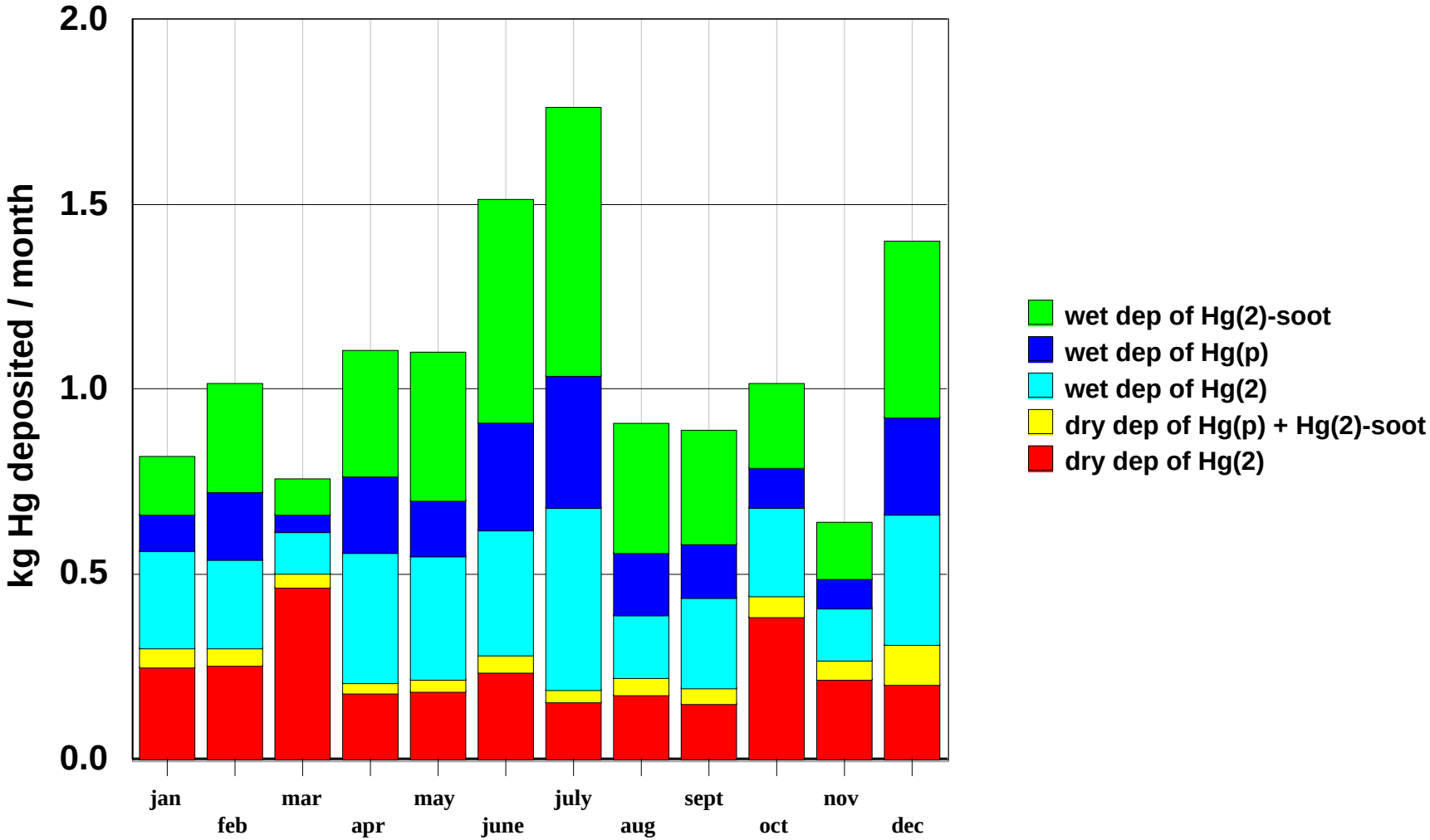
Geographical Distribution of Atmospheric Deposition Contributions to Lake Champlain Arising from Anthropogenic Sources in the U.S. and Canada During 1996



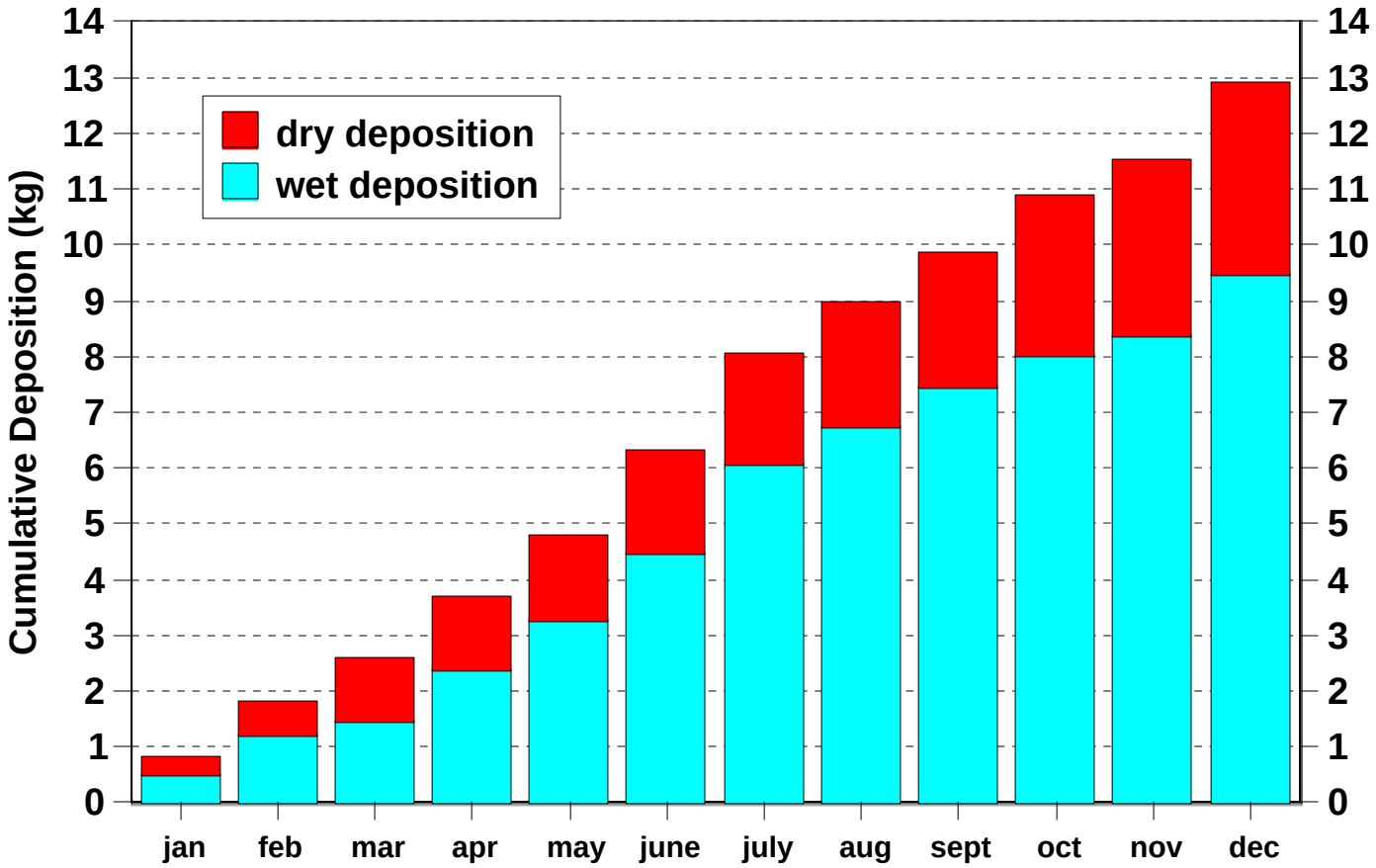

Lake Champlain
ug/km2-yr
0 - 10
10 - 20
20 - 40
40 - 70
70 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 2000
2000 - 4000
4000 - 7000
7000 - 10000
10000 - 100000
100000 - 1000000
1000000 - 3000000
500
0
500 Kilometers


ug/km2-yr
0 - 10
10 - 20
20 - 40
40 - 70
70 - 100
100 - 200
200 - 400
400 - 700
700 - 1000
1000 - 2000
2000 - 4000
4000 - 7000
7000 - 10000
10000 - 100000
100000 - 1000000
1000000 - 3000000
100 0 100 Kilometers

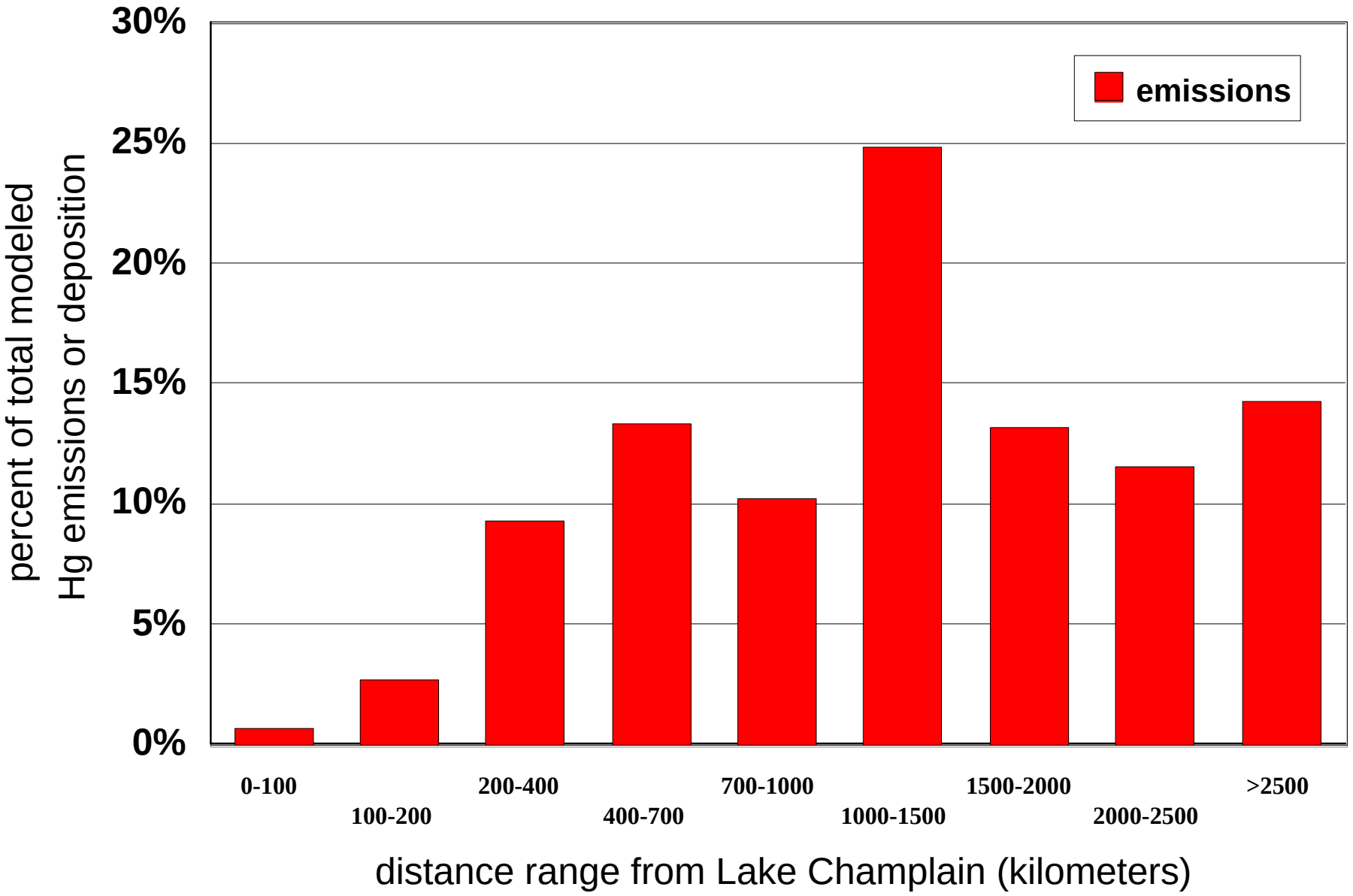
Monthly Model-Estimated Wet and Dry Deposition of Different Forms of Mercury to Lake Champlain During 1996 Arising from U.S. and Canadian Anthropogenic Emissions



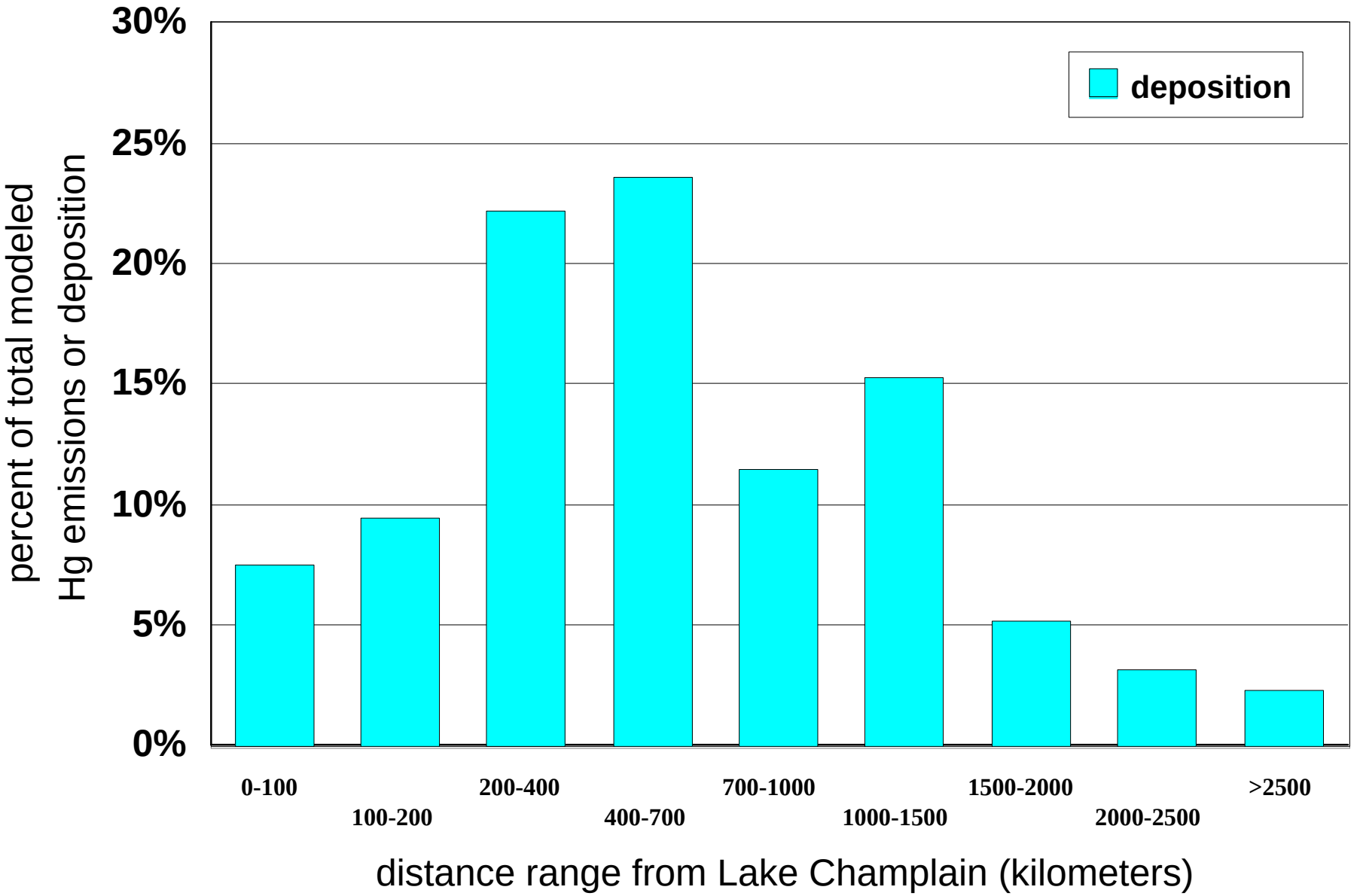
Cumulative Model-Estimated Wet and Dry Deposition of Mercury to Lake Champlain During 1996 Arising from U.S. and Canadian Anthropogenic Emissions



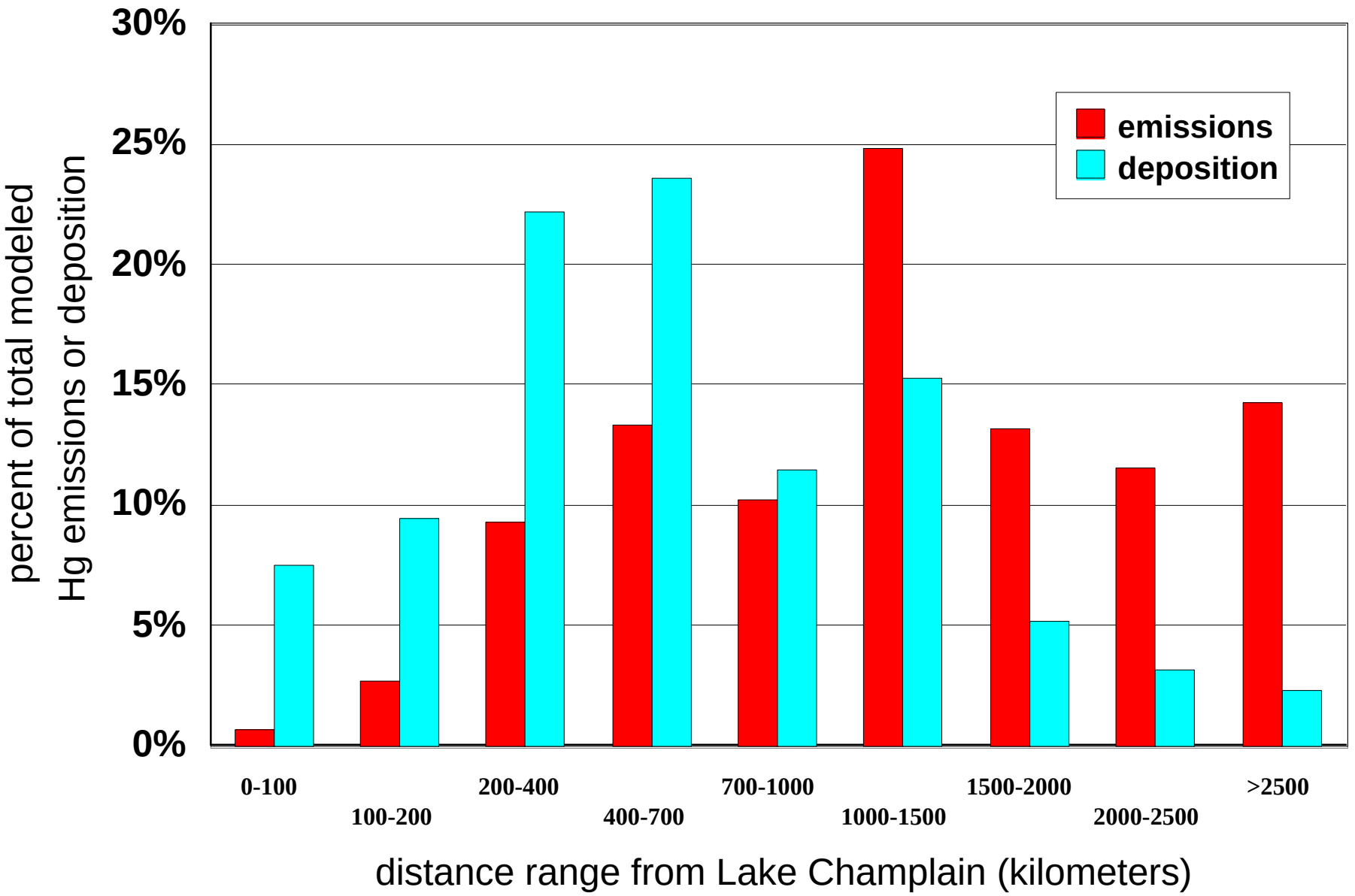
Percent of modeled **EMISSIONS** of mercury to Lake Champlain from different distance ranges away from the lake



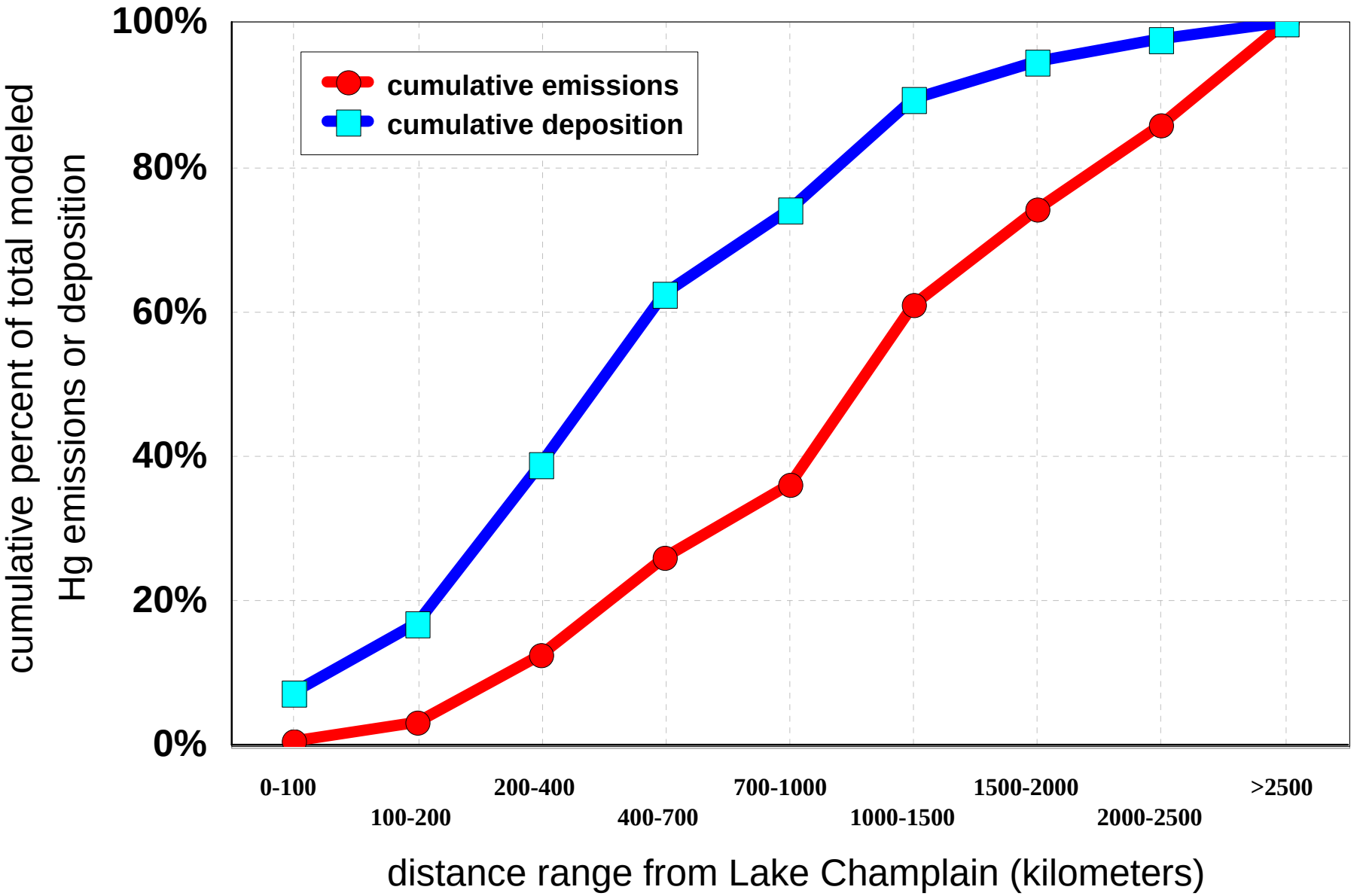
Percent of modeled **DEPOSITION** of mercury to Lake Champlain from different distance ranges away from the lake



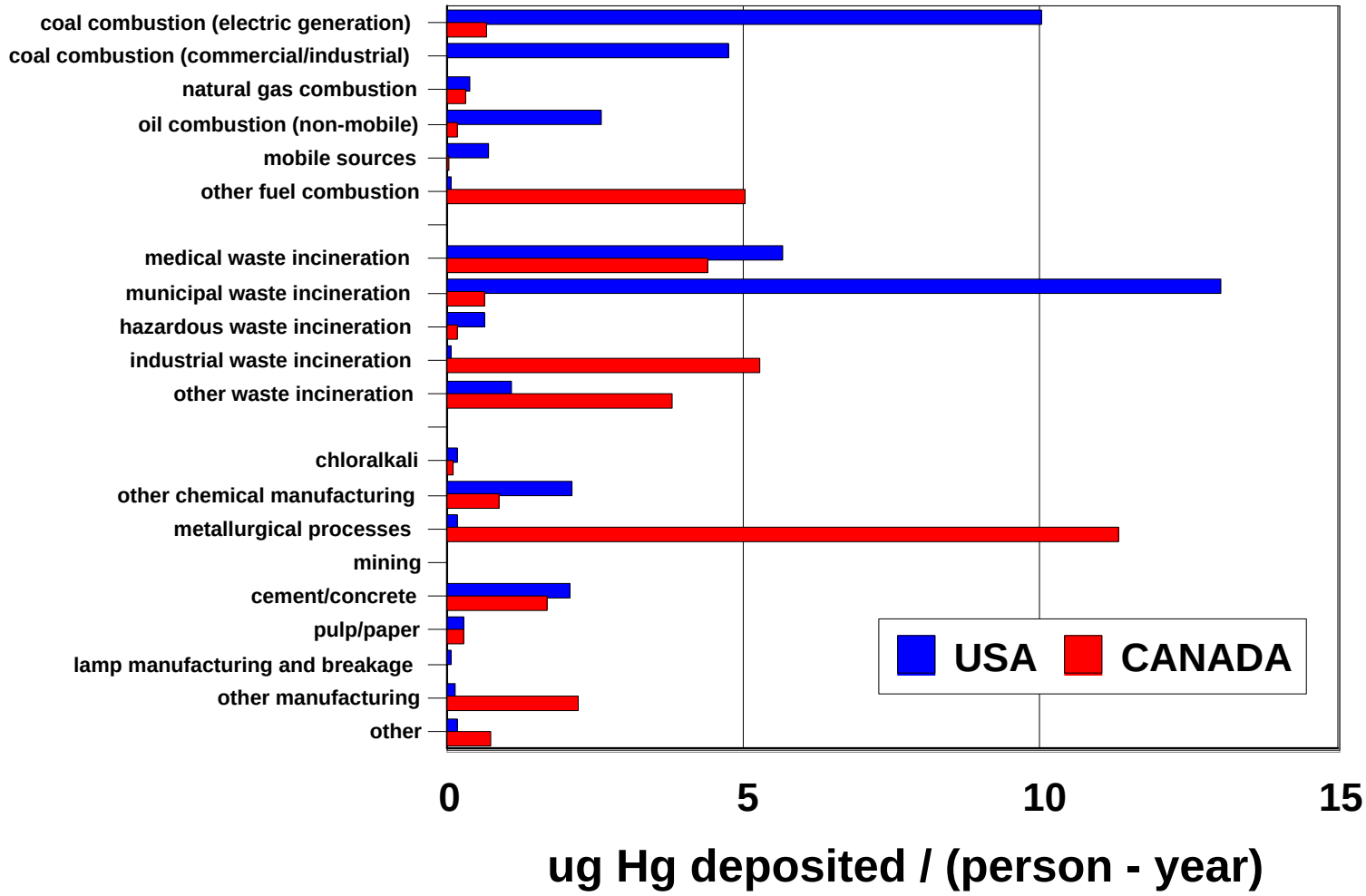
Percent of modeled **emissions** and **deposition** of mercury to Lake Champlain from different distance ranges away from the lake



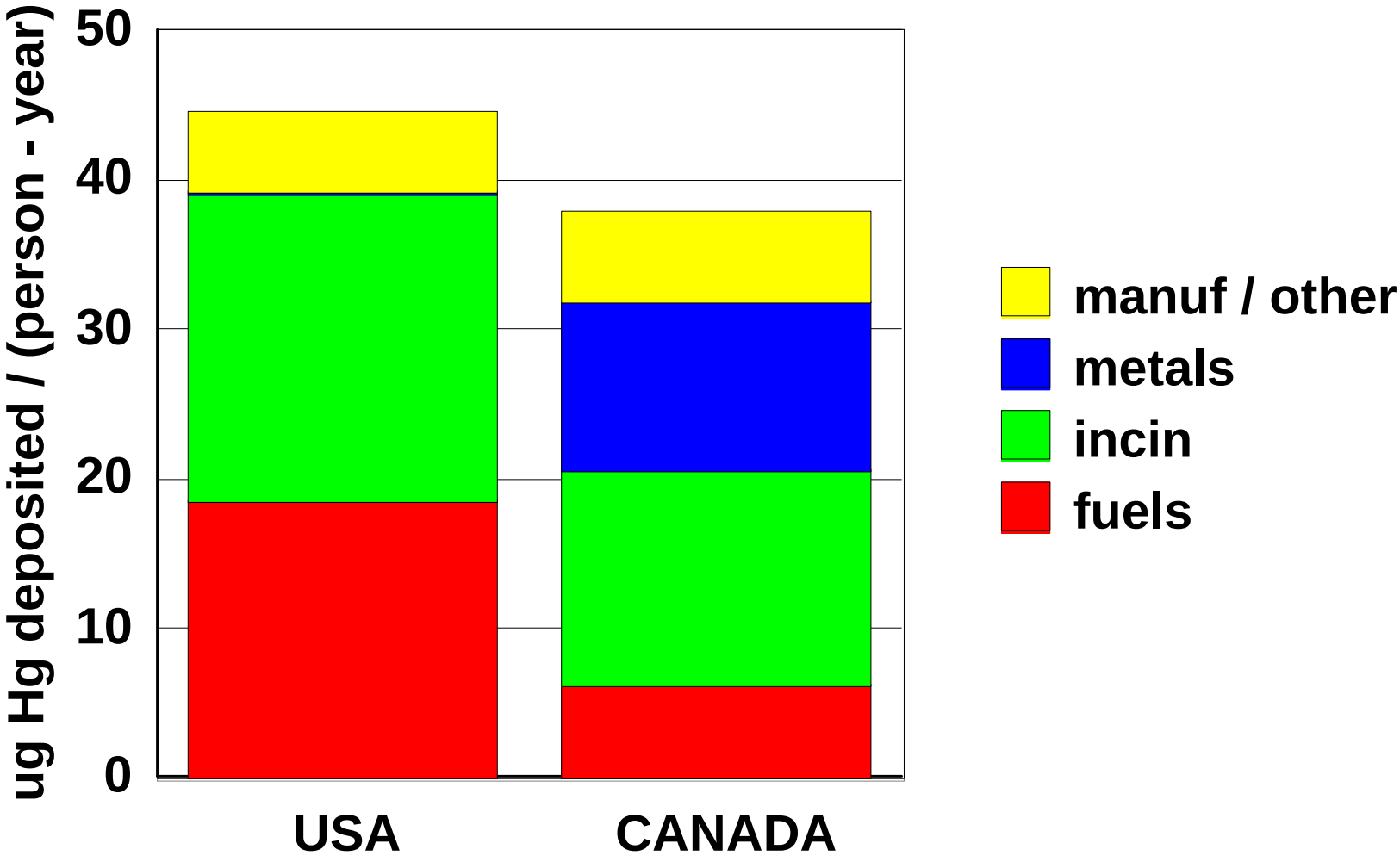
Cumulative percent of modeled **emissions** and **deposition** of mercury to Lake Champlain from different distance ranges away from the lake



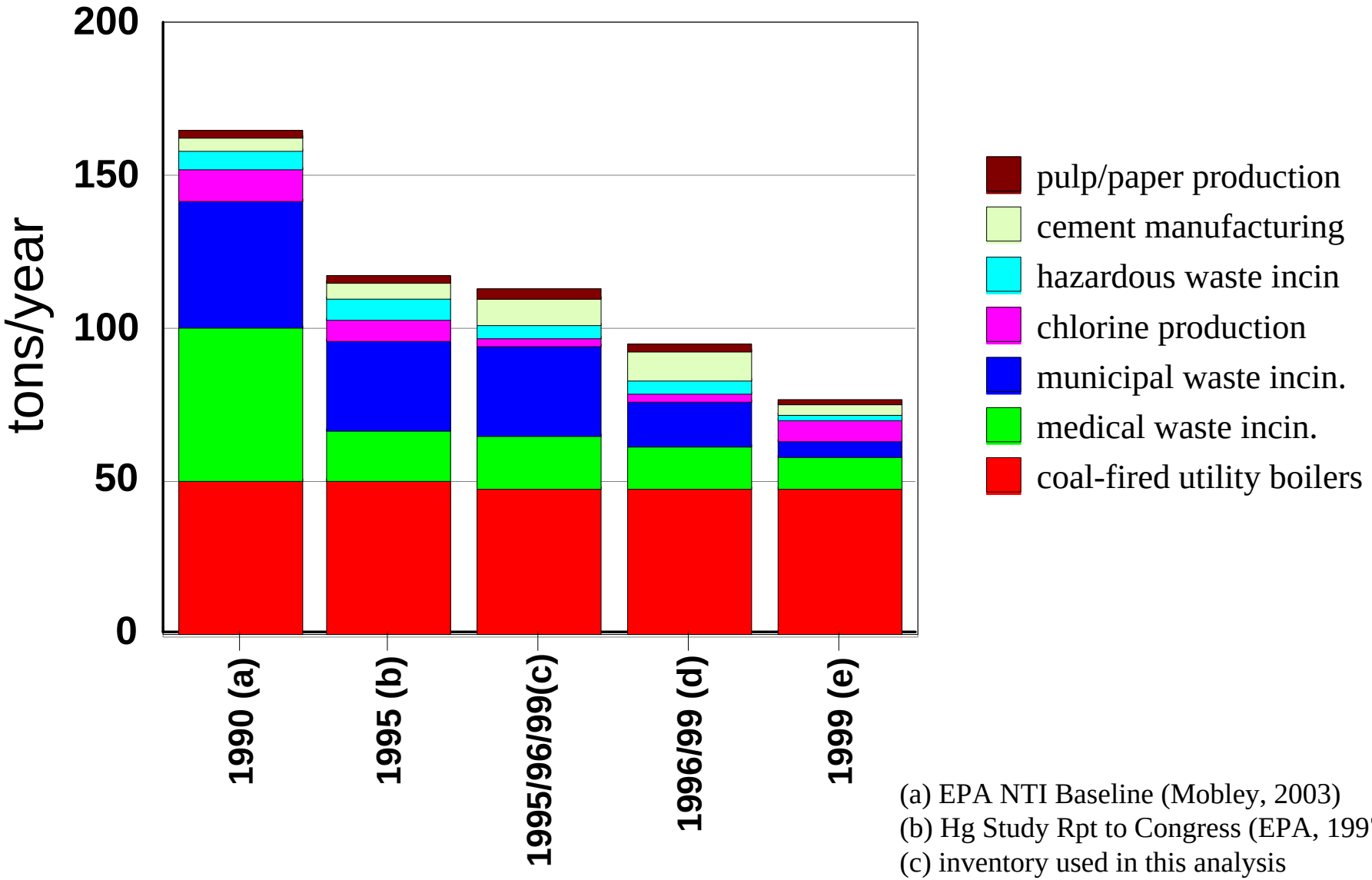
Per Capita Mercury Atmospheric Mercury Deposition Contributions to Lake Champlain from U.S. and Canadian *Detailed* Source Categories (1996)



Per Capita Mercury Atmospheric Mercury Deposition Contributions to Lake Champlain from U.S. and Canadian Aggregated Source Categories (1996)

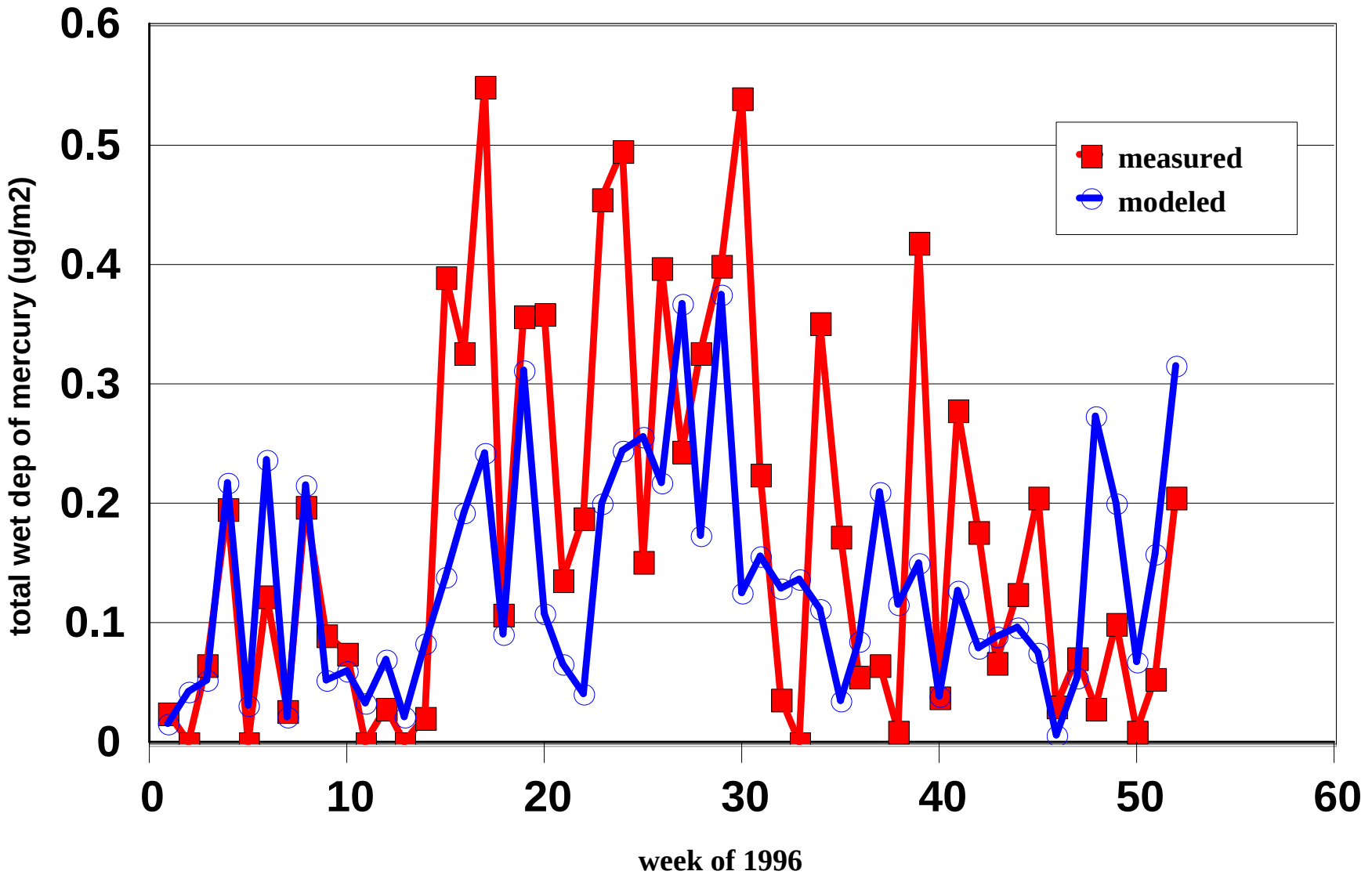


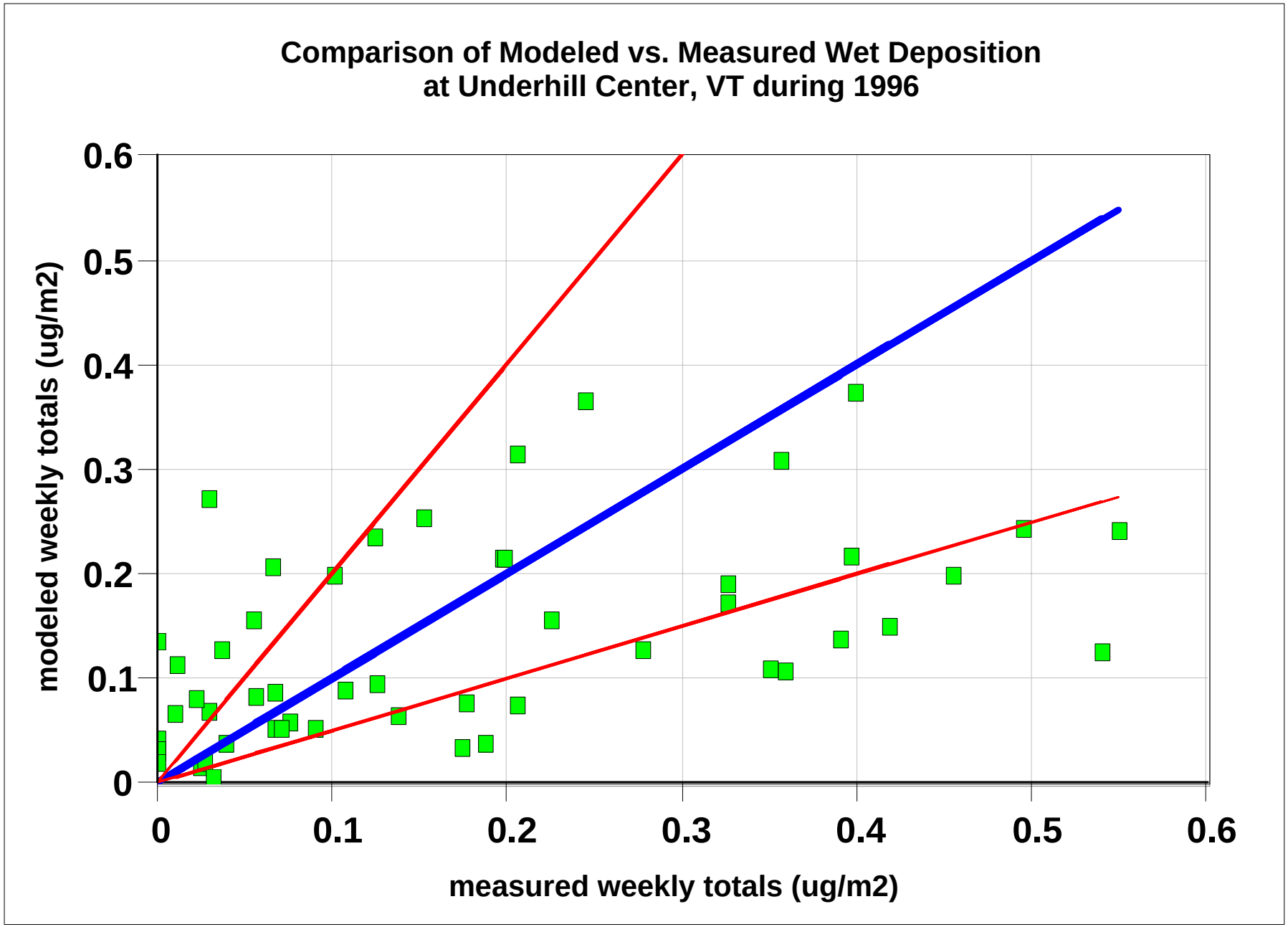
Reported trends in U.S. atmospheric mercury emissions 1990-1999 (selected source categories)

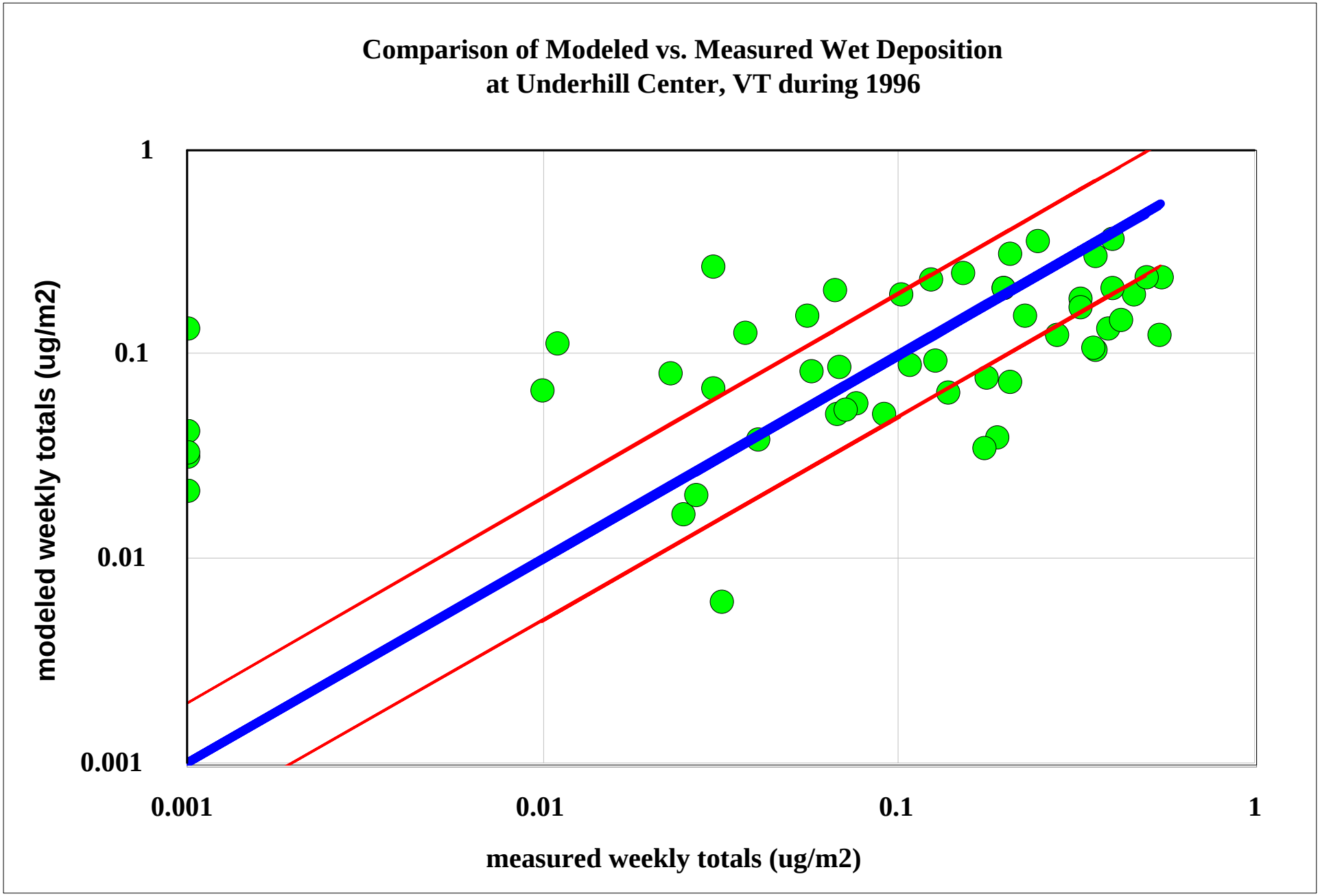


(a) EPA NTI Baseline (Mobley, 2003)
(b) Hg Study Rpt to Congress (EPA, 1997)
(c) inventory used in this analysis
(d) EPA 96/99 Inventory (Ryan, 2001)
(e) EPA NEI 1999 (draft) (Mobley, 2003)

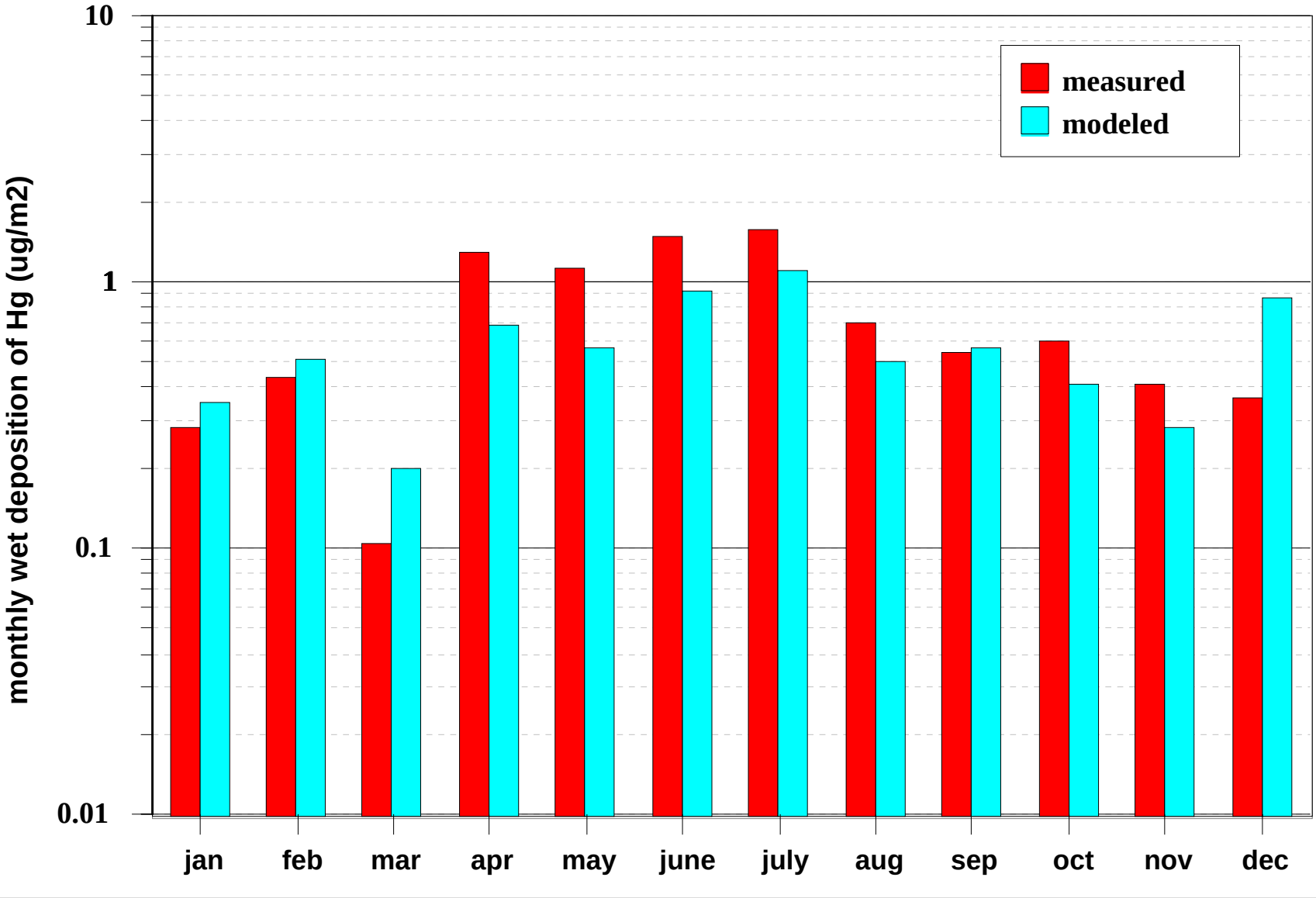
Comparison of Modeled vs. Measured Wet Deposition at Underhill Center, VT during 1996

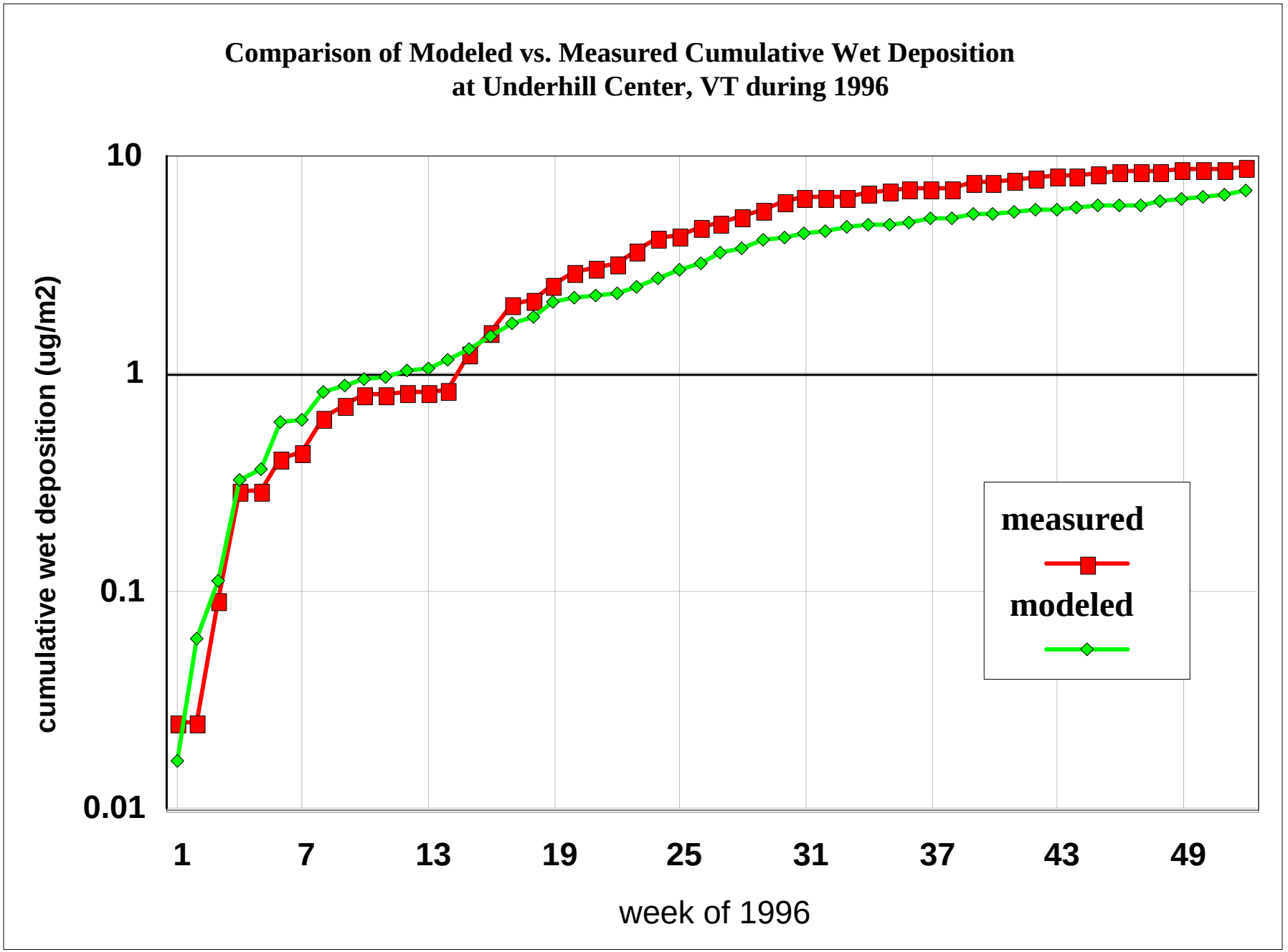



Comparison of Modeled vs. Measured Wet Deposition
at Underhill Center, VT during 1996
modeled weekly totals (ug/m2)
measured weekly totals (ug/m2)
0
0.1
0.2
0.3
0.4
0.5
0.6


Comparison of Modeled vs. Measured Wet Deposition
at Underhill Center, VT during 1996
modeled weekly totals (ug/m2)
1
0.1
0.01
0.001
0.001
0.01
0.1
1
measured weekly totals (ug/m2)

Comparison of Modeled vs. Measured Wet Deposition at Underhill Center, VT during 1996




Comparison of Modeled vs. Measured Cumulative Wet Deposition
at Underhill Center, VT during 1996
cumulative wet deposition (ug/m2)
10
1
0.1
0.01
1
7
13
19
25
31
37
43
49
week of 1996
measured
modeled

Some Limitations of this Modeling Analysis

Uncertainties in emissions (speciation, amount, temporal variations)

Uncertainties in atmospheric chemistry of mercury

Uncertainties in simulating wet and dry deposition phenomena

Only direct deposition to lake surface considered; deposition to watershed and subsequent entry into the lake not yet included in modeling

Coarse meteorological data grid (180 km)

Only U.S. and Canadian anthropogenic sources have been included; need to add global sources, natural sources, and anthropogenic mercury re-emitted after initially deposited

Assuming net deposition of Hg^0 is zero – essentially that natural emissions and re-emitted mercury sort of balance out Hg^0 deposition, so that net flux ~ 0

This is probably not true for Lake Champlain (or most lakes), as there is probably a net evasion of Hg^0, as a response to the deposition of Hg(II) and Hg(p). In this modeling (to date), we have only estimated this downward flux of Hg(II) and Hg(p).