

# **Modeling the Atmospheric Deposition of Mercury to Lake Champlain**

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



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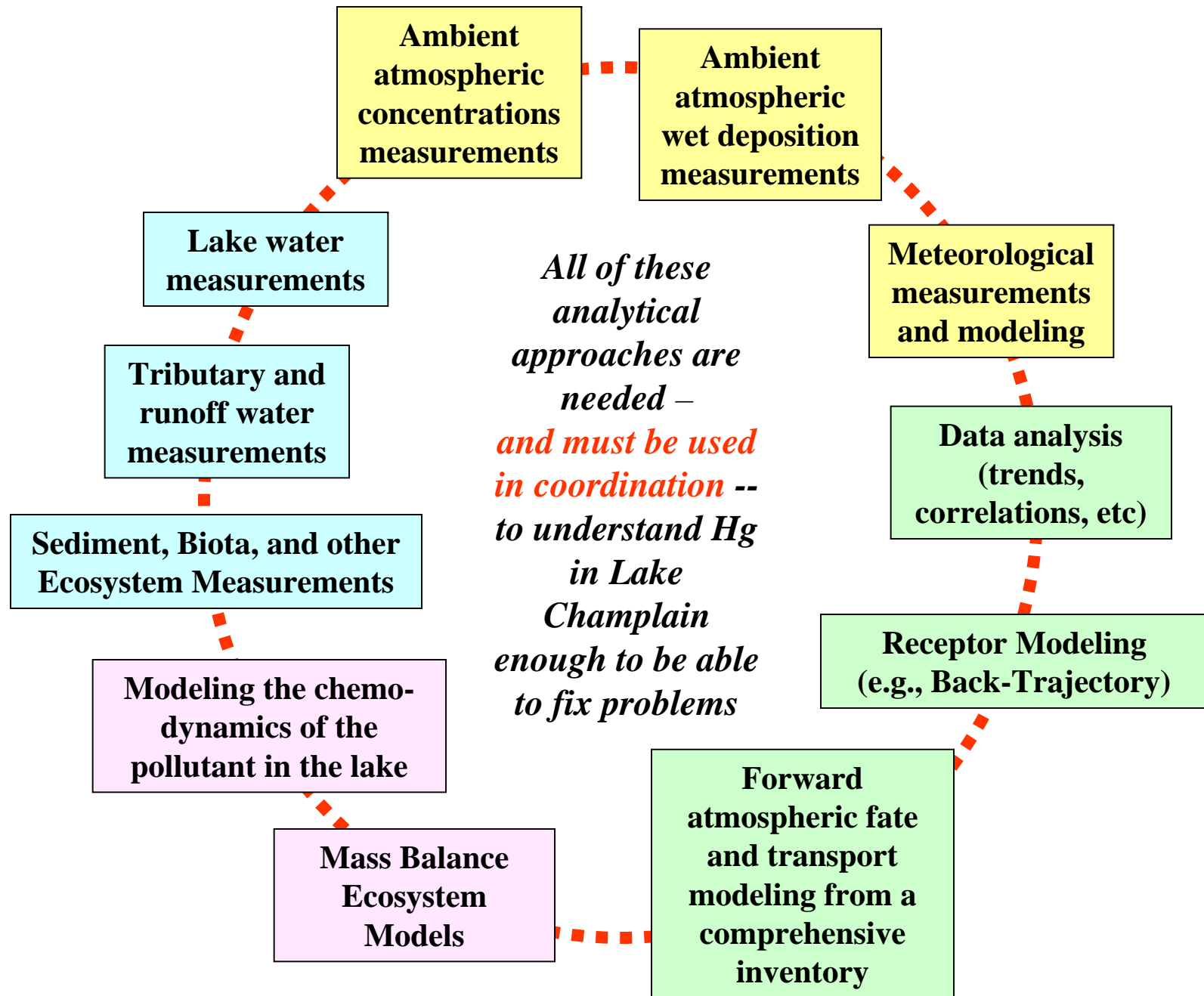


Presentation at the Workshop on  
**Coordination of Atmospheric Deposition Research  
in the Lake Champlain Basin**  
**June 5-6, 2003**  
**Bishop Booth Conference Center**  
**Burlington, Vermont**

***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.***



# Three “forms” of atmospheric mercury



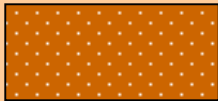
## Elemental Mercury: Hg(0)

- ~ 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)
- HgCl<sub>2</sub>, others species?
- somewhat operationally defined by measurement method
- *very* water soluble
- short atmospheric lifetime (~ 1 week or less);
- more local and regional effects

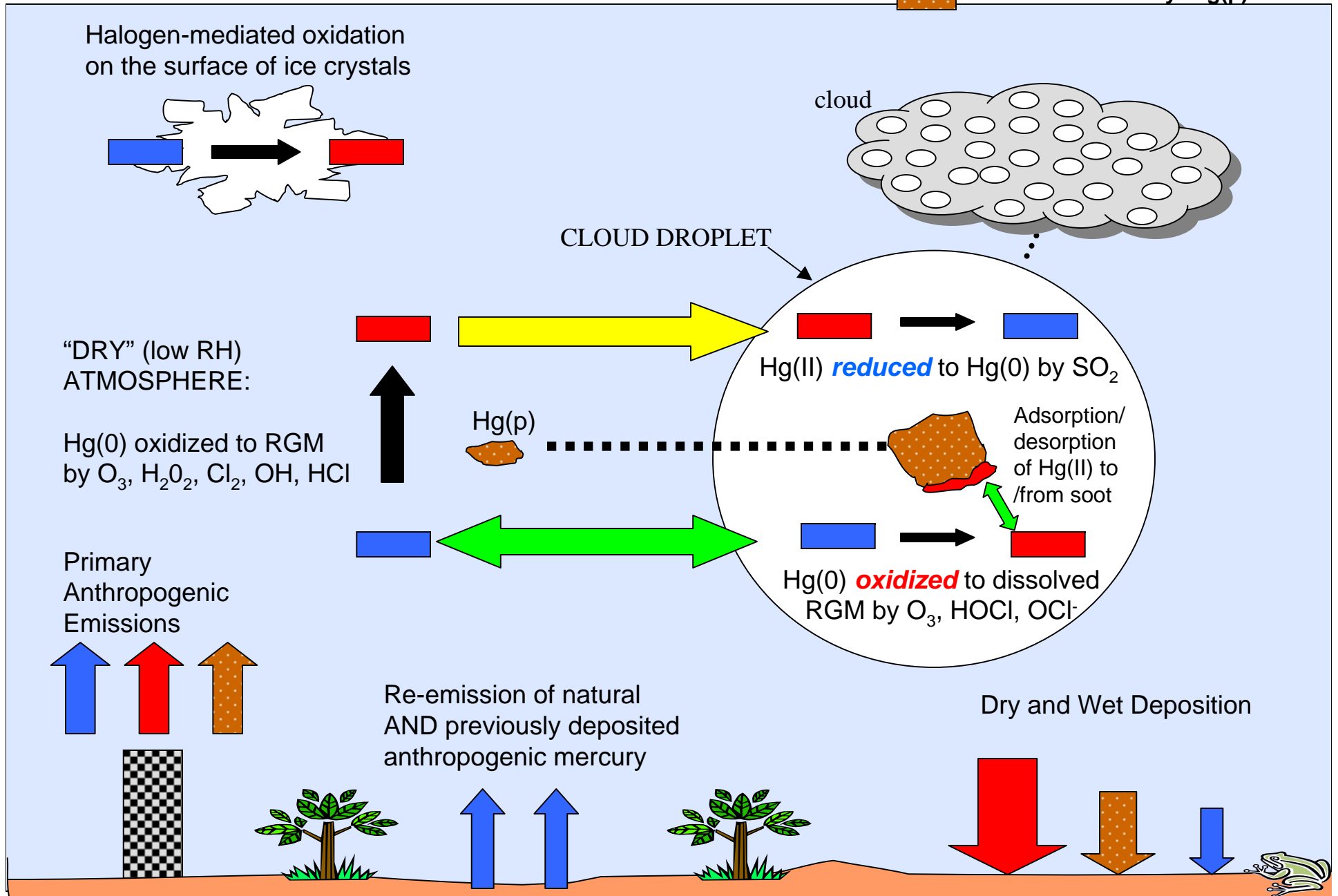


## Particulate Mercury (Hg(p))

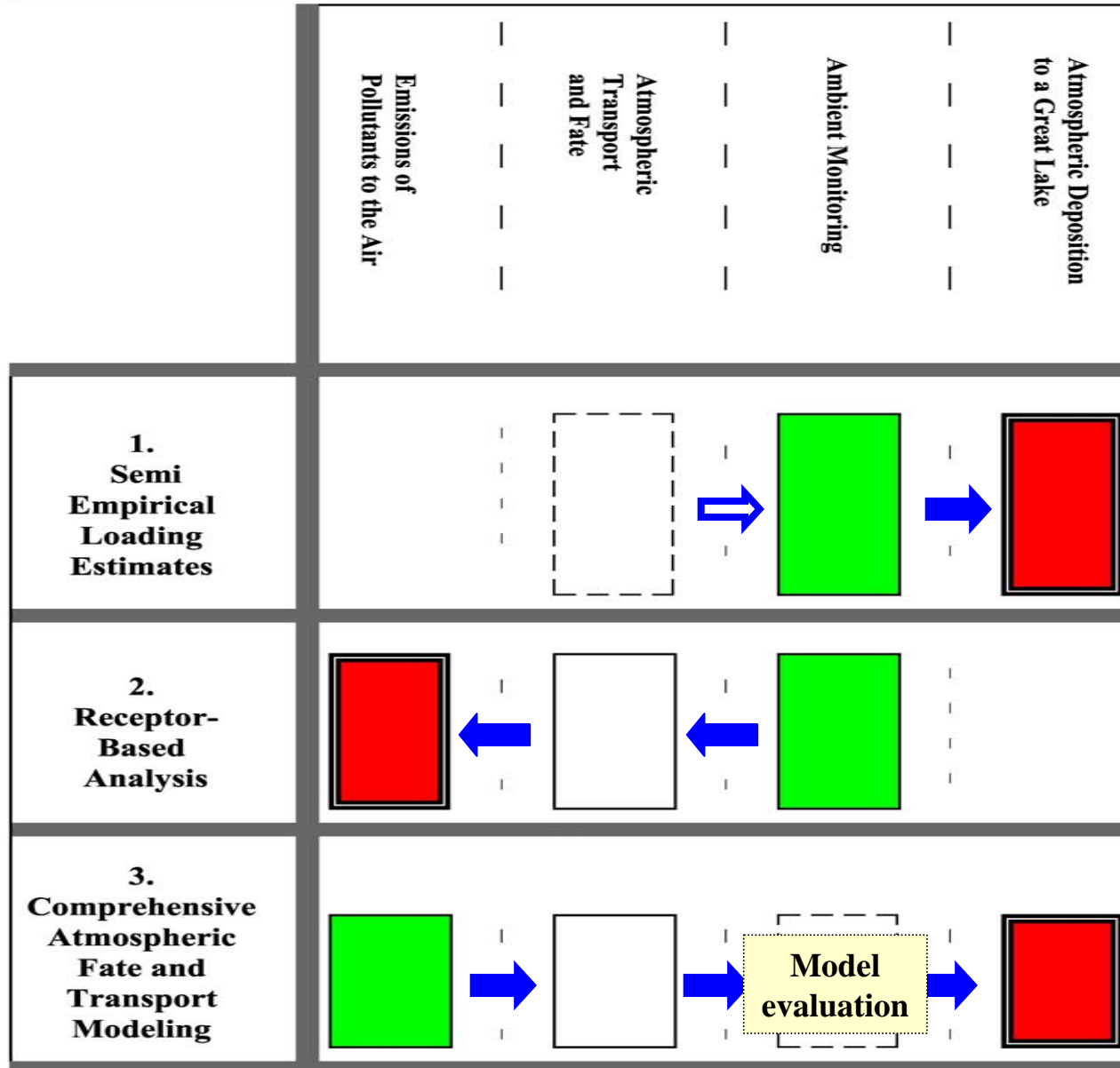
- a few percent of total Hg in atmosphere
- not pure particles of mercury...  
(Hg compounds associated with atmospheric particulate)
- species largely unknown (in some cases, may be HgO?)
- moderate atmospheric lifetime (perhaps 1~ 2 weeks)
- local and regional effects
- bioavailability?

# Atmospheric Fate Processes for Hg

- Elemental Mercury: Hg(0)
- Reactive Gaseous Mercury: RGM
- Particulate Mercury: Hg(p)



## 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...

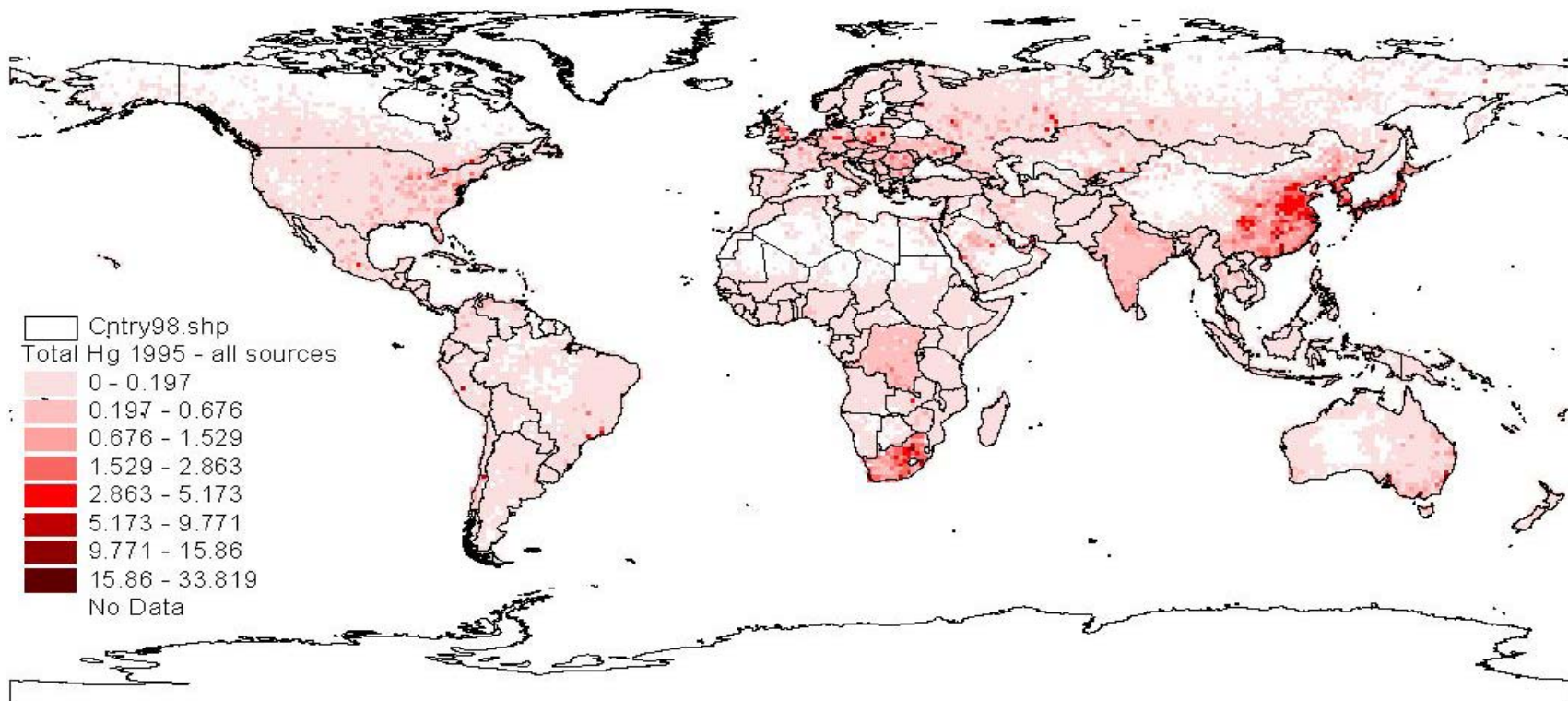
❑ We are interested in the deposition to an *entire* water body, or to a particular ecosystem

❑ *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

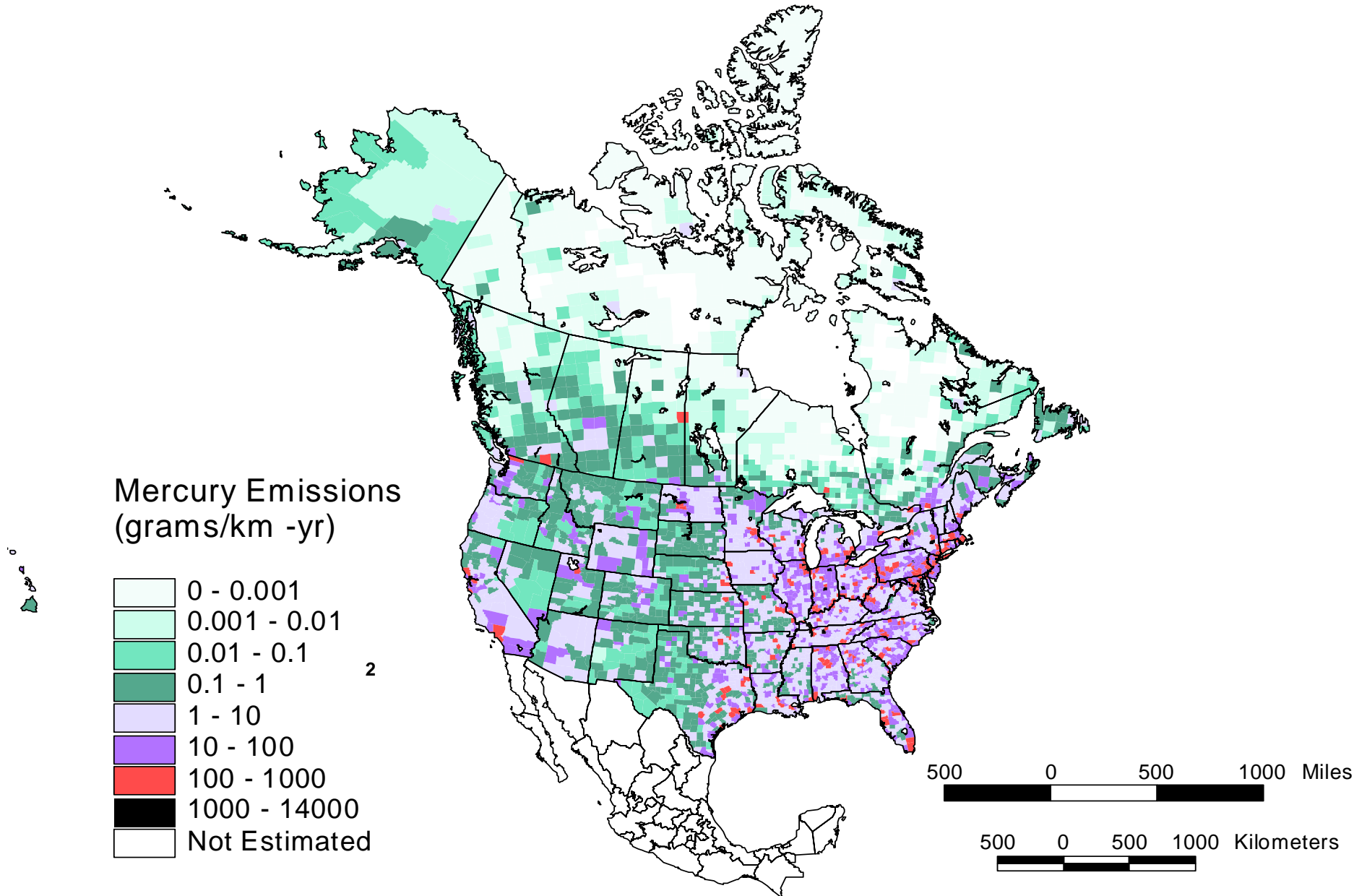
- **Start with atmospheric mercury *emissions inventory***
- **Perform *atmospheric fate and transport modeling* of these emissions (using a modified version of NOAA's HSYPLIT model)**
- **Keep track of *source-receptor information* during the modeling**
- **Evaluate the modeling by *comparison* of the 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)**



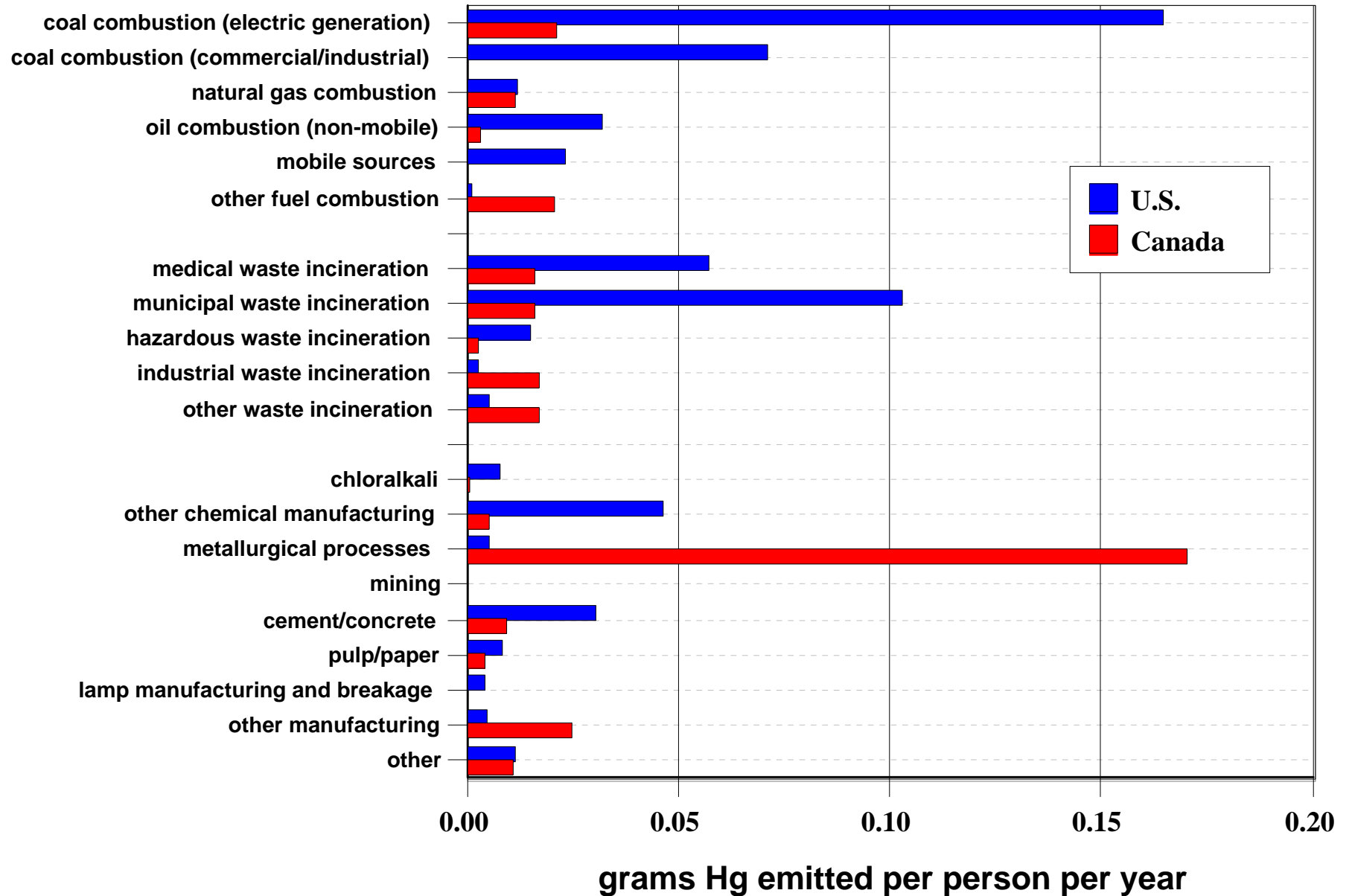
# 1995 Global Hg Emissions Inventory

Josef Pacyna, NILU, Norway (2001)

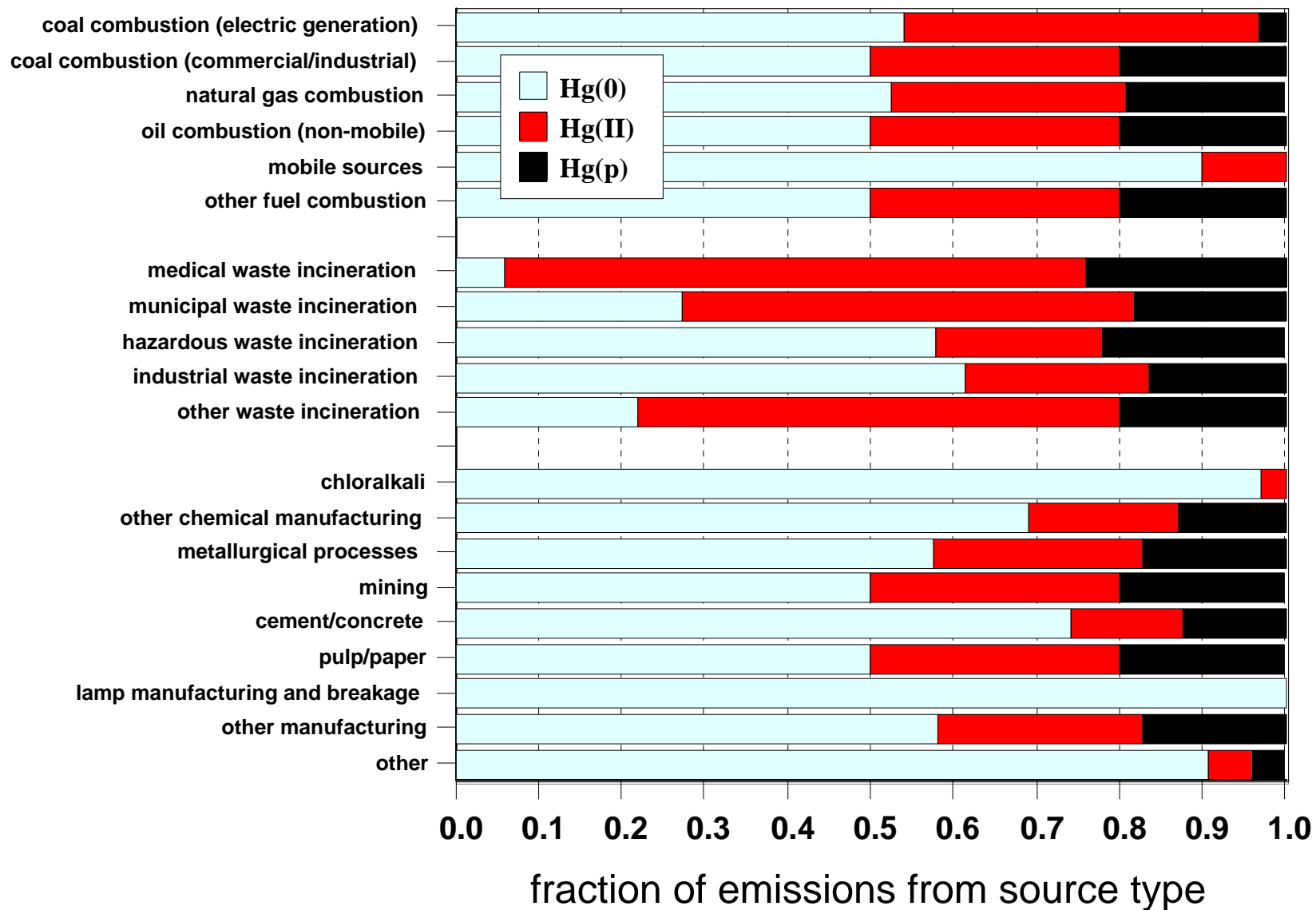
# Geographic Distribution of Estimated Anthropogenic Mercury Emissions in the U.S. and Canada for 1995-1996



# 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

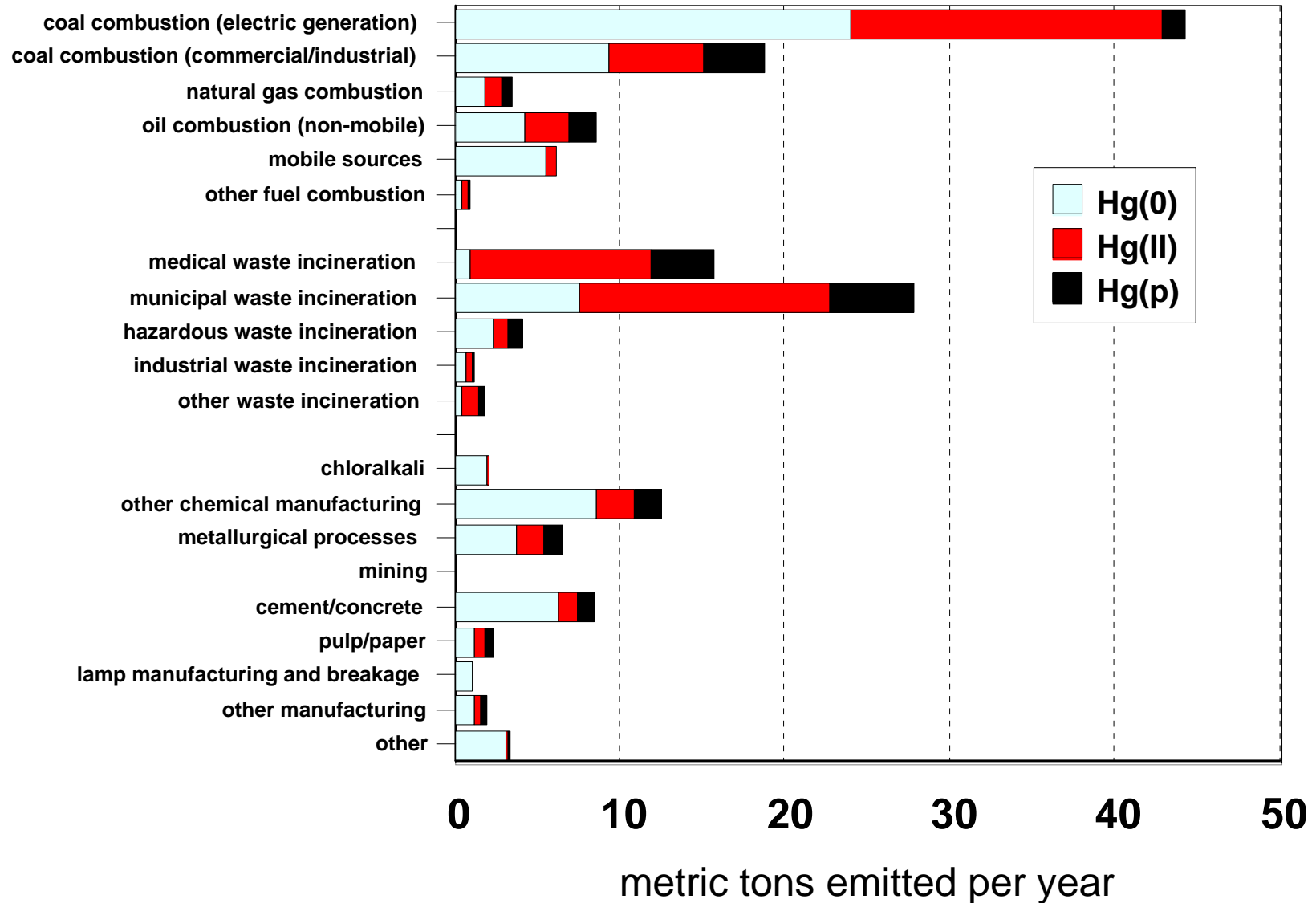
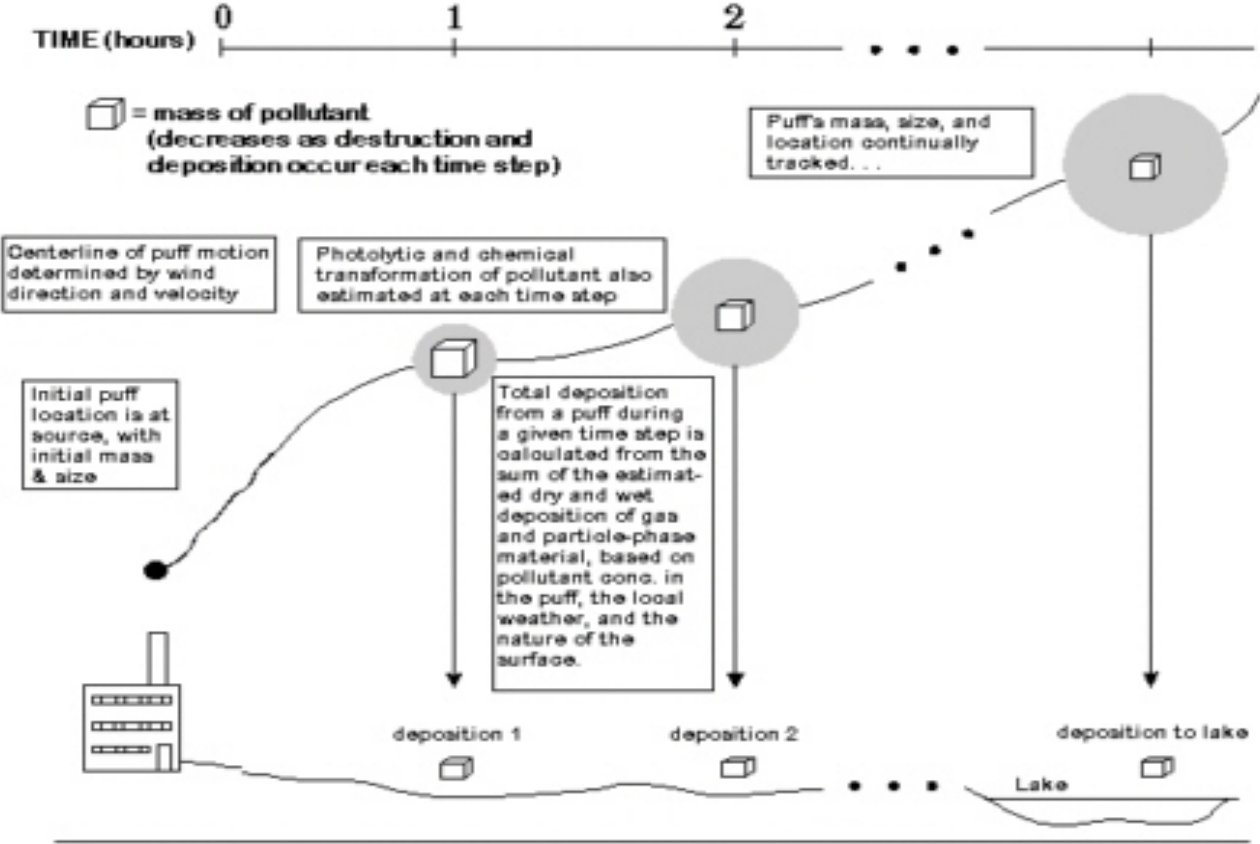
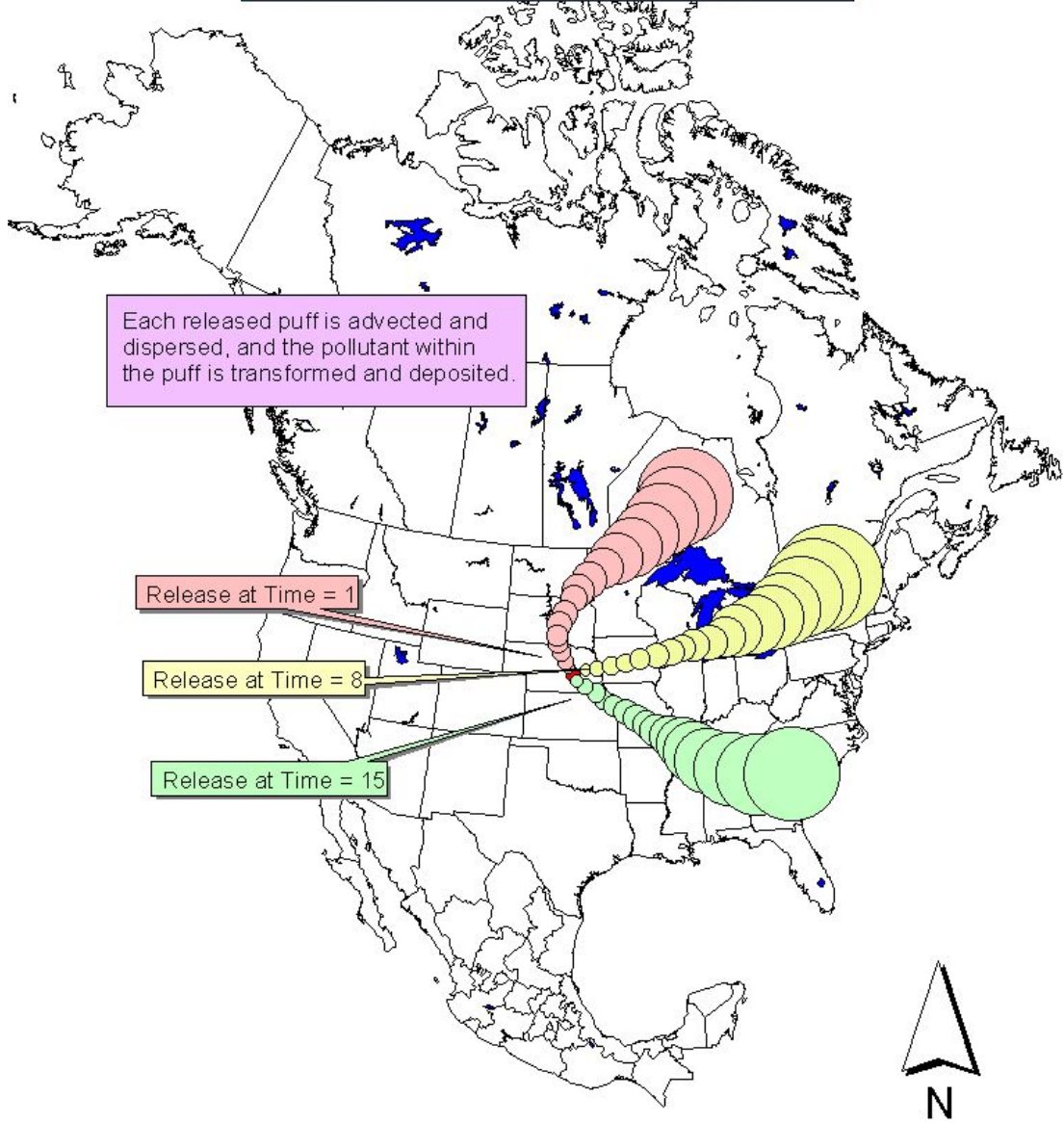


Figure 1. Lagrangian Puff Air Transport and Deposition Model



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

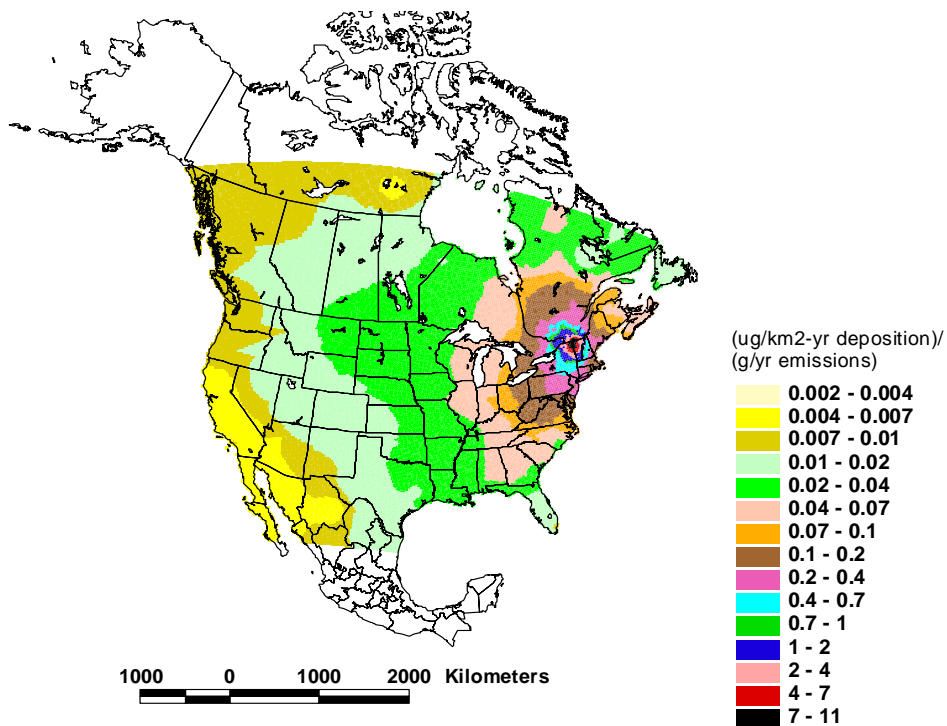




at any given location,  
the *transfer coefficient*  
is defined as the amount  
that would be deposited  
in the given receptor  
(in this case, Lake Champlain)  
*if* there were emissions  
at that location.

## Transfer Coefficients

- refer to hypothetical emissions;  
*are independent of actual emissions*
- can be formulated with different units  
*[total Hg deposition flux (ug/km<sup>2</sup>-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]*



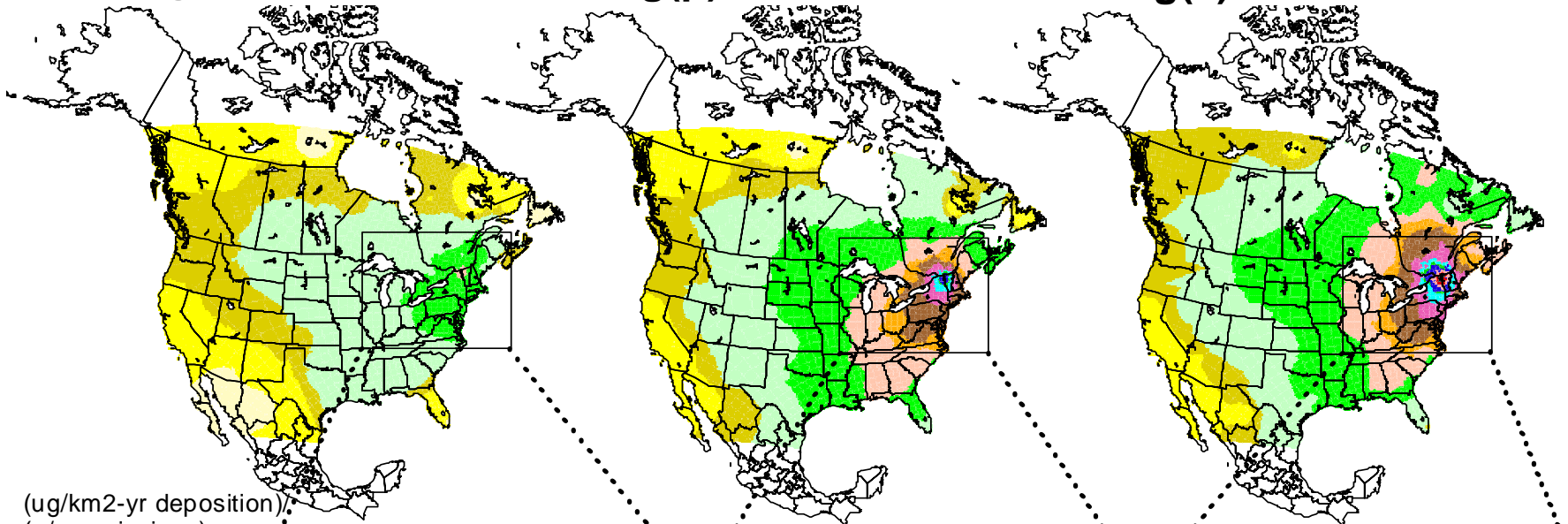
# Annual 1996 Transfer Coefficients for Different Forms of Mercury to Lake Champlain

1000 0 1000 Kilometers

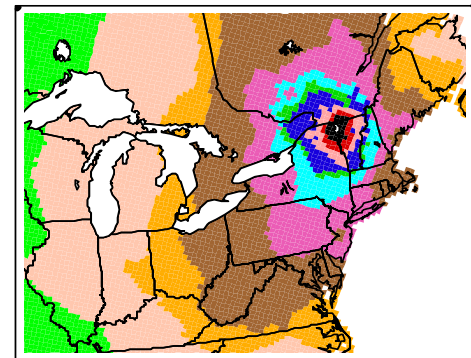
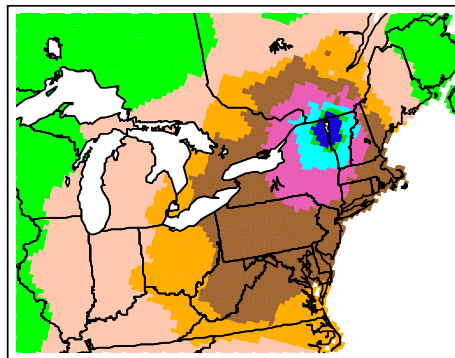
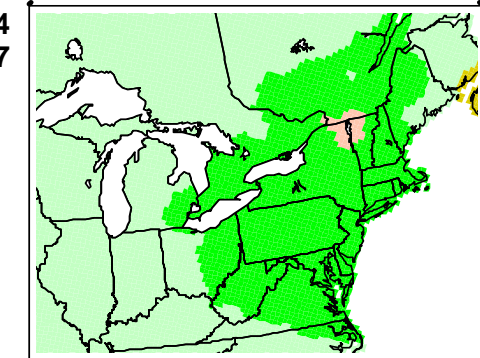
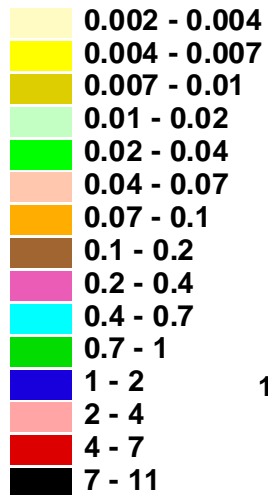
Hg(0)

Hg(p)

Hg(II)

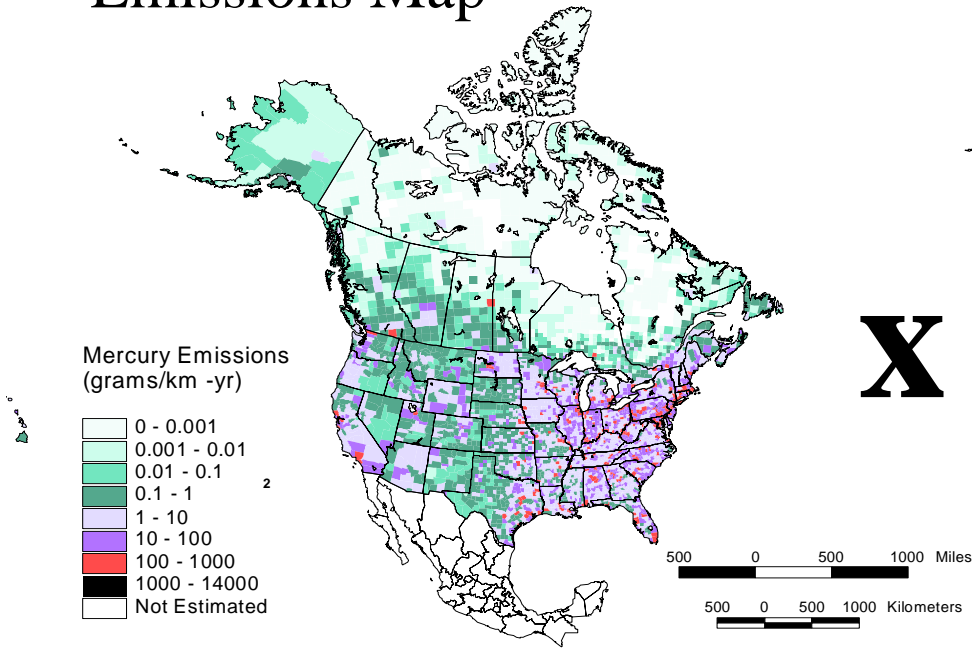


(ug/km<sup>2</sup>-yr deposition),  
(g/yr emissions)

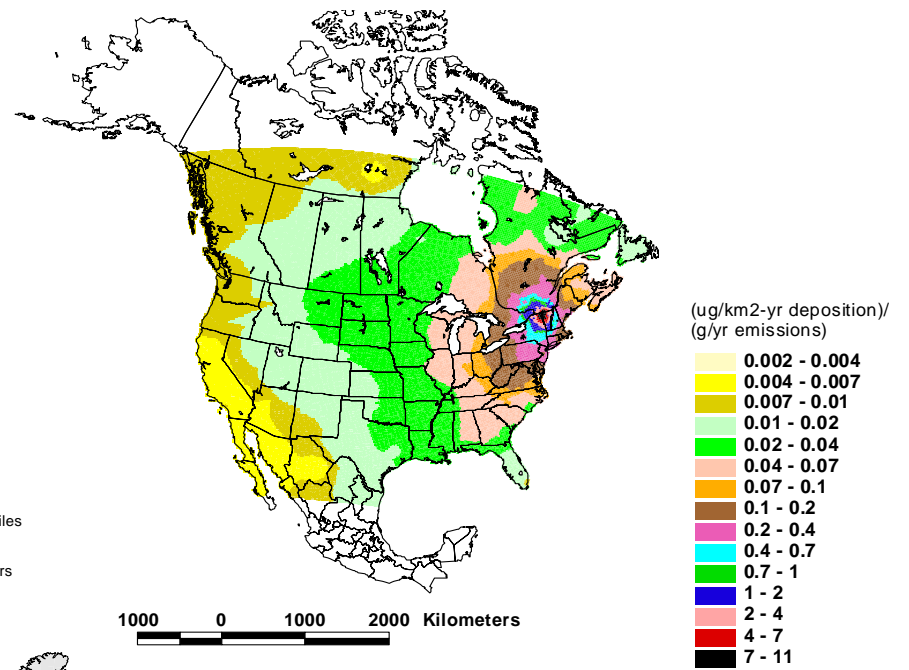


1000 0 1000 Kilometers

# Emissions Map

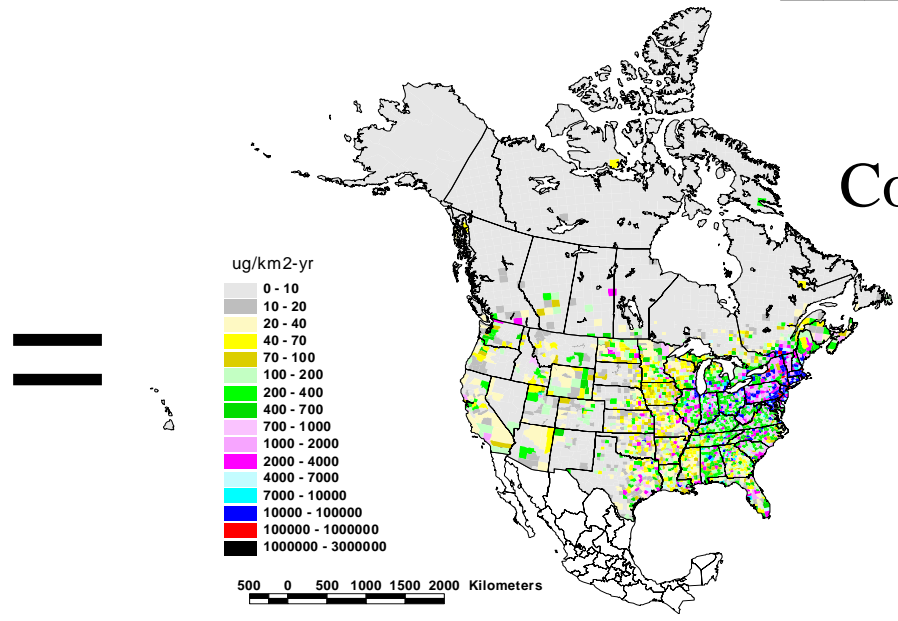


# Transfer Coefficient Map



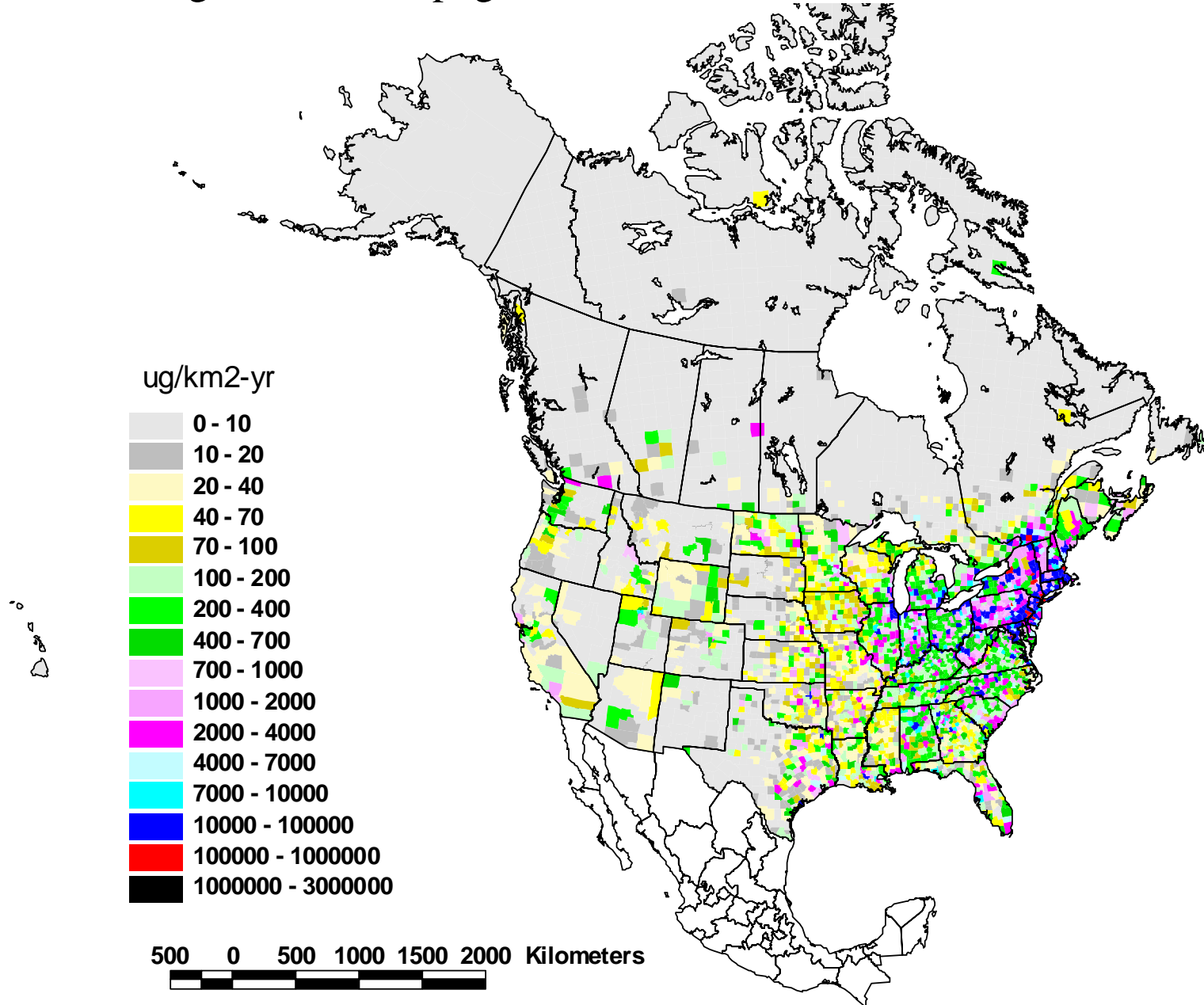
**X**

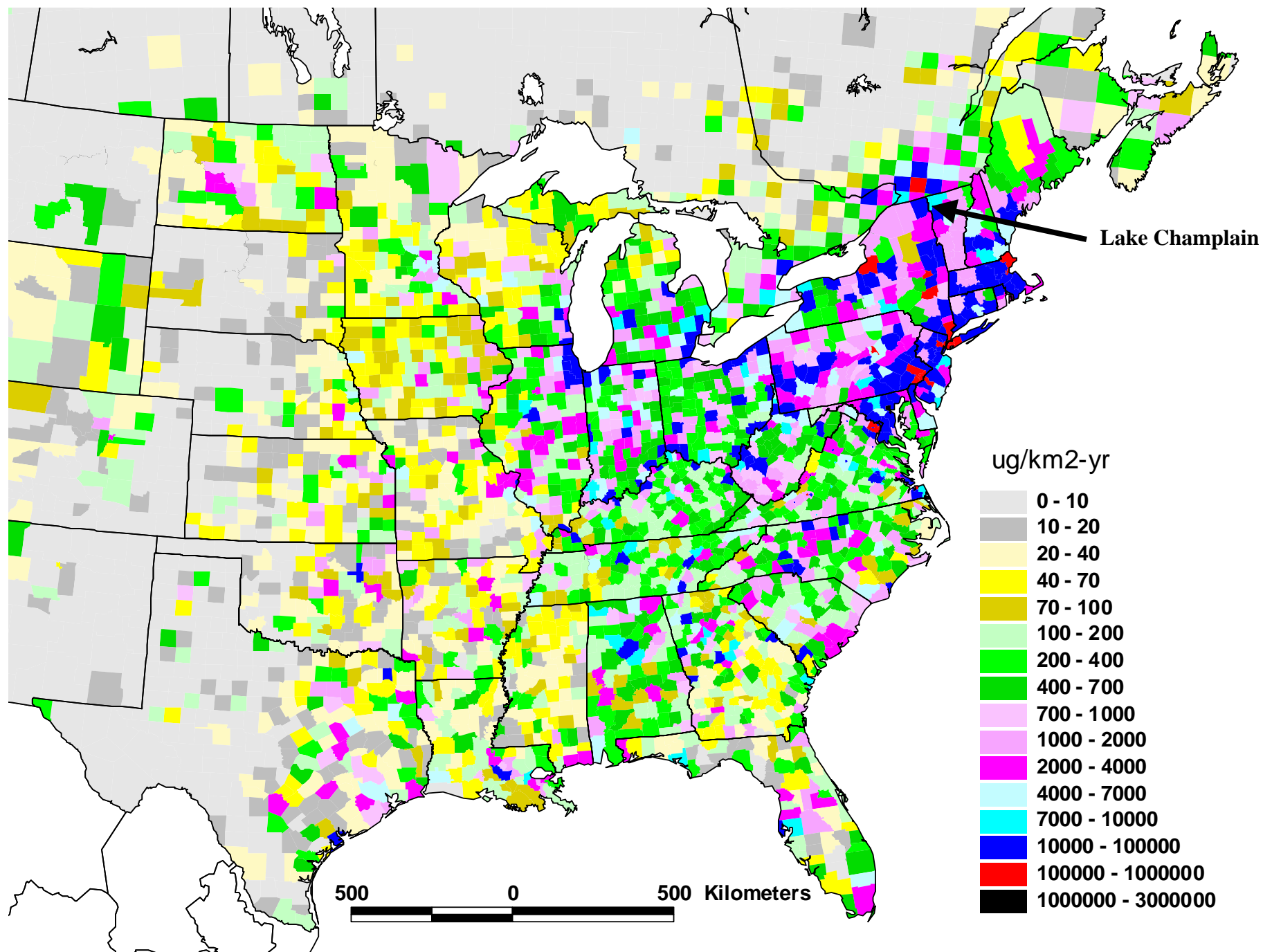
# Contribution Map

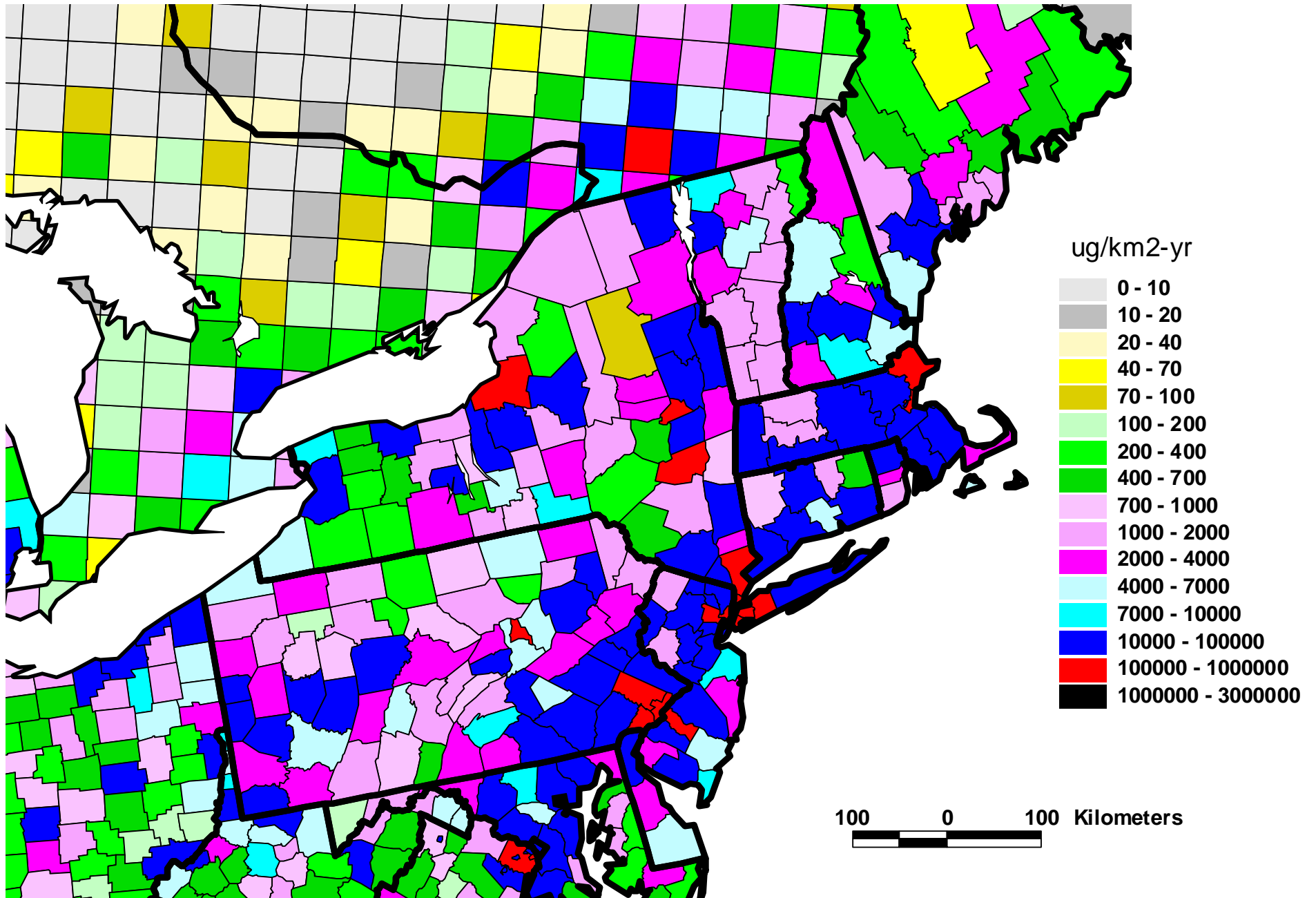


**=**

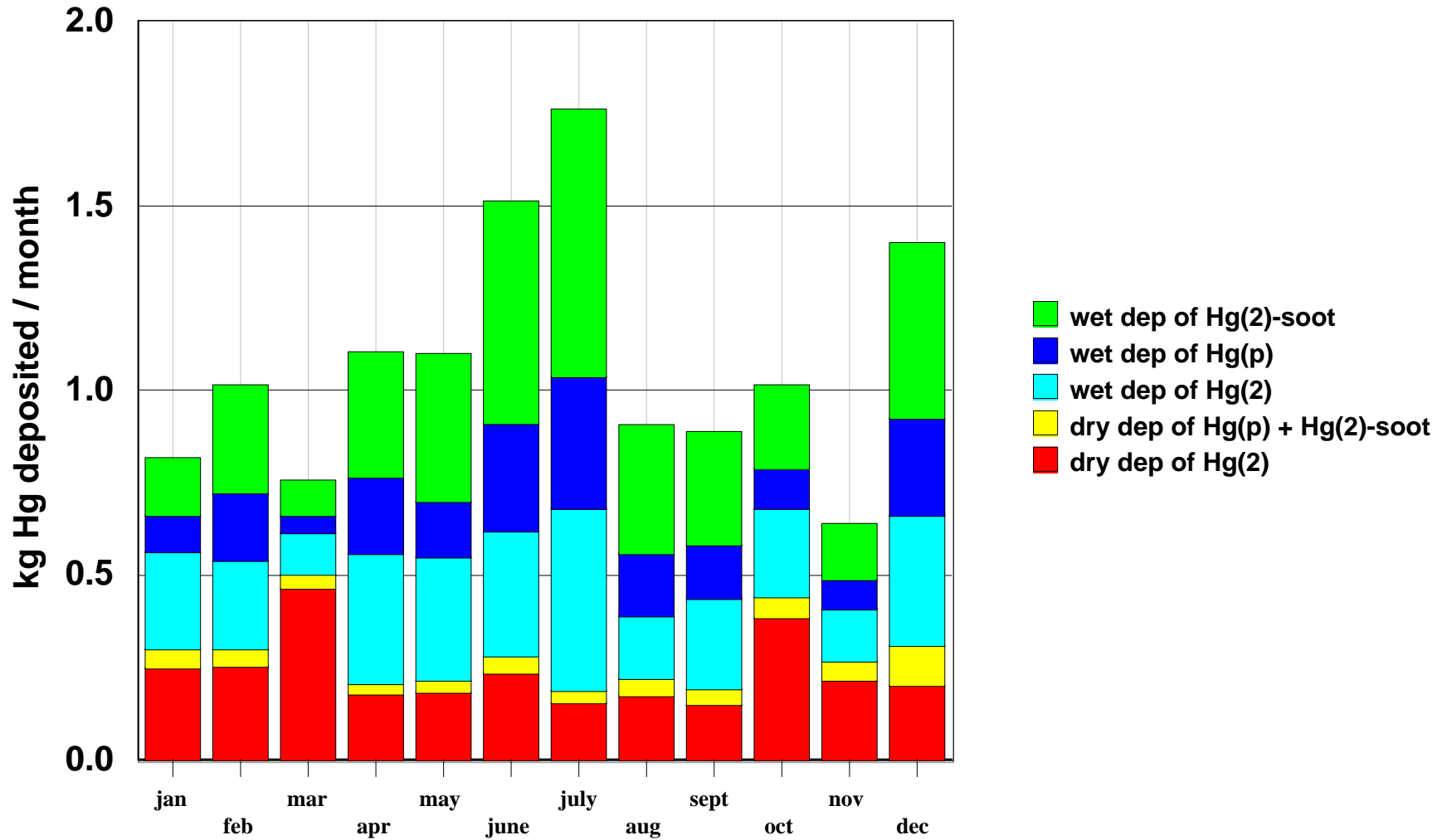
# Geographical Distribution of Atmospheric Deposition Contributions to Lake Champlain Arising from Anthropogenic Sources in the U.S. and Canada During 1996



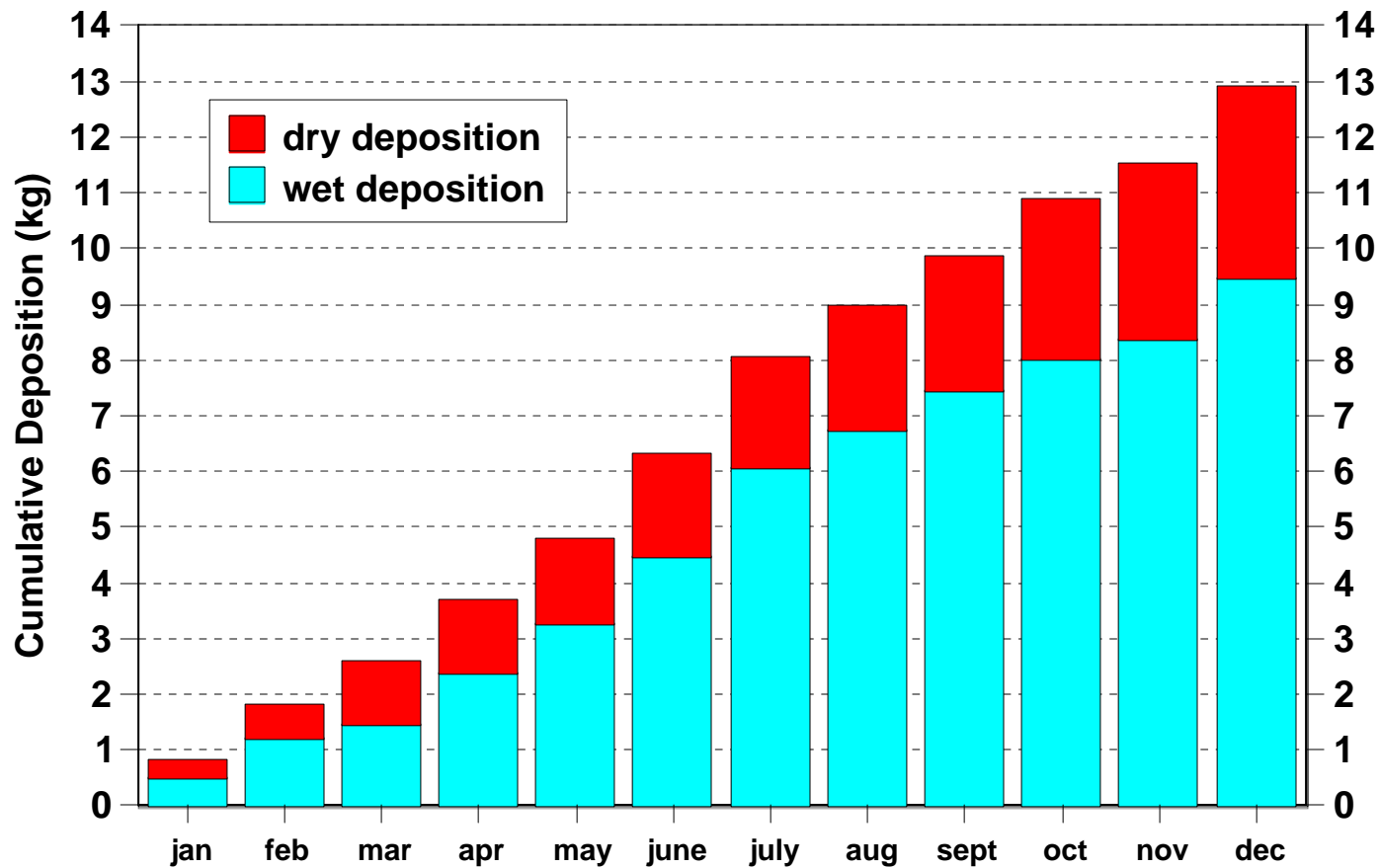




# Monthly Model-Estimated Wet and Dry Deposition of Different Forms of Mercury to Lake Champlain During 1996 Arising from U.S. and Canadian Anthropogenic Emissions

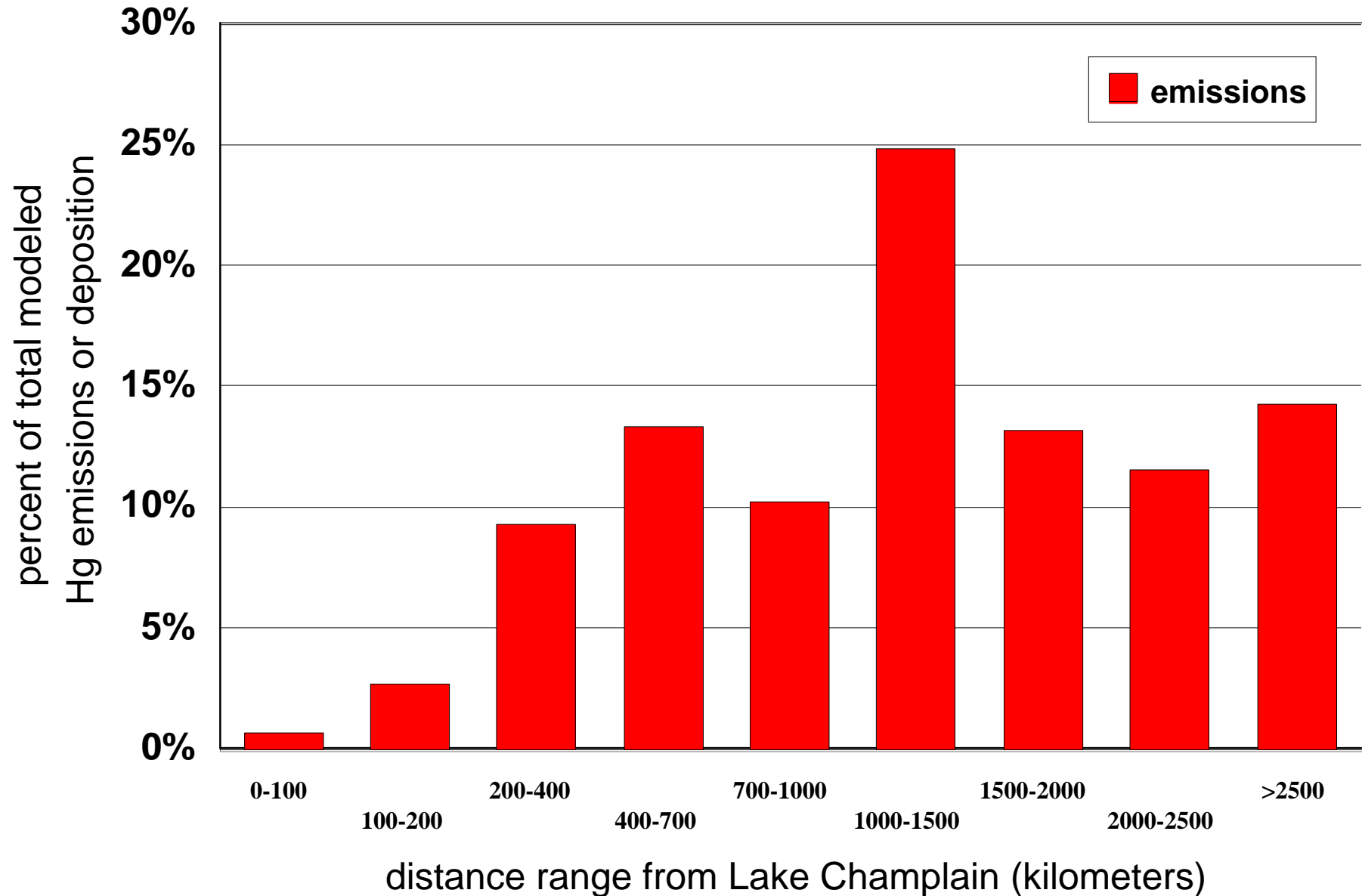


# Cumulative Model-Estimated Wet and Dry Deposition of Mercury to Lake Champlain During 1996 Arising from U.S. and Canadian Anthropogenic Emissions

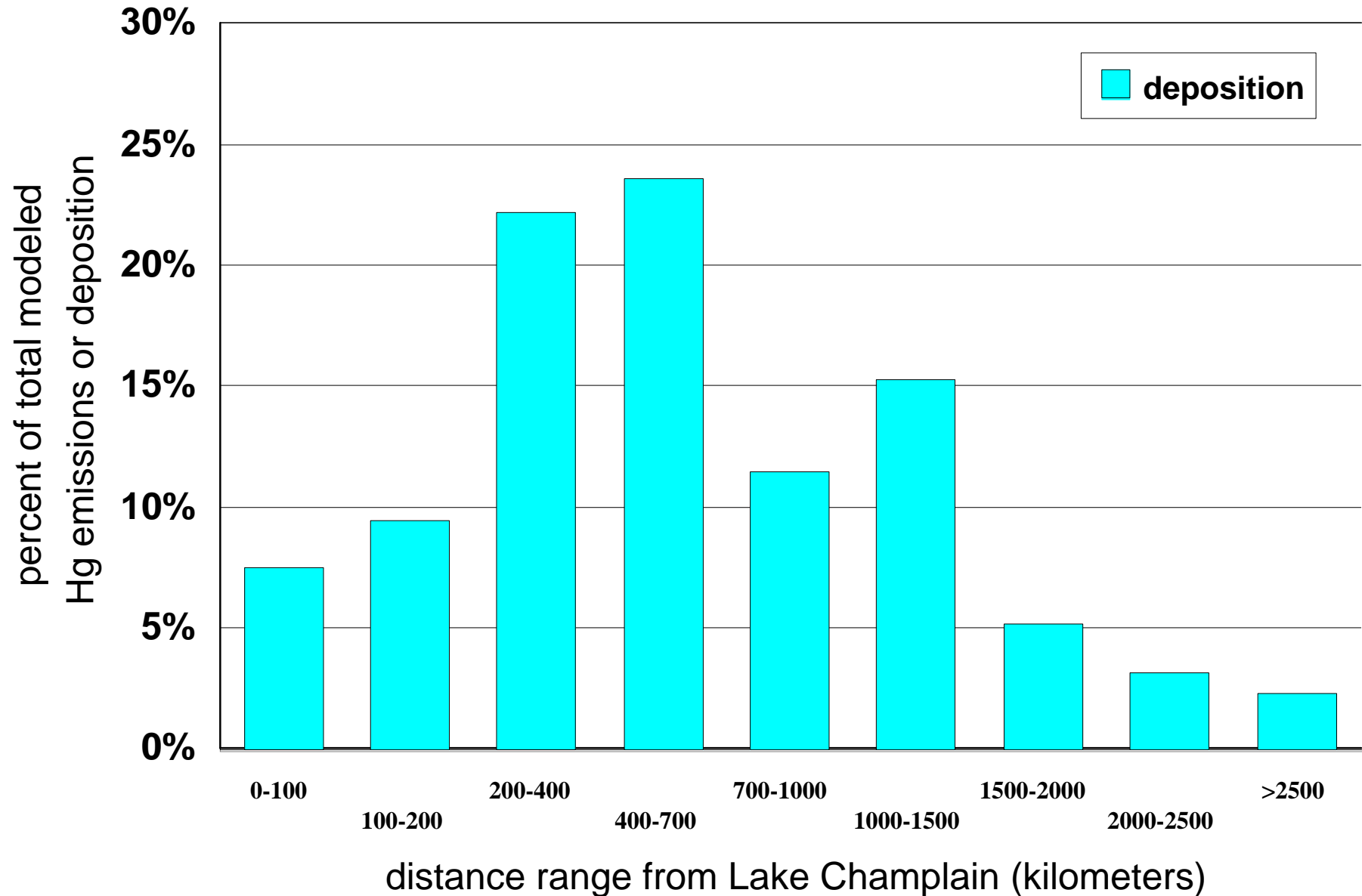




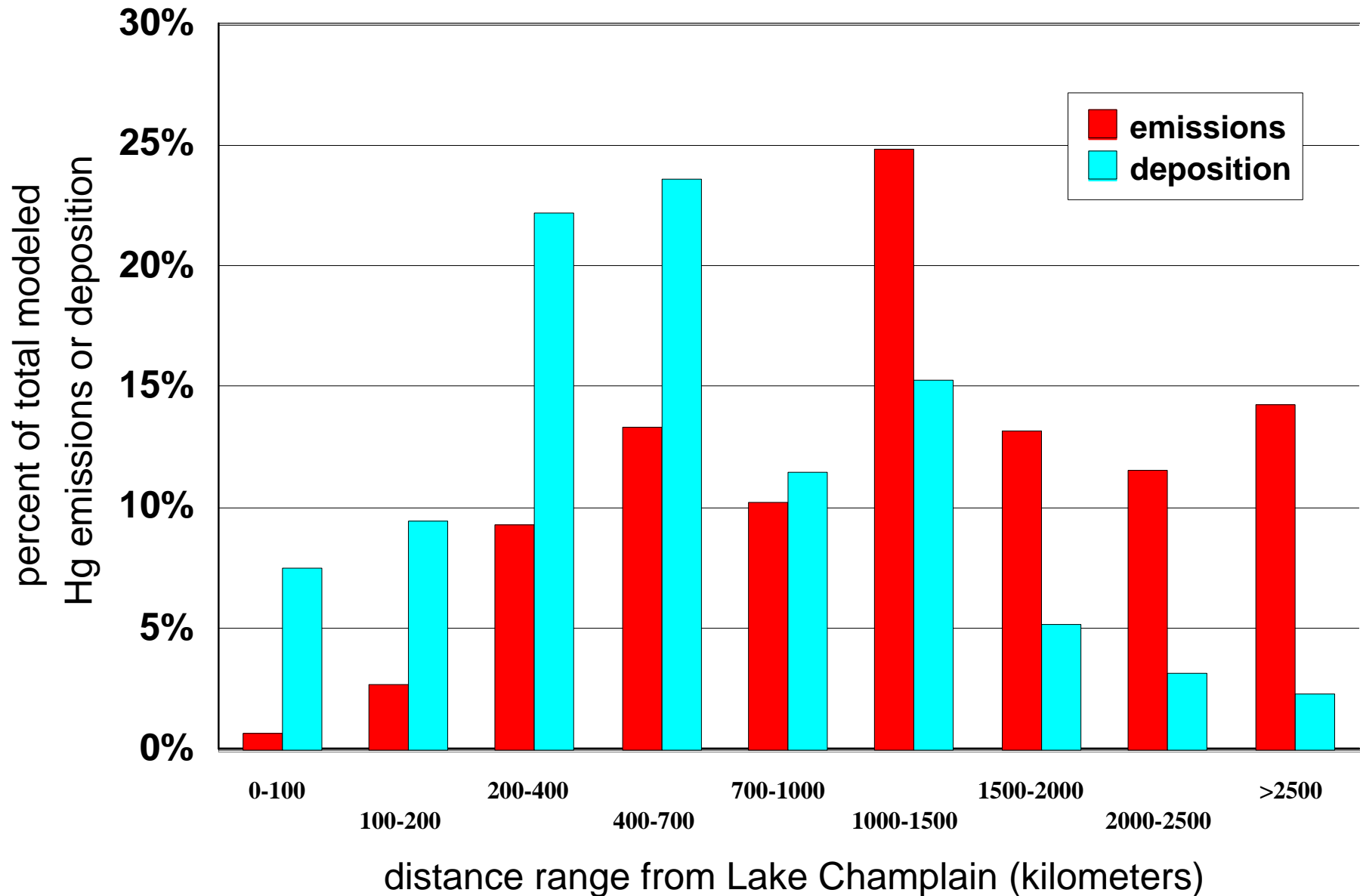
Percent of modeled **EMISSIONS** of mercury to Lake Champlain from different distance ranges away from the lake



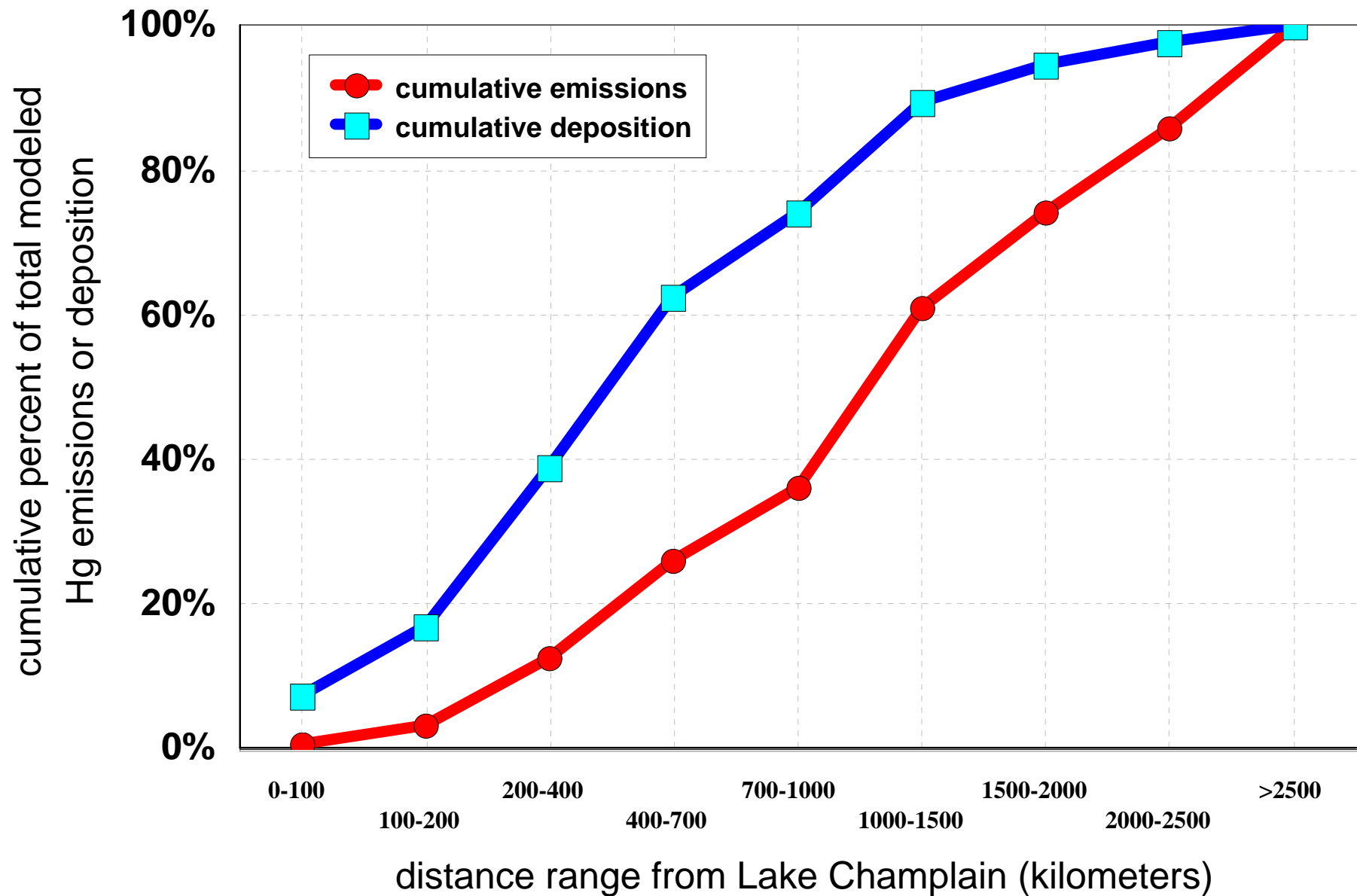
Percent of modeled **DEPOSITION** of mercury to Lake Champlain from different distance ranges away from the lake



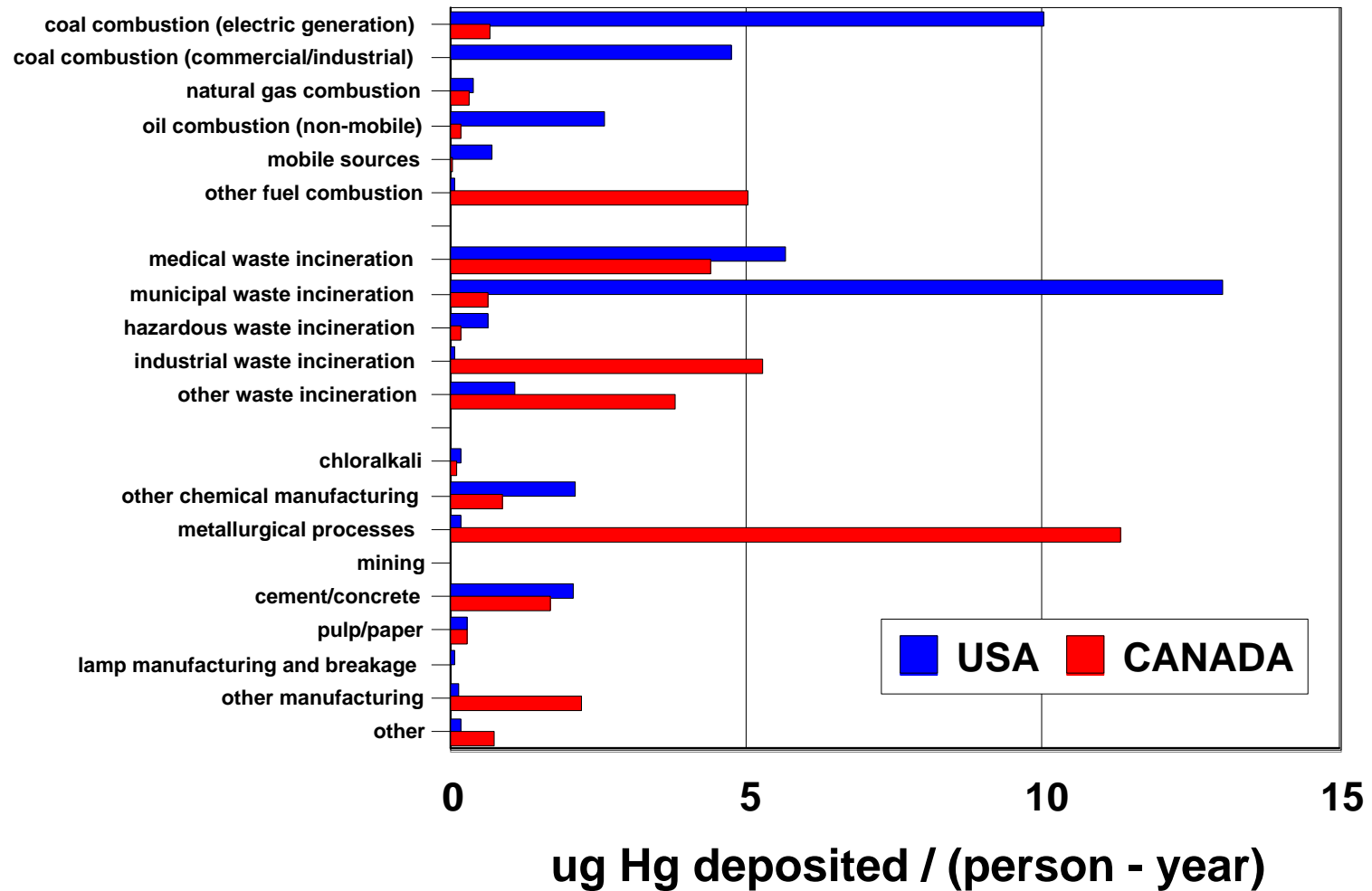
Percent of modeled **emissions** and **deposition** of mercury to Lake Champlain from different distance ranges away from the lake



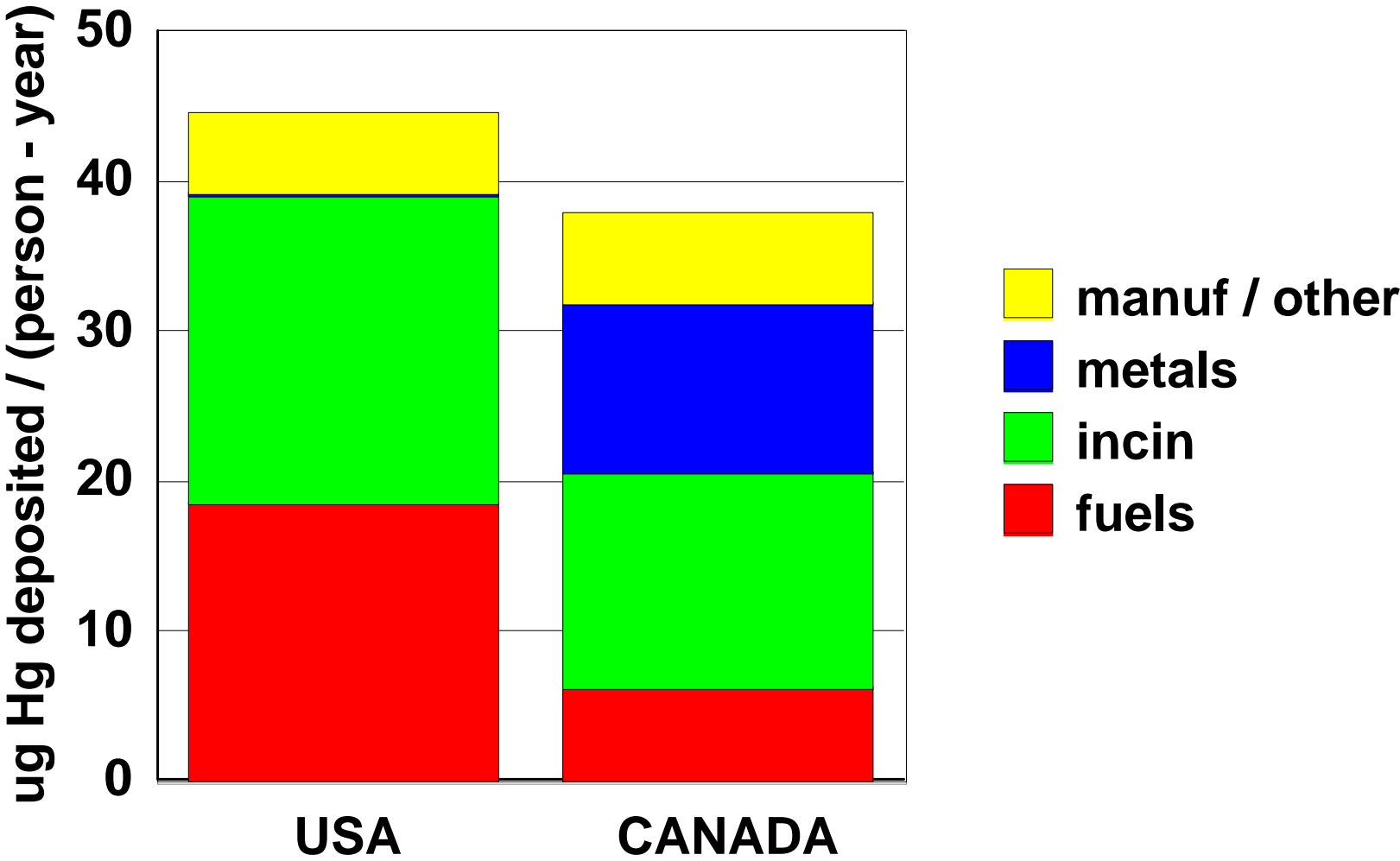
*Cumulative* percent of modeled **emissions** and **deposition** of mercury to Lake Champlain from different distance ranges away from the lake



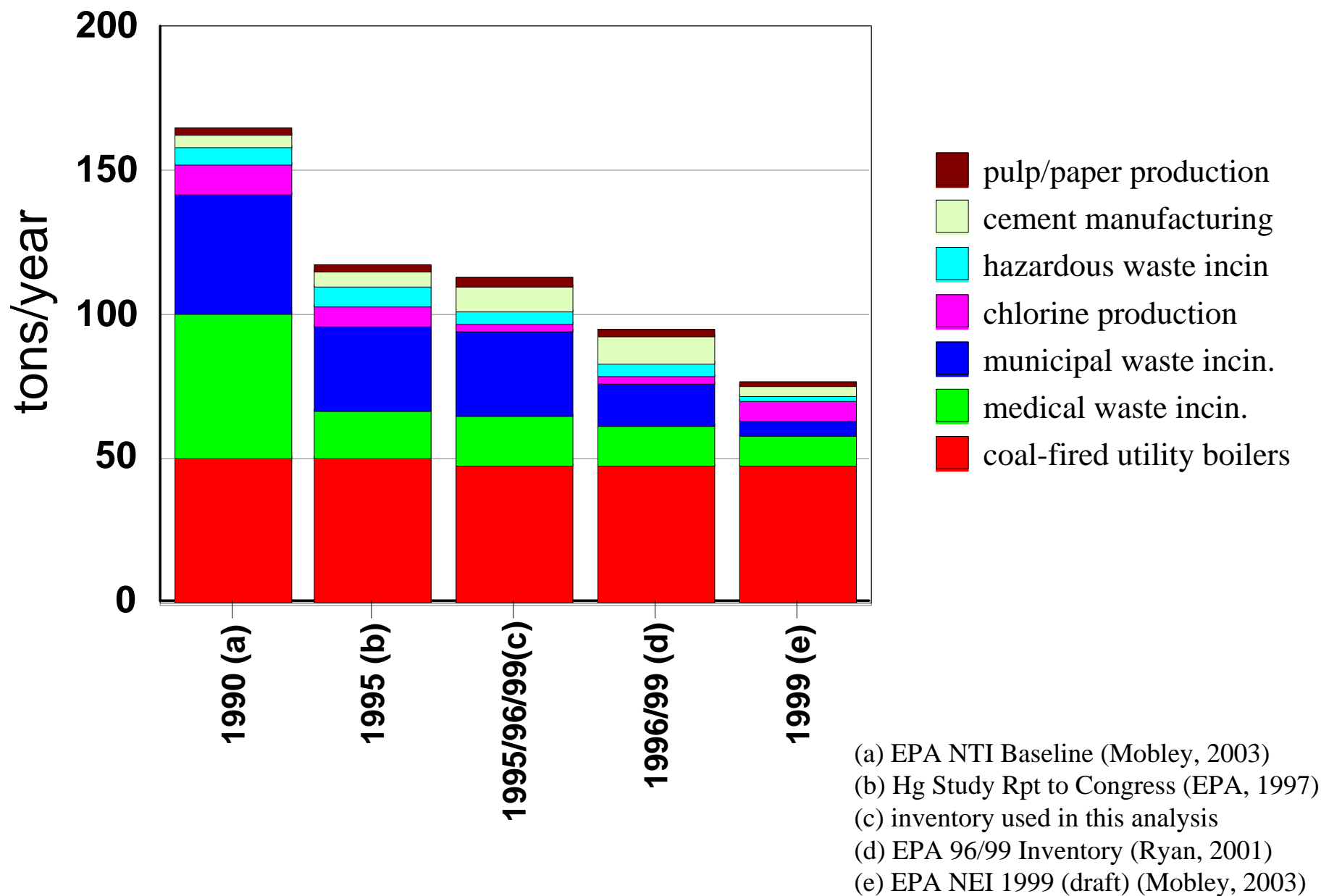
# Per Capita Mercury Atmospheric Mercury Deposition Contributions to Lake Champlain from U.S. and Canadian *Detailed* Source Categories (1996)



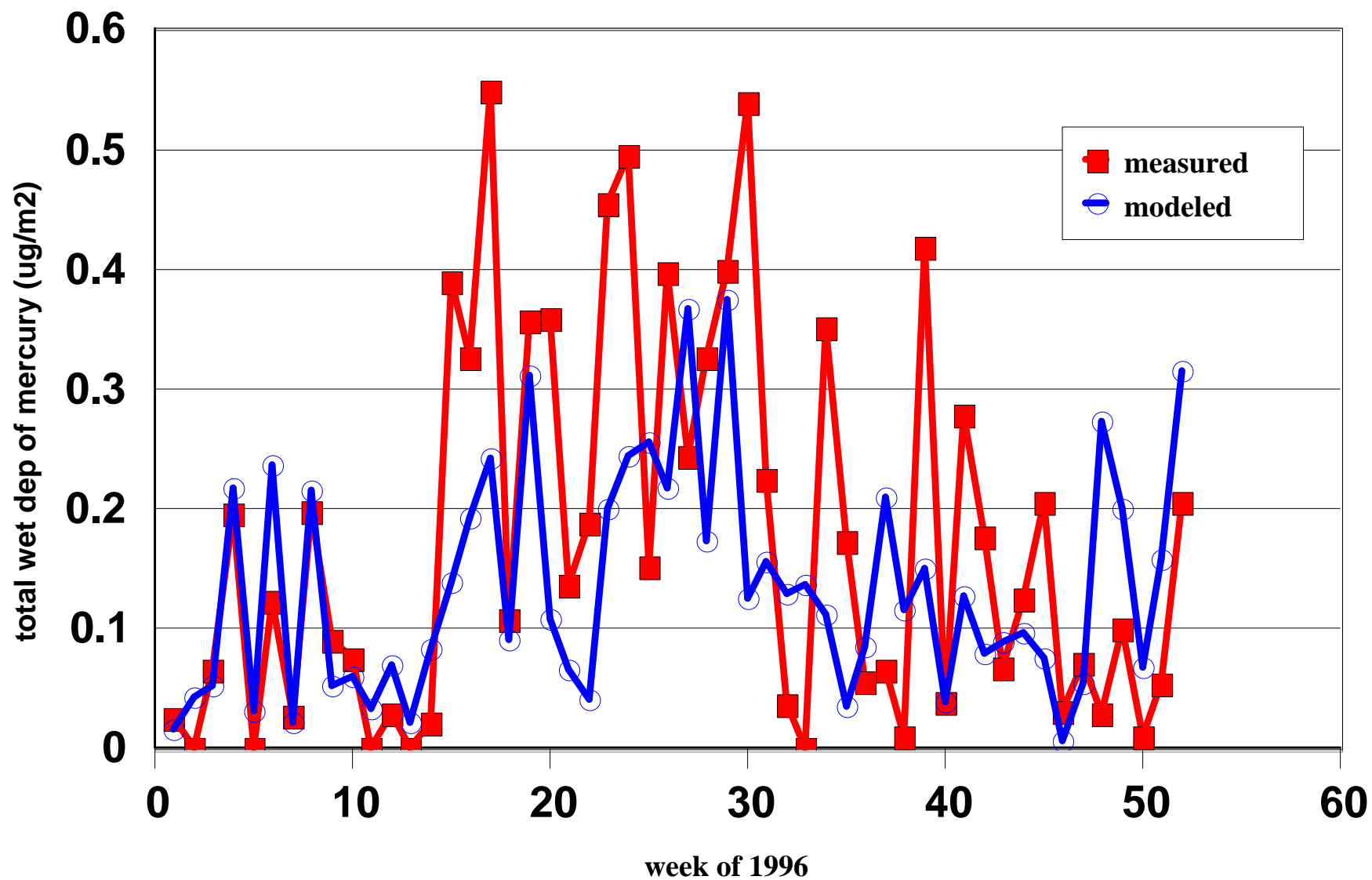
**Per Capita Mercury Atmospheric Mercury Deposition  
Contributions to Lake Champlain from U.S. and  
Canadian Aggregated Source Categories (1996)**



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

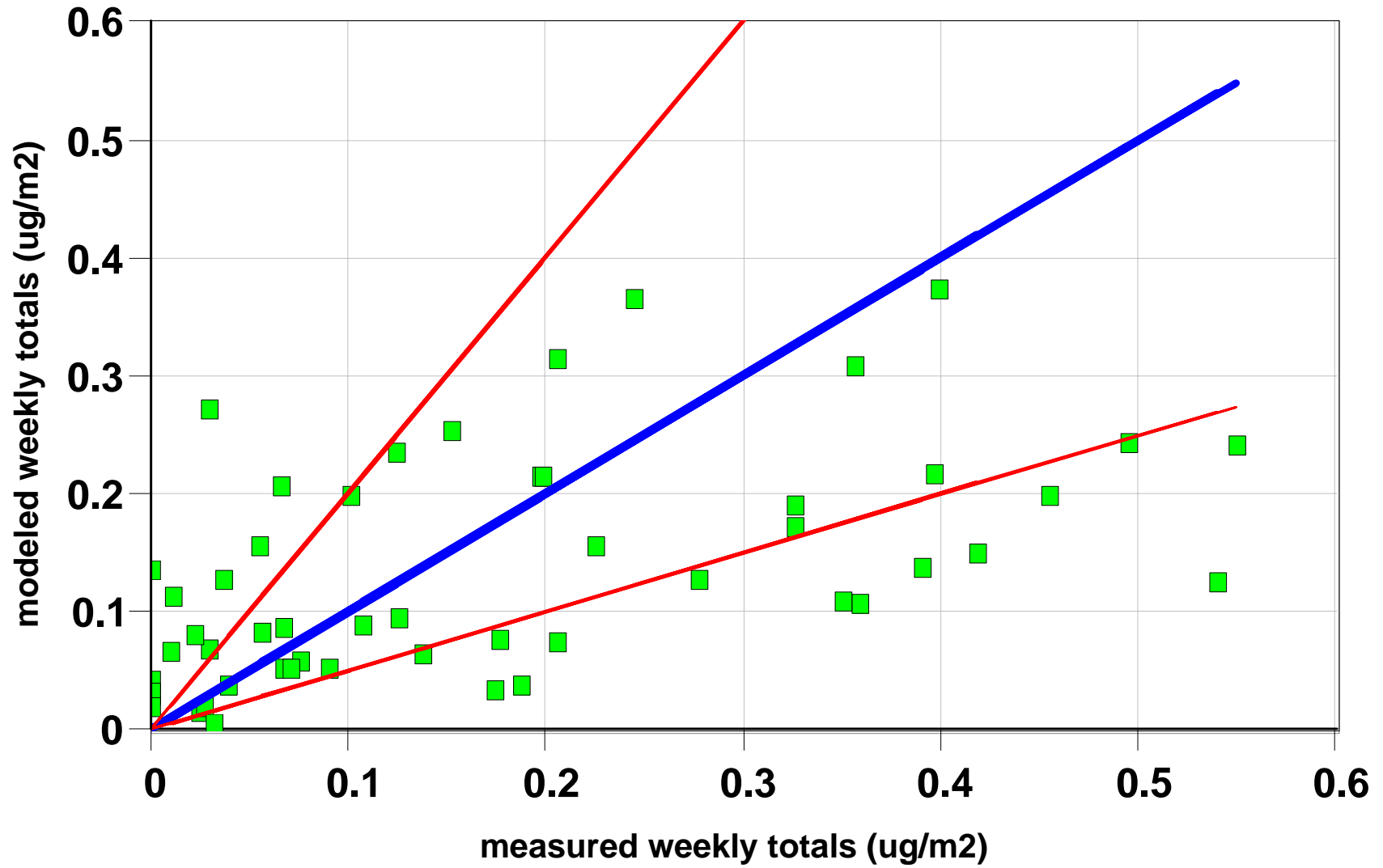


Comparison of Modeled vs. Measured Wet Deposition at Underhill Center, VT during 1996

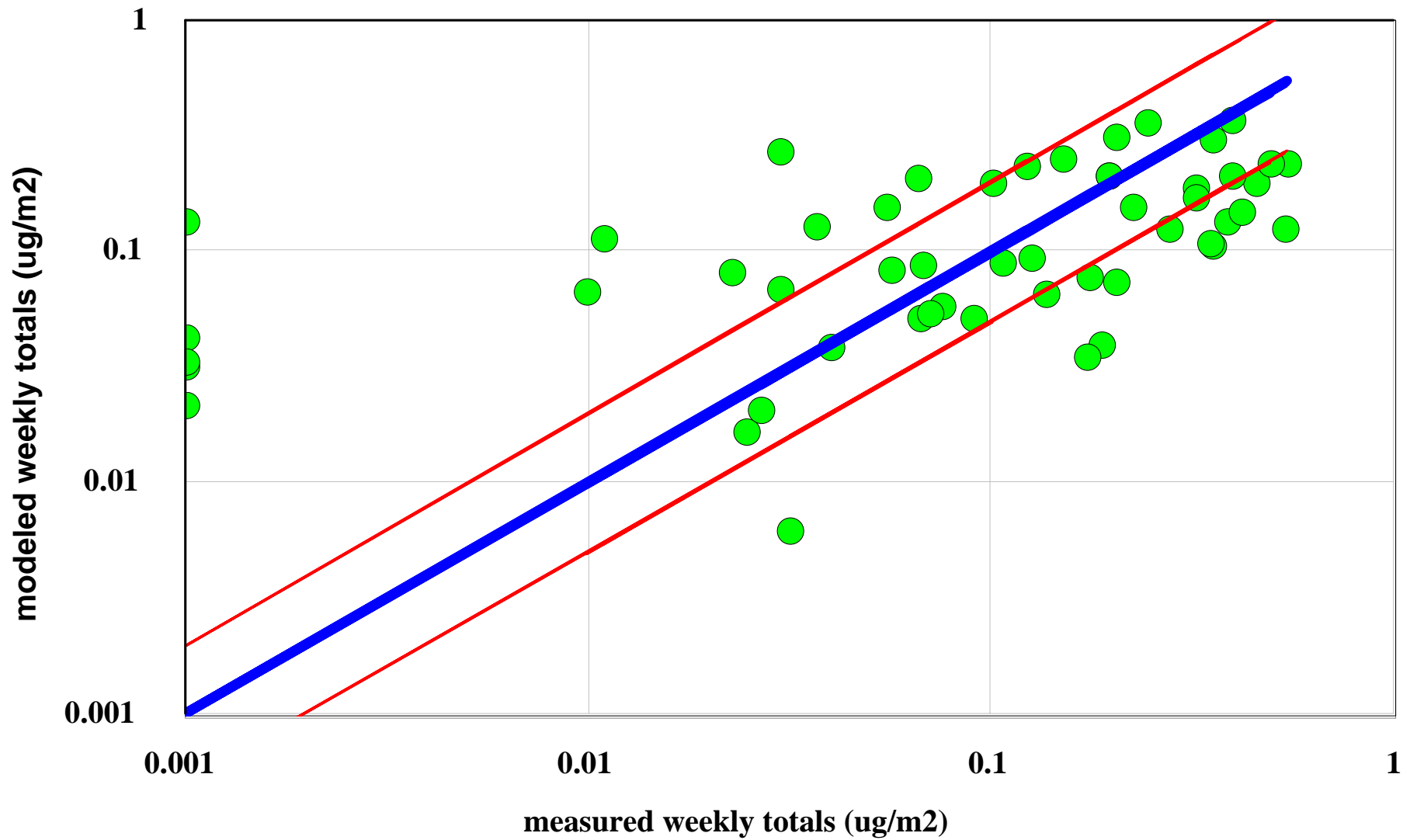




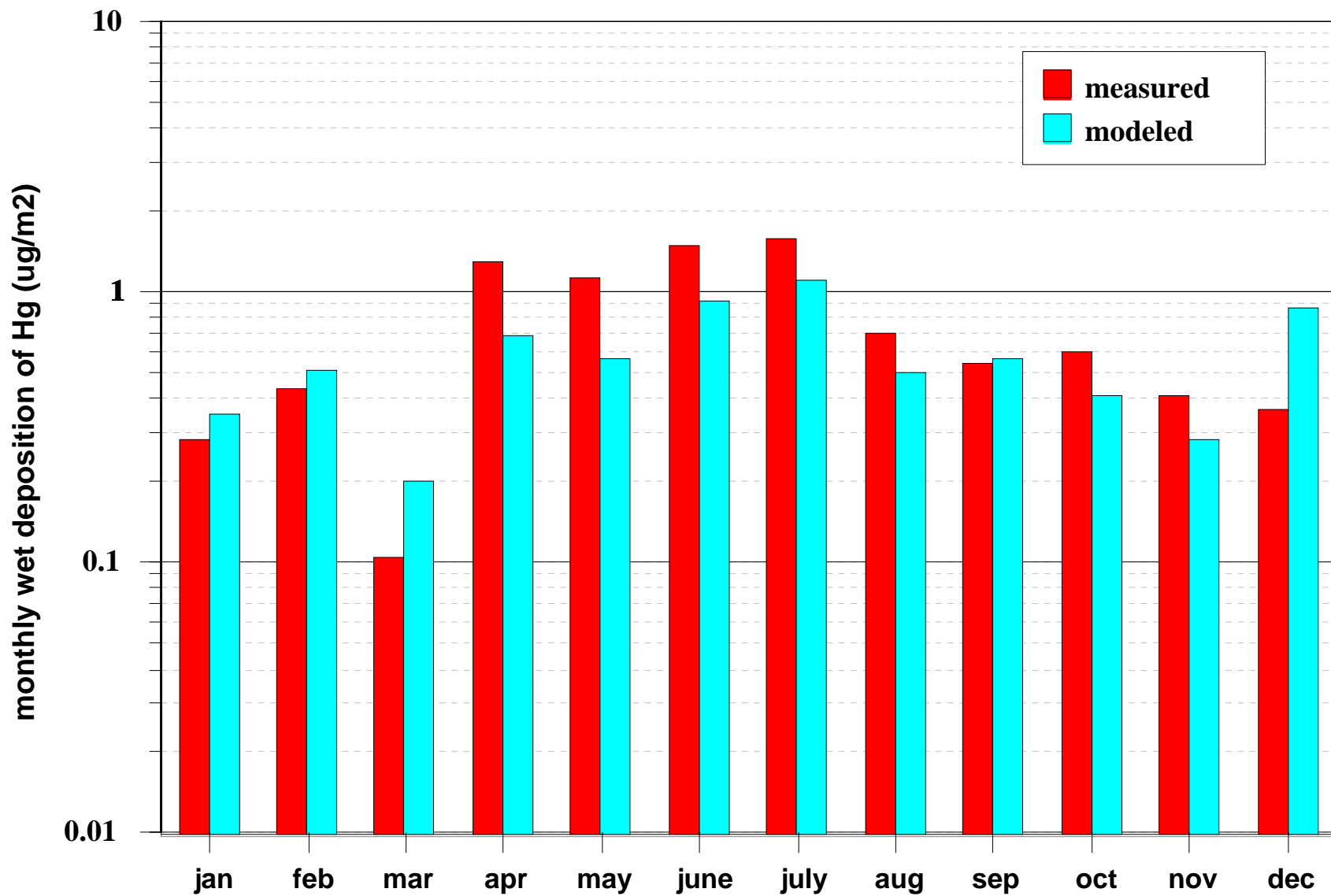
**Comparison of Modeled vs. Measured Wet Deposition  
at Underhill Center, VT during 1996**



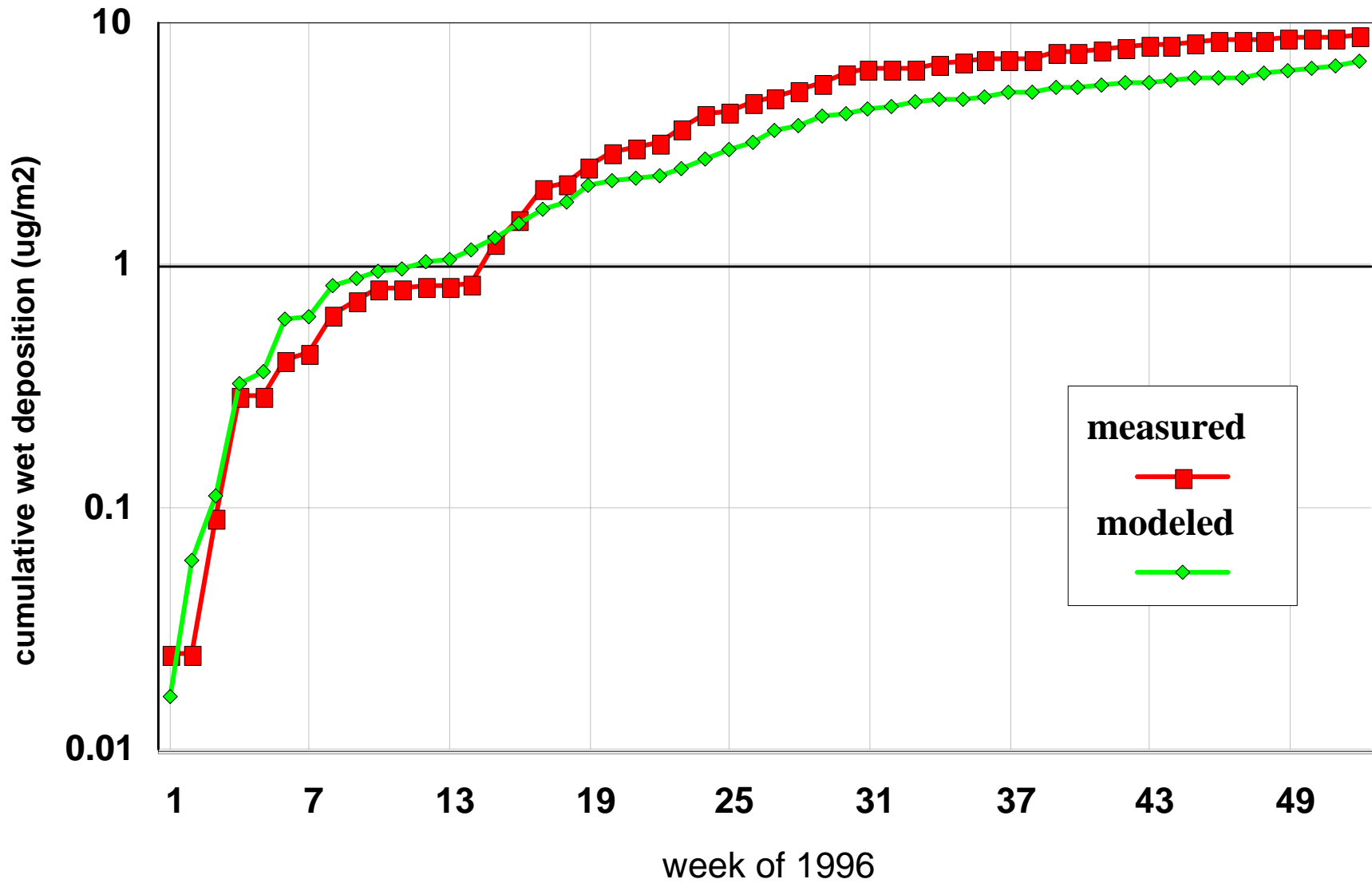
**Comparison of Modeled vs. Measured Wet Deposition  
at Underhill Center, VT during 1996**



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



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



# 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  $\text{Hg}^0$  is zero – essentially that natural emissions and re-emitted mercury sort of balance out  $\text{Hg}^0$  deposition, so that net flux  $\sim 0$**

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