

Atmospheric Mercury Measurements and Modeling at the Grand Bay NERR

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<http://www.arl.noaa.gov/mercury.php>

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Acknowledgements

- ❑ Jake Walker, Mark Woodrey, Glen Ruple (*Grand Bay National Estuarine Research Reserve*)
- ❑ Yerramilli Anjaneyulu, Venkatesh Dodla, Jerzy Lesczynski, Hari Dasari, & others (*Jackson State Univ.*)
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- ❑ Glenn Rolph, Barbara Stunder, Ariel Stein, Steve Fine (*NOAA Air Resources Laboratory*)
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- ❑ *U.S. Fish and Wildlife Service -- Grand Bay National Wildlife Refuge*



MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

1. Measurements

- A. Site Location and Setting
- B. Current Suite of Measurements
- C. Wet Deposition (soon)
- D. Intensive (this Summer)
- E. Data – some examples

2. Modeling

- A. Episodes for Model Evaluation
- B. Met Data -- Fine-Scale
- C. Evolution of the HYSPLIT-Hg Model
- D. Emissions Information and Data?

MEASUREMENTS

MODELING

Site

Current

Wet Dep

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Data

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Evolution

Emissions?

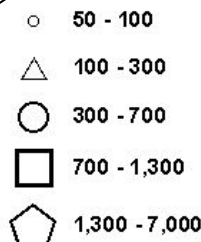
Canaan Valley Institute
[CVI-NOAA]

Allegheny Portage
[CVI-PA-NOAA]

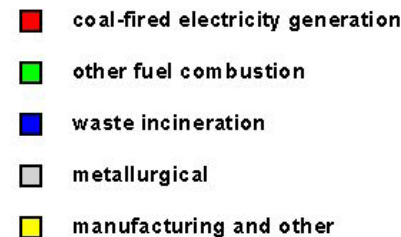
Beltsville
[EPA-NOAA]

Grand Bay NERR
NOAA-MSDEP

Emissions (kg/year)

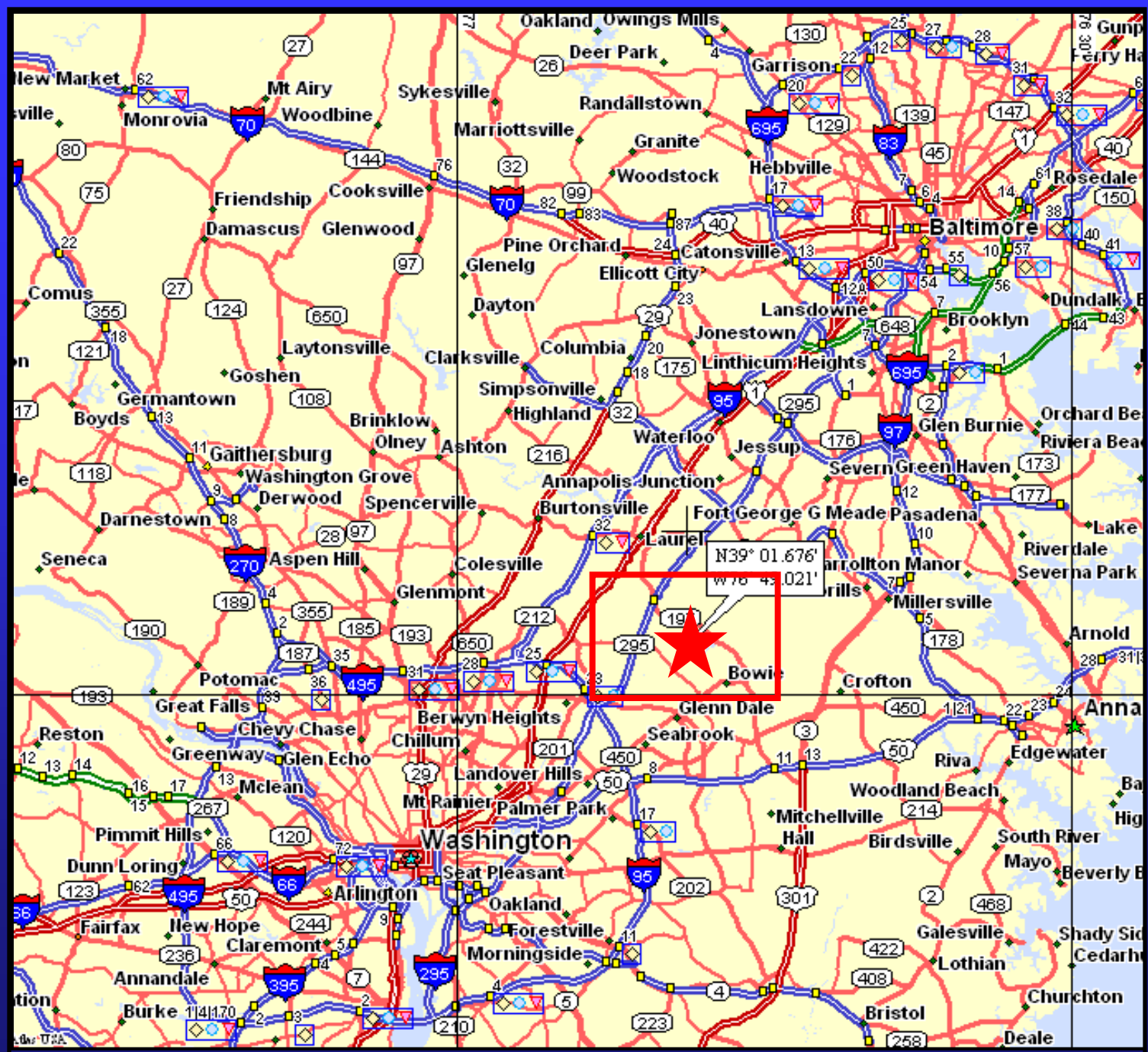


Type of Emissions Source

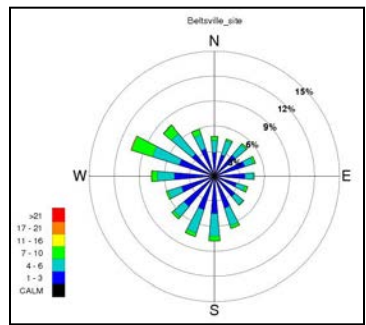


Four NOAA associated sites
committed to emerging inter-
agency speciated mercury ambient
concentration measurement
network

(comparable to Mercury Deposition
Network (MDN) for wet deposition,
but for air concentrations)



Beltsville monitoring site



Brunner Island

Large Incinerators:
3 medical waste,
1 MSW, 1 haz waste
(Total Hg ~ 500 kg/yr)

Harford County MSW Incin

Brandon Shores and H.A. Wagner

Monitoring sites

- rural AQS
- other AQS
- ★ NADP/MDN
- CASTNet
- ▲ Hg site
- ⬠ IMPROVE

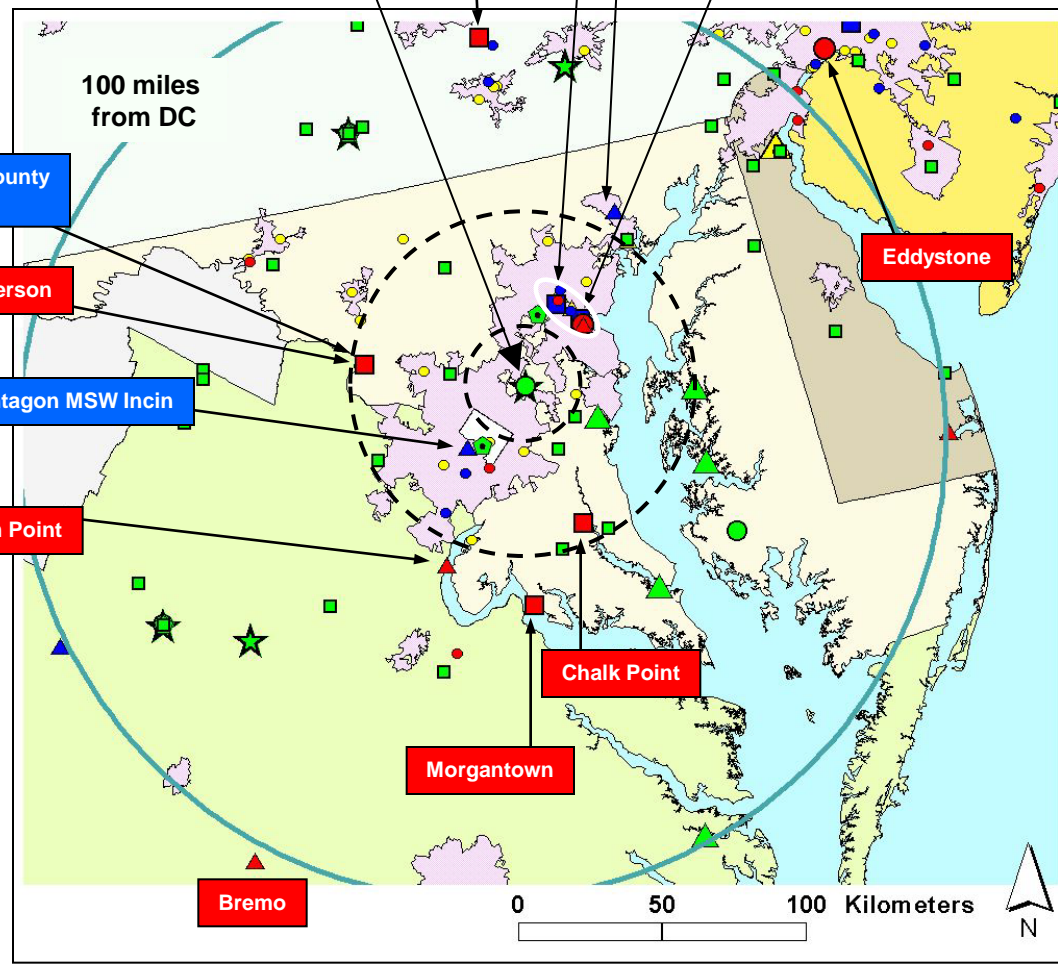
Symbol color indicates type of mercury source

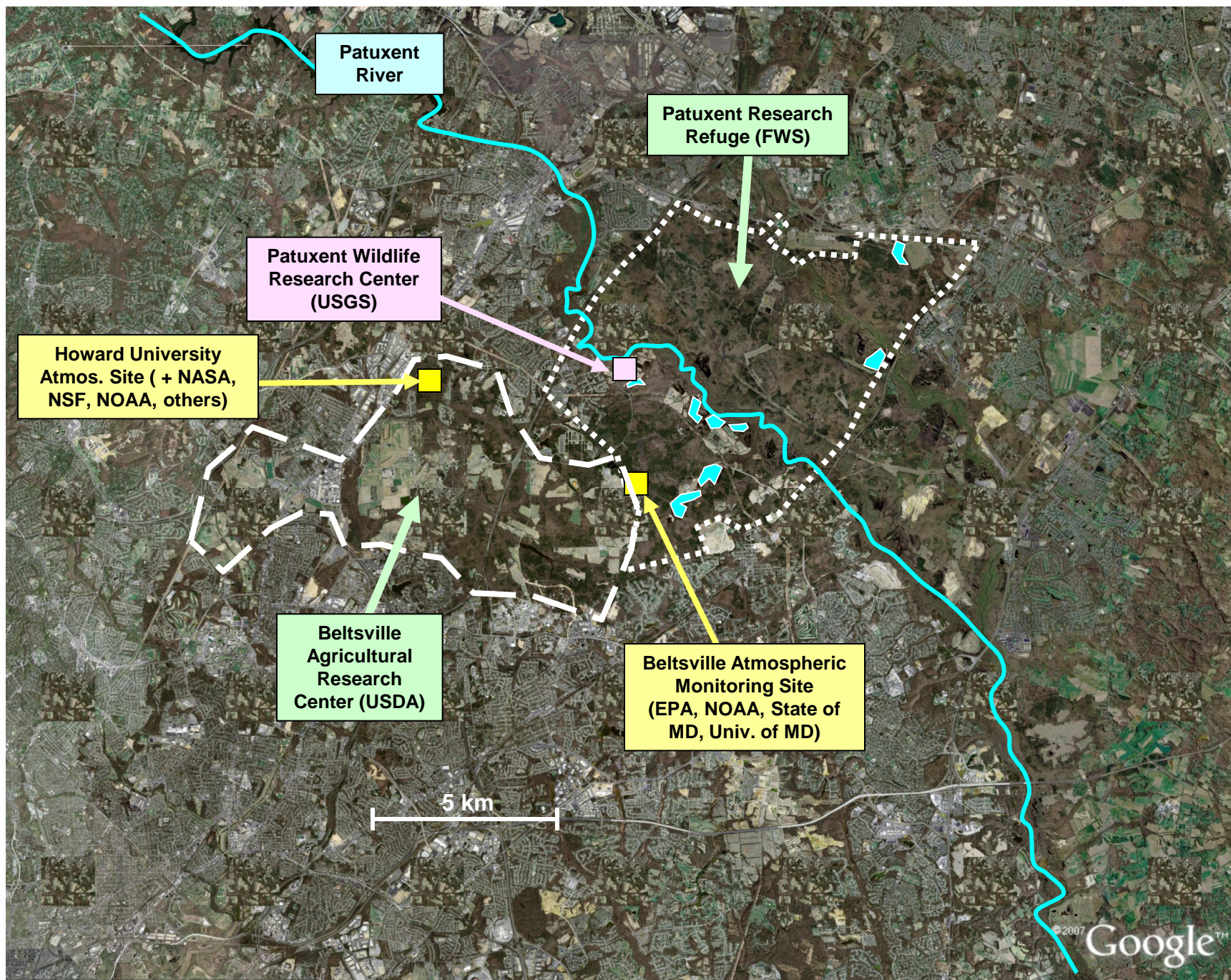
- coal
- incinerator
- metals
- manuf/other

Symbol size and shape indicates 1999 mercury emissions, kg/yr

- 1 - 50
- △ 50 - 100
- 100 - 200
- 200 - 400
- △ 400 - 700
- ⬠ 700 - 1000
- > 1000

the region between the 20 km and 60 km radius circles displayed around the monitoring site might be considered the "ideal" location for sources to be investigated by the site





Atmospheric Mercury Measurement Site at Beltsville, MD



ARL's Winston Luke working with RGM and Hg(p) collectors



Precipitation measurements (left to right):
Mercury Deposition Network,
Major Ions (e.g. "acid rain"),
Precipitation Amount



mercury and trace gas
monitoring tower (10 meters)



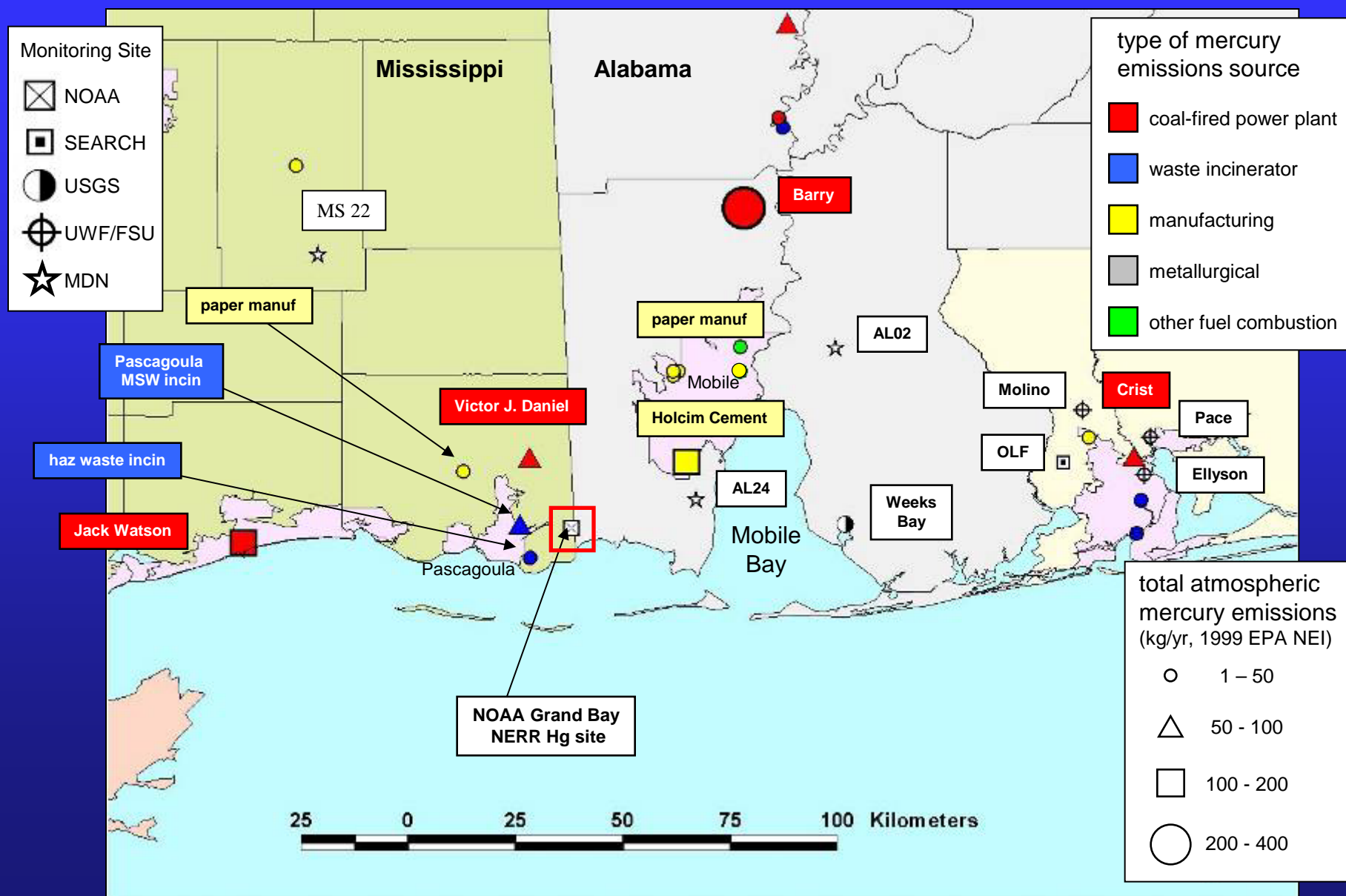
Top of tower (close-up)
with two sets of RGM
and Hg(p) collectors



After RGM and Hg(p) is
collected, it is desorbed
and analyzed inside the
trailer, along with Hg(0)

Atmospheric Mercury Measurements and Modeling at the Grand Bay National Estuarine Research Reserve

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Atmospheric Mercury Measurements and Modeling at the Grand Bay National Estuarine Research Reserve

MEASUREMENTS

MODELING

Site

Current

Wet Dep

Intensive

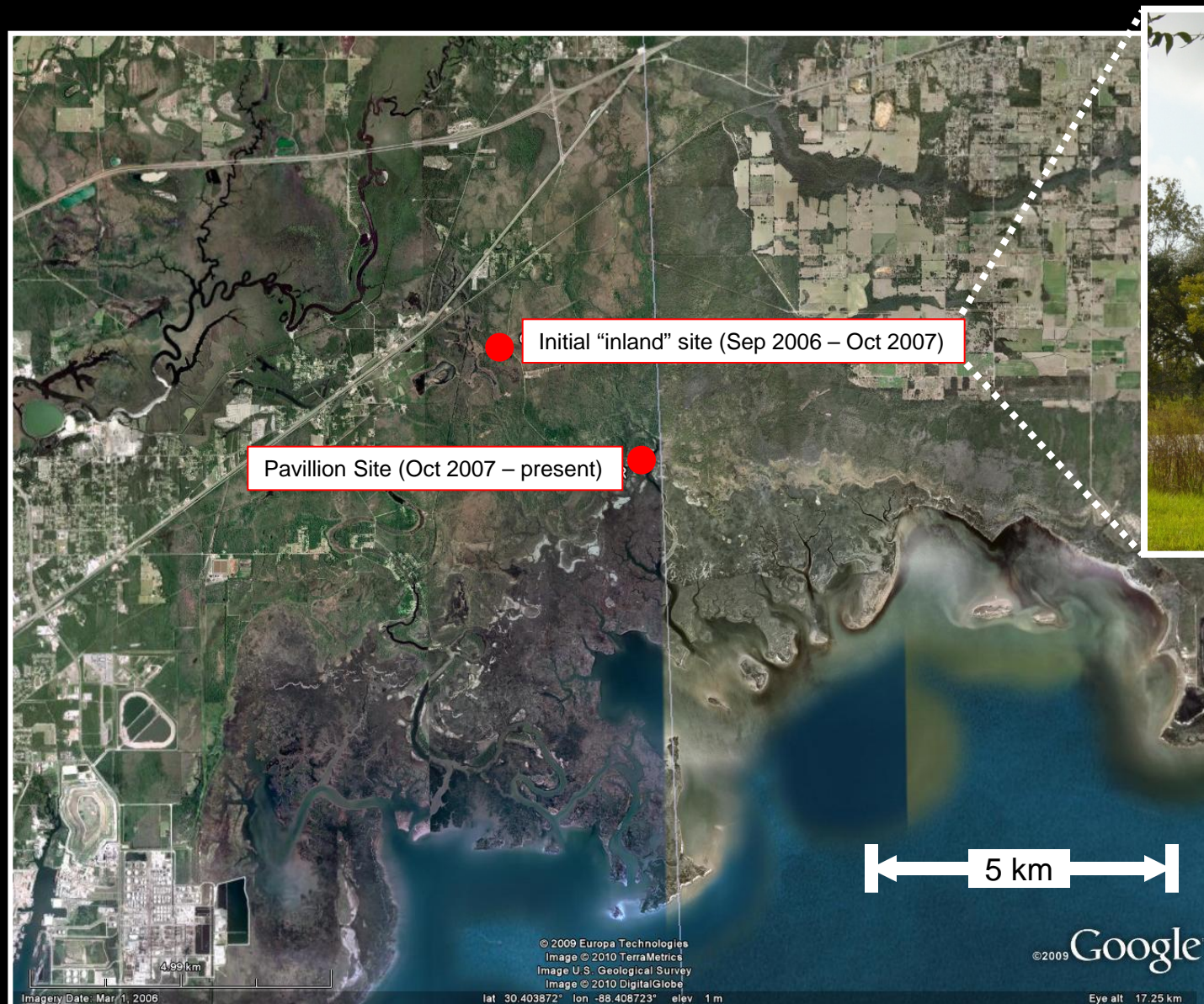
Data

Episodes

Met Data

Evolution

Emissions?



MEASUREMENTS

MODELING

Site

Current

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Intensive

Data

Episodes

Met Data

Evolution

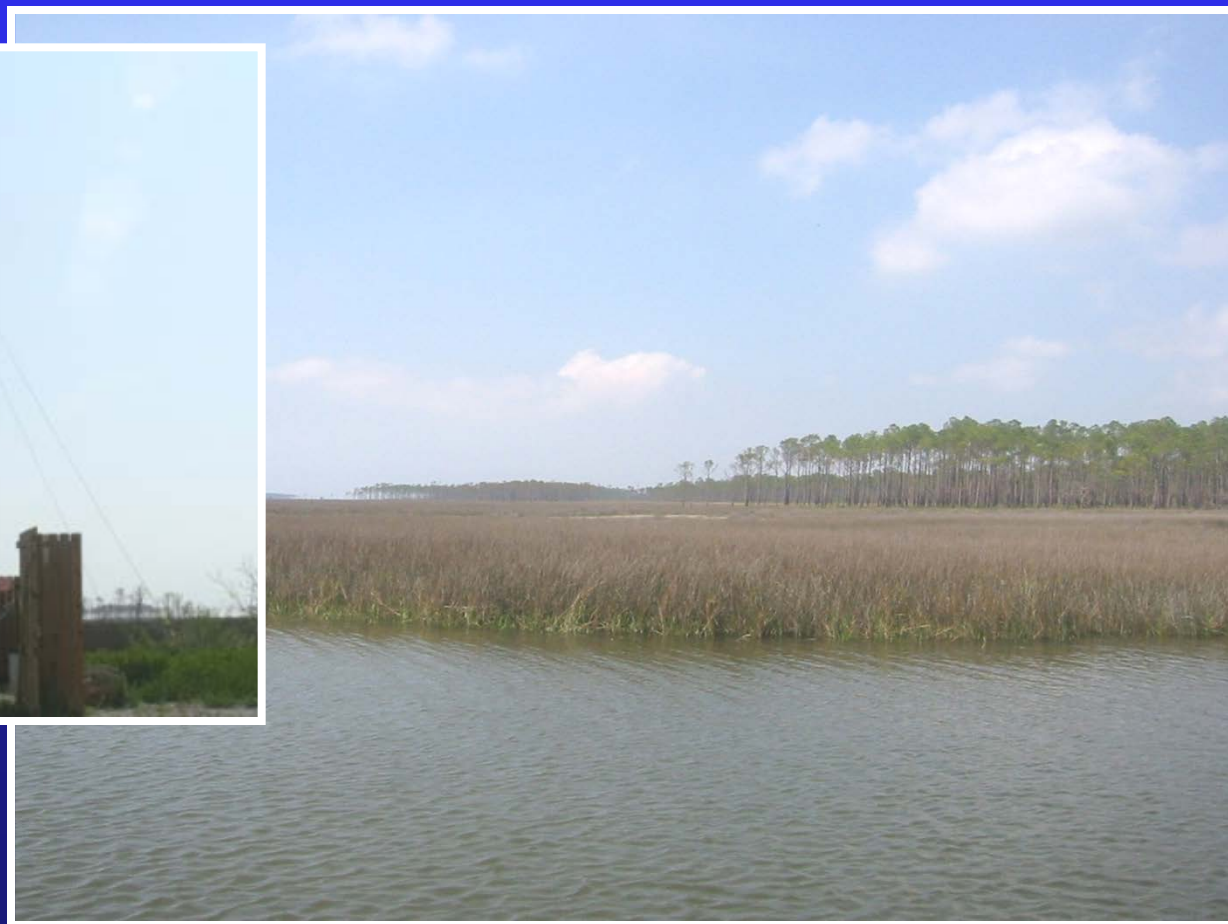
Emissions?

Current Location of Site

view from top of the tower



mercury and trace gas
monitoring tower
(10 meters)



MEASUREMENTS					MODELING			
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“Hmmm...maybe it would work better if we have the tower go vertical?”

Winston Luke (Principal Investigator, NOAA – Air Resources Laboratory) and **Jake Walker** (site operator, Grand Bay National Estuarine Research Reserve)

MEASUREMENTS

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Emissions?



Paul Kelley
NOAA – ARL

instrument engineer,
data management
and acquisition

MEASUREMENTS

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Jake Walker
Grand Bay NERR

Site operator

MEASUREMENTS					MODELING			
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Current Atmospheric Measurements

Elemental mercury (two instruments)	}	“Speciated” Atmospheric Mercury Concentrations
Fine particulate mercury (two instruments)		
Reactive gaseous mercury (two instruments)		
Sulfur dioxide	}	Trace gases and other measurements to help understand and interpret mercury data
Ozone		
Carbon Monoxide		
Nitrogen Oxides (NO, NOy)		
Aerosol Black Carbon	}	Meteorological Data
Wind speed, Wind Direction		
Temperature, Relative Humidity		
Precipitation Amount		

MEASUREMENTS

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**Instrumentation inside the trailer
at the Grand Bay NERR site**



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Sept 2006:
Speciated Hg,
SO₂, O₃, and CO
measurements begin
at “inland” site

2005-2006:
site selection

Aug 21 – Oct 5, 2008:
site shut down due to
threat of hurricanes

Summer 2010:
Field Intensive
(being planned)

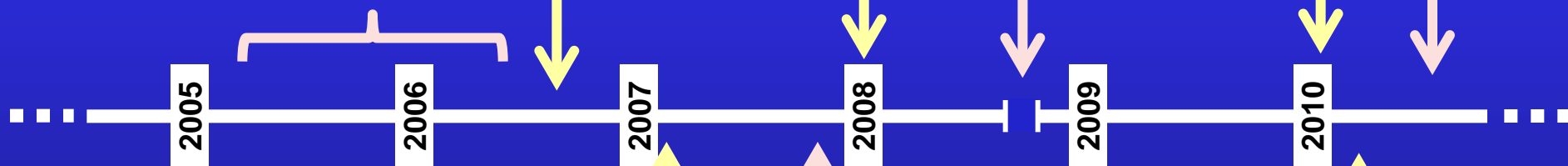
Jan 2008:
NO/NO_y
added

Jan 2010:
Black
Carbon
added

Feb 2007: Meteorological
measurements added

Oct 2007:
Move to “coastal”
site near Pavillion;
2nd Tekran speciated Hg
measurement suite added

2010: Wet
Deposition
measurements
being added



MEASUREMENTS					MODELING			
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**Wet Deposition Measurements Being Added in 2010
by the Mississippi Department of Environmental Protection
(Henry Folmar, Becky Comyns, others), with funding from the EPA**

Precipitation

Continuous digital measurement of
precipitation amount

Major Ions

pH, SO_4^{-2} , NO_3^- , PO_4^{-3} , Cl^- ,
 NH_4^+ , Ca^{+2} , Mg^{+2} , K^+ , Na^+

Weekly measurements of concentrations
in precipitation (NADP-NTN)

Total Mercury

Weekly measurements of concentration
in precipitation (NADP-MDN)

Methyl Mercury

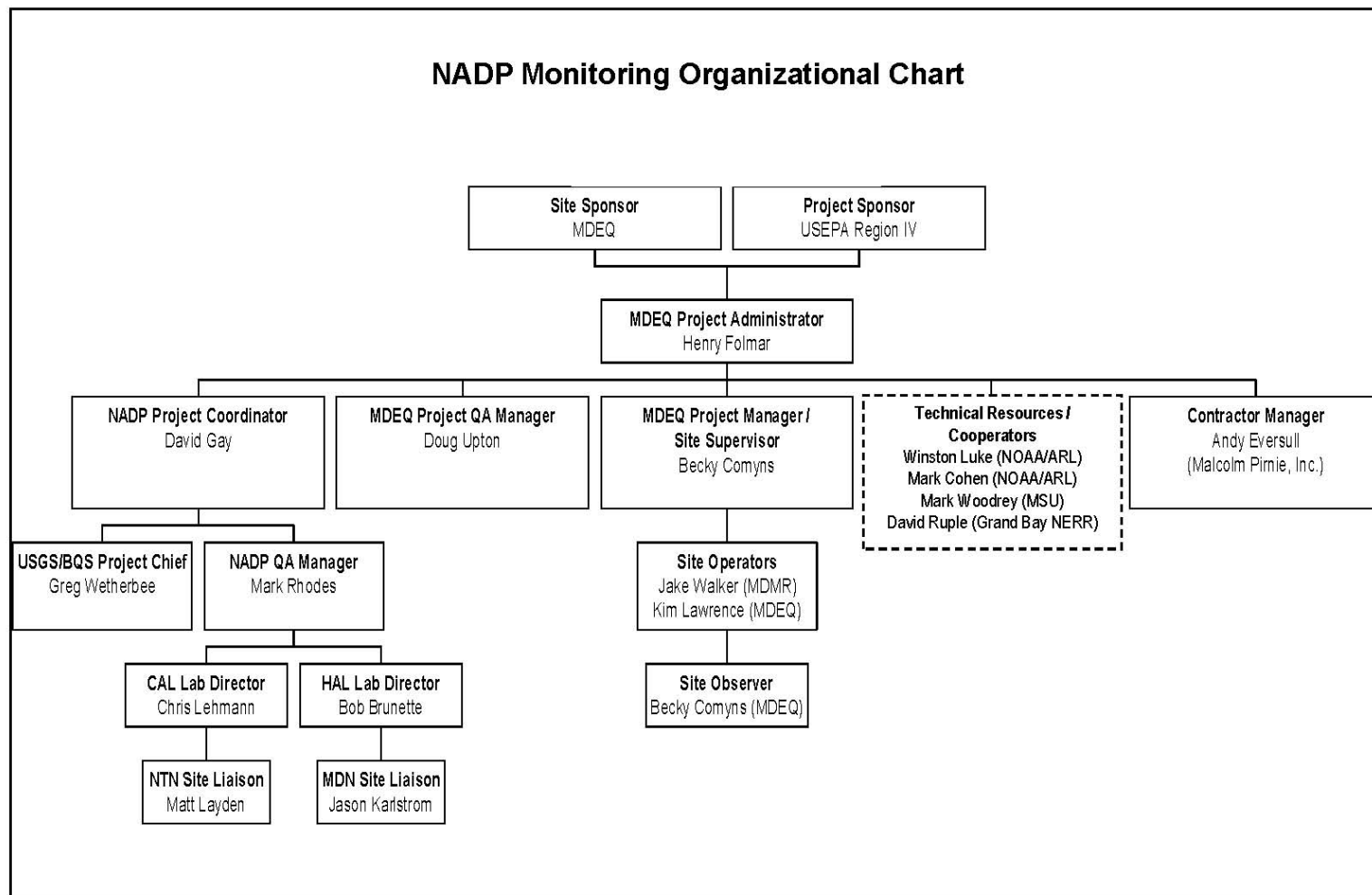
Monthly measurements of concentration
in precipitation (composite)

Selected Trace Metals

As, Cd, Cr, Cu, Pb, Ni, Se, Zn

Weekly measurements of concentrations
in precipitation

MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?



Organizational Chart
NADP Monitoring QAPP
Mississippi Department of Environmental Quality

July 2009


Figure 1

MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?



Approximate location
of the three wet
deposition collectors

Photo courtesy of MDEQ

	Location of Sample Collectors at Grand Bay, MS NADP Monitoring QAPP Mississippi Department of Environmental Quality	July 2009
		Figure 3

MEASUREMENTS

MODELING

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Evolution

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*For the record,
when we first
picked the
Grand Bay
NERR site, there
were no large
houses nearby*

MEASUREMENTS

MODELING

Site

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Data

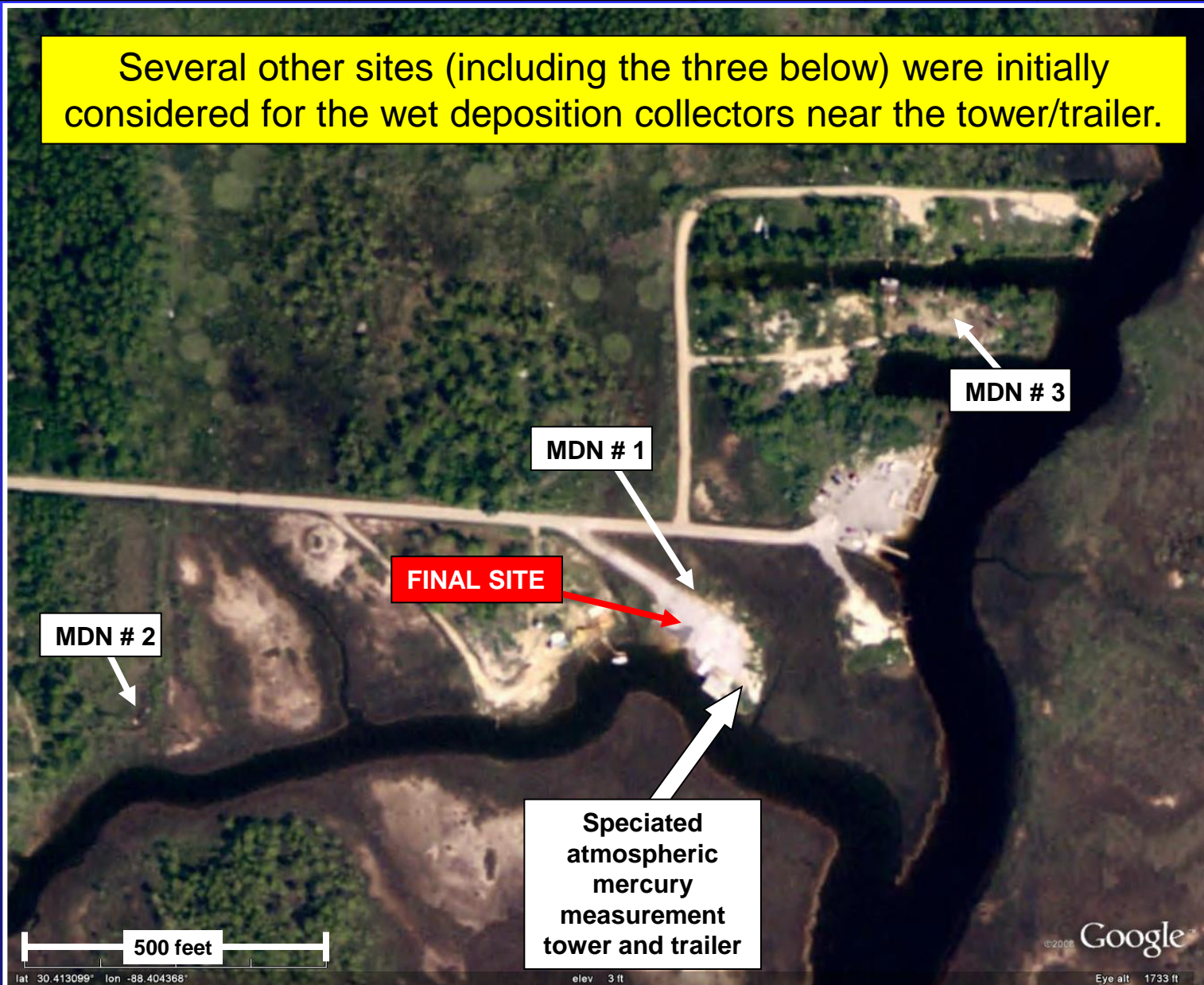
Episodes

Met Data

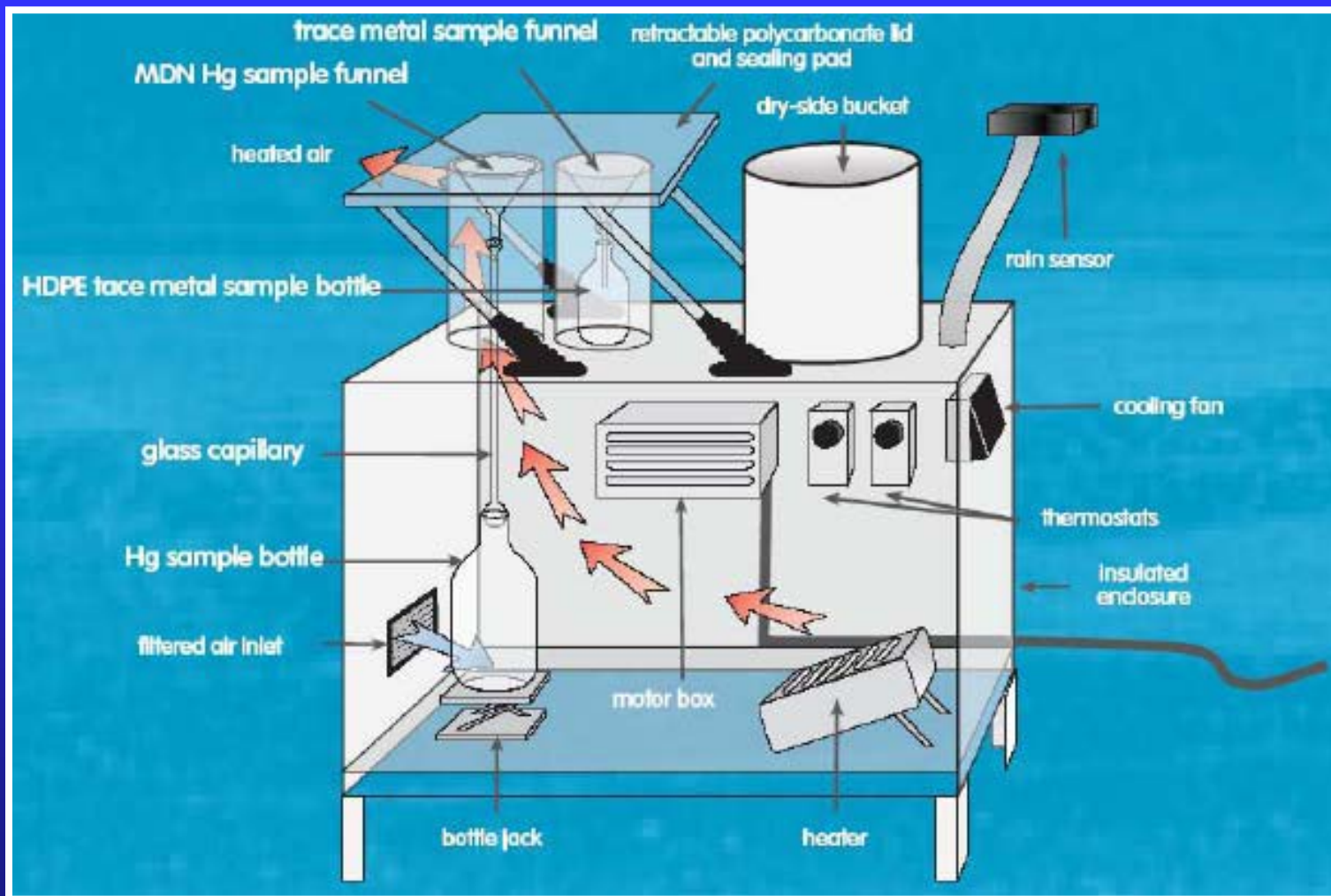
Evolution

Emissions?

Several other sites (including the three below) were initially considered for the wet deposition collectors near the tower/trailer.



MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?



Schematic of the modified Aerochem Metrics wet/dry precipitation collector used for MDN monitoring of mercury, methyl mercury, and trace metals. Photo courtesy of the *New Initiative for the MDN (NADP, 2004)*.

MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

Details are still being worked out...

Emerging Plans for Atmospheric Field Intensive Scheduled for July-Aug 2010

Ground-Based Measurements

(ongoing) mercury, trace gas, black carbon, meteorological measurements	<ul style="list-style-type: none"> • Winston Luke and Paul Kelley (NOAA ARL), • Jake Walker (Grand Bay NERR)
(ongoing) wet deposition: major ions, mercury, methylmercury, trace metals <i>Will try to switch to event-based during intensive</i>	<ul style="list-style-type: none"> • Mississippi State Dept of Env Protection/EPA • Jake Walker (Grand Bay NERR)
ambient concentrations of BrO at the surface via Chemical Ionization Mass Spectrometry (possibly other Br compounds, e.g., Br ₂ , BrCl, and HOBr)	<ul style="list-style-type: none"> • Greg Huey (Georgia Tech)
isotopic mercury analysis of event-based precipitation and aerosols	<ul style="list-style-type: none"> • Bill Landing, Flip Froelich (Florida State Univ)
trace metal analysis of size-segregated aerosol <i>Spring 2010 and possibly during intensive</i>	<ul style="list-style-type: none"> • Mark Engle (USGS)

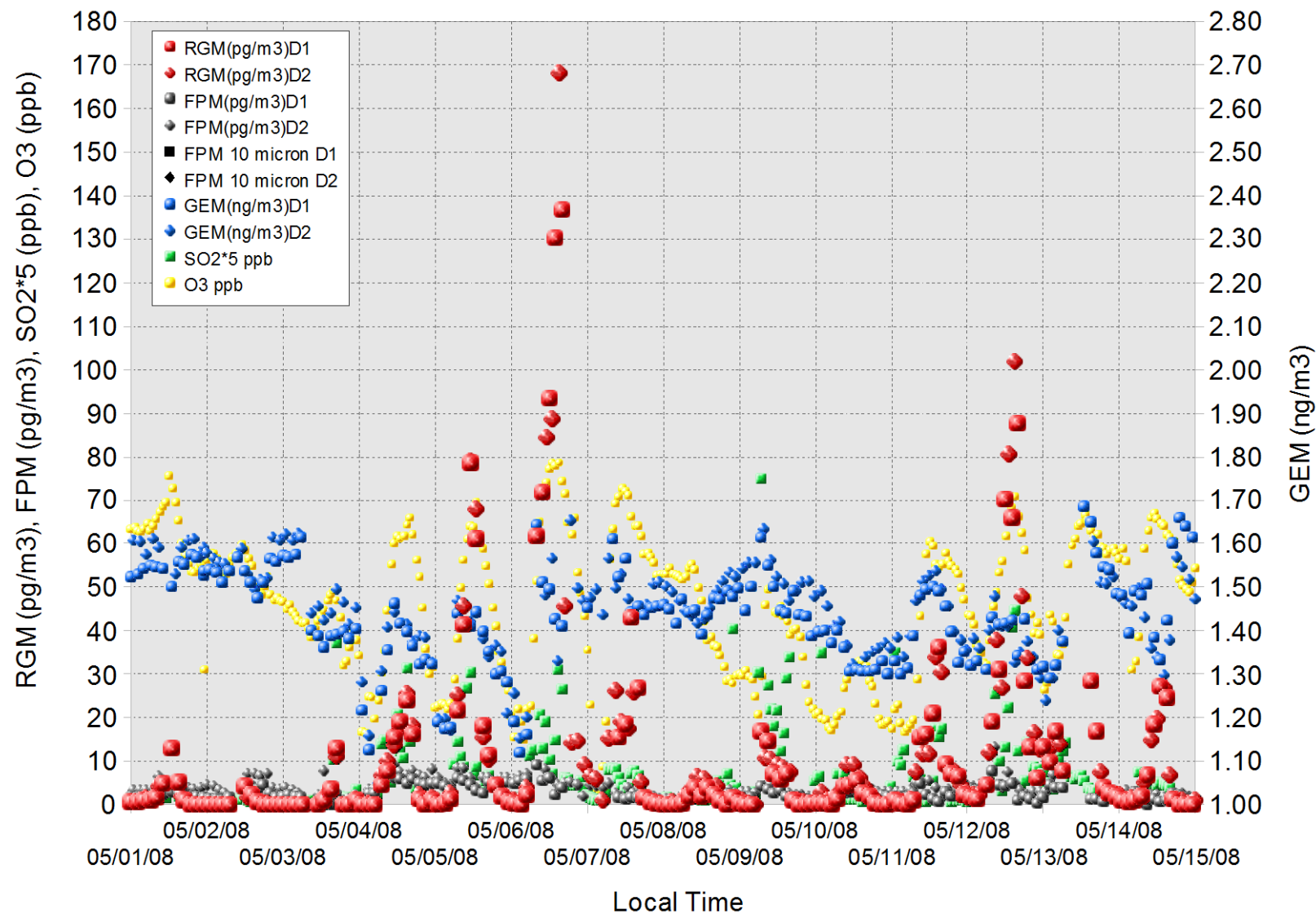
Aircraft and Above Surface Measurements

aircraft flights measuring concentrations of Hg ⁰ (Tekran), total and “speciated” RGM (coated/uncoated denuders), O ₃ , SO ₂ , and particle count	<ul style="list-style-type: none"> • Stephen Corda, John Muratore, & colleagues (Univ. of Tennessee Space Institute – UTSI) • Hynes and Swartzendruber (Univ of Miami) • Luke and Kelley (NOAA ARL)
vertical distribution of O ₃ and met data above the site (ozonesondes)	<ul style="list-style-type: none"> • Luke and Kelley (NOAA ARL) • Jake Walker (Grand Bay NERR)

MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

Speciated Atmospheric Mercury and Selected Trace Gas Concentration Measurements at Grand Bay NERR

Courtesy of Winston Luke and Paul Kelley (NOAA ARL) and Jake Walker (Grand Bay NERR) (Preliminary Values)



MEASUREMENTS

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Atmospheric Mercury Initiative

<http://nadp.sws.uiuc.edu/AMN/>

Alert, Nunavut

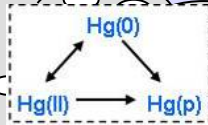
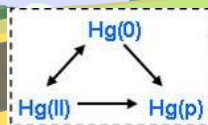
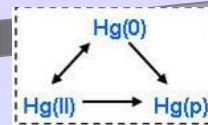
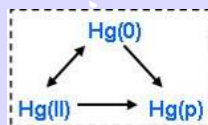
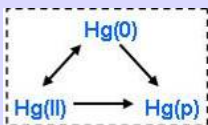
Atmospheric Mercury Sites

- ★ Current Network Sites
- Interested Sites
- Other Atm. Mercury Sites
- CAMNET

Four NOAA-associated sites committed to emerging inter-agency speciated mercury ambient concentration measurement network

(comparable to Mercury Deposition Network (MDN) for wet deposition, but for air concentrations)

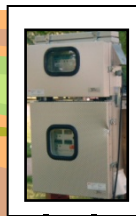
**Hg from
other sources:
local, regional
& more distant**



**Measurement
of wet
deposition**



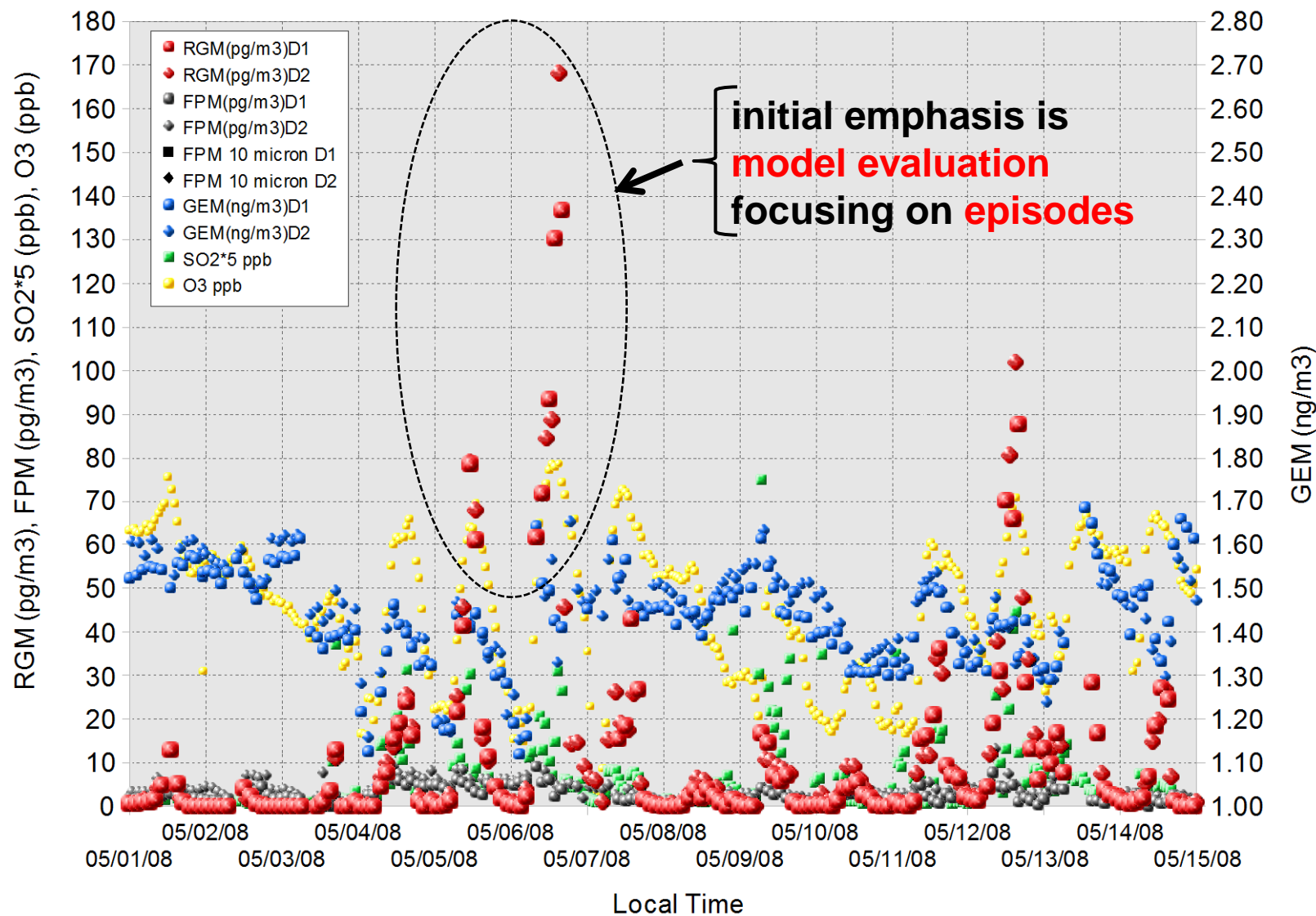
**Measurement
of ambient air
concentrations**



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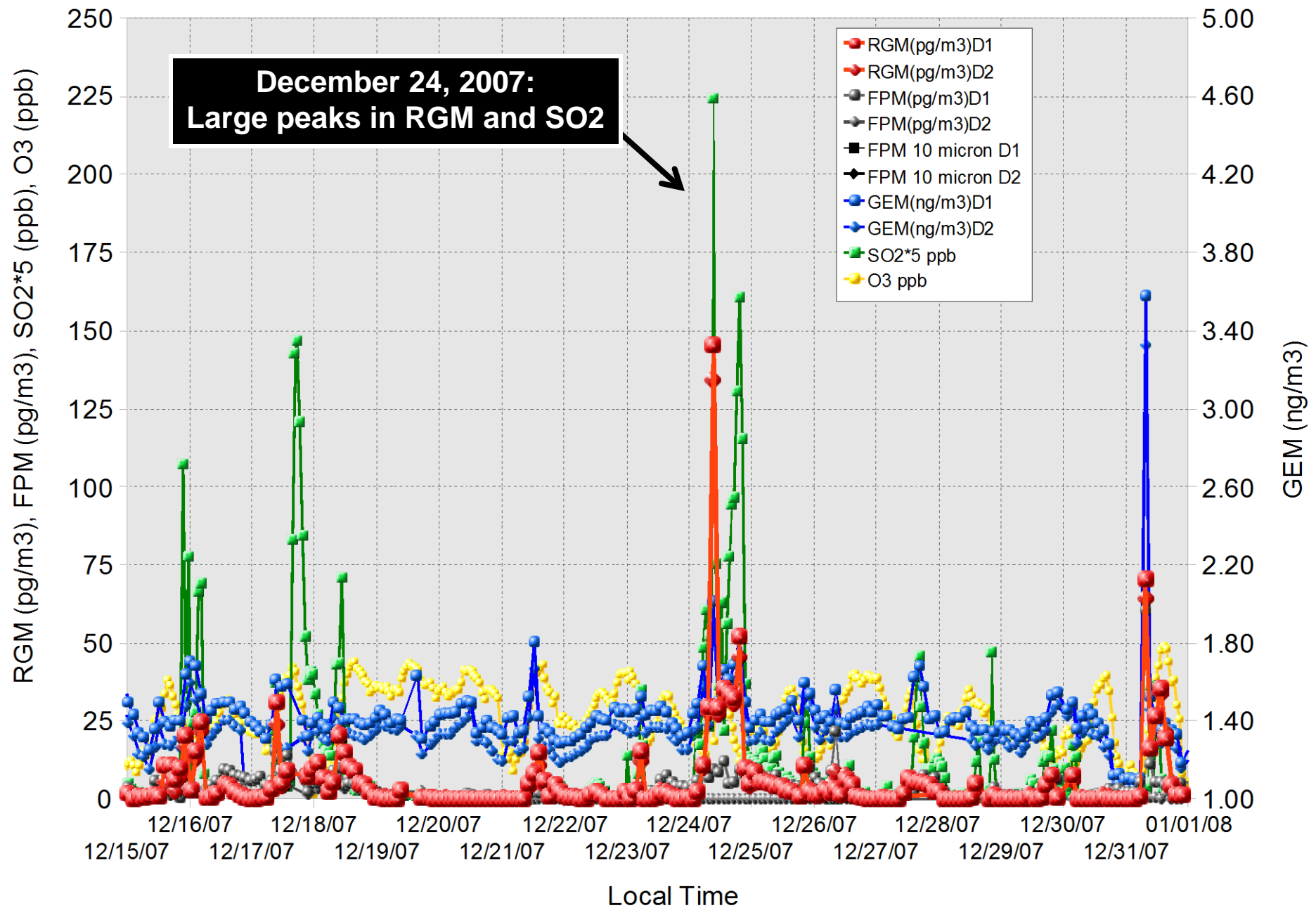
Speciated Atmospheric Mercury and Selected Trace Gas Concentration Measurements at Grand Bay NERR

Courtesy of Winston Luke and Paul Kelley (NOAA ARL) and Jake Walker (Grand Bay NERR) (Preliminary Values)



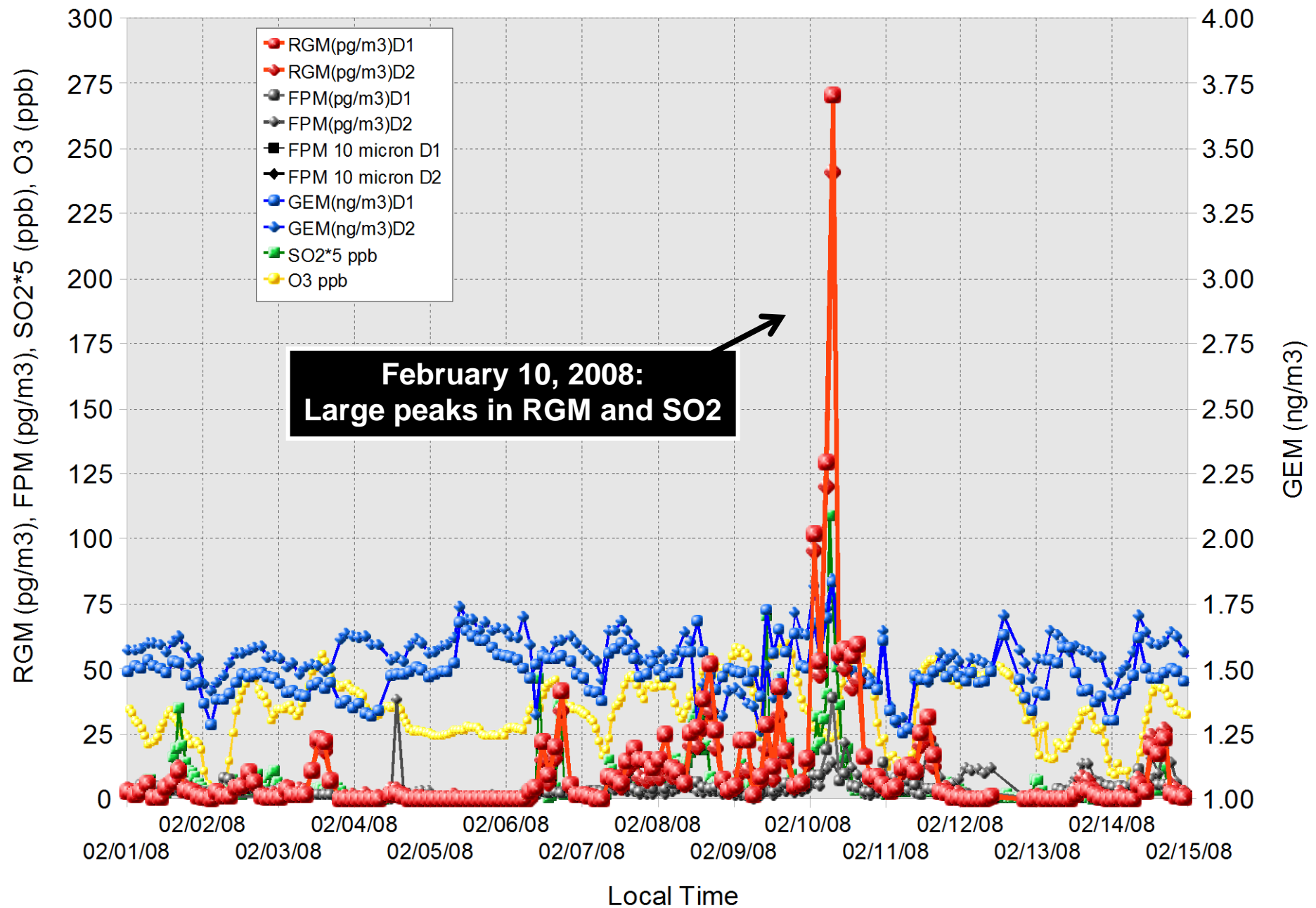
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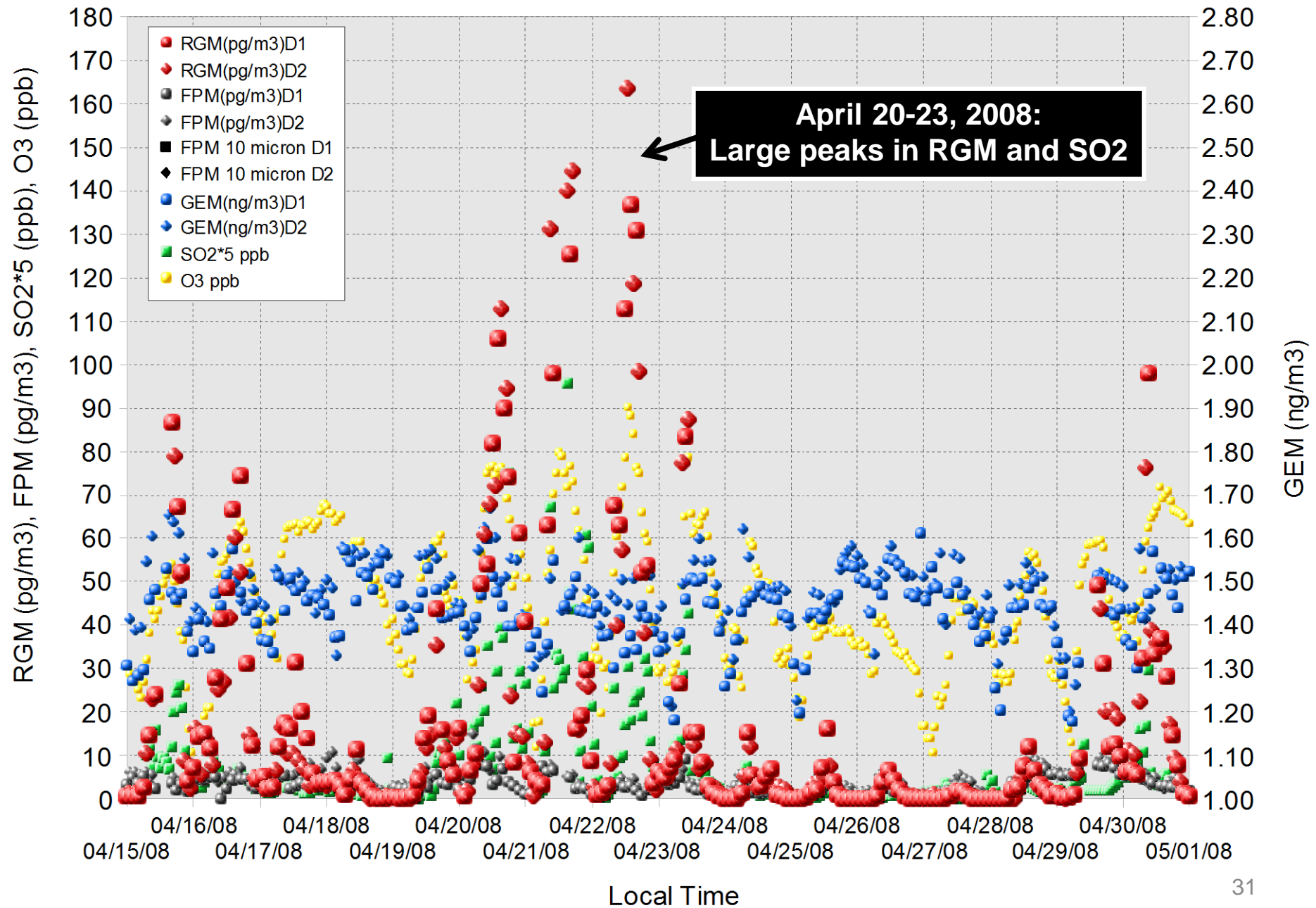
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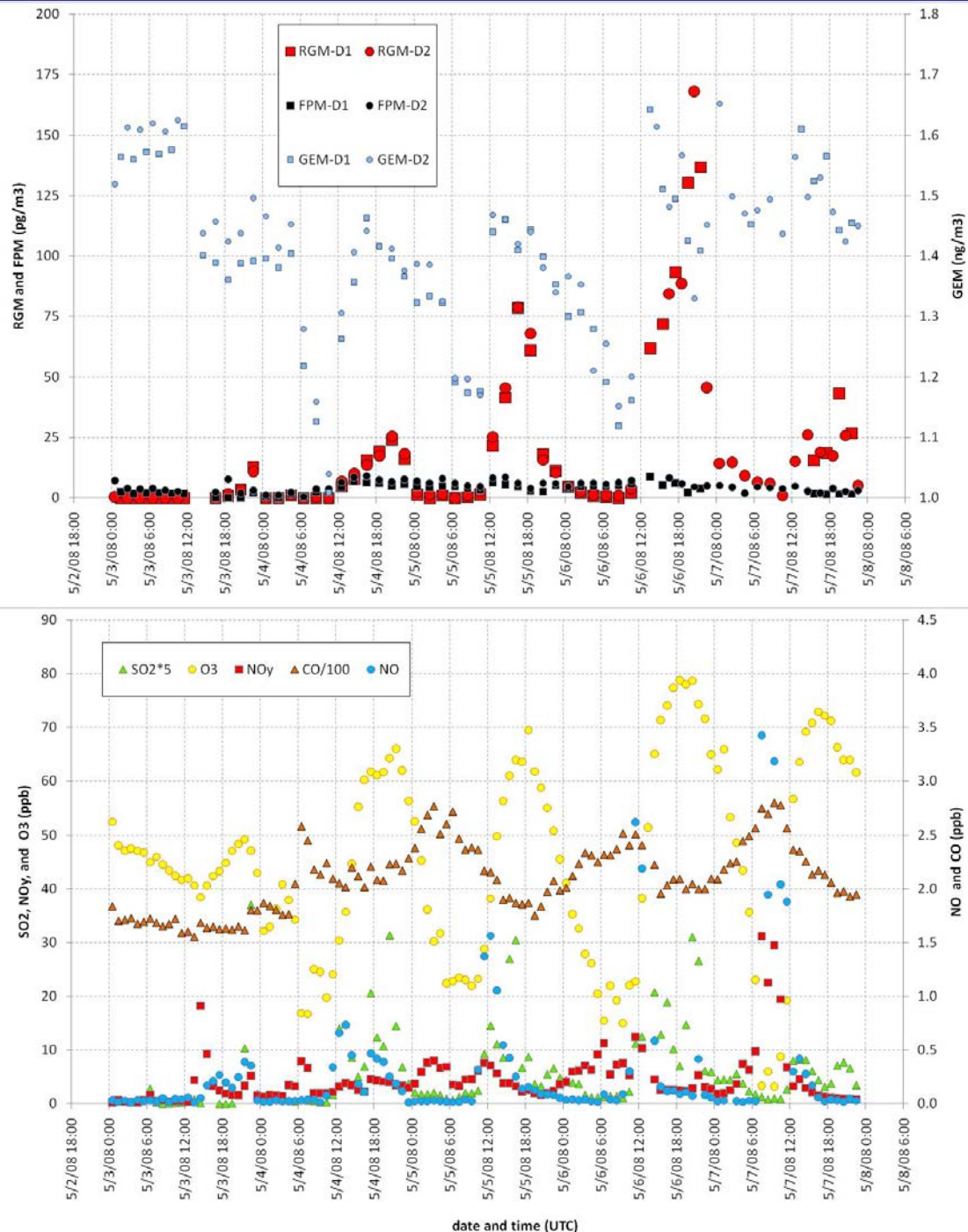


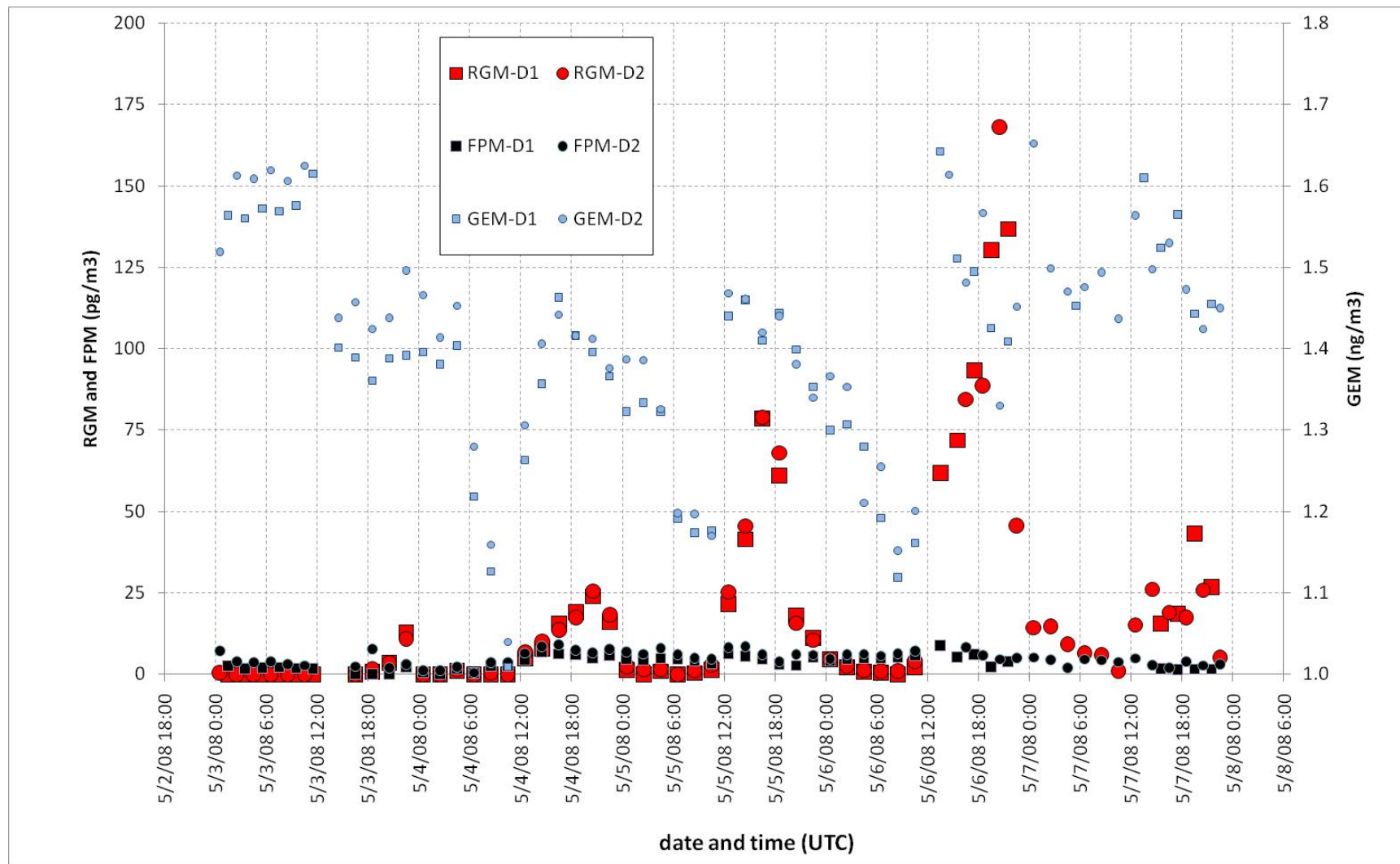
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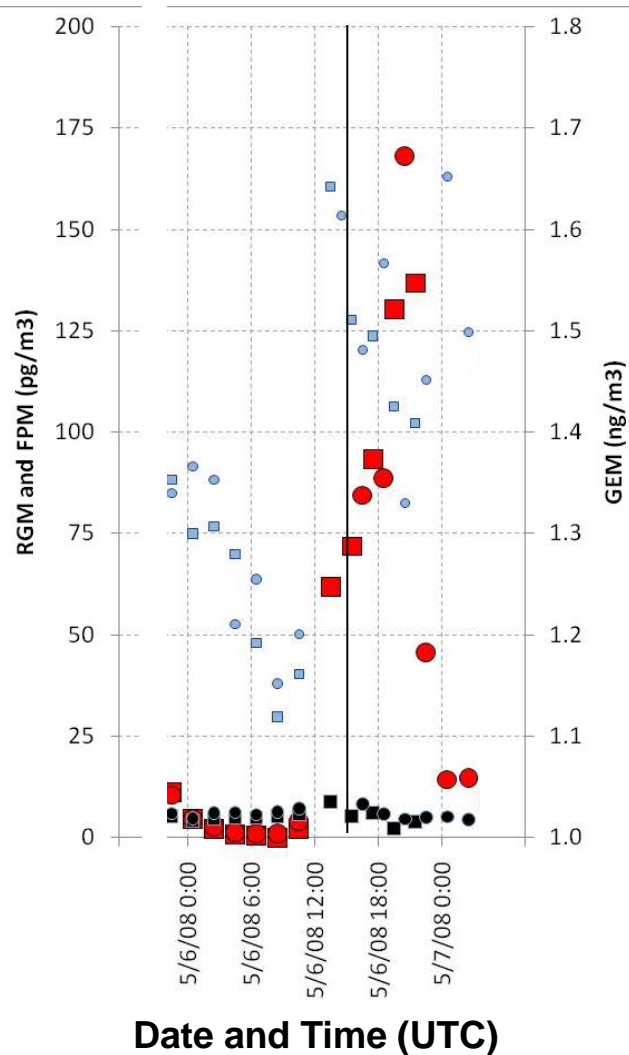
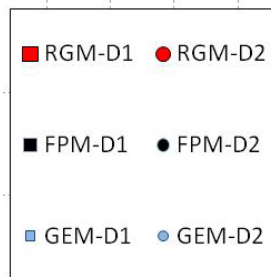


Time series of Reactive Gaseous Mercury (RGM), Fine Particulate Mercury (FPM) and Gaseous Elemental Mercury (GEM) from two co-located instruments (D1 and D2) (top graph) and of SO₂, O₃, NO, NO_y, and CO (bottom graph) measured at the Grand Bay NERR from May 3-8, 2008

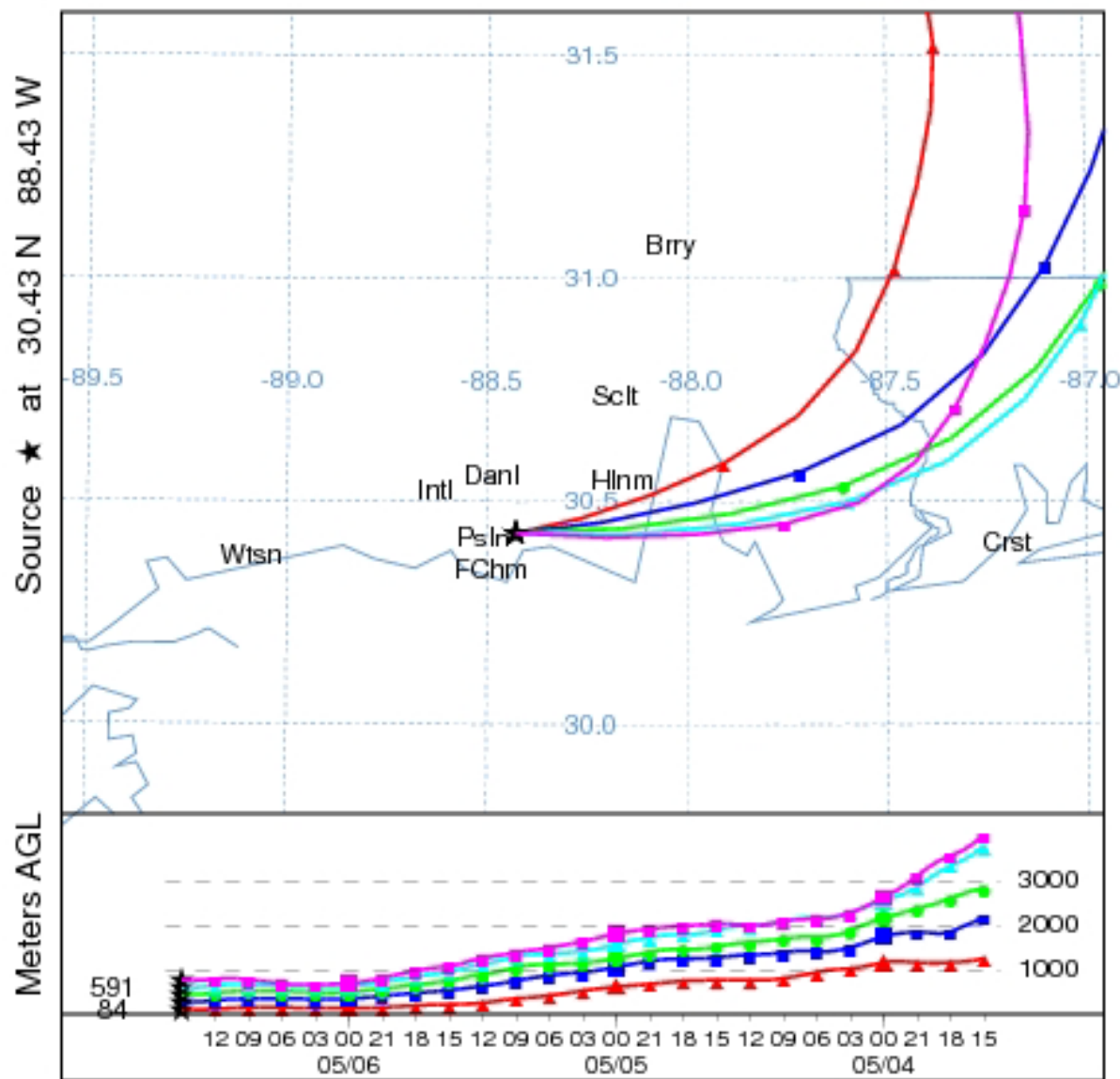


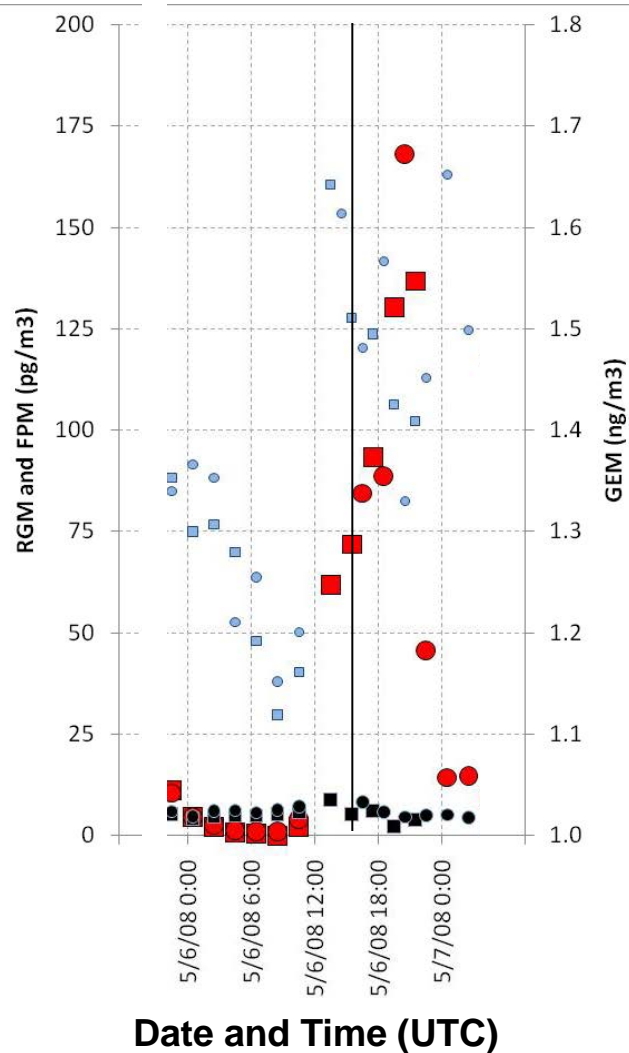
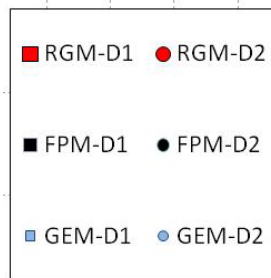


Time series of Reactive Gaseous Mercury (RGM), Fine Particulate Mercury (FPM) and Gaseous Elemental Mercury (GEM) from two co-located instruments (D1 and D2) at the Grand Bay NERR from May 3-8, 2008

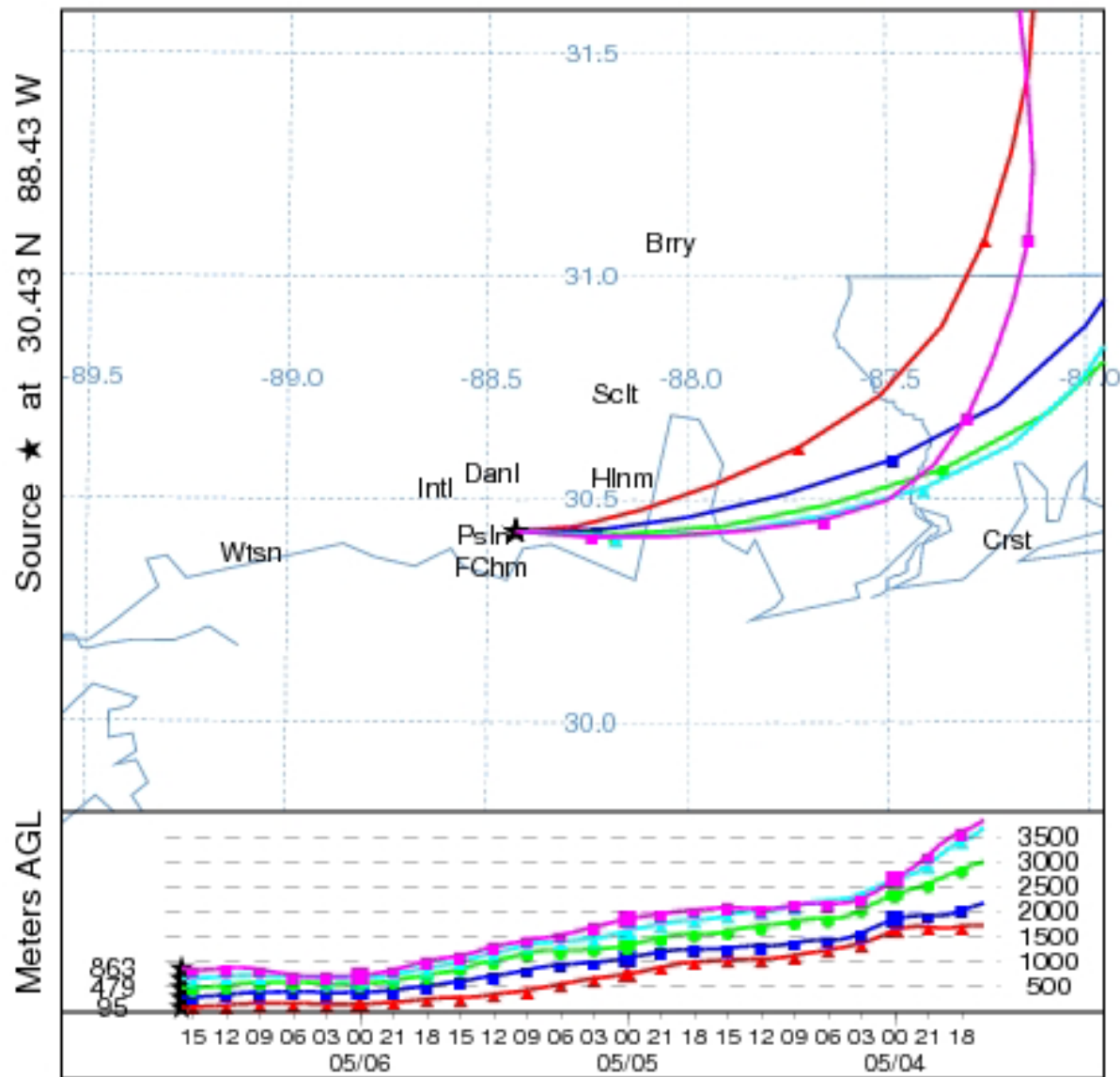


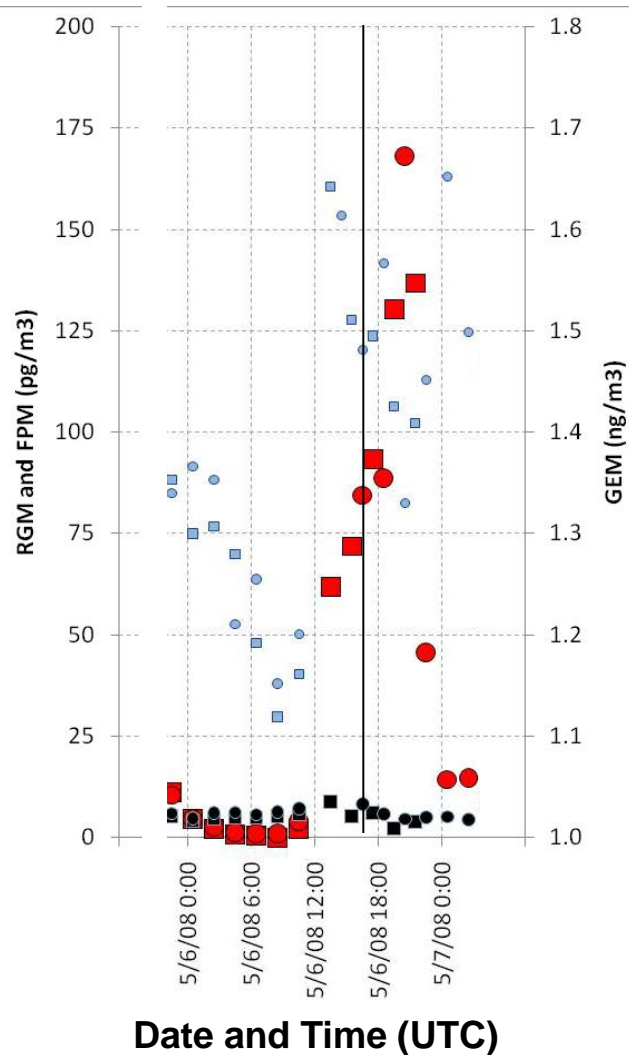
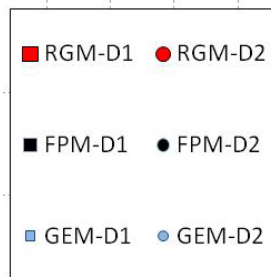
NOAA HYSPLIT MODEL Backward trajectories ending at 15 UTC 06 May 08 NAM Meteorological Data



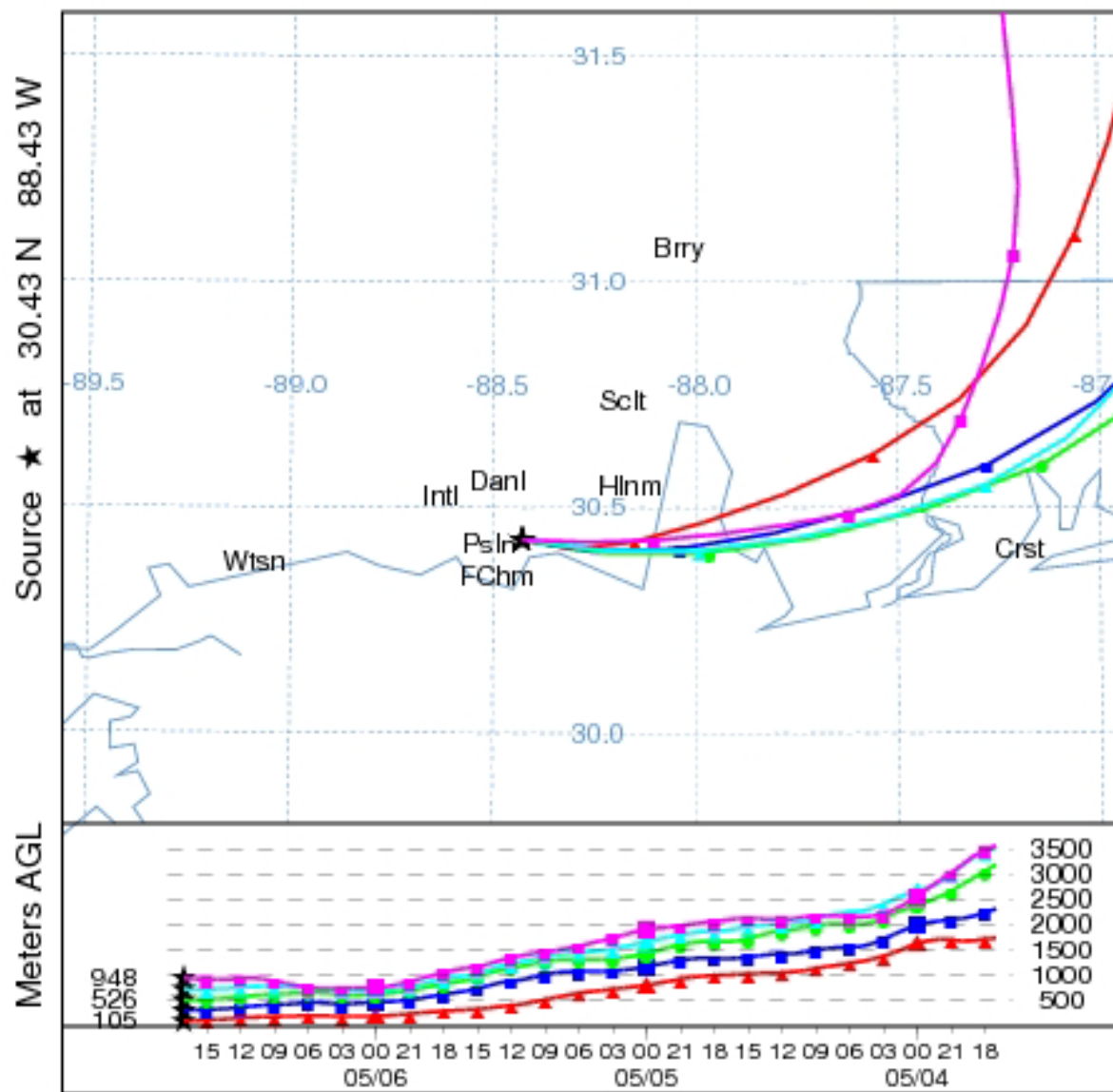


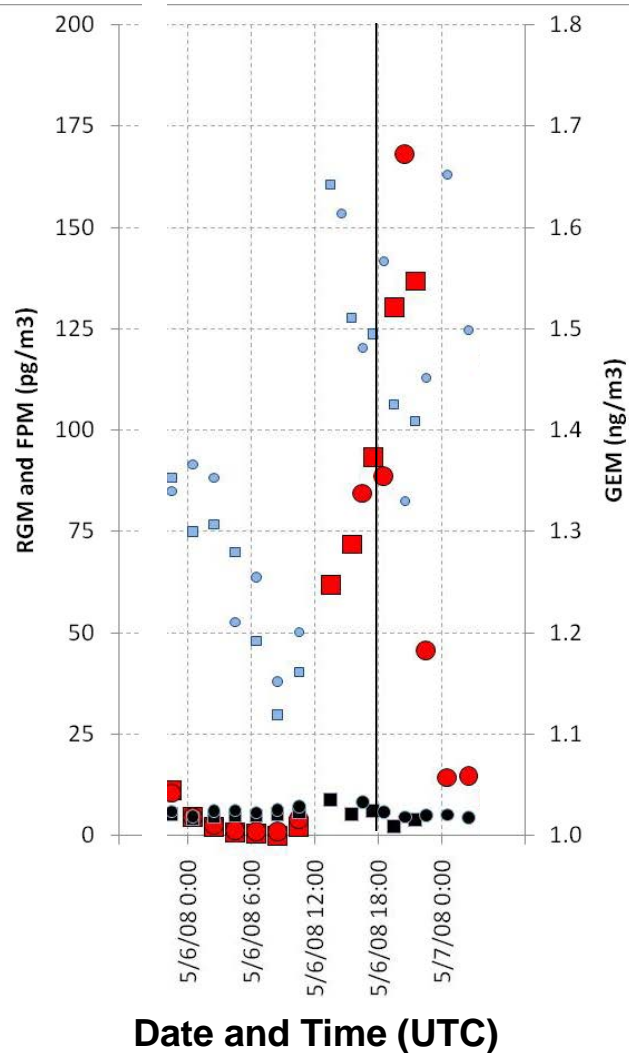
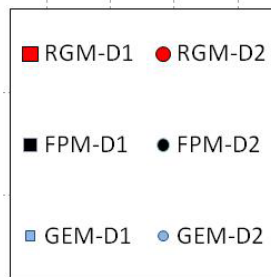
NOAA HYSPLIT MODEL Backward trajectories ending at 16 UTC 06 May 08 NAM Meteorological Data



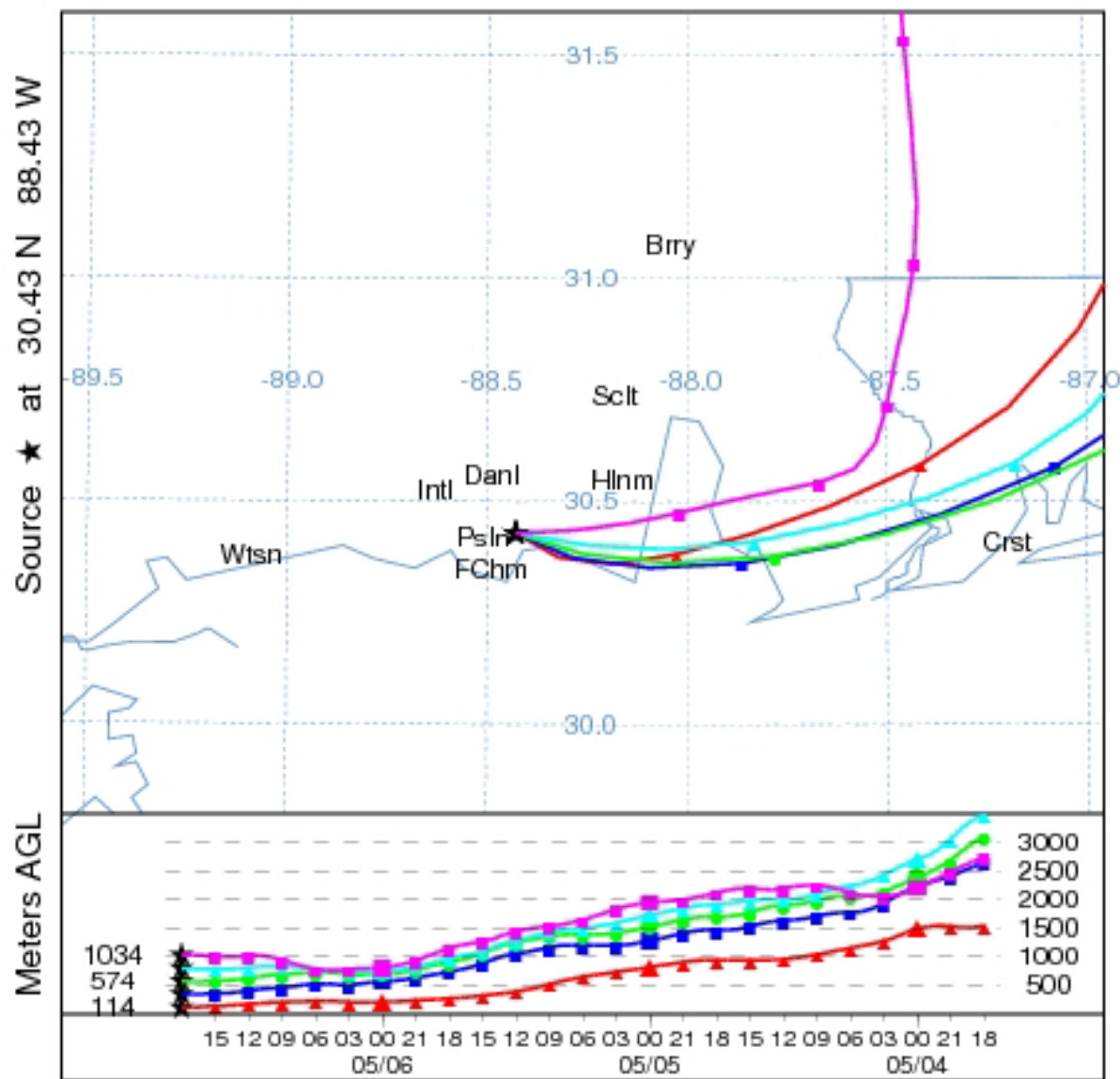


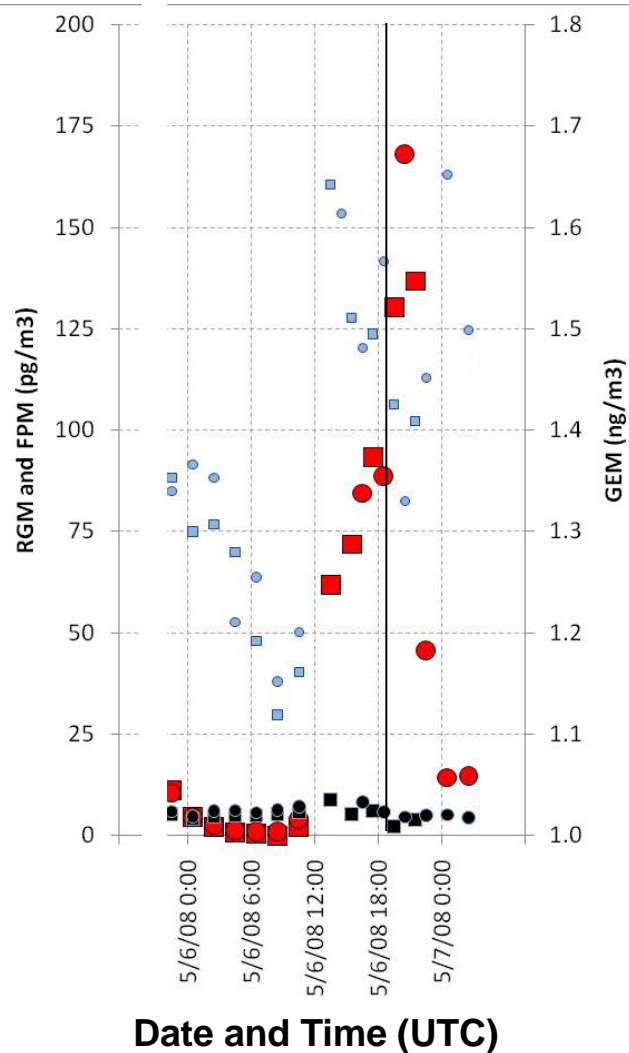
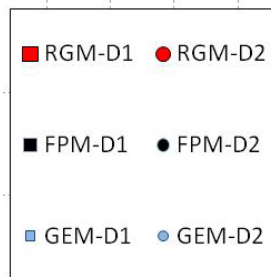
NOAA HYSPLIT MODEL Backward trajectories ending at 17 UTC 06 May 08 NAM Meteorological Data



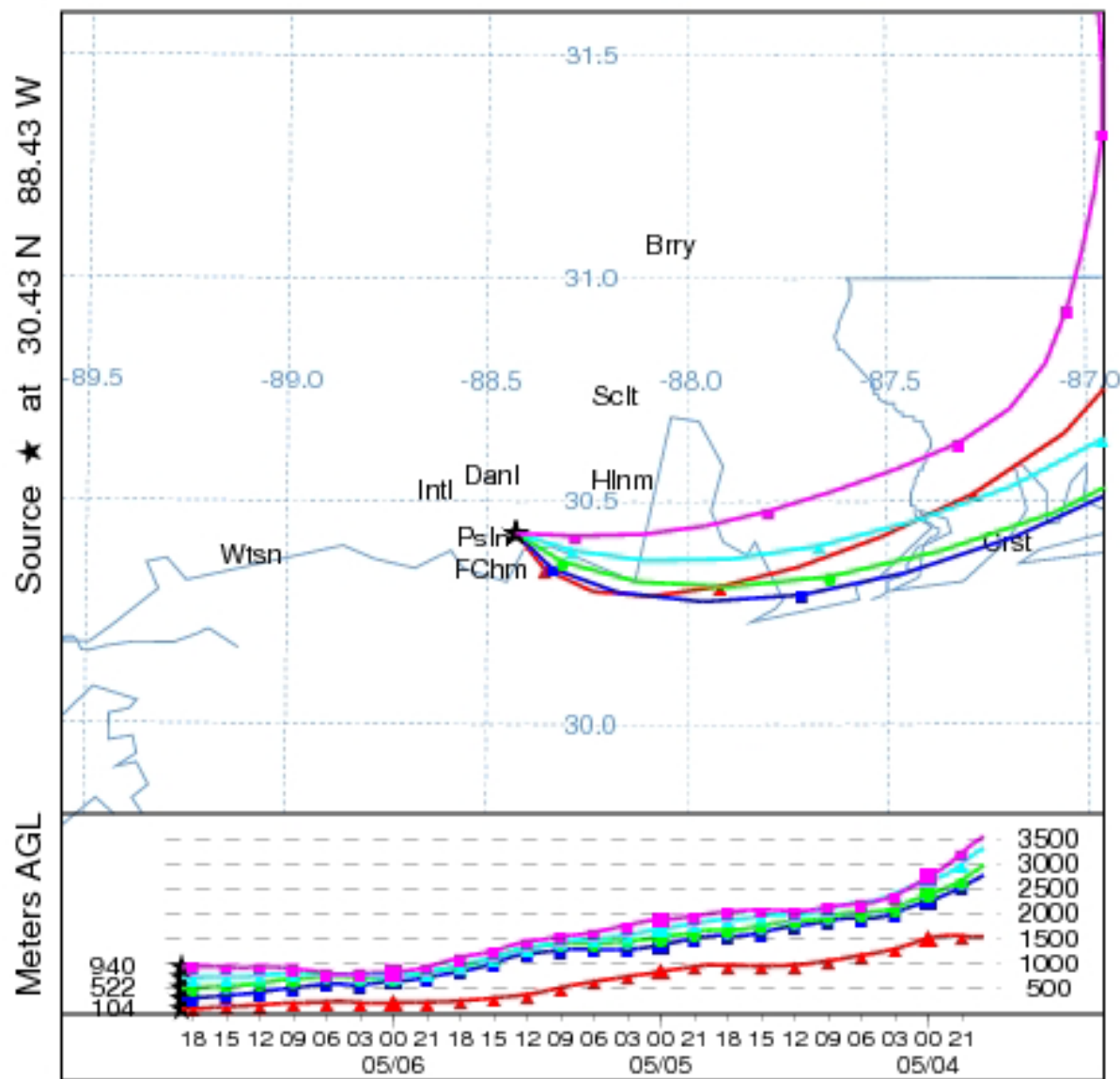


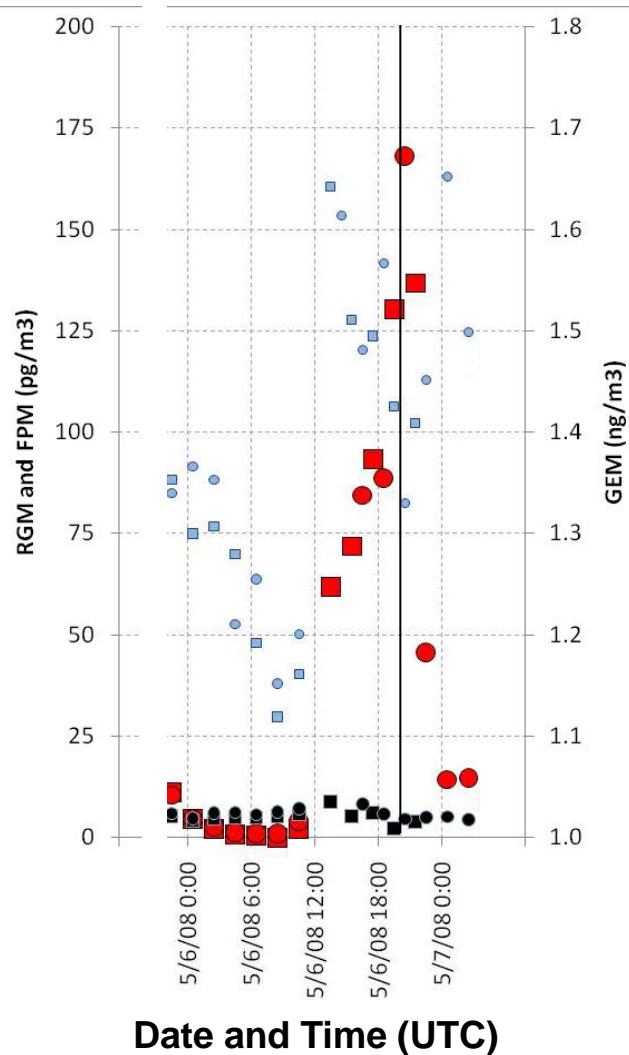
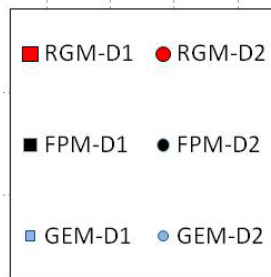
NOAA HYSPLIT MODEL Backward trajectories ending at 18 UTC 06 May 08 NAM Meteorological Data



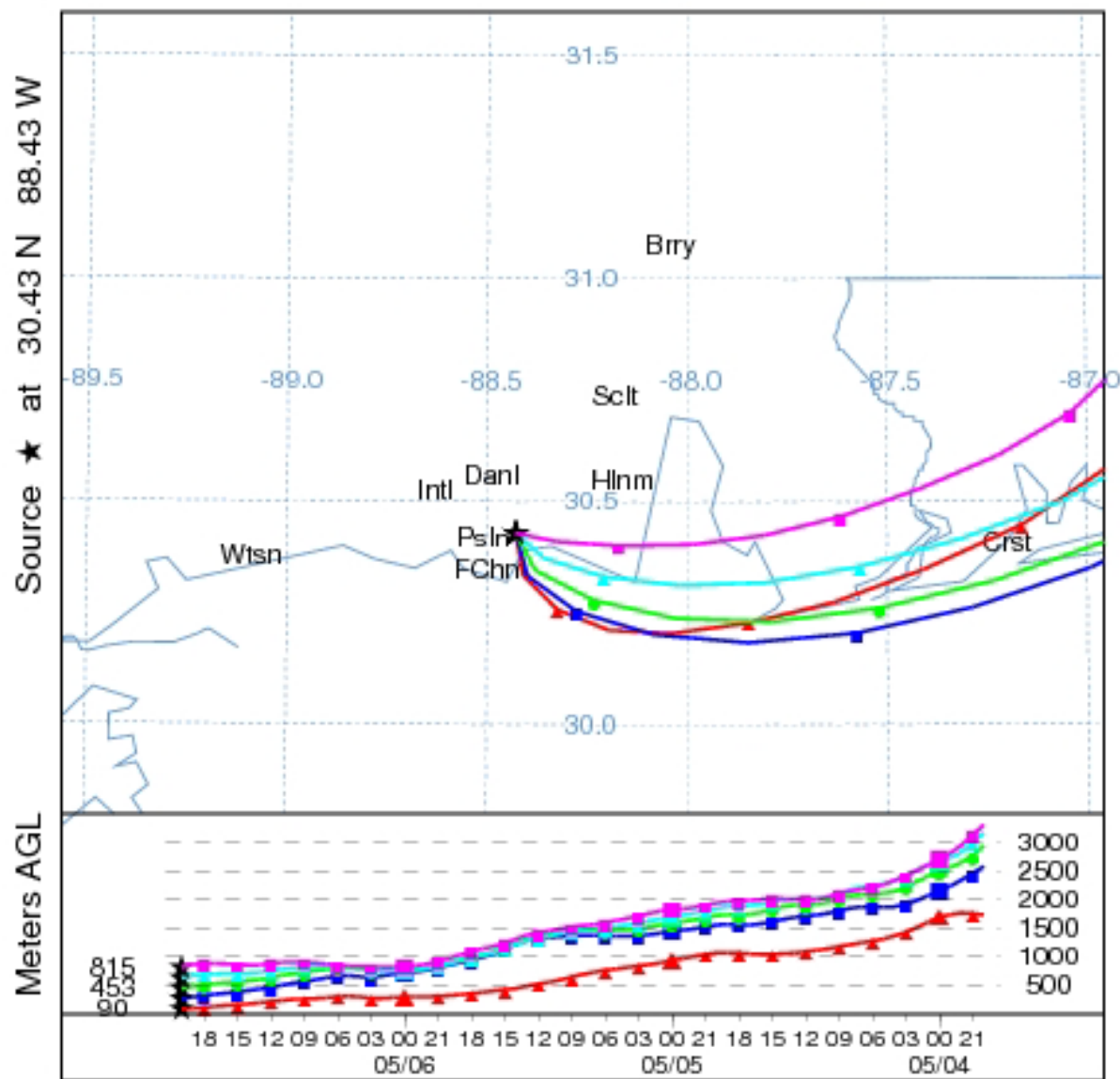


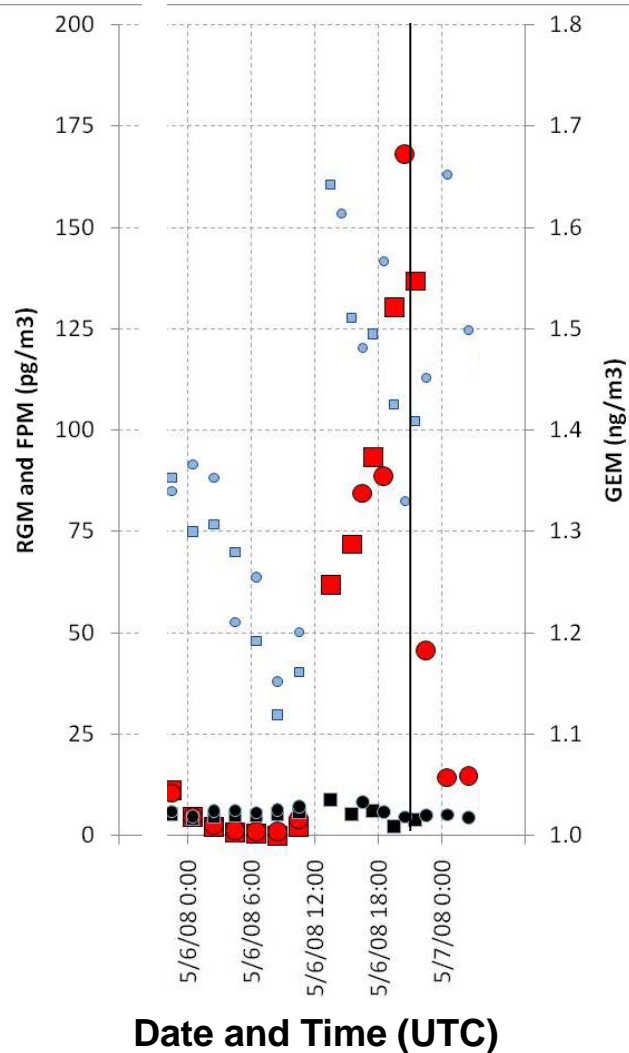
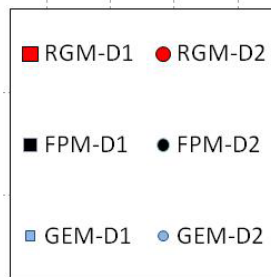
NOAA HYSPLIT MODEL Backward trajectories ending at 19 UTC 06 May 08 NAM Meteorological Data



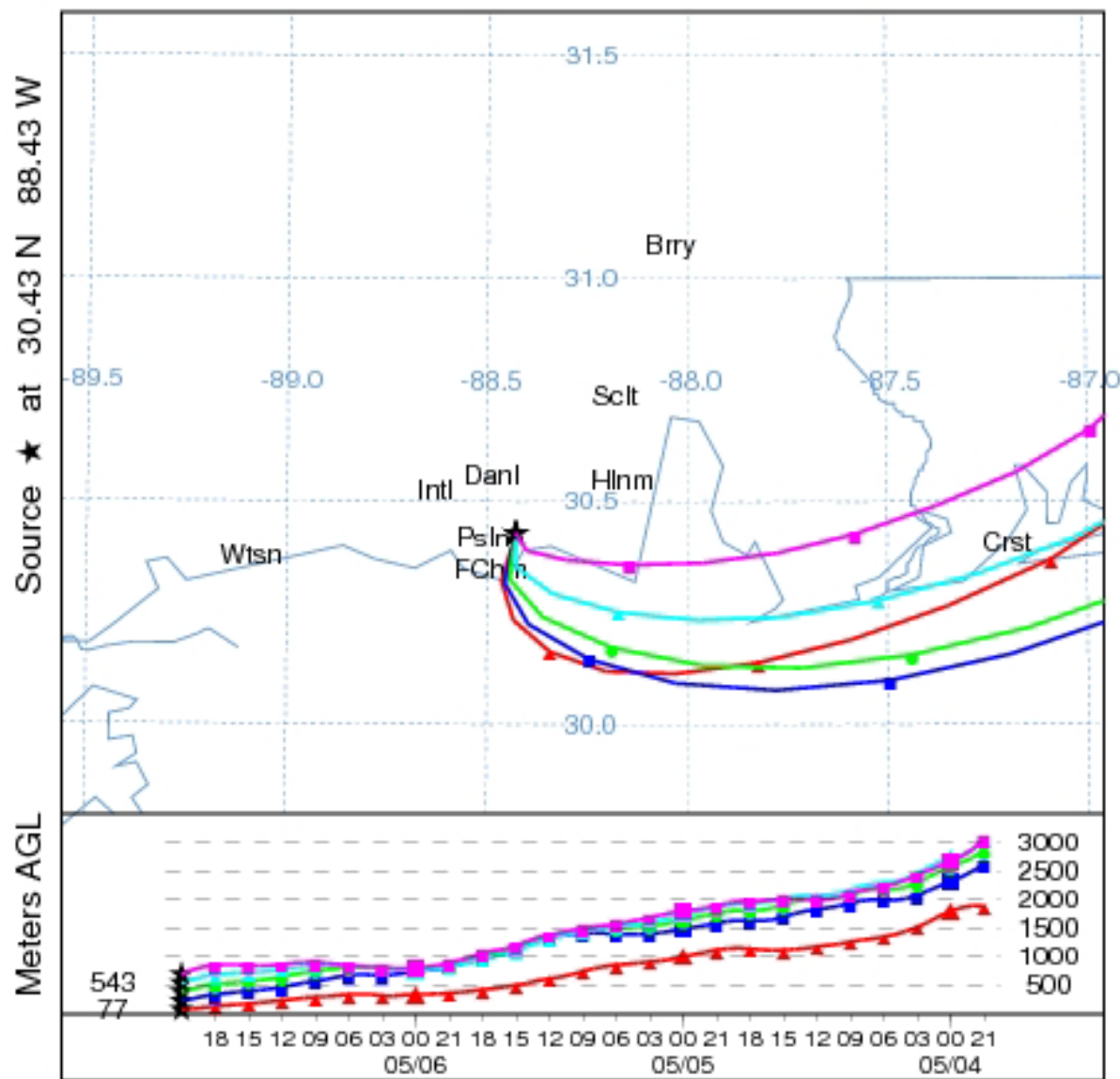


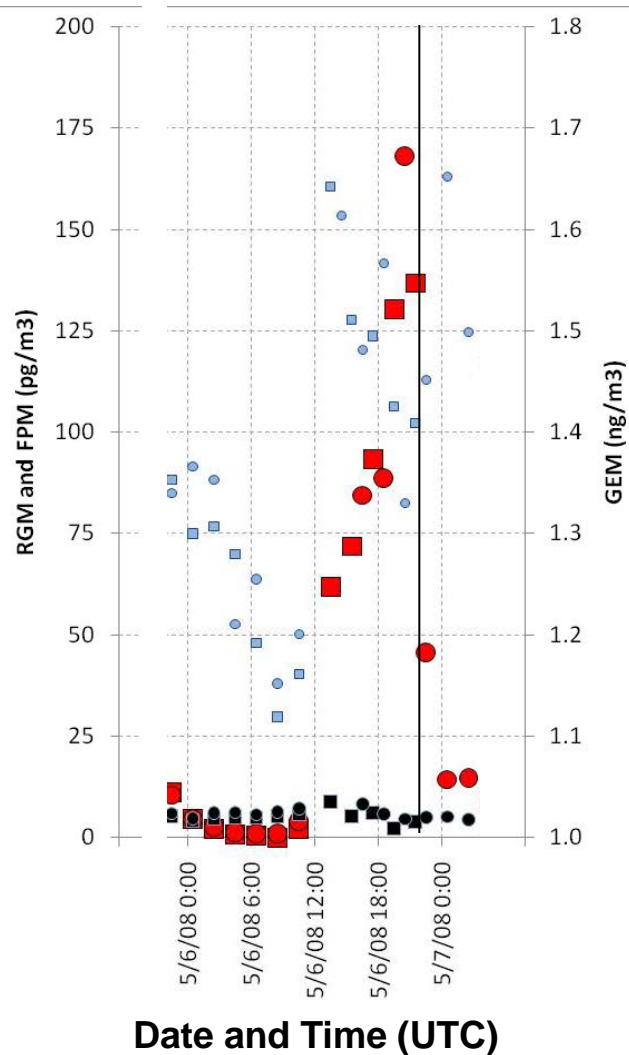
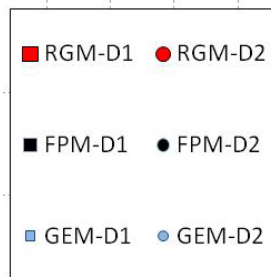
NOAA HYSPLIT MODEL Backward trajectories ending at 20 UTC 06 May 08 NAM Meteorological Data



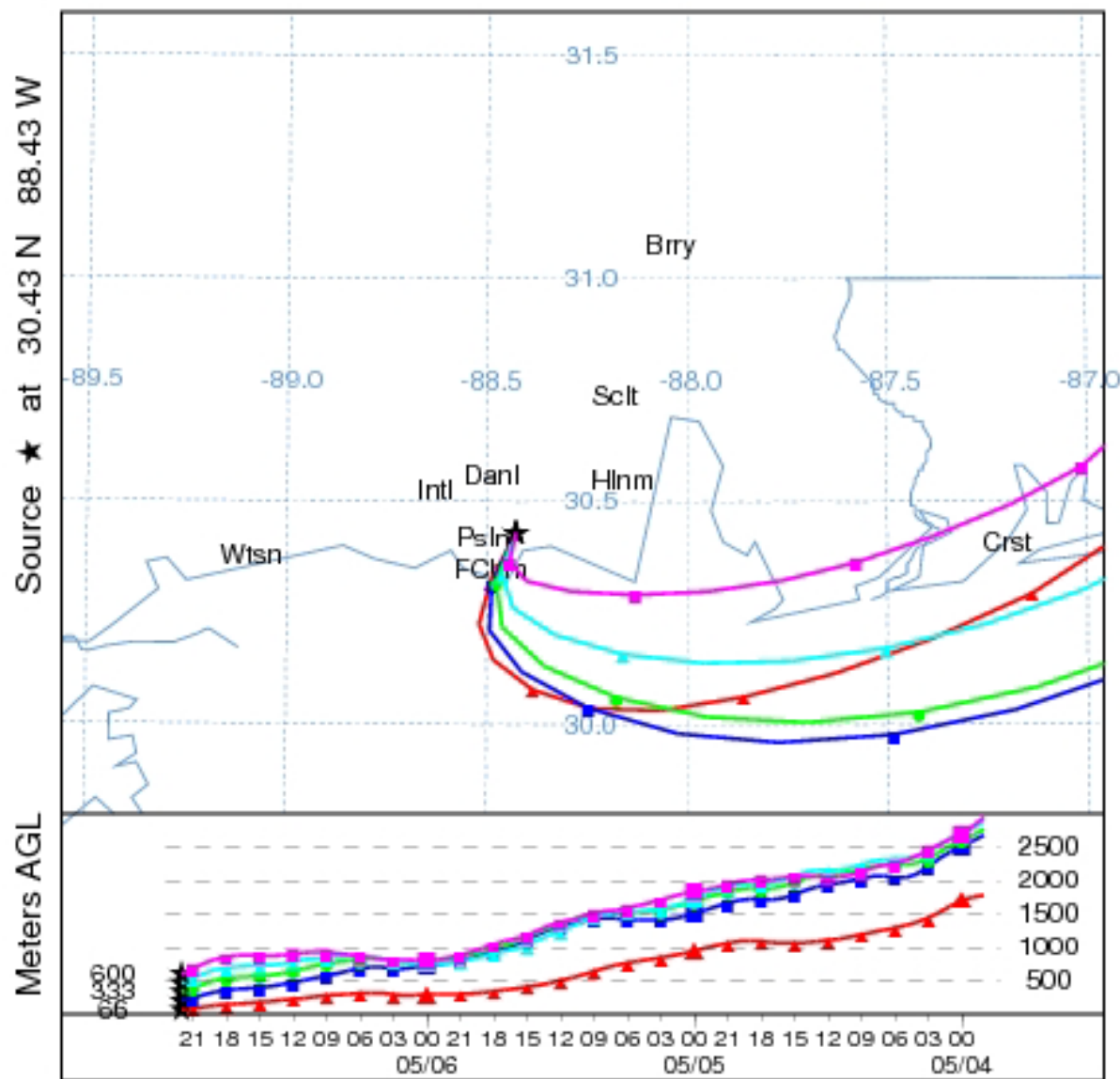


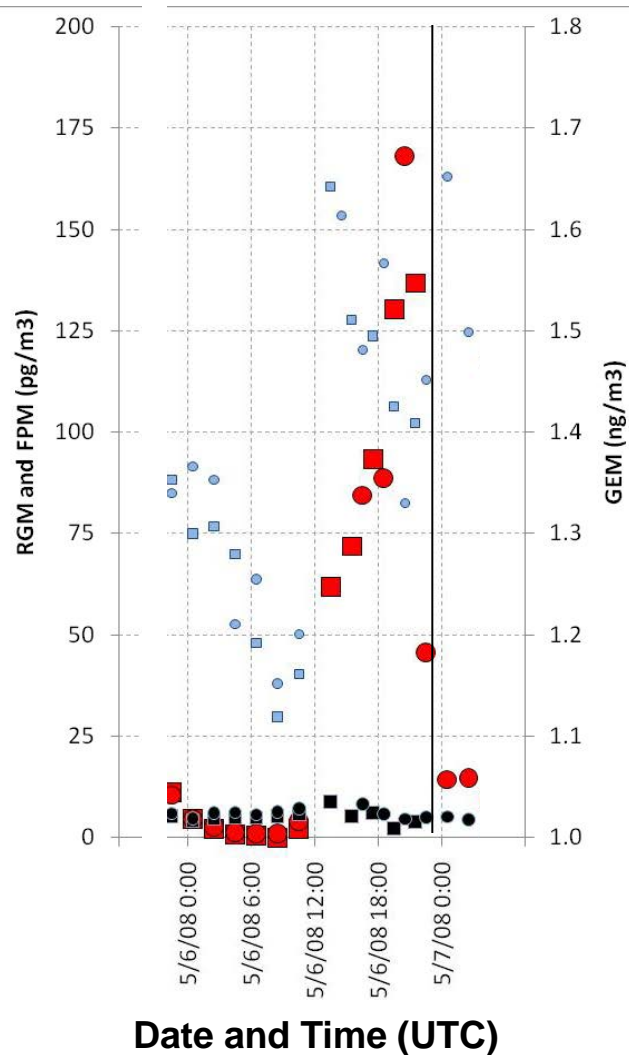
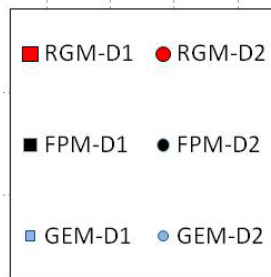
NOAA HYSPLIT MODEL Backward trajectories ending at 21 UTC 06 May 08 NAM Meteorological Data



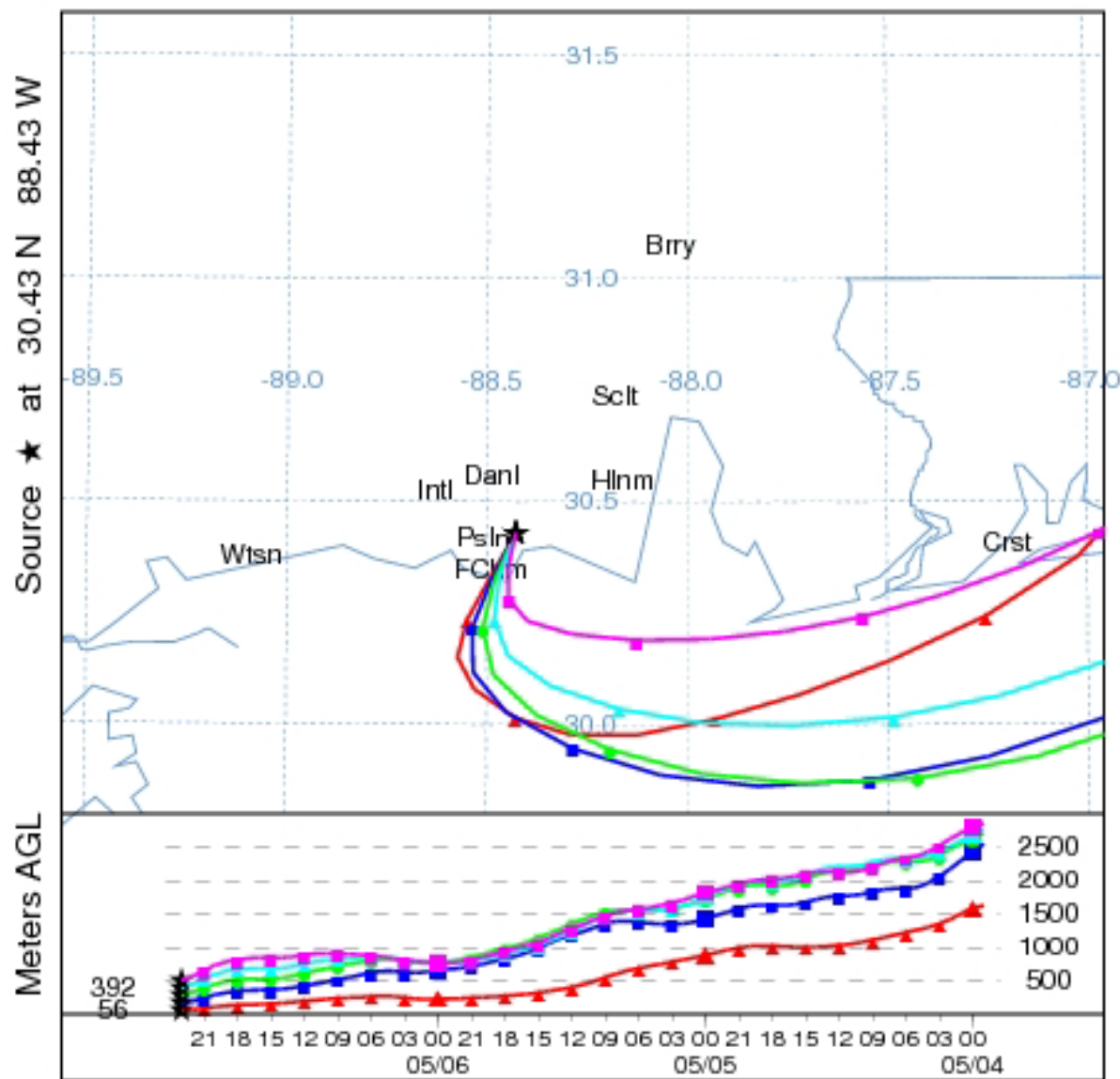


NOAA HYSPLIT MODEL Backward trajectories ending at 22 UTC 06 May 08 NAM Meteorological Data



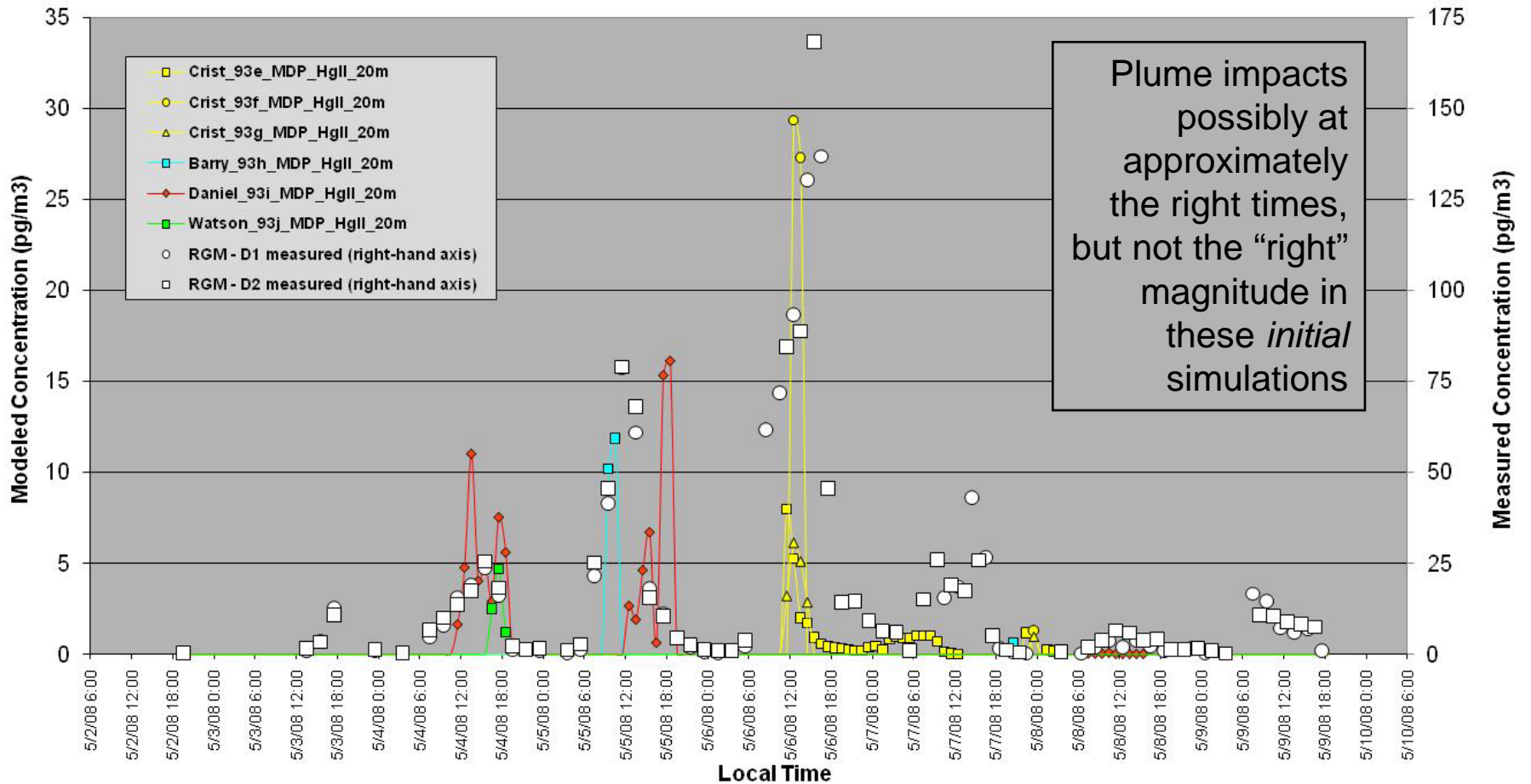


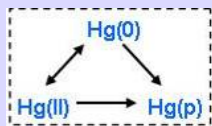
NOAA HYSPLIT MODEL Backward trajectories ending at 23 UTC 06 May 08 NAM Meteorological Data



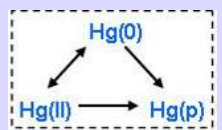
Some very preliminary results for May 5-6 2008 Episode
simple puff version of HYSPLIT-Hg, EDAS 40km met data

RGM -- May 5-6, 2008 Grand Bay NERR

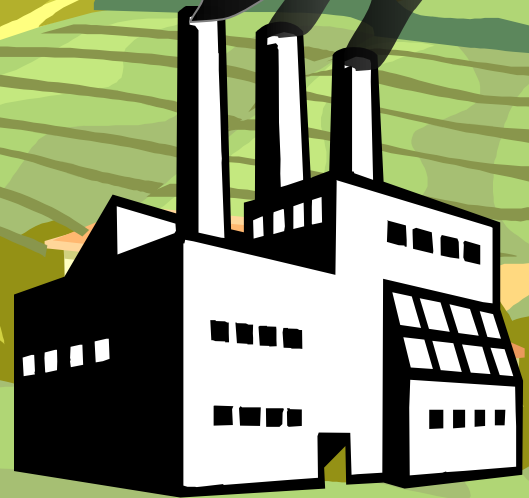
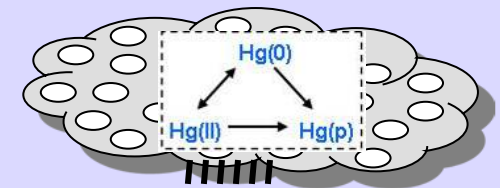
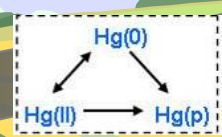
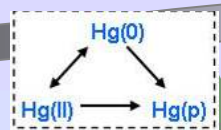




Hg from other sources: local, regional & more distant



One critical factor in carrying out a meaningful model evaluation in cases where local/regional sources may be important is to have accurate meteorological data to drive the model



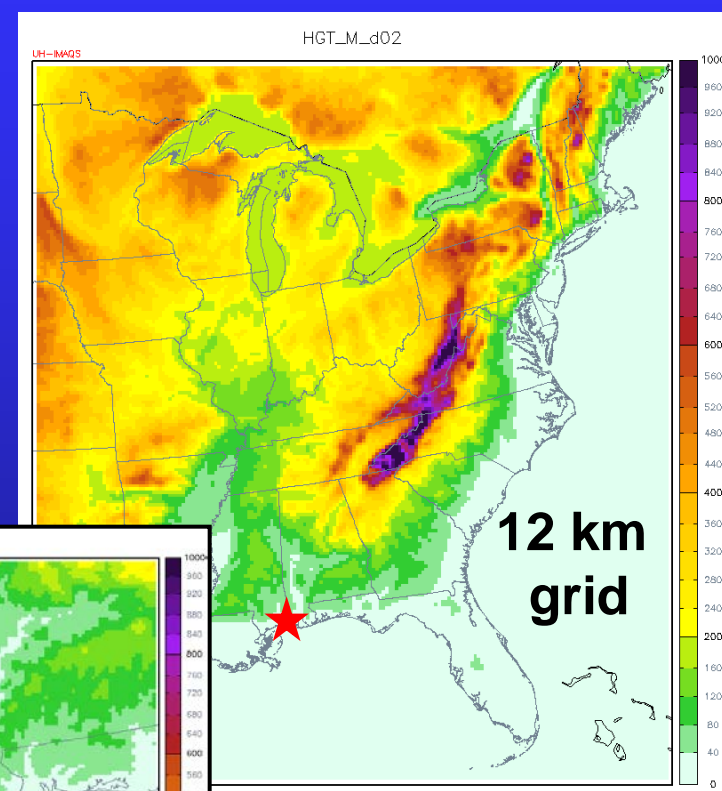
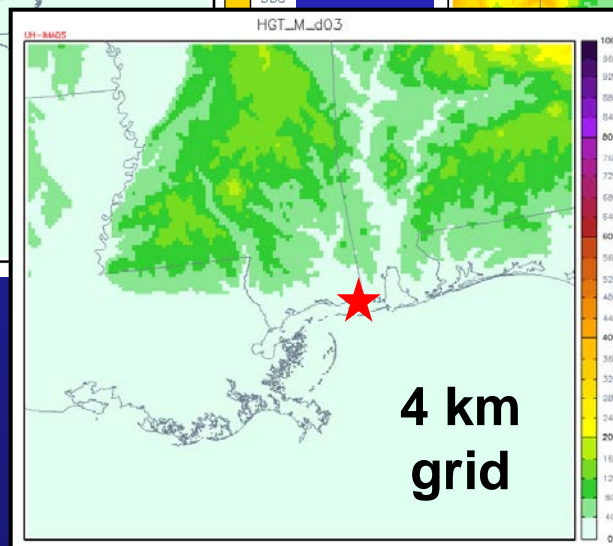
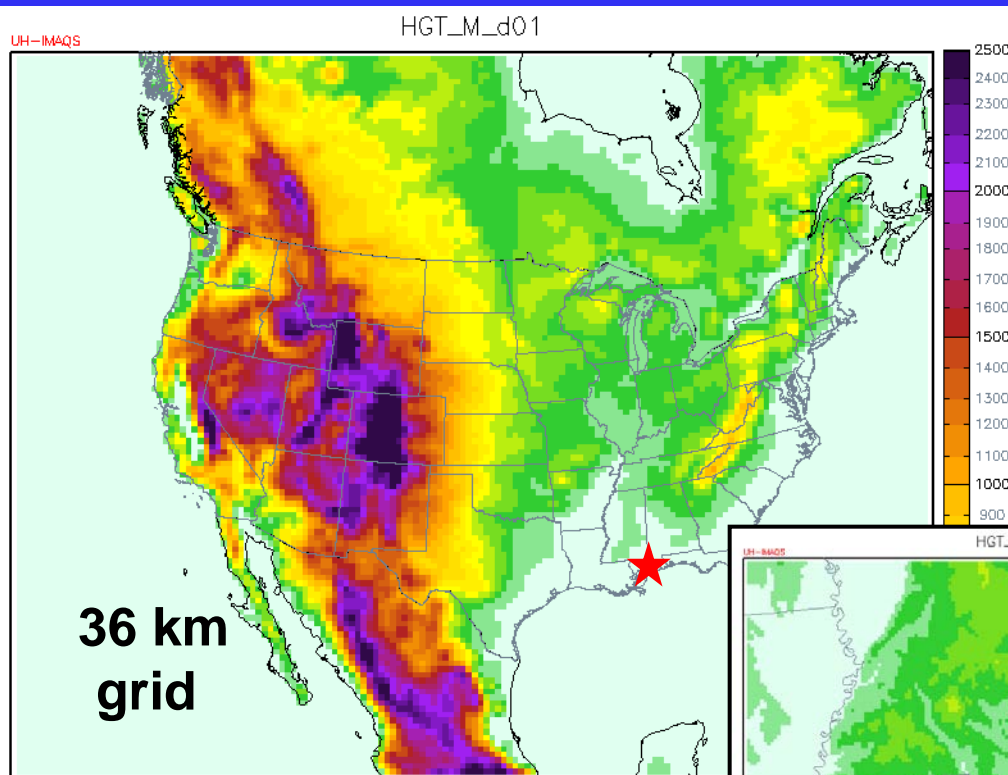
MEASUREMENTS

Site	Current	Wet Dep	Intensive	Data
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MODELING

Episodes	Met Data	Evolution	Emissions?
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High-resolution meteorological simulations being carried out for episodes at the Grand Bay NERR [★] by Dr. Fantine Ngan, a post-doc at NOAA ARL



*Terrain height
of 3 domains*

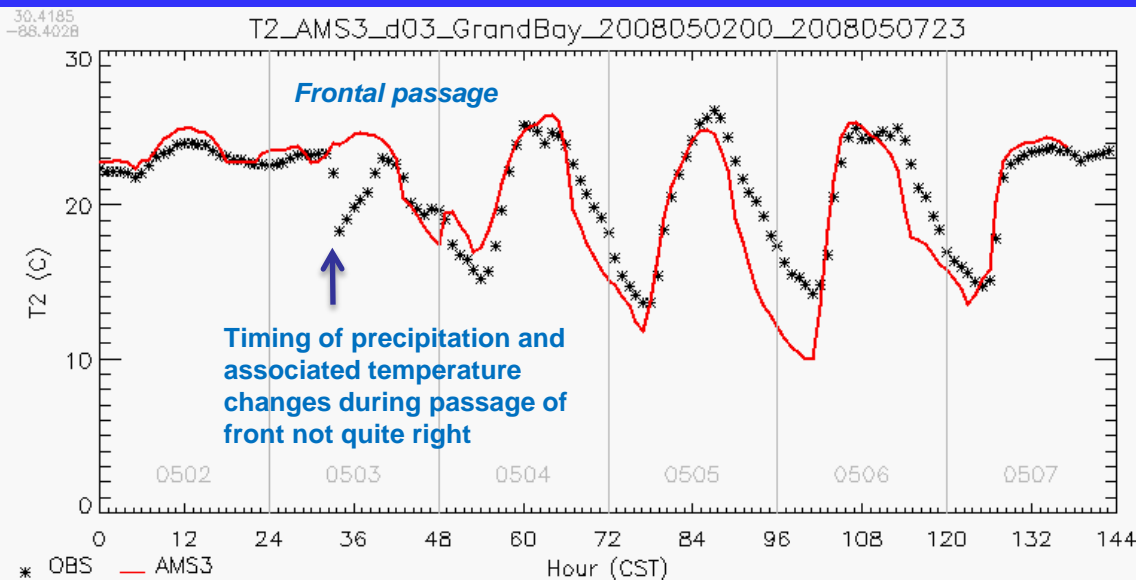
Fantine Ngan, NOAA ARL

MEASUREMENTS

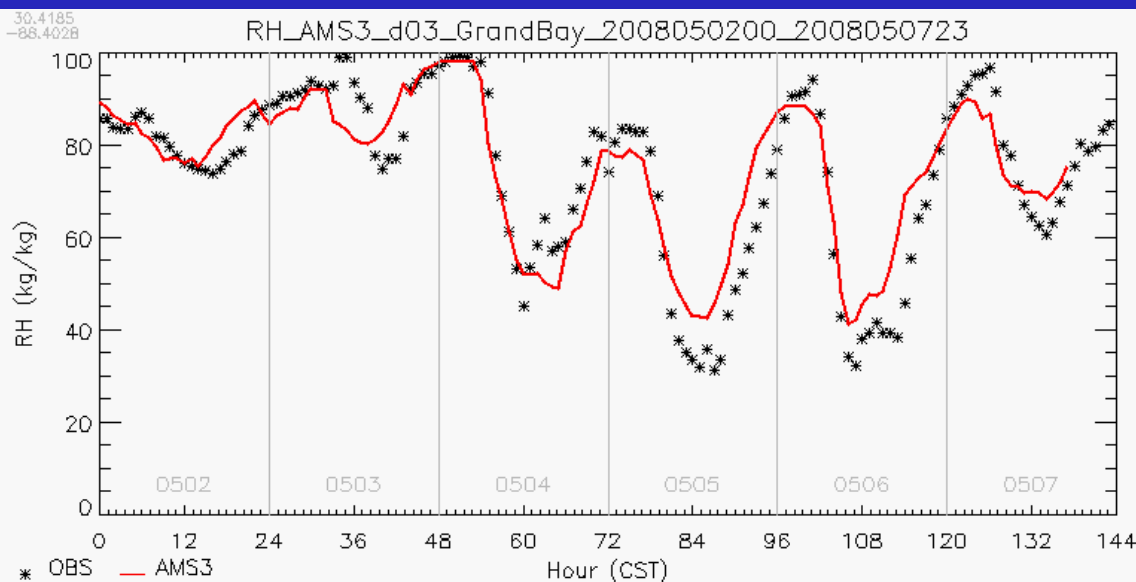
Site	Current	Wet Dep	Intensive	Data
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MODELING

Episodes	Met Data	Evolution	Emissions?
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**Modeled vs. Measured
Temperature at 2 m,
May 2-7, 2008**



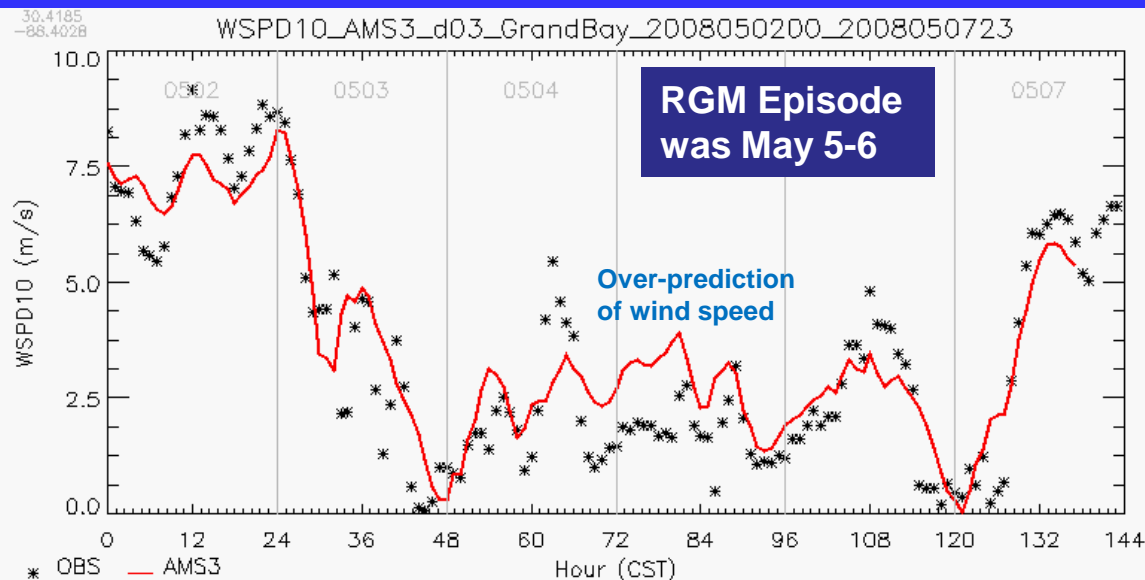
**Modeled vs. Measured
Relative Humidity,
May 2-7, 2008**

MEASUREMENTS

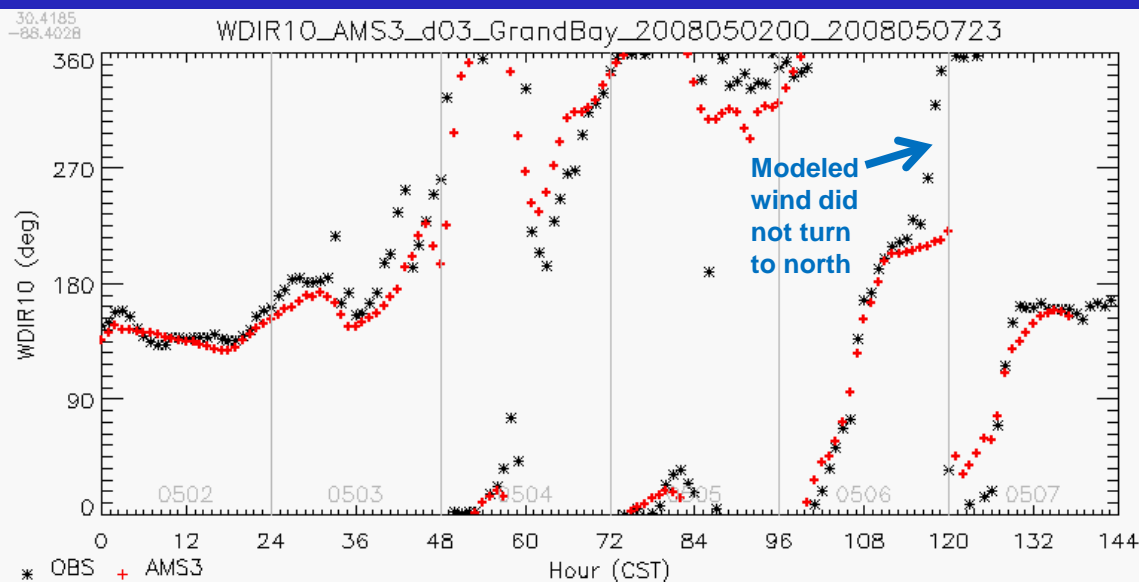
MODELING

Site	Current	Wet Dep	Intensive	Data
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Episodes	Met Data	Evolution	Emissions?
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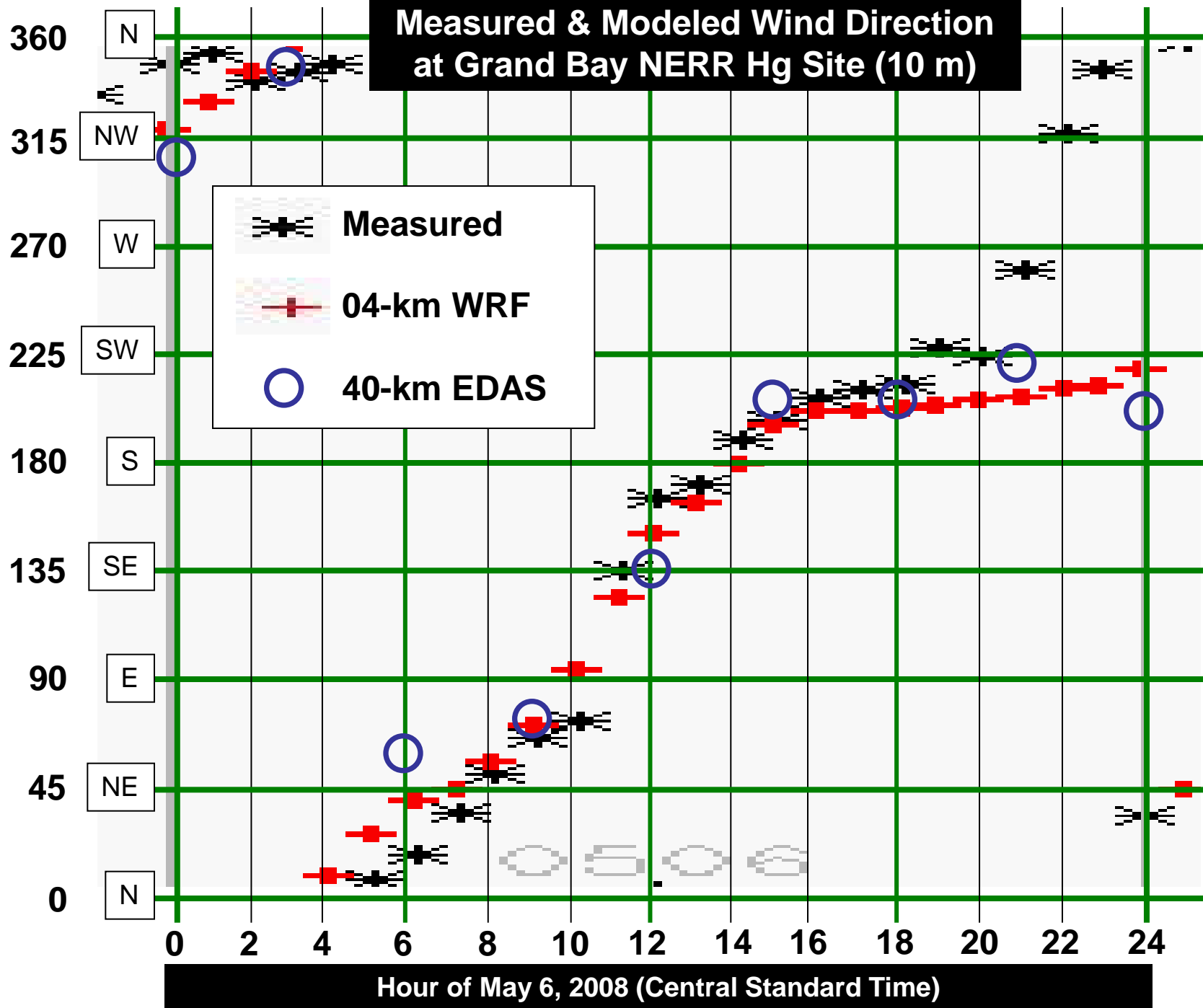
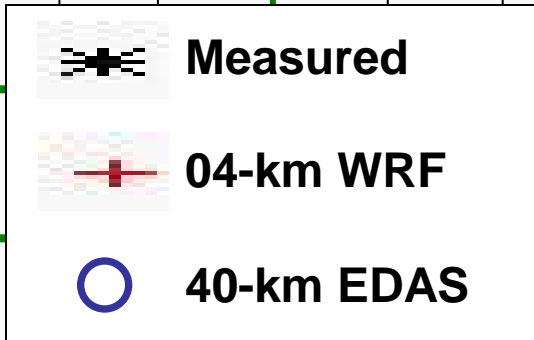
Modeled vs. Measured
Wind Speed at 10 m,
May 2-7, 2008



Modeled vs. Measured
Wind Direction at 10 m,
May 2-7, 2008

Wind Direction (degrees)

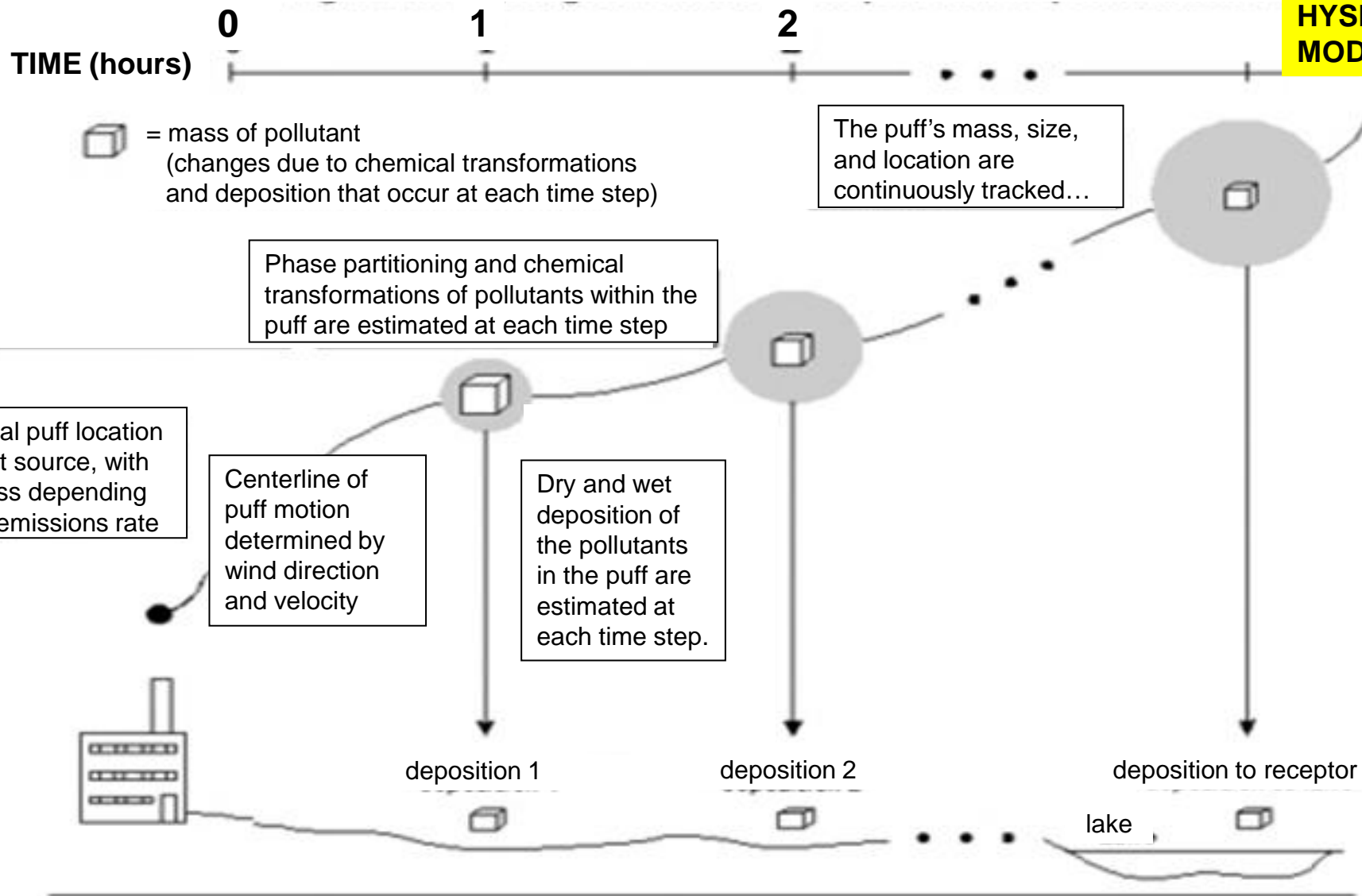
Measured & Modeled Wind Direction at Grand Bay NERR Hg Site (10 m)



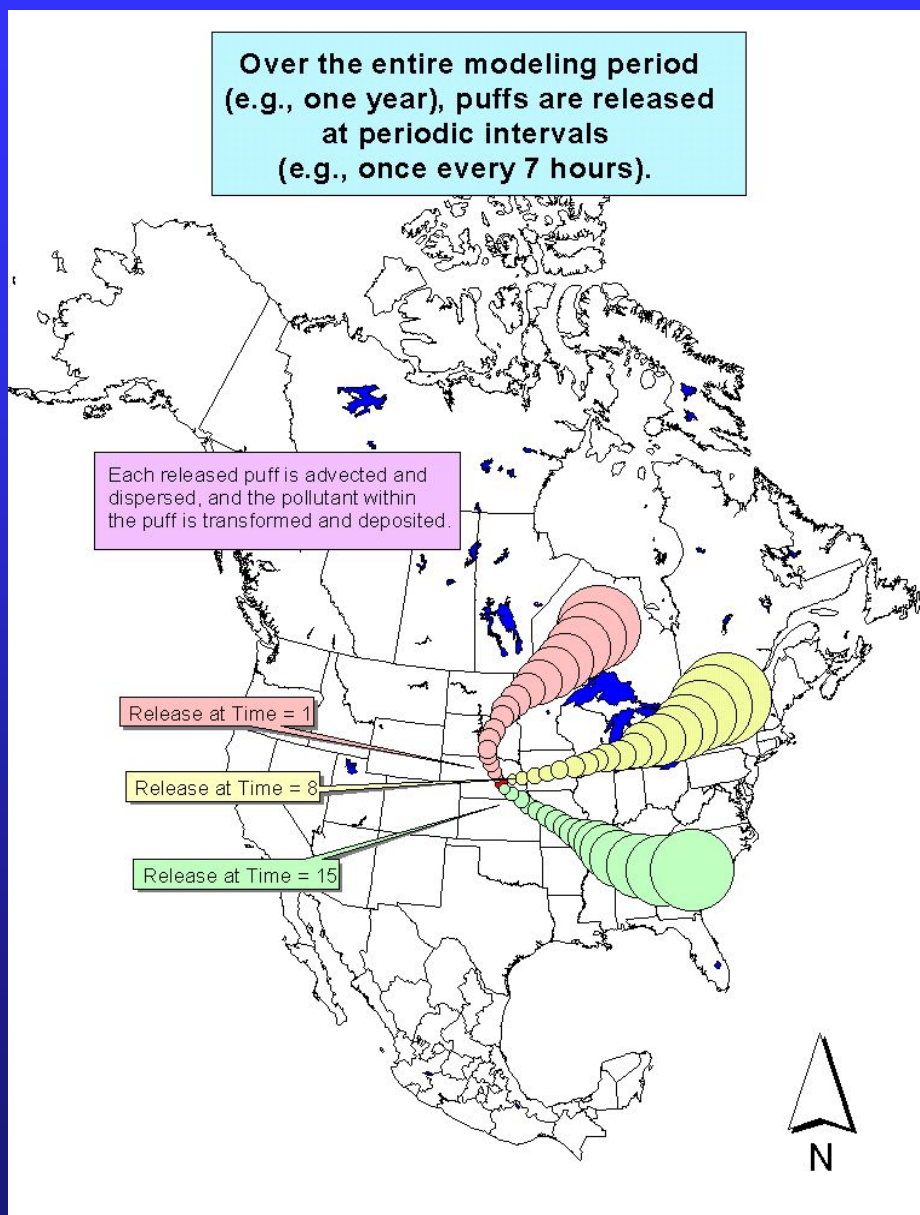
MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

Lagrangian Puff Atmospheric Fate and Transport Model

**NOAA
HYSPLIT
MODEL**



MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?



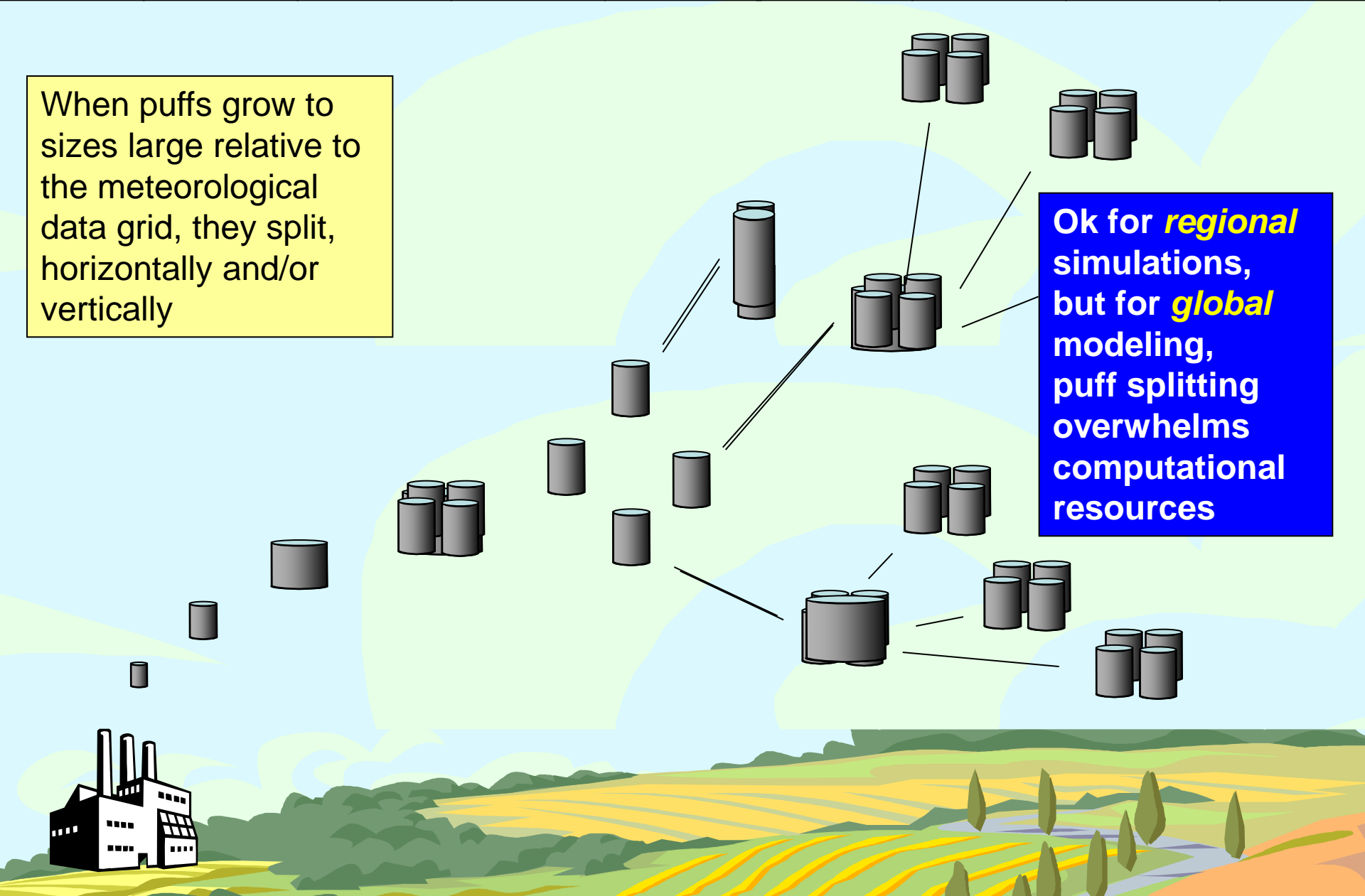
(Evolving) Atmospheric Chemical Reaction Scheme for Mercury

Reaction	Rate	Units	Reference
GAS PHASE REACTIONS			
? $\text{Hg}^0 + \text{O}_3 \rightarrow \text{Hg(p)}$	3.0E-20	$\text{cm}^3/\text{molec-sec}$	Hall (1995)
$\text{Hg}^0 + \text{HCl} \rightarrow \text{HgCl}_2$	1.0E-19	$\text{cm}^3/\text{molec-sec}$	Hall and Bloom (1993)
$\text{Hg}^0 + \text{H}_2\text{O}_2 \rightarrow \text{Hg(p)}$	8.5E-19	$\text{cm}^3/\text{molec-sec}$	Tokos et al. (1998) (upper limit based on experiments)
$\text{Hg}^0 + \text{Cl}_2 \rightarrow \text{HgCl}_2$	4.0E-18	$\text{cm}^3/\text{molec-sec}$	Calhoun and Prestbo (2001)
? $\text{Hg}^0 + \text{OH} \rightarrow \text{Hg(p)}$	8.7E-14	$\text{cm}^3/\text{molec-sec}$	Sommar et al. (2001)
new $\text{Hg}^0 + \text{Br} \rightarrow \text{HgBr}_2$			
AQUEOUS PHASE REACTIONS			
$\text{Hg}^0 + \text{O}_3 \rightarrow \text{Hg}^{+2}$	4.7E+7	$(\text{molar-sec})^{-1}$	Munthe (1992)
$\text{Hg}^0 + \text{OH} \rightarrow \text{Hg}^{+2}$	2.0E+9	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1997)
$\text{HgSO}_3 \rightarrow \text{Hg}^0$	$T^*e^{((31.971^*T)-12595.0)/T}$ sec ⁻¹ [T = temperature (K)]		Van Loon et al. (2002)
? $\text{Hg(II)} + \text{HO}_2 \rightarrow \text{Hg}^0$	~ 0	$(\text{molar-sec})^{-1}$	Gardfeldt & Jonnson (2003)
$\text{Hg}^0 + \text{HOCl} \rightarrow \text{Hg}^{+2}$	2.1E+6	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1998)
$\text{Hg}^0 + \text{OCl}^{-1} \rightarrow \text{Hg}^{+2}$	2.0E+6	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1998)
$\text{Hg(II)} \leftrightarrow \text{Hg(II)}_{(\text{soot})}$	9.0E+2	liters/gram; t = 1/hour	eqbrm: Seigneur et al. (1998) rate: Bullock & Brehme (2002).
$\text{Hg}^{+2} + \text{h}\nu \rightarrow \text{Hg}^0$	6.0E-7	$(\text{sec})^{-1}$ (maximum)	Xiao et al. (1994); Bullock and Brehme (2002)

MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

When puffs grow to sizes large relative to the meteorological data grid, they split, horizontally and/or vertically

Ok for *regional* simulations, but for *global* modeling, puff splitting overwhelms computational resources



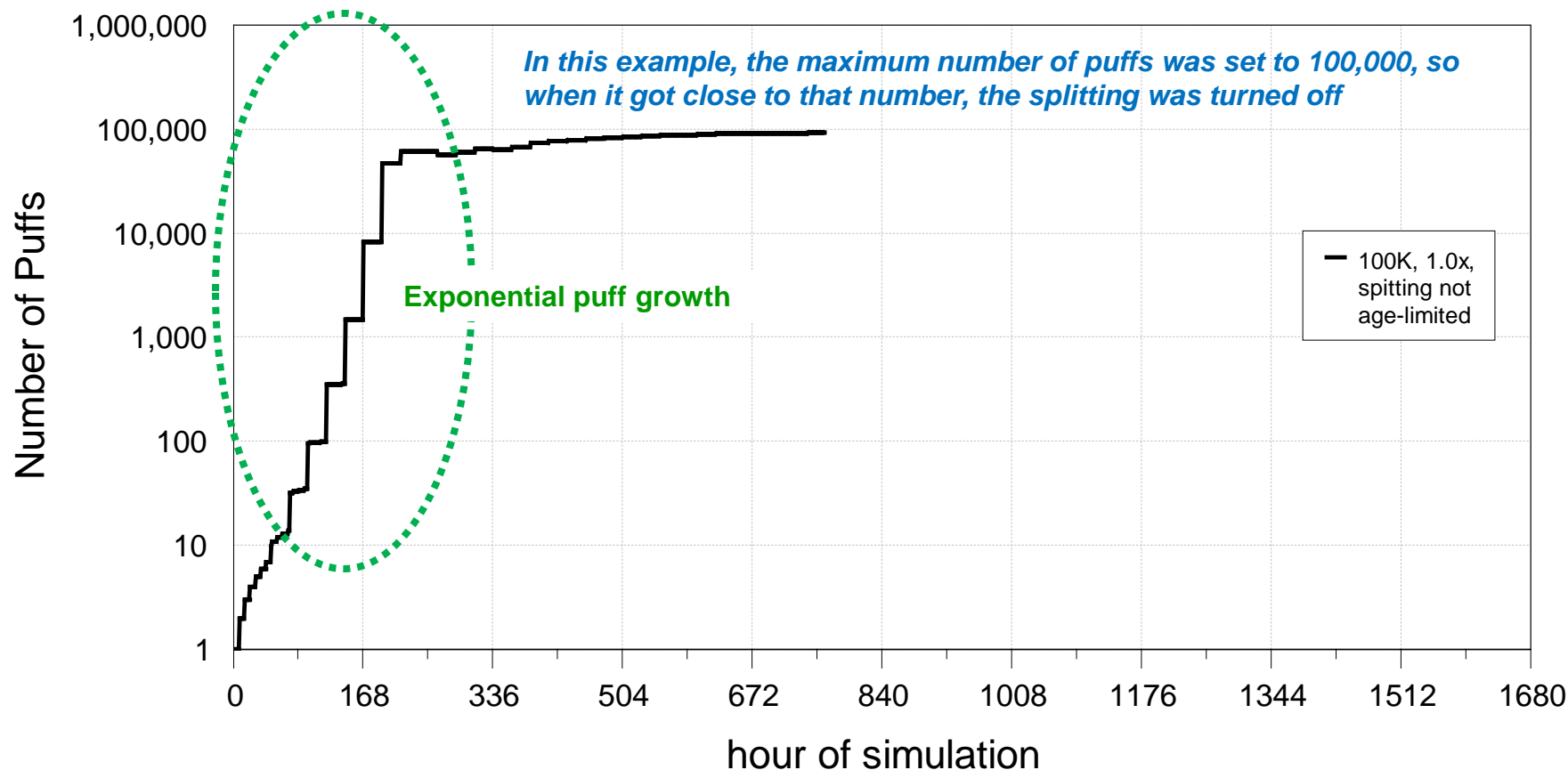
MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

Due to puff splitting, the number of puffs quickly overwhelms numerical resources

Evolution of Number of Puffs

as a function of MAXPAR and merge parameter multiplication factor

elem emit; growth not stopped; splitting not age-limited; source at lat = 30, long = 105 (China)



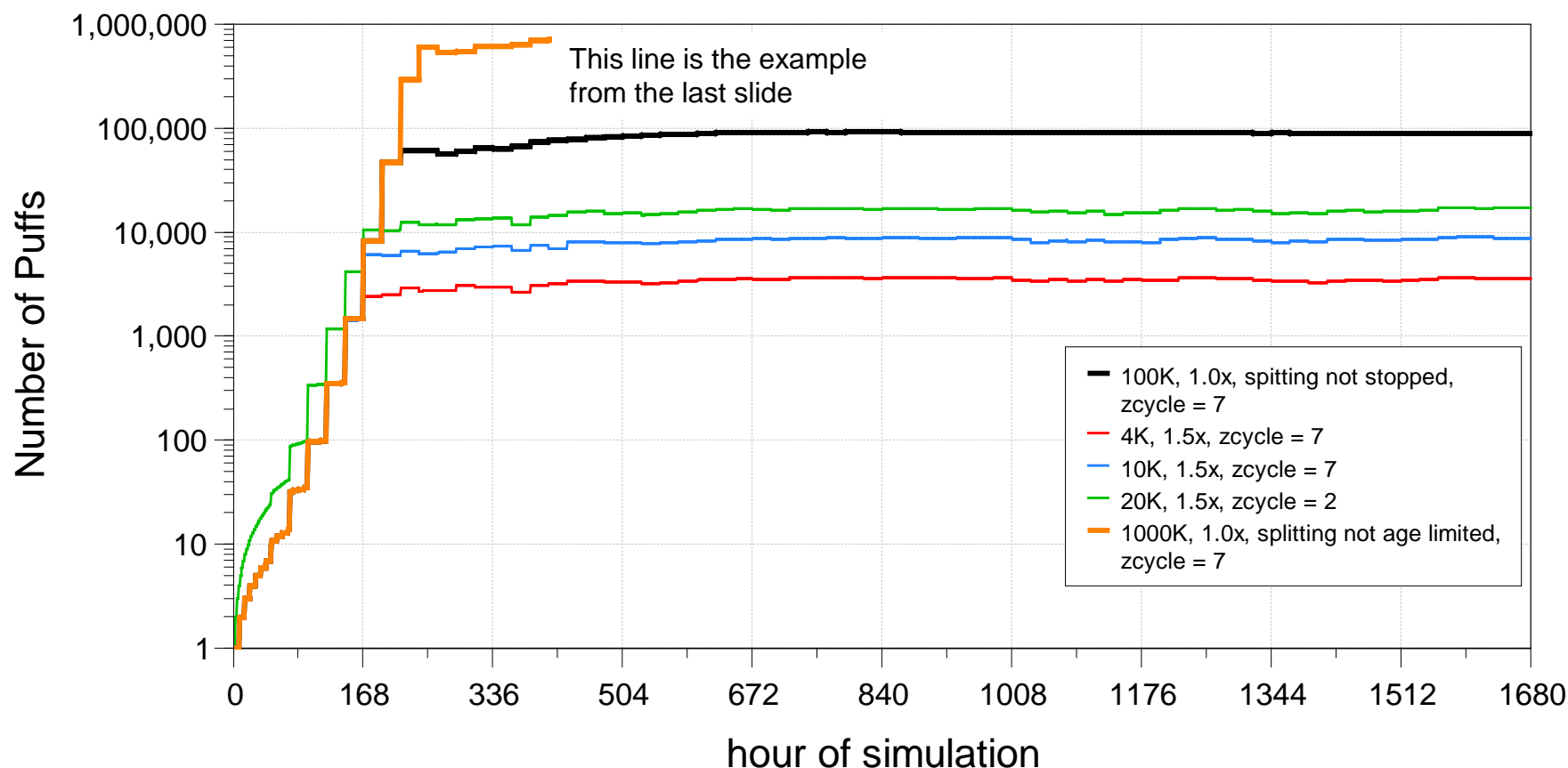
MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

*In each test, the number of puffs rises to the maximum allowable within ~ one week
(and then splitting stops...)*

Evolution of Number of Puffs

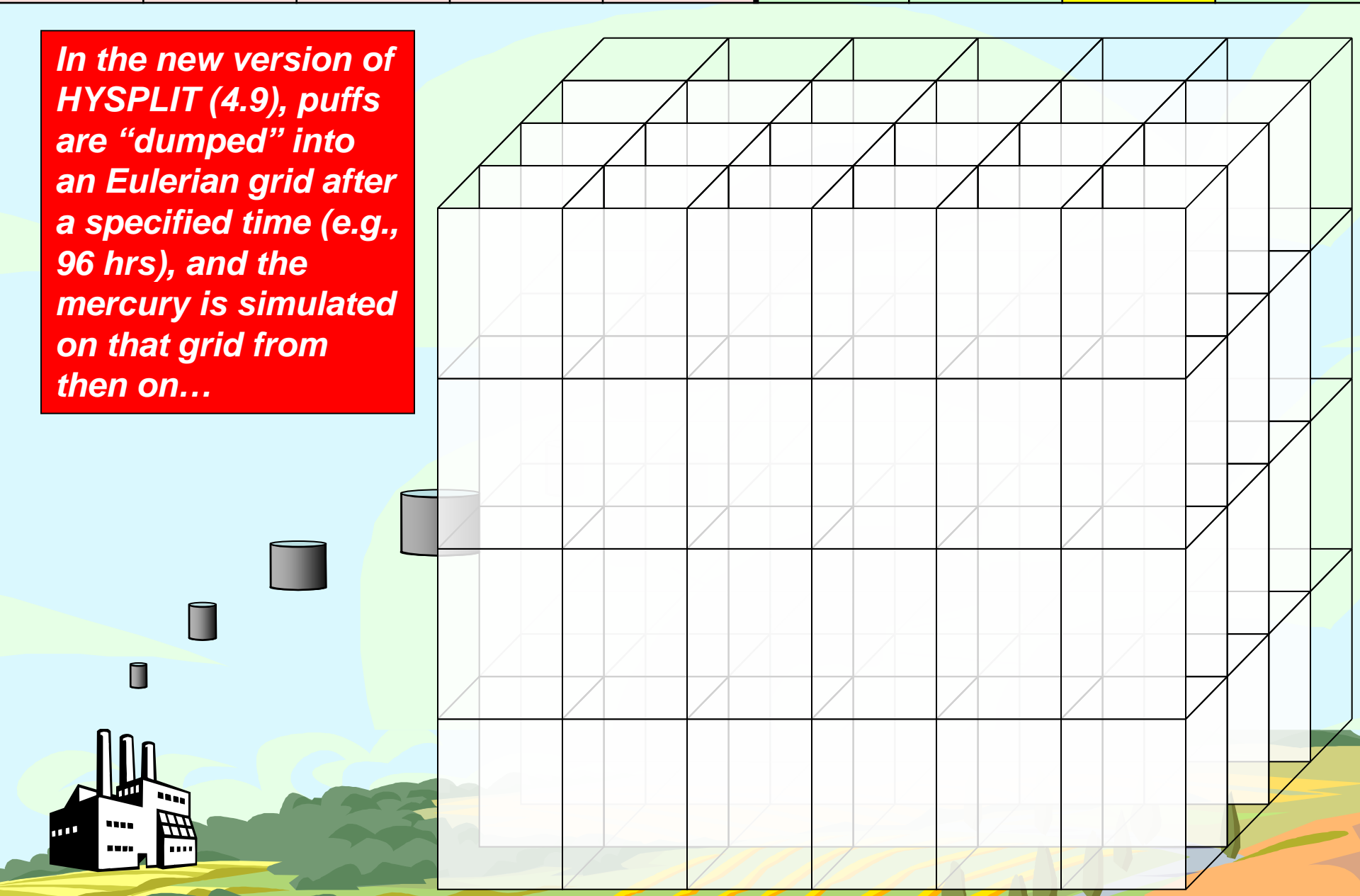
as a function of MAXPAR and merge parameter multiplication factor

elem emit; growth not stopped; splitting stopped after 168 hours; source at lat = 30, long = 105 (China)



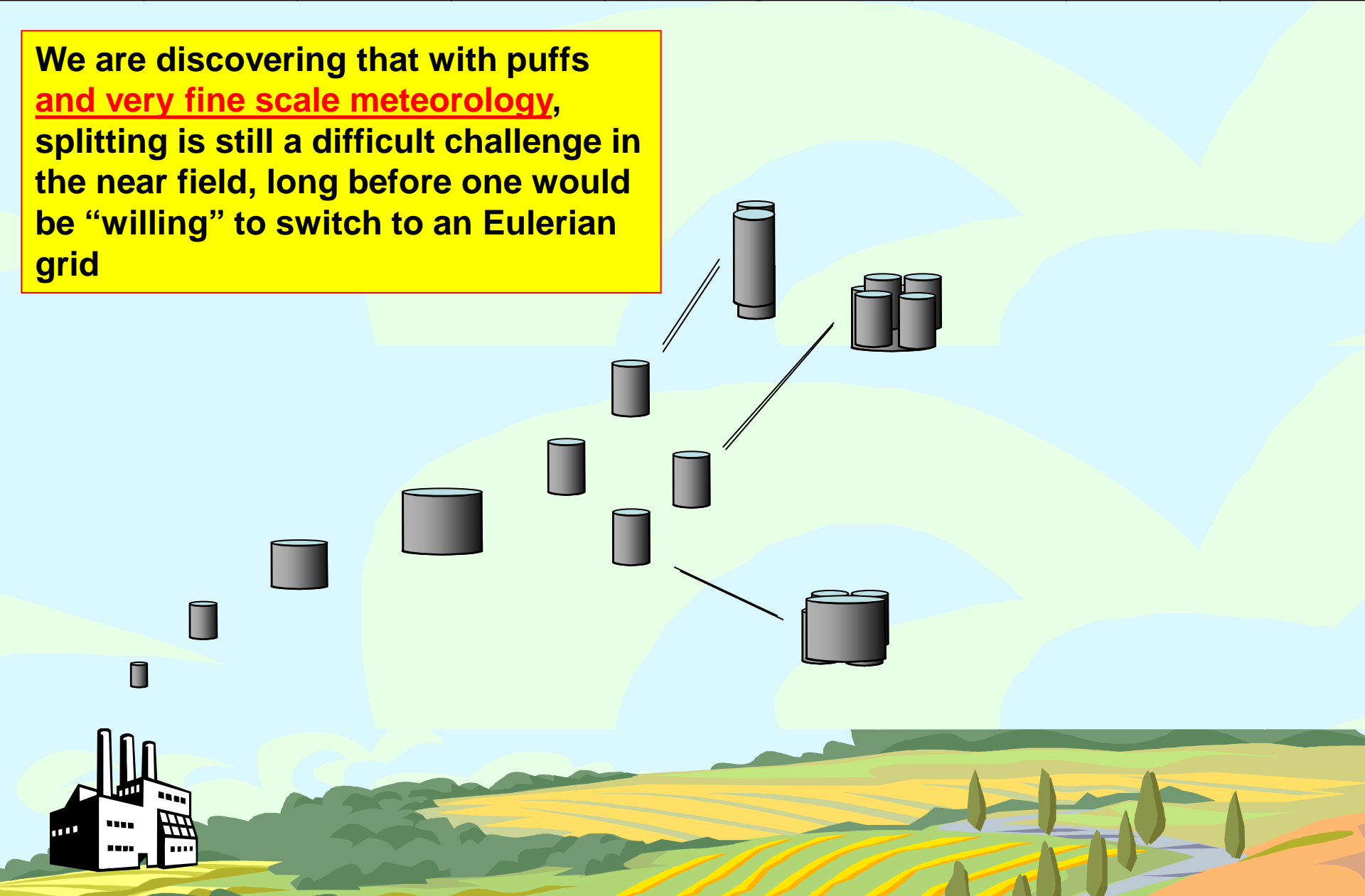
MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

In the new version of HYSPLIT (4.9), puffs are “dumped” into an Eulerian grid after a specified time (e.g., 96 hrs), and the mercury is simulated on that grid from then on...



MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

**We are discovering that with puffs
and very fine scale meteorology,
splitting is still a difficult challenge in
the near field, long before one would
be “willing” to switch to an Eulerian
grid**



MEASUREMENTS

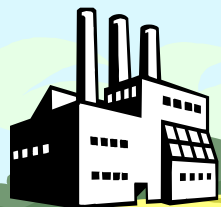
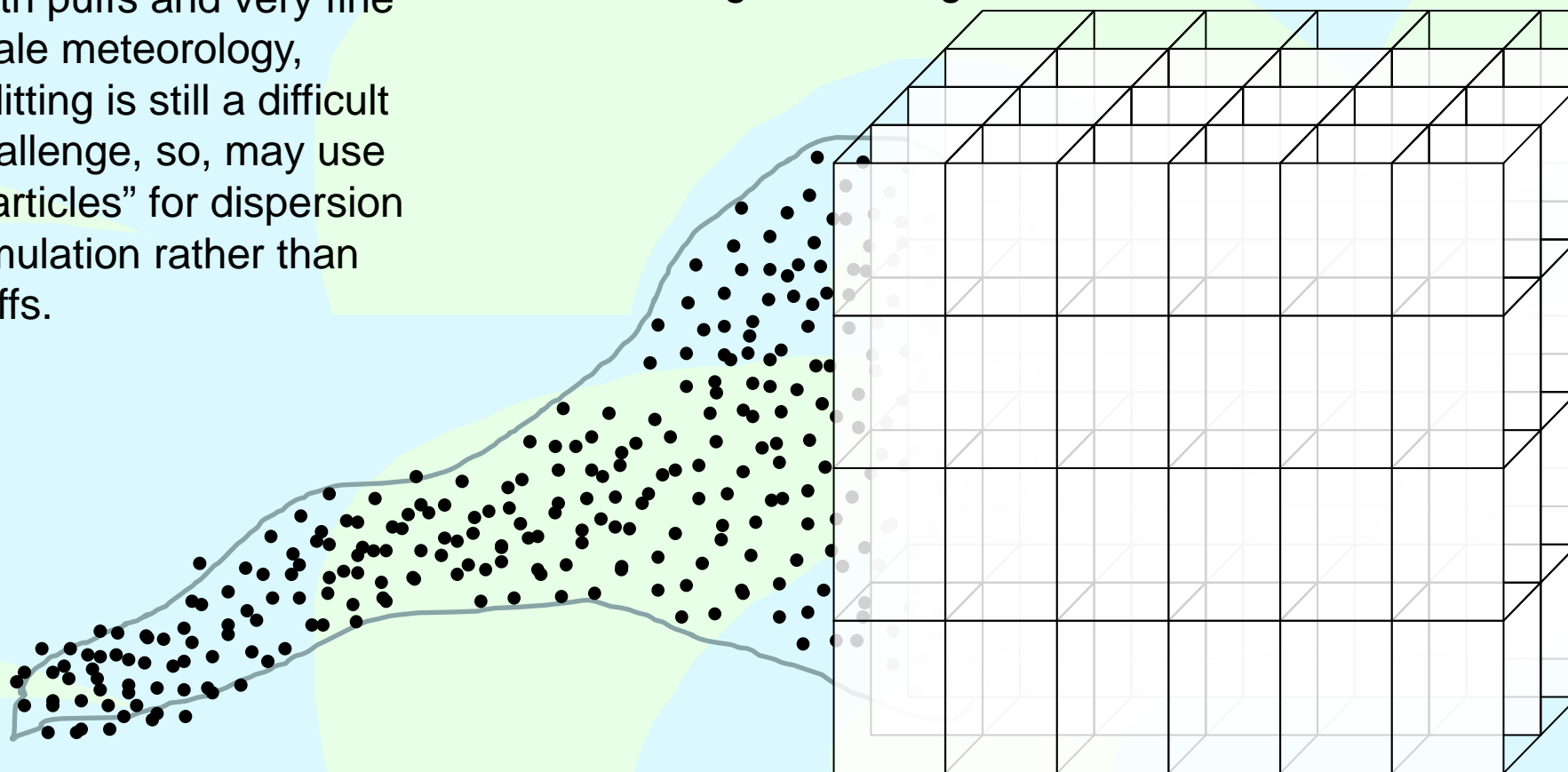
Site	Current	Wet Dep	Intensive	Data
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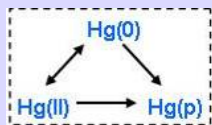
MODELING

Episodes	Met Data	Evolution	Emissions?
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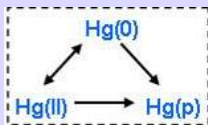
With puffs and very fine scale meteorology, splitting is still a difficult challenge, so, may use “particles” for dispersion simulation rather than puffs.

The particles would still be transferred to an Eulerian grid after a given time



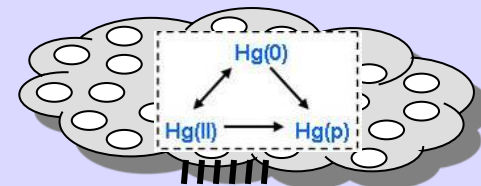
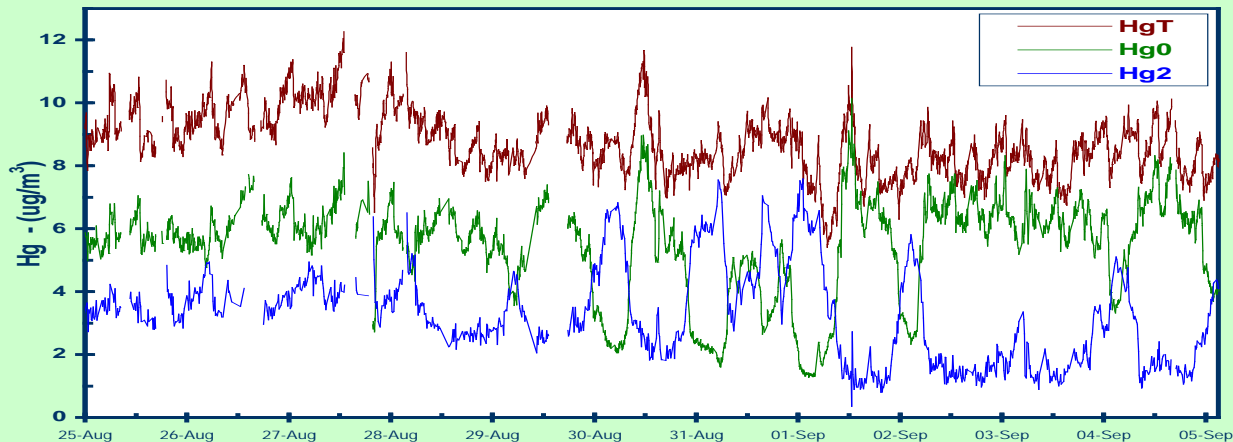


**Hg from
other sources:
local, regional
& more distant**



Series 3300 CEM - Continuous Speciated Mercury Data

Resolution: 2.5 min Duration: 11 Days



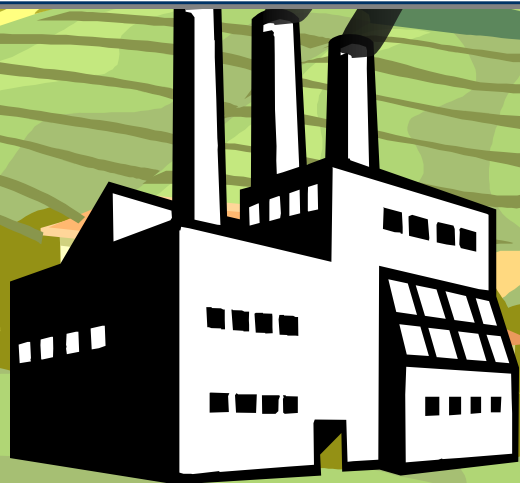
atmospheric
deposition
to the water
surface

Measurement
of wet
deposition

Measurement
of ambient air
concentrations



**Another essential factor in
carrying out a meaningful
model evaluation in cases
where local/regional sources
may be important is to have
accurate emissions data for
local/regional sources, valid
for the time of the episode
being studied**



MEASUREMENTS					MODELING			
Site	Current	Wet Dep	Intensive	Data	Episodes	Met Data	Evolution	Emissions?

For episode analysis, emissions information needed for major sources in the vicinity of the site, e.g., coal fired power plants such as: Daniel, Watson, Barry, Crist, ...

- ☐ EPA NEI 2002 (or 2005) is very out of date...
- ☐ Ideally we'd like accurate, speciated emissions estimates for each key source that might have contributed to a given episode at the site, for the period of the episode, but, we are also interested in more "basic" things like:
 - Was a given plant operating normally during any given episode?
 - Were there any process or control changes at a given plant since the last available inventory information that would have significantly changed the emissions (or speciation of the emissions)

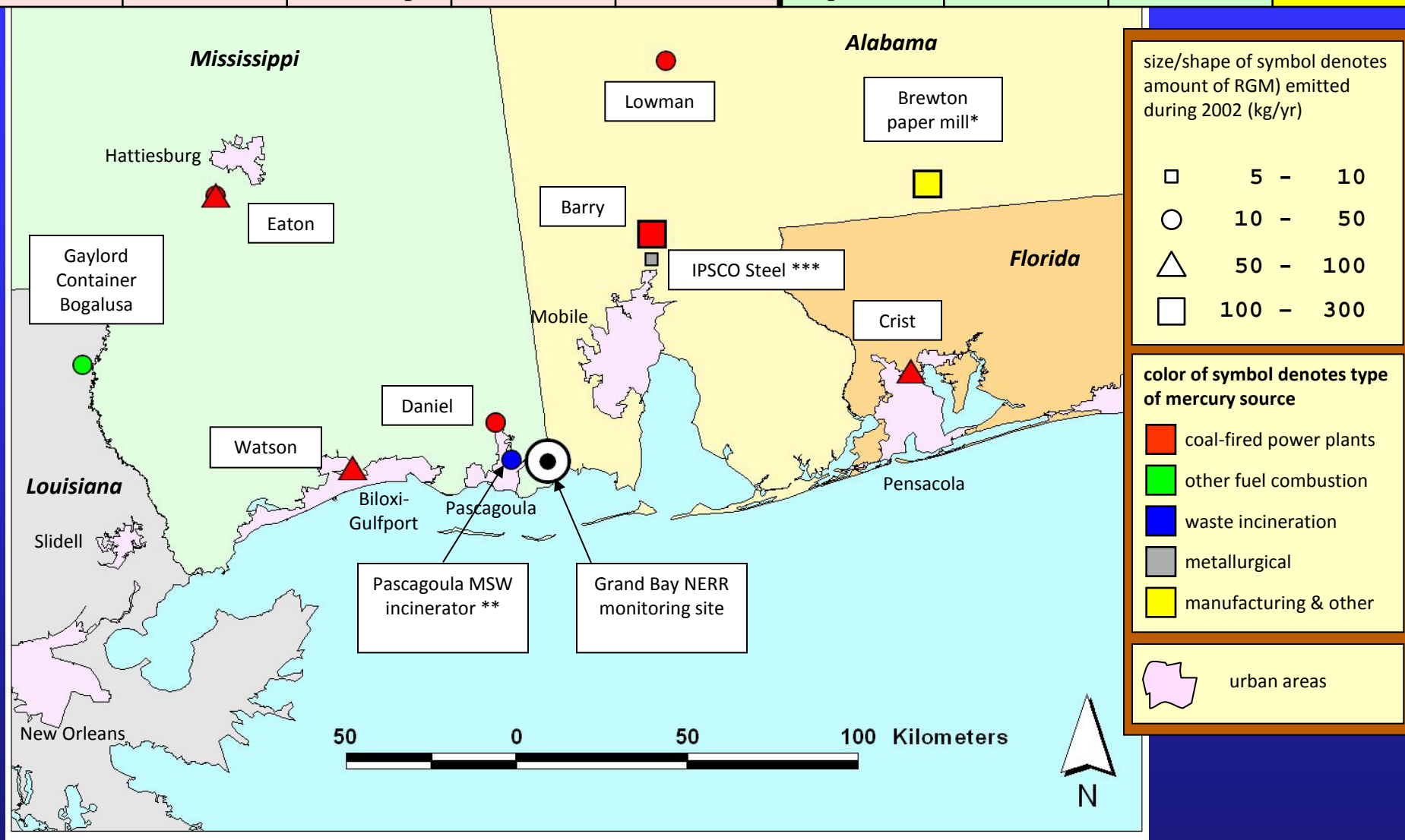
Atmospheric Mercury Measurements and Modeling at the Grand Bay National Estuarine Research Reserve

MEASUREMENTS

Site	Current	Wet Dep	Intensive	Data
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MODELING

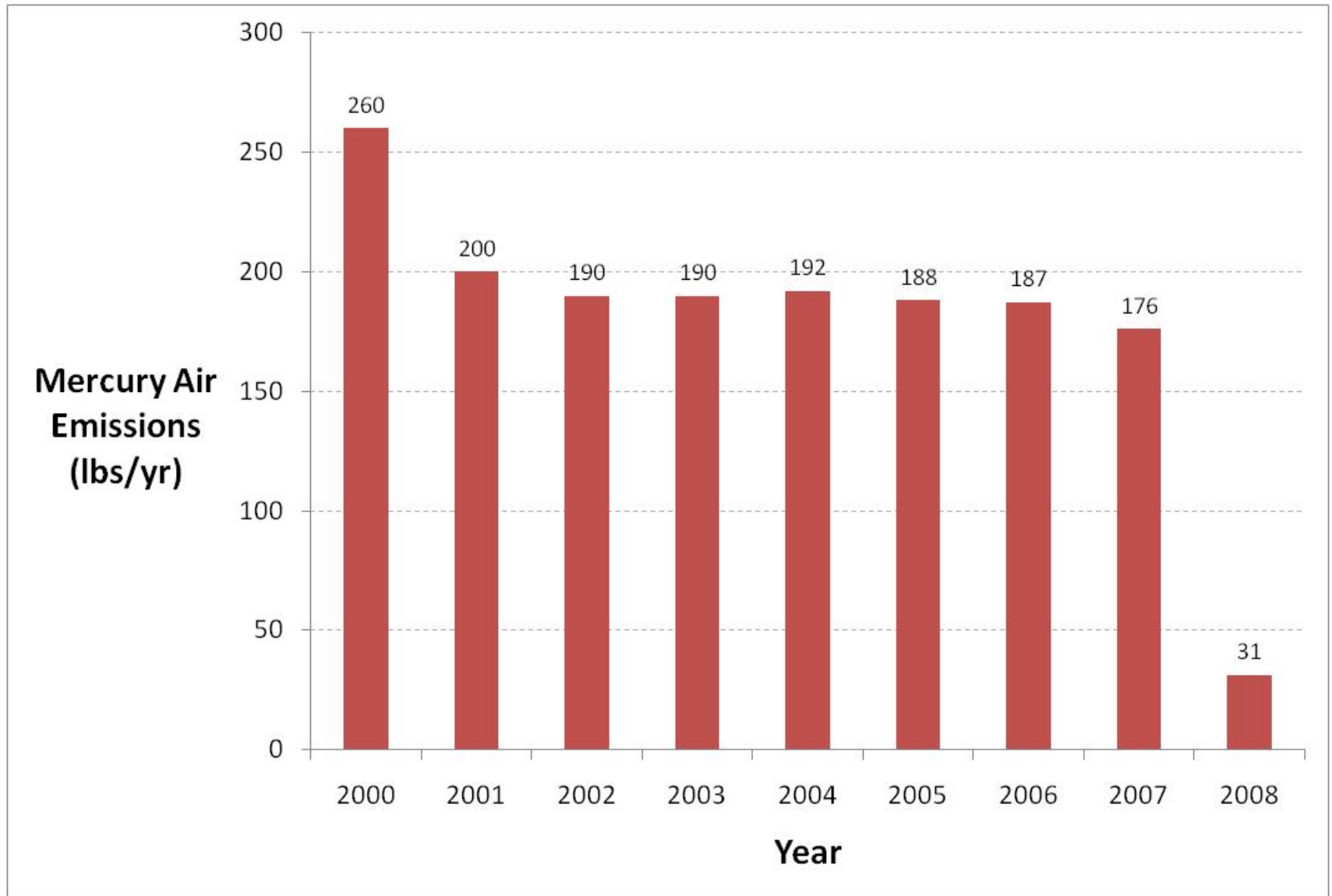
Episodes	Met Data	Evolution	Emissions?
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Location of the Grand Bay NERR sampling site, along with large point sources of RGM in the region, based on the EPA's 2002 National Emissions Inventory

* Hg emissions included in 2002 NEI, but do not appear to be in 2000-2008 TRI
 ** Hg emissions included in 2002 NEI but incineration ceased in January 2001
 *** Significant Hg emissions in 2002 NEI, but negligible emissions reported in 2008 TRI

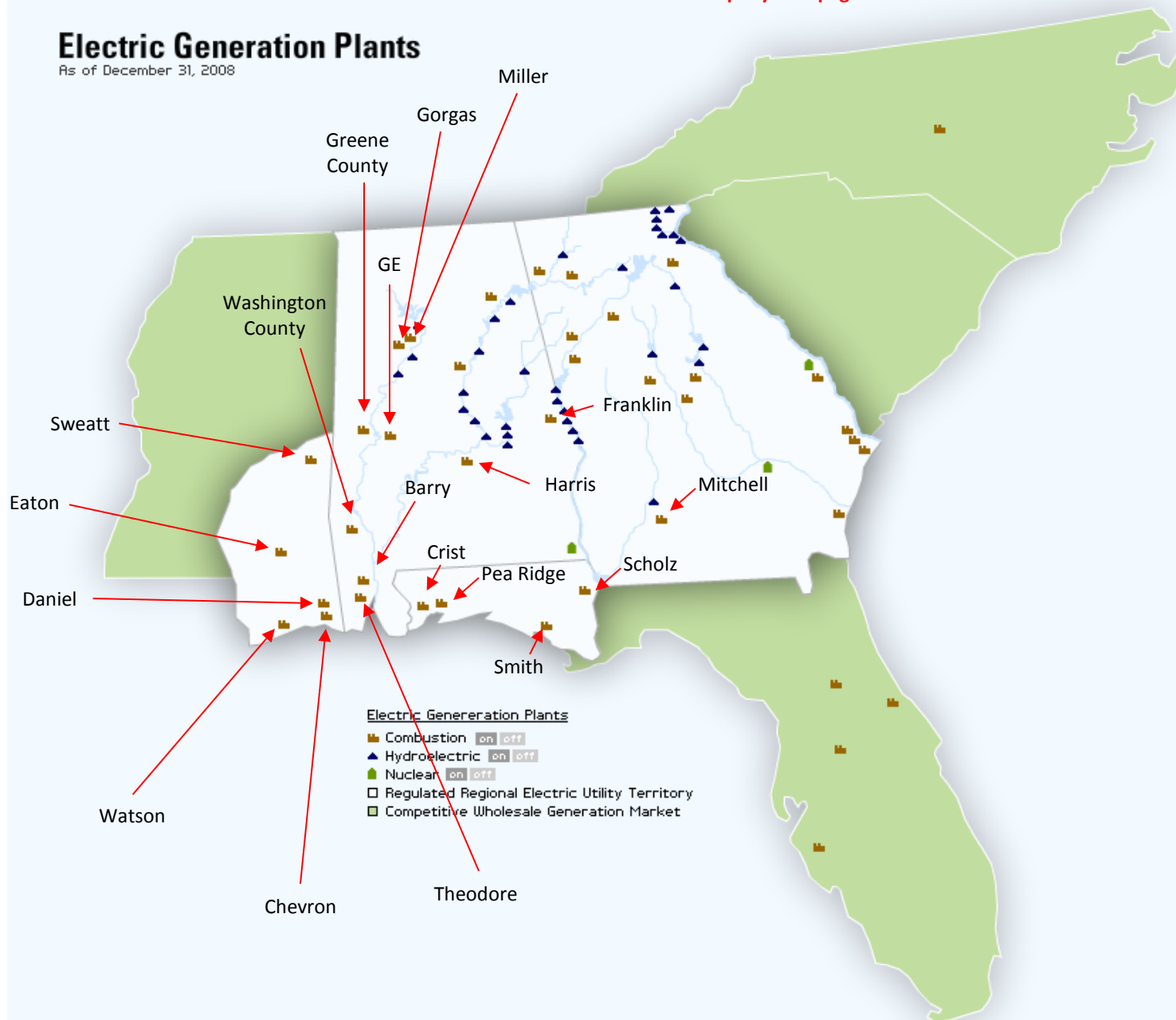
Mercury Air Emissions from Charles R. Lowman Power Plant as reported to the Toxic Release Inventory



Information from Southern Company web page

Electric Generation Plants

As of December 31, 2008



A satellite image of the Gulf of Mexico, showing the coastline of North America to the north and west, and Central America to the south. The ocean is a deep blue, with a prominent white, swirling cloud pattern in the lower-left quadrant. The word "Thanks!" is centered in white text.

Thanks!

