Atmospheric Mercury Model Intercomparisons

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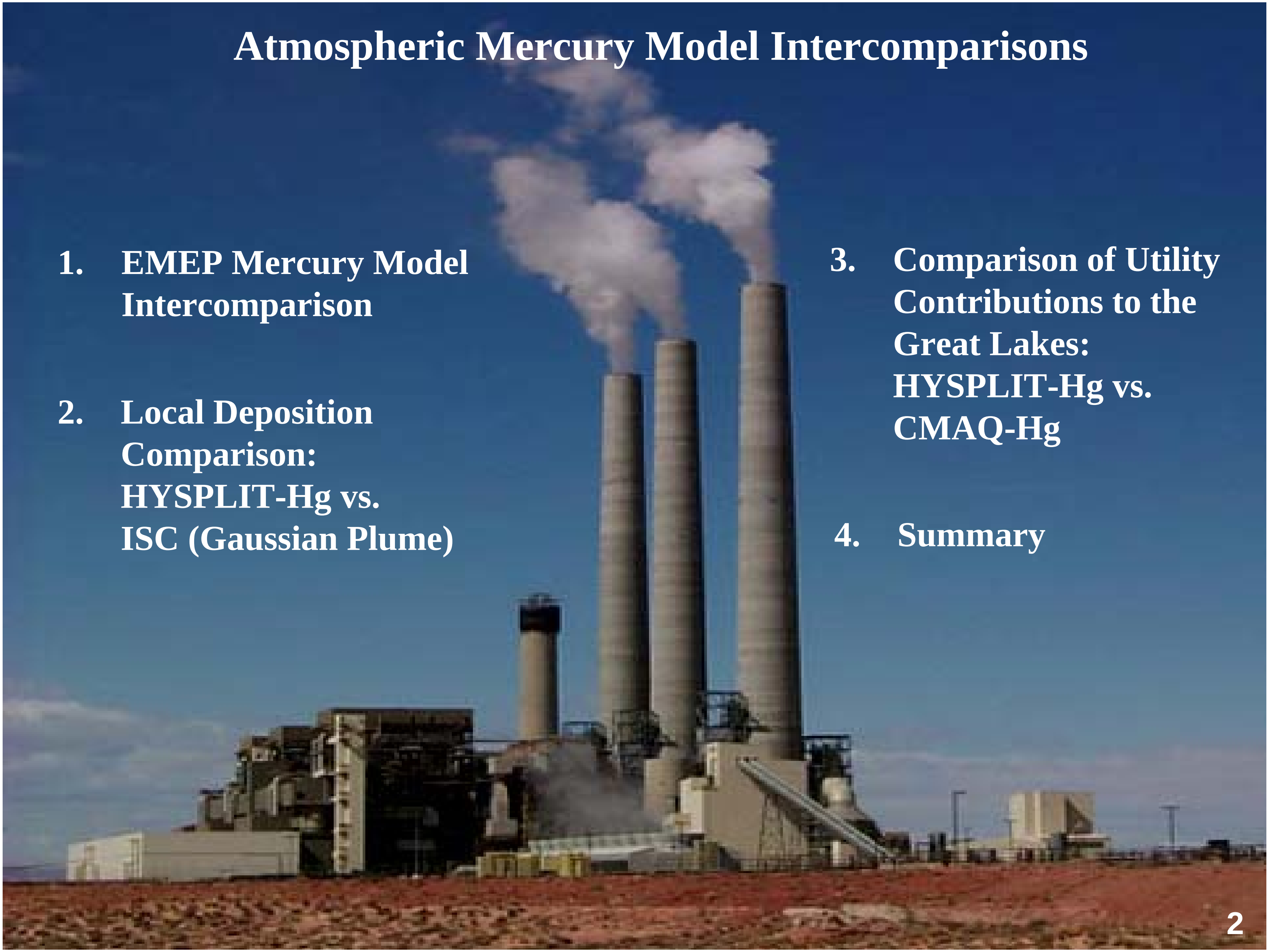
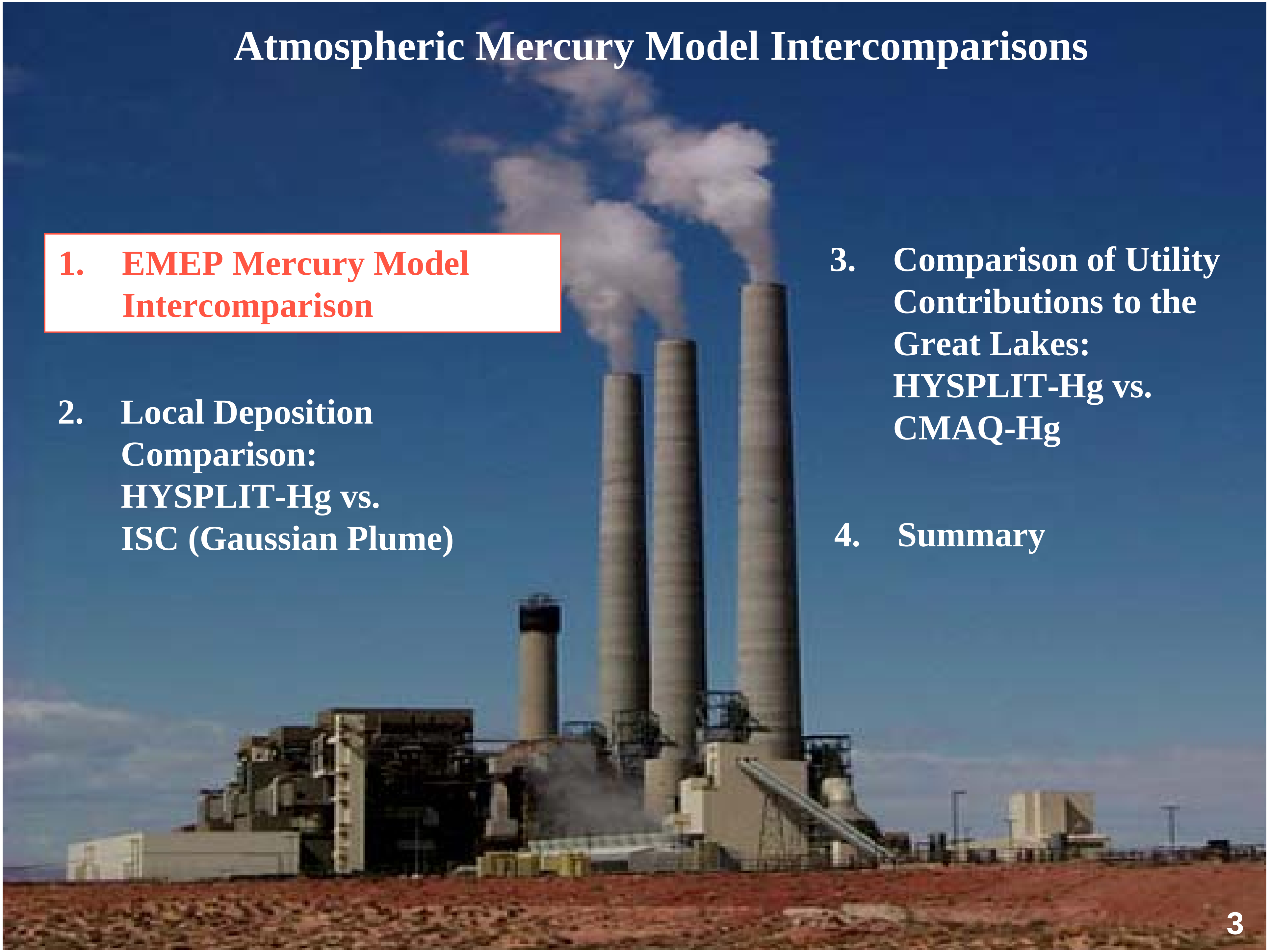
Presentation at
Collaborative Meeting on Modeling Mercury
in Freshwater Environments
Niagara Falls, NY, January 19-20, 2006



Atmospheric Mercury Model Intercomparisons

1. **EMEP Mercury Model Intercomparison**
2. **Local Deposition Comparison: HYSPLIT-Hg vs. ISC (Gaussian Plume)**
3. **Comparison of Utility Contributions to the Great Lakes: HYSPLIT-Hg vs. CMAQ-Hg**
4. **Summary**



Atmospheric Mercury Model Intercomparisons

1. EMEP Mercury Model Intercomparison

2. Local Deposition Comparison: HYSPLIT-Hg vs. ISC (Gaussian Plume)

3. Comparison of Utility Contributions to the Great Lakes: HYSPLIT-Hg vs. CMAQ-Hg

4. Summary

Participants

D. Syrakov ………………………………. **Bulgaria….NIMH**

A. Dastoor, D. Davignon ………………. **Canada...... MSC-Can**

J. Christensen …………………………… **Denmark…NERI**

G. Petersen, R. Ebinghaus ……………..... **Germany…GKSS**

J. Pacyna ……………………………….. **Norway…..NILU**

J. Munthe, I. Wängberg ……………….... **Sweden….. IVL**

R. Bullock ………………………………**USA………EPA**

M. Cohen, R. Artz, R. Draxler …………**USA………NOAA**

C. Seigneur, K. Lohman ………………...**USA……... AER/EPRI**

A. Ryaboshapko, I. Ilyin, O.Travnikov…**EMEP……MSC-E**

Intercomparison Conducted in 3 Stages

I. Comparison of chemical schemes for a cloud environment

II. Air Concentrations in Short Term Episodes

III. Long-Term Deposition and Source-Receptor Budgets

Participating Models

Model Acronym	***Model Name*** **and Institution**	**Stage**		
		I	**II**	**III**
CAM	*Chemistry of Atmos. Mercury model*, Environmental Institute, Sweden			
MCM	*Mercury Chemistry Model*, Atmos. & Environmental Research, USA			
CMAQ	*Community Multi-Scale Air Quality model*, US EPA			
ADOM	*Acid Deposition and Oxidants Model*, GKSS Research Center, Germany			
MSCE-HM	*MSC-E heavy metal regional model*, EMEP MSC-E			
GRAHM	*Global/Regional Atmospheric Heavy Metal model*, Environment Canada			
EMAP	*Eulerian Model for Air Pollution*, Bulgarian Meteo-service			
DEHM	*Danish Eulerian Hemispheric Model*, National Environmental Institute			
HYSPLIT	*Hybrid Single Particle Lagrangian Integrated Trajectory model*, US NOAA			
MSCE-HM-Hem	*MSC-E heavy metal hemispheric model*, EMEP MSC-E			

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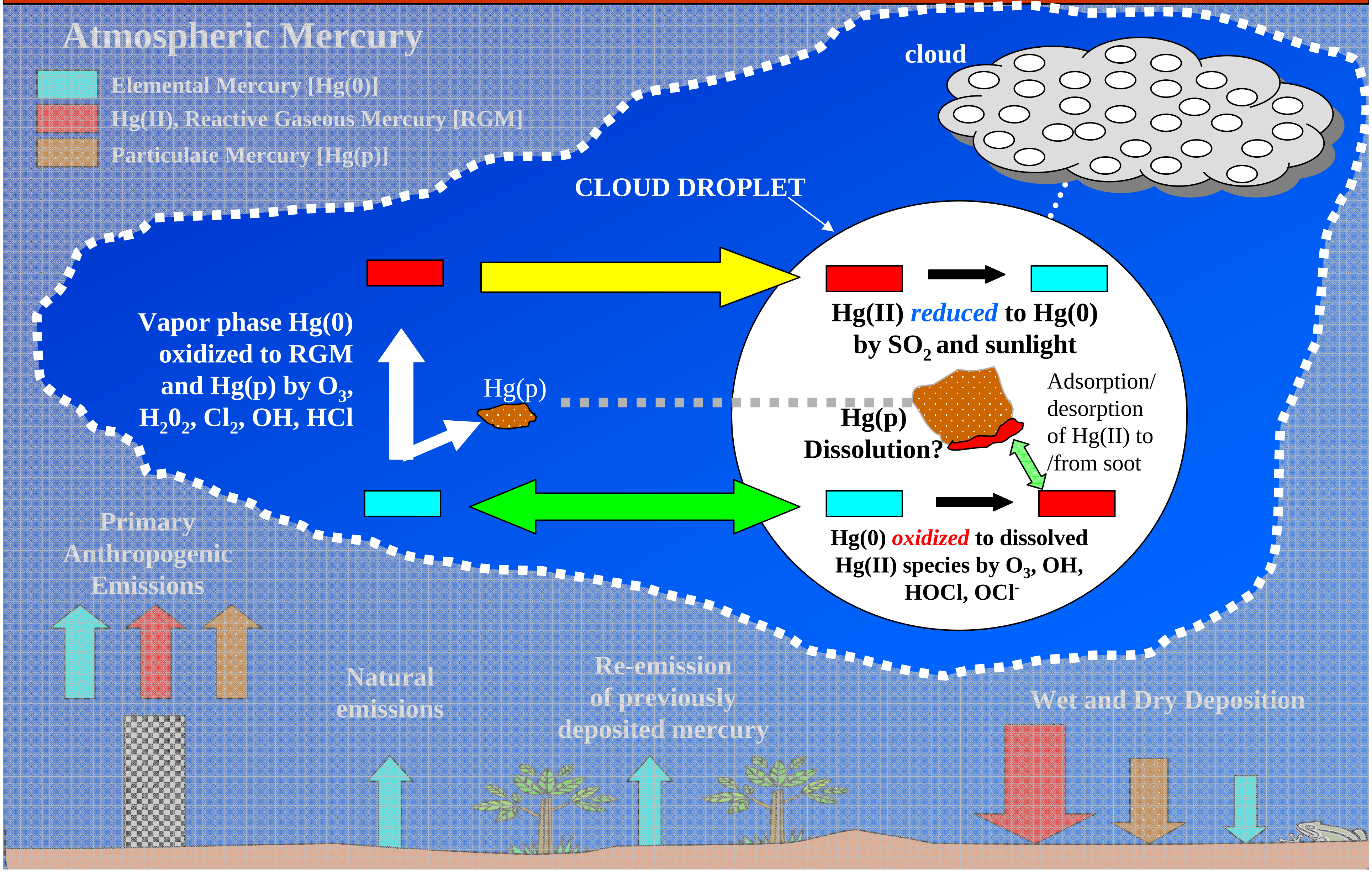
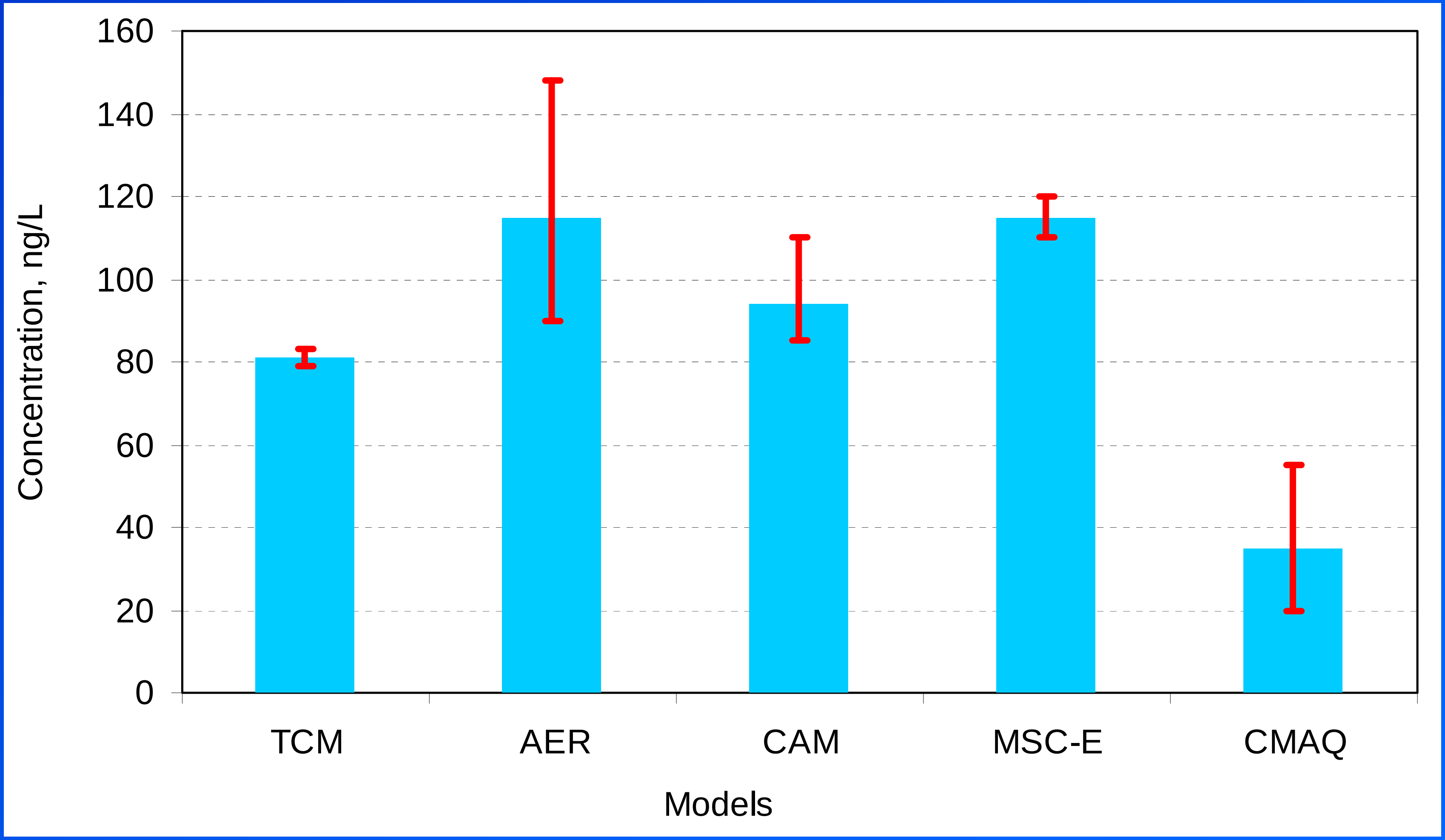

Atmospheric Mercury
Elemental Mercury [Hg(0)]
Hg(II), Reactive Gaseous Mercury [RGM]
Particulate Mercury [Hg(p)]
cloud
CLOUD DROPLET
Vapor phase Hg(0) oxidized to RGM and Hg(p) by O_3, H_2O_2, Cl_2, OH, HCl
Hg(p)
Hg(II) *reduced* to Hg(0) by SO_2 and sunlight
Hg(p) Dissolution?
Adsorption/ desorption of Hg(II) to /from soot
Hg(0) *oxidized* to dissolved Hg(II) species by O_3, OH, HOCl, OCl^-
Primary Anthropogenic Emissions
Natural emissions
Re-emission of previously deposited mercury
Wet and Dry Deposition

Variation of Hg concentrations (ng/L)



Stage I Publications:

2001 Ryaboshapko, A., Ilyin, I., Bullock, R., Ebinghaus, R., Lohman, K., Munthe, J., Petersen, G., Seigneur, C., Wangberg, I. *Intercomparison Study of Numerical Models for Long Range Atmospheric Transport of Mercury*. ***Stage I. Comparisons of Chemical Modules for Mercury Transformations in a Cloud/Fog Environment***. Meteorological Synthesizing Centre – East, Moscow, Russia.

2002 Ryaboshapko, A., Bullock, R., Ebinghaus, R., Ilyin, I., Lohman, K., Munthe, J., Petersen, G., Seigneur, C., Wangberg, I. ***Comparison of Mercury Chemistry Models***. Atmospheric Environment **36**, 3881-3898.

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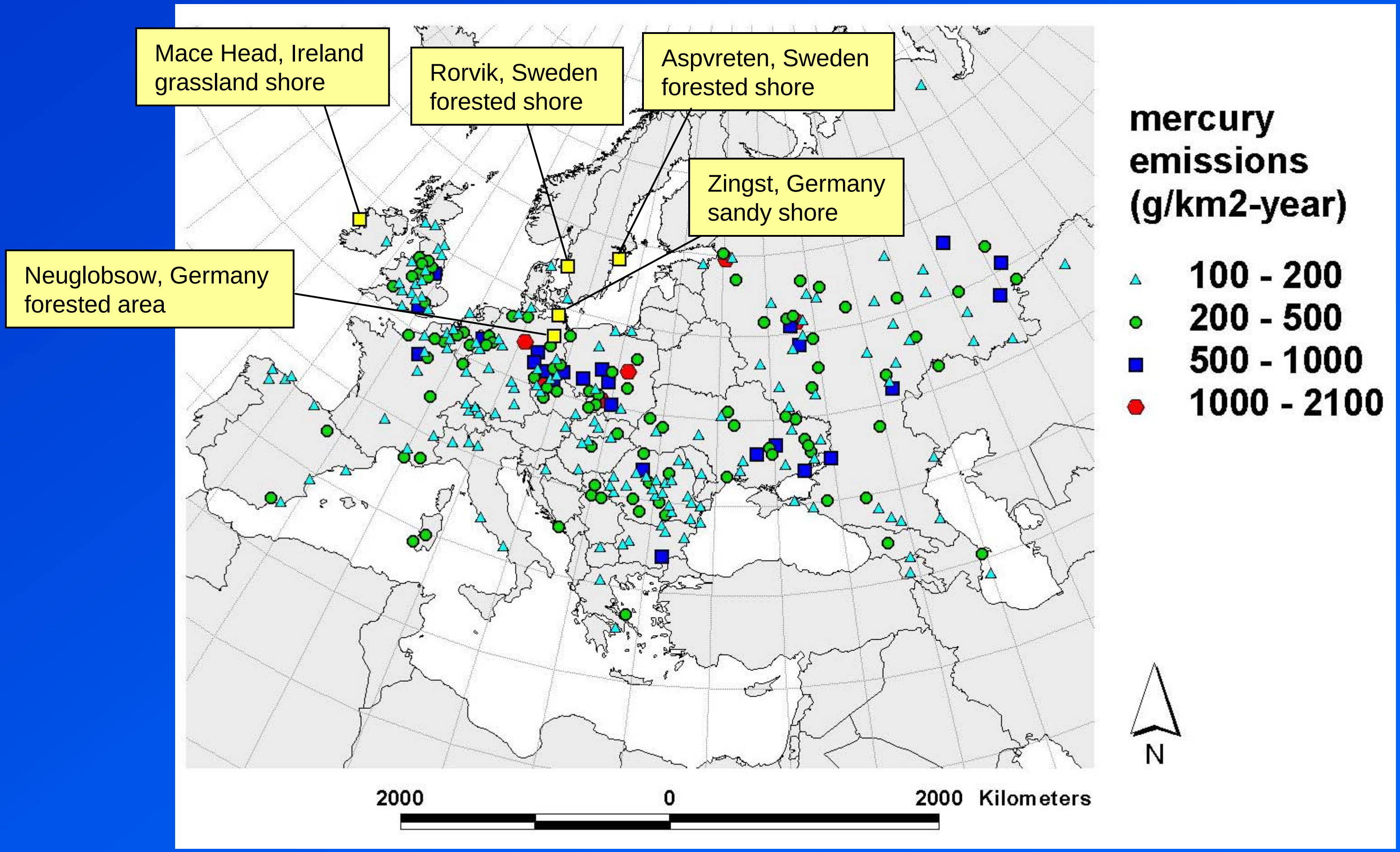
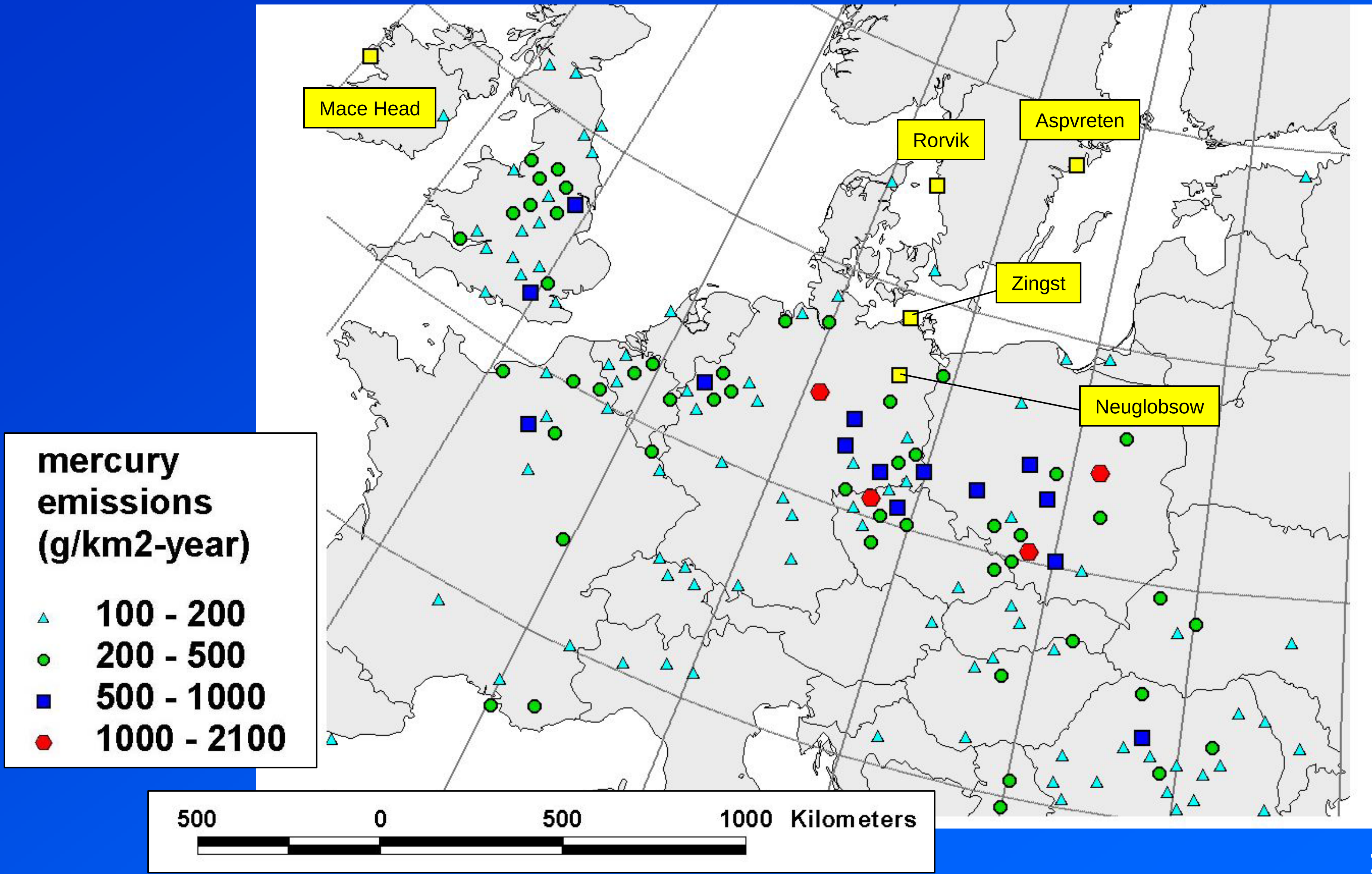
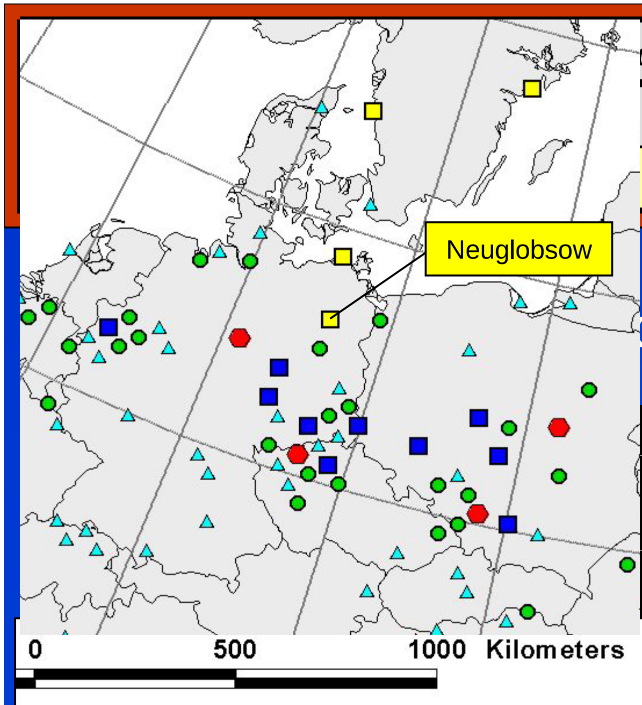
Model	**CMAQ-Hg**	**ADOM**	**HYSPLIT**	**EMAP**	**GRAHM**	**DEHM**	**MSCE-Hg**
Model type	Eulerian	Eulerian	Lagrangian	Eulerian	Eulerian	Eulerian	Eulerian
Scale/ Domain	regional/ Central and Northern Europe	regional/ Central Europe	regional/ EMEP	regional/ EMEP	global	Hemispheric	regional/ EMEP
Source of meteorological data	ECMWF TOGA reanalysis (MM5)	HIRLAM	NCEP/NCAR (MM-5)	SDA, NCEP/NCAR reanalysis	Canadian Meteorolo-gical Centre	NCEP / NCAR reanalysis	SDA, NCEP/NCAR reanalysis
Model top height (km)	15	10	15	5	30	15	3.9
Horizontal resolution (km, unless noted differently)	36 x 36	55 x 55	36 x 36, 108 x 108	50 x 50	1º x 1º	50 x 50 150 x 150	50 x 50
Hg(0) boundary condition (ng/m^3)	1.7	1.5	1.5	1.5	No	1.5	1.6 - 1.7
RGM boundary condition (pg/m^3)	17	2	5	10	none	0	0
TPM boundary condition (pg/m^3)	17	20	10	10	none	0	20
Gas-phase oxidation agents	O_3, H_2O_2, Cl_2, $OH^{\cdot}$	O_3	O_3, H_2O_2, Cl_2, HCl	O_3, $OH^{\bullet}$	O_3	O_3	O_3 (f)
Liquid-phase oxidation agents	O_3, $OH^{\bullet}$, HOCl, OCl^-	O_3	O_3, $OH^{\bullet}$, HOCl, OCl^-	O_3	O_3	O_3	O_3
Liquid-phase reduction agents	$SO_3^=$, hv, HO_2	$SO_3^=$	$SO_3^=$, HO_2	$SO_3^=$	$SO_3^=$	$SO_3^=$	$SO_3^=$, HO_2

Anthropogenic Mercury Emissions Inventory and Monitoring Sites for Phase II

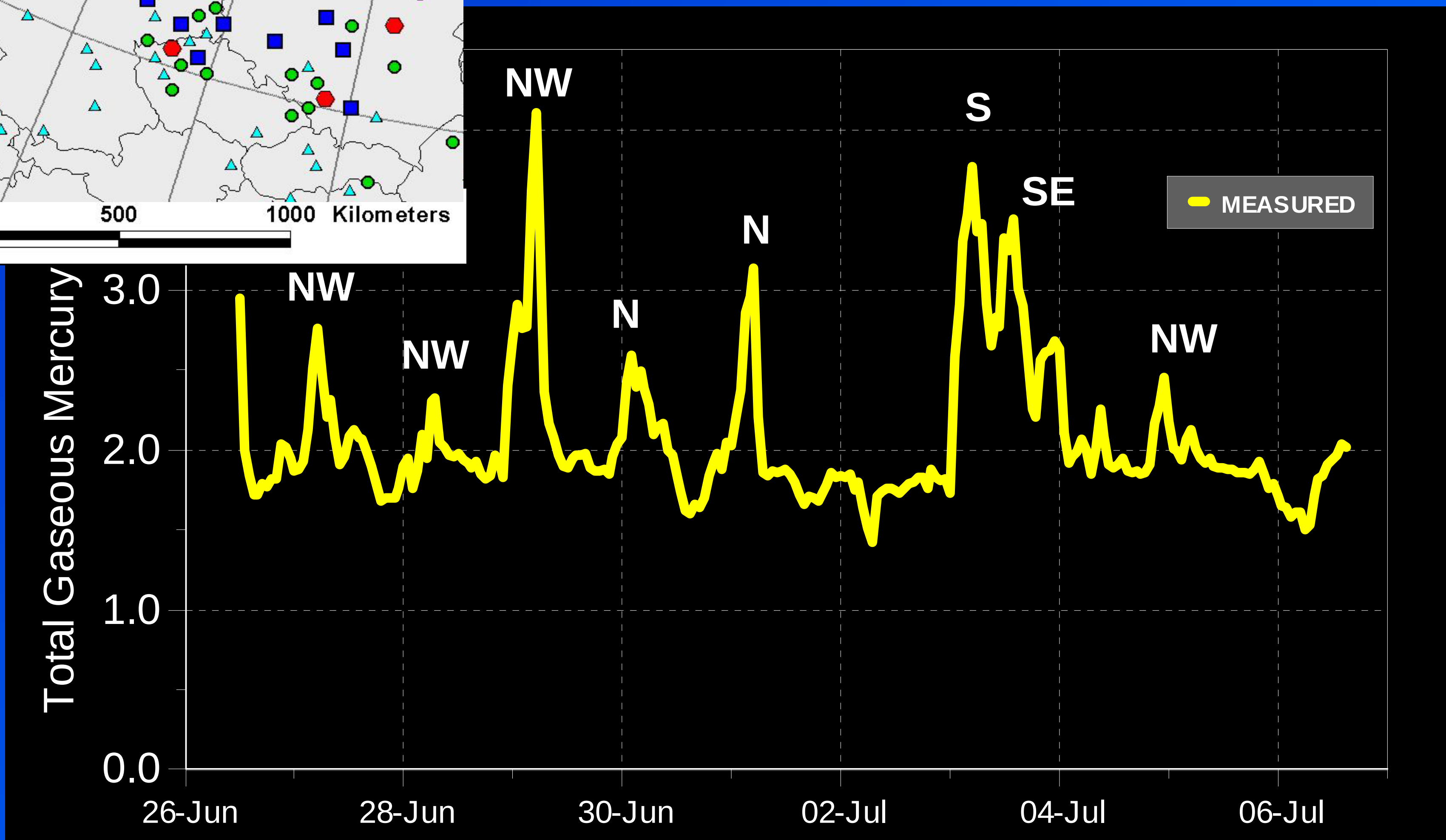
(note: only showing largest emitting grid cells)



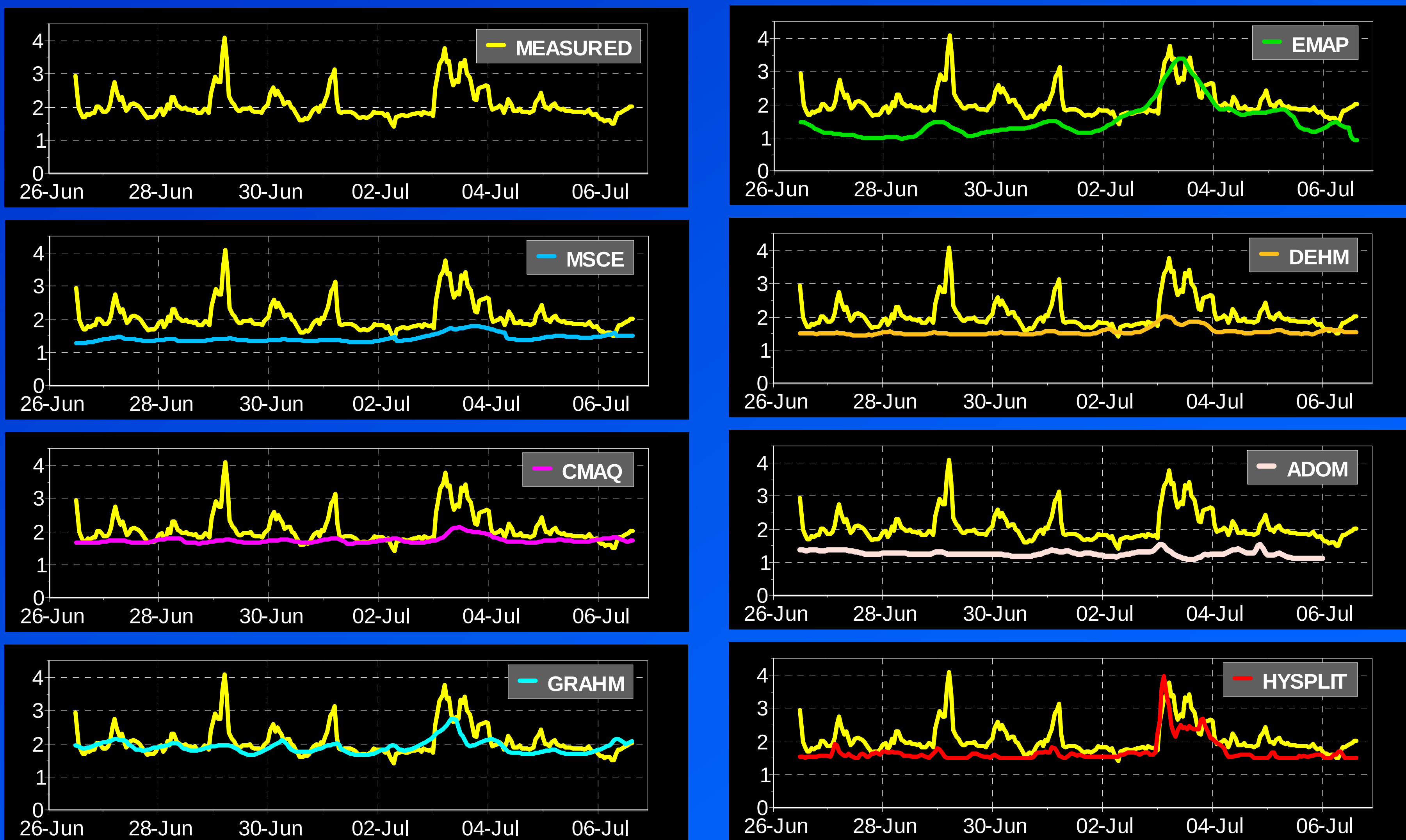

Mace Head
Rorvik
Aspvreten
Zingst
Neuglobsow
mercury emissions (g/km2-year)
100 - 200
200 - 500
500 - 1000
1000 - 2100
500
0
500
1000 Kilometers



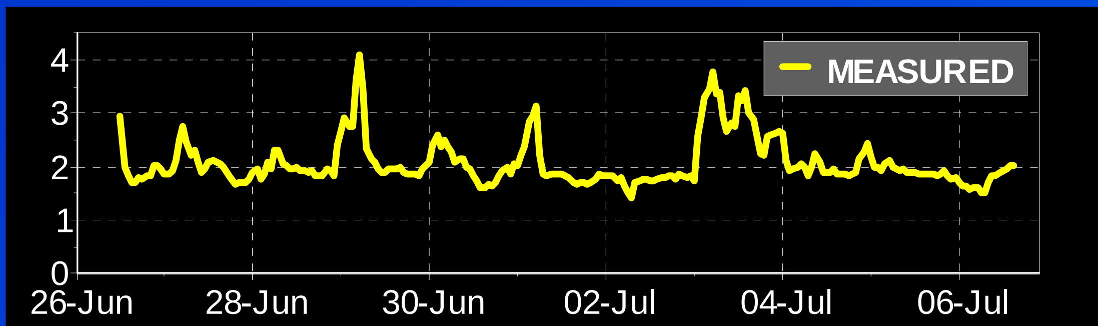
cury at Neuglobsow: June 26 – July 6, 1995



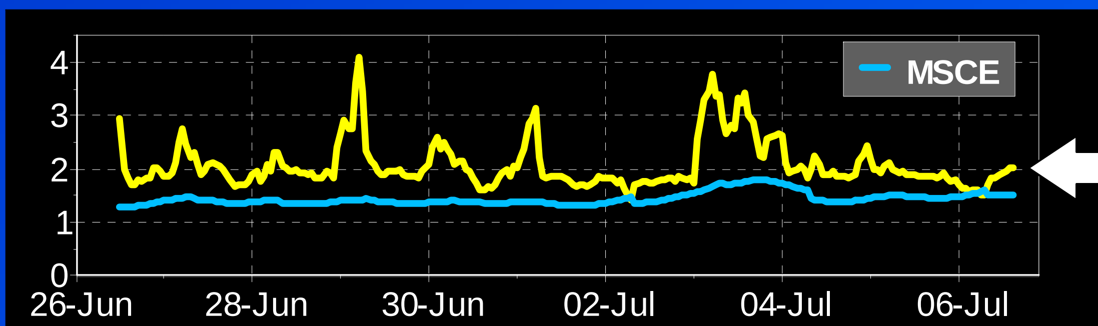
Total Gaseous Mercury (ng/m^3) at Neuglobsow: June 26 – July 6, 1995



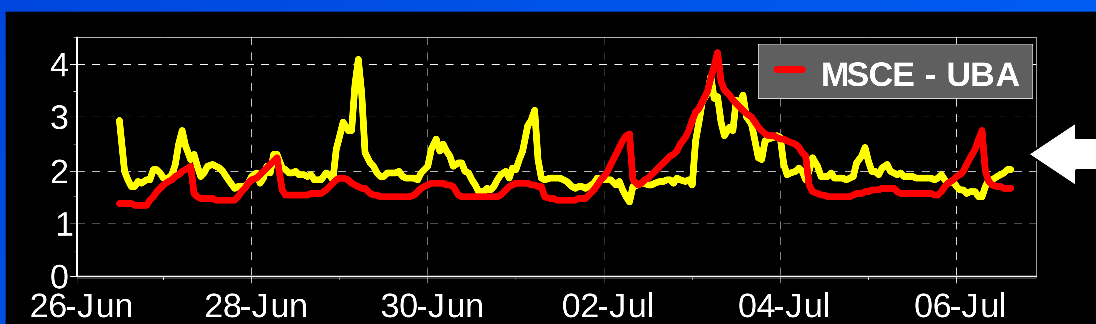
Total Gaseous Mercury (ng/m^3) at Neuglobsow: June 26 – July 6, 1995



The emissions inventory is a critical input to the models…

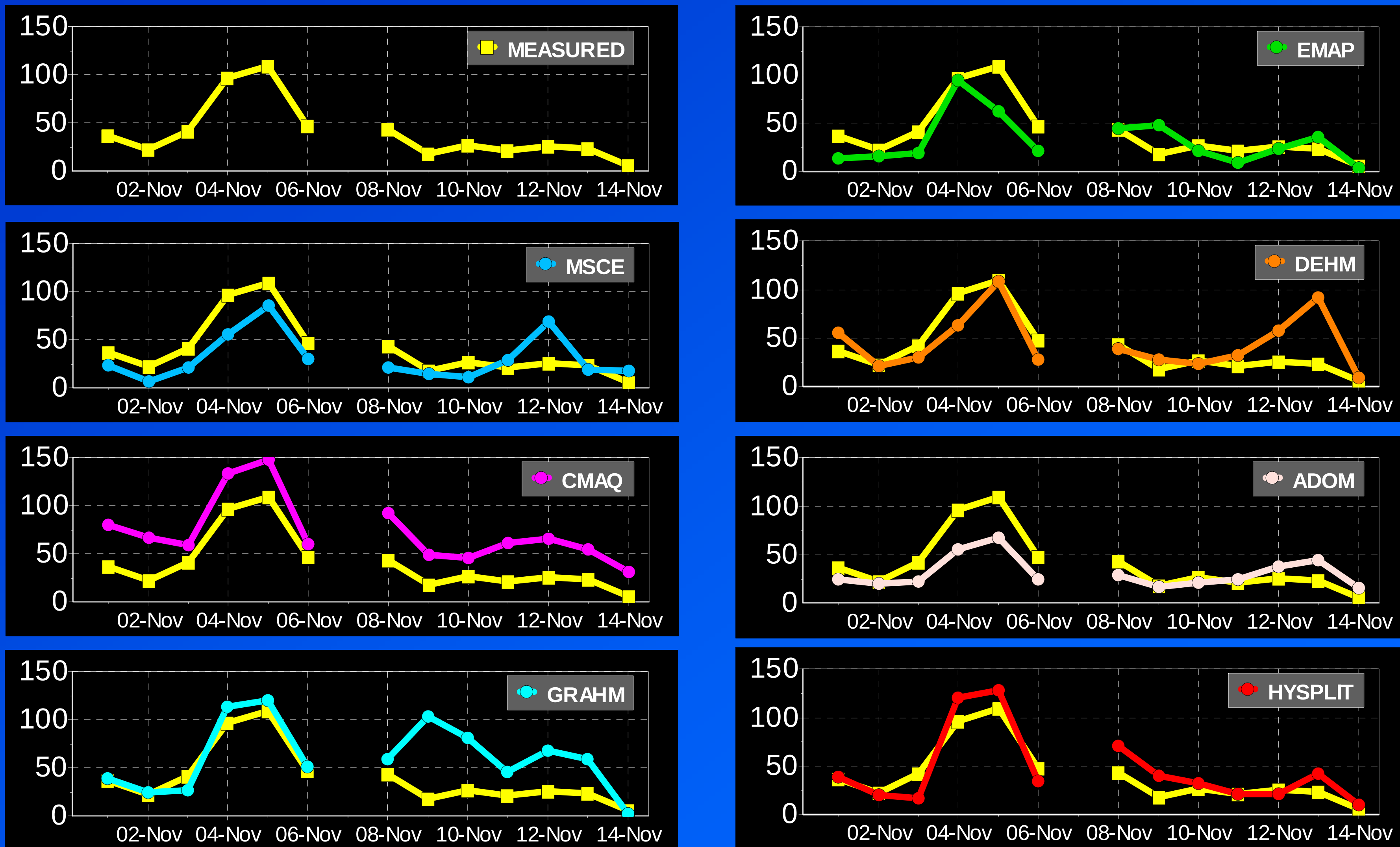


Using default emissions inventory

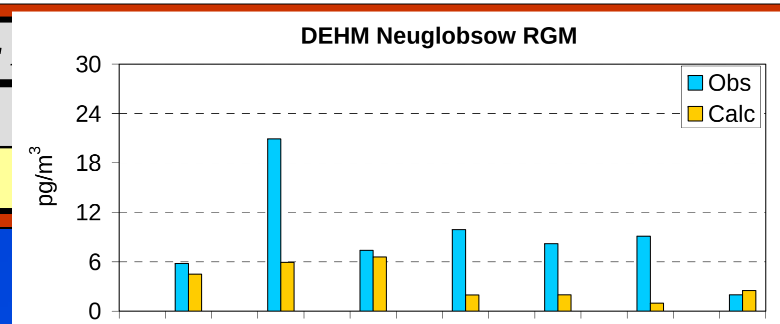


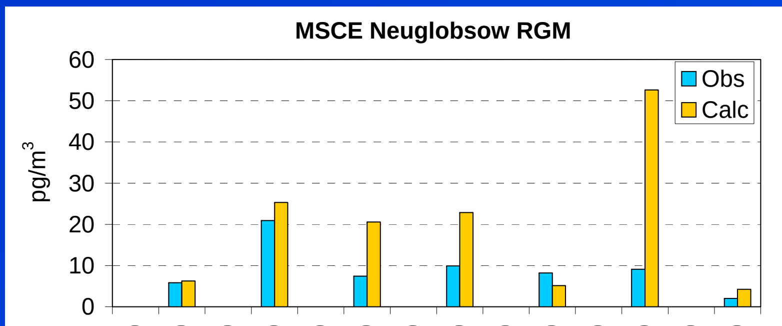
Using alternative emissions inventory

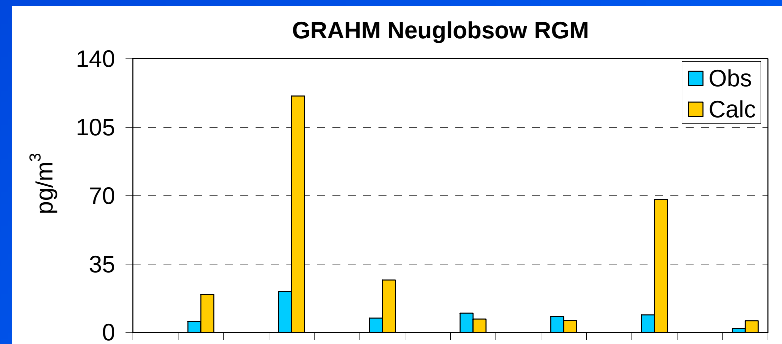
Total *Particulate* Mercury (pg/m^3) at Neuglobsow, Nov 1-14, 1999

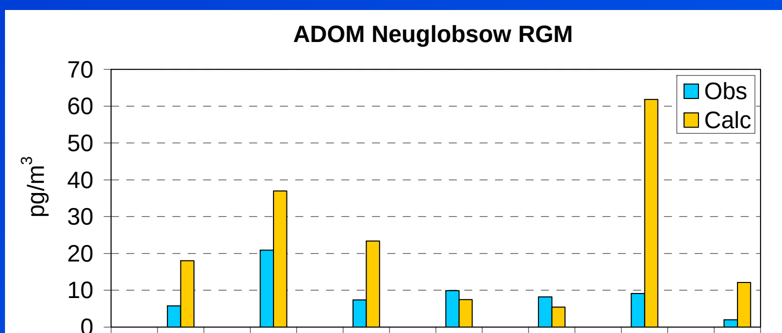


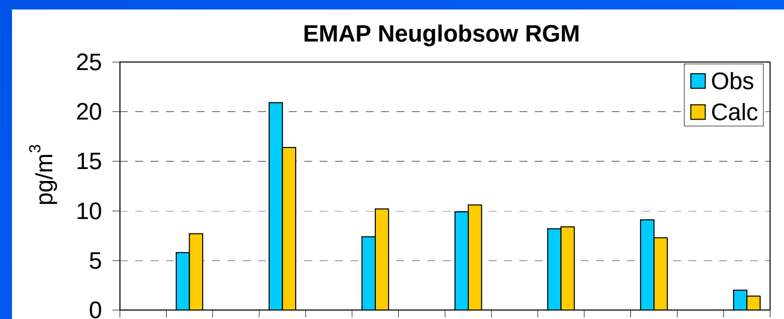
Reactive Gaseous Mercury at Neuglobsow, Nov 1-14, 1999

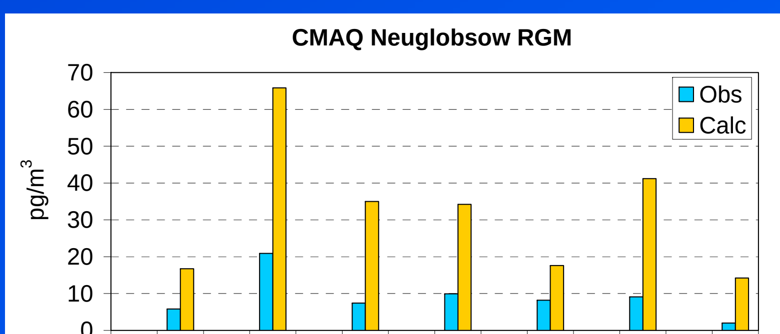


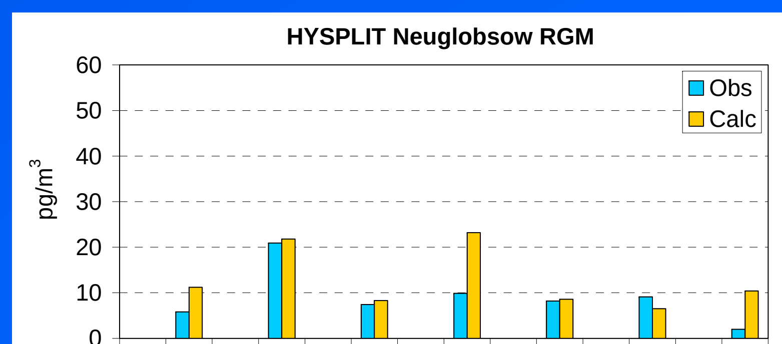




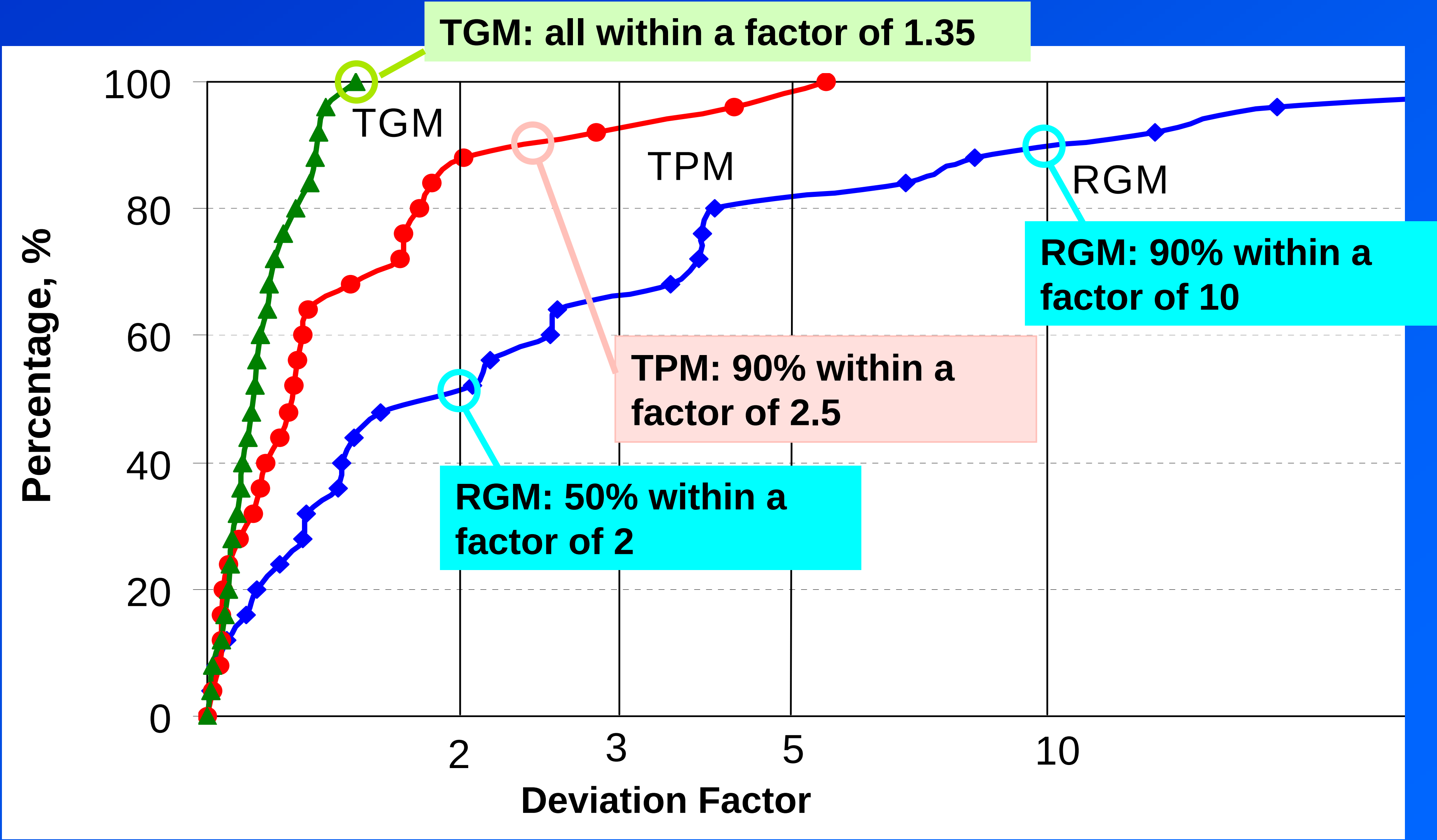








Overall Phase II statistics for 2-week episode means



Stage II Publications:

2003 Ryaboshapko, A., Artz, R., Bullock, R., Christensen, J., Cohen, M., Dastoor, A., Davignon, D., Draxler, R., Ebinghaus, R., Ilyin, I., Munthe, J., Petersen, G., Syrakov, D. *Intercomparison Study of Numerical Models for Long Range Atmospheric Transport of Mercury*. ***Stage II. Comparisons of Modeling Results with Observations Obtained During Short Term Measuring Campaigns***. Meteorological Synthesizing Centre – East, Moscow, Russia.

2005 Ryaboshapko, A., Bullock, R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen, G., Syrakov, D., Artz, R., Davignon, D., Draxler, R., and Munthe, J. *Intercomparison Study of Atmospheric Mercury Models*. ***Phase II. Comparison of Models with Short-Term Measurements***. Submitted to Atmospheric Environment.

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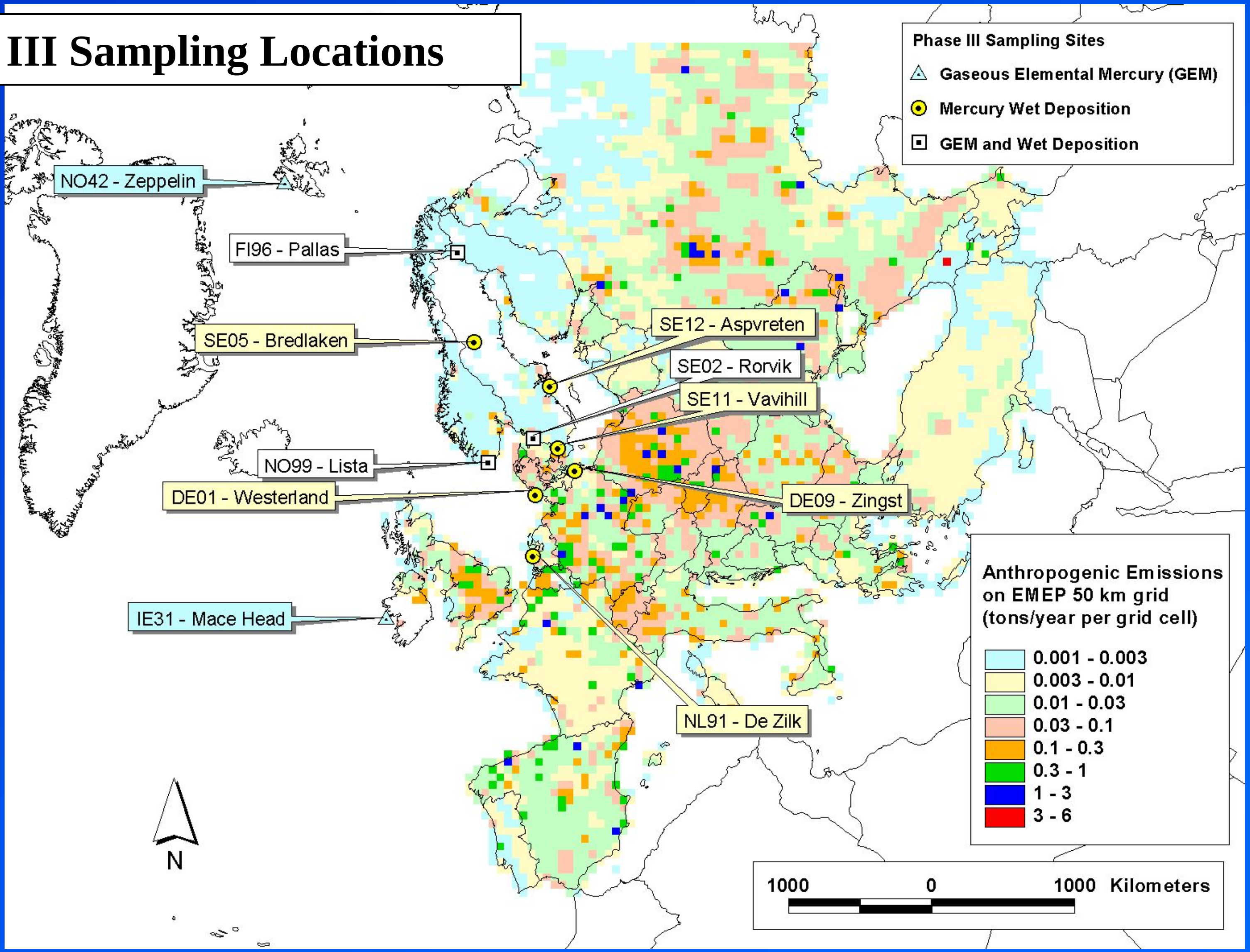
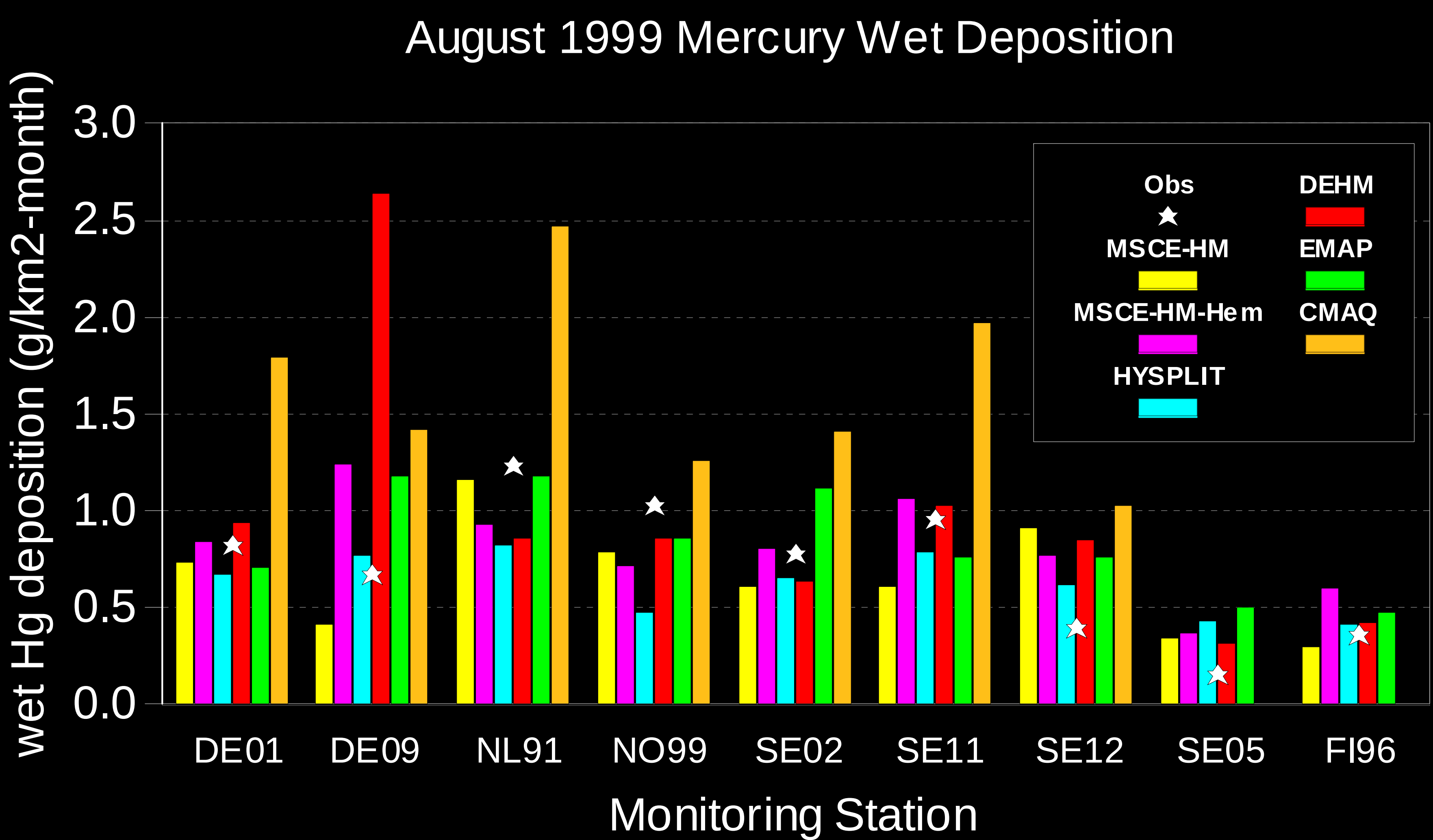
Phase III Sampling Locations



Due to resource constraints, not all models simulated the entire year 1999…

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
CMAQ												
HYSPLIT												
ADOM												
MSCE-HM												
MSCE-HEM												
DEHM												
EMAP												



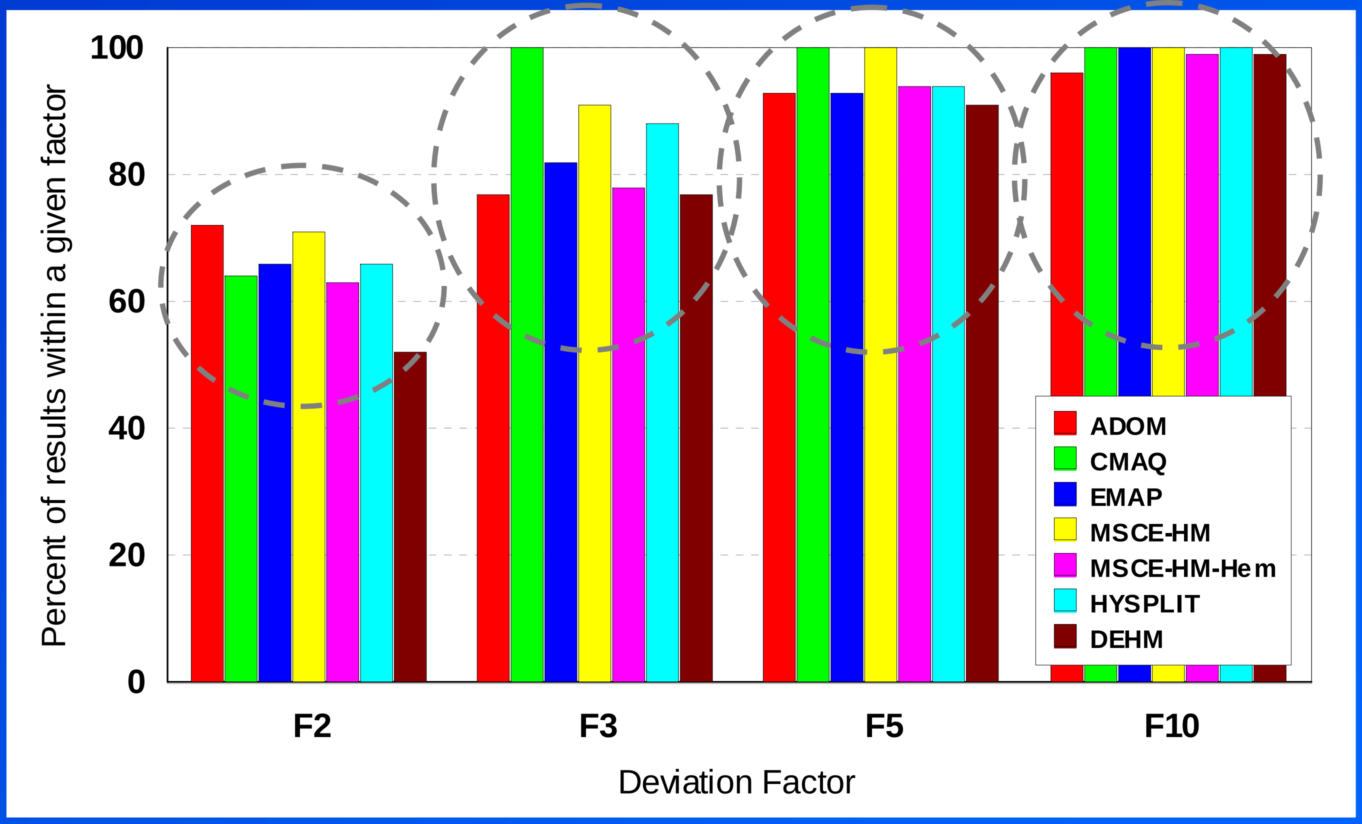
August 1999 Mercury Wet Deposition
wet Hg deposition (g/km2-month)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station
Obs
DEHM
MSCE-HM
EMAP
MSCE-HM-Hem
CMAQ
HYSPLIT

~60% within a factor of 2

~80% within a factor of 3

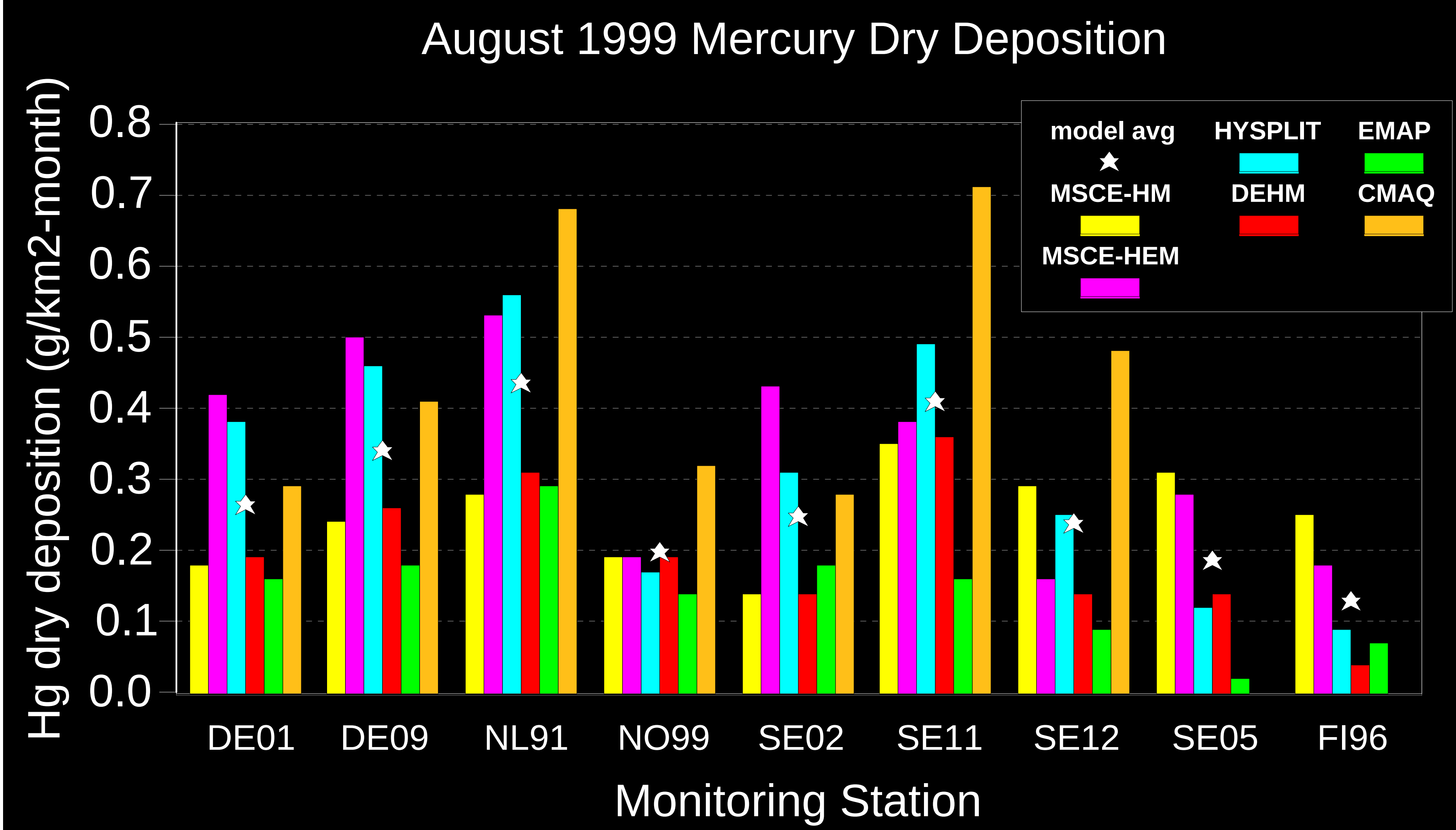
~90% within a factor of 5

~100% within a factor of 10

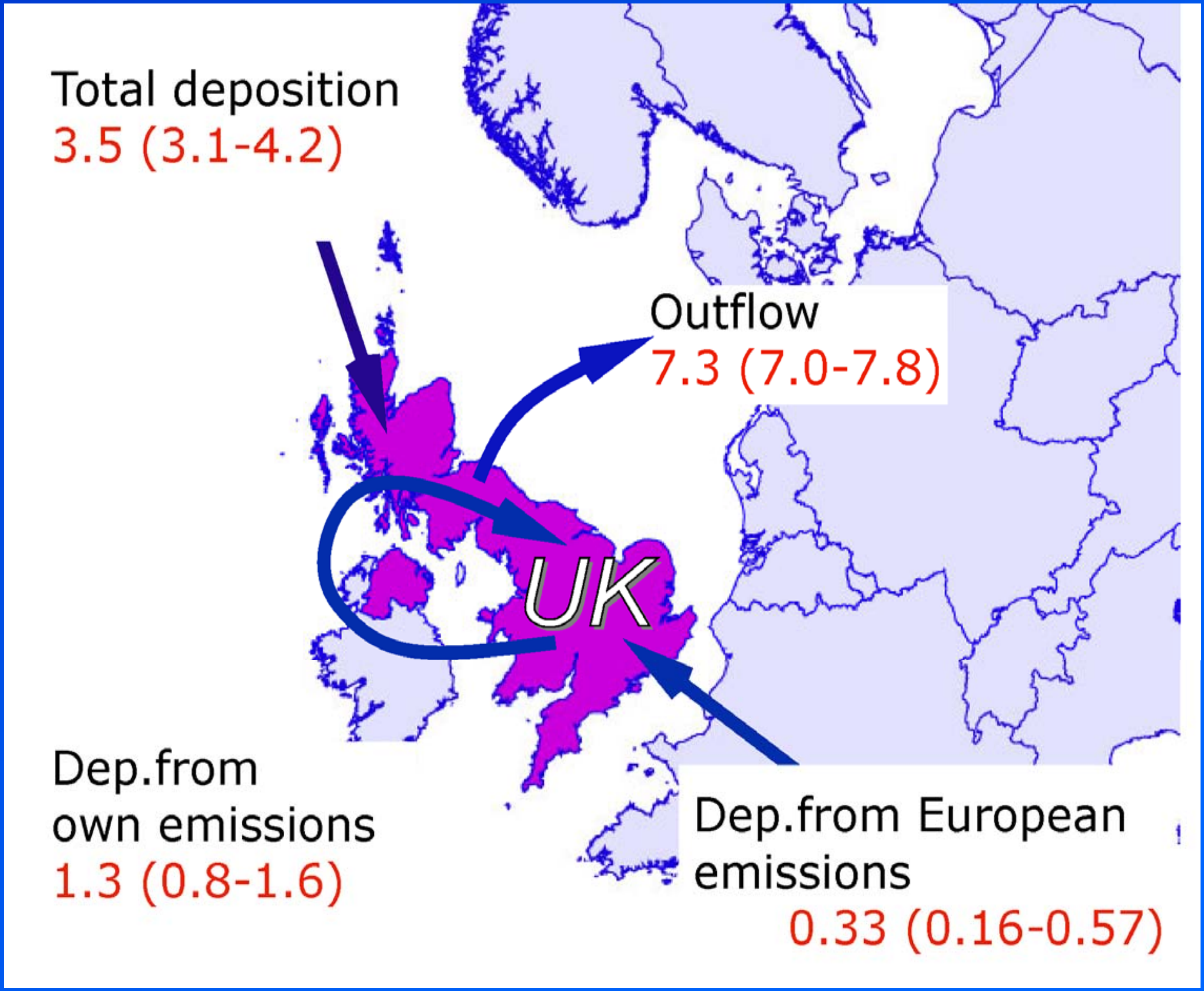


Wet Deposition Summary

- **For *dry deposition*, there are no measurement results to compare the models against;**
- **However, the models can be compared against *each other*…**


August 1999 Mercury Dry Deposition
Hg dry deposition (g/km2-month)
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
model avg
HYSPLIT
EMAP
MSCE-HM
DEHM
CMAQ
MSCE-HEM
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station

Main items of mercury atmospheric balance for the UK in 1999, t/yr

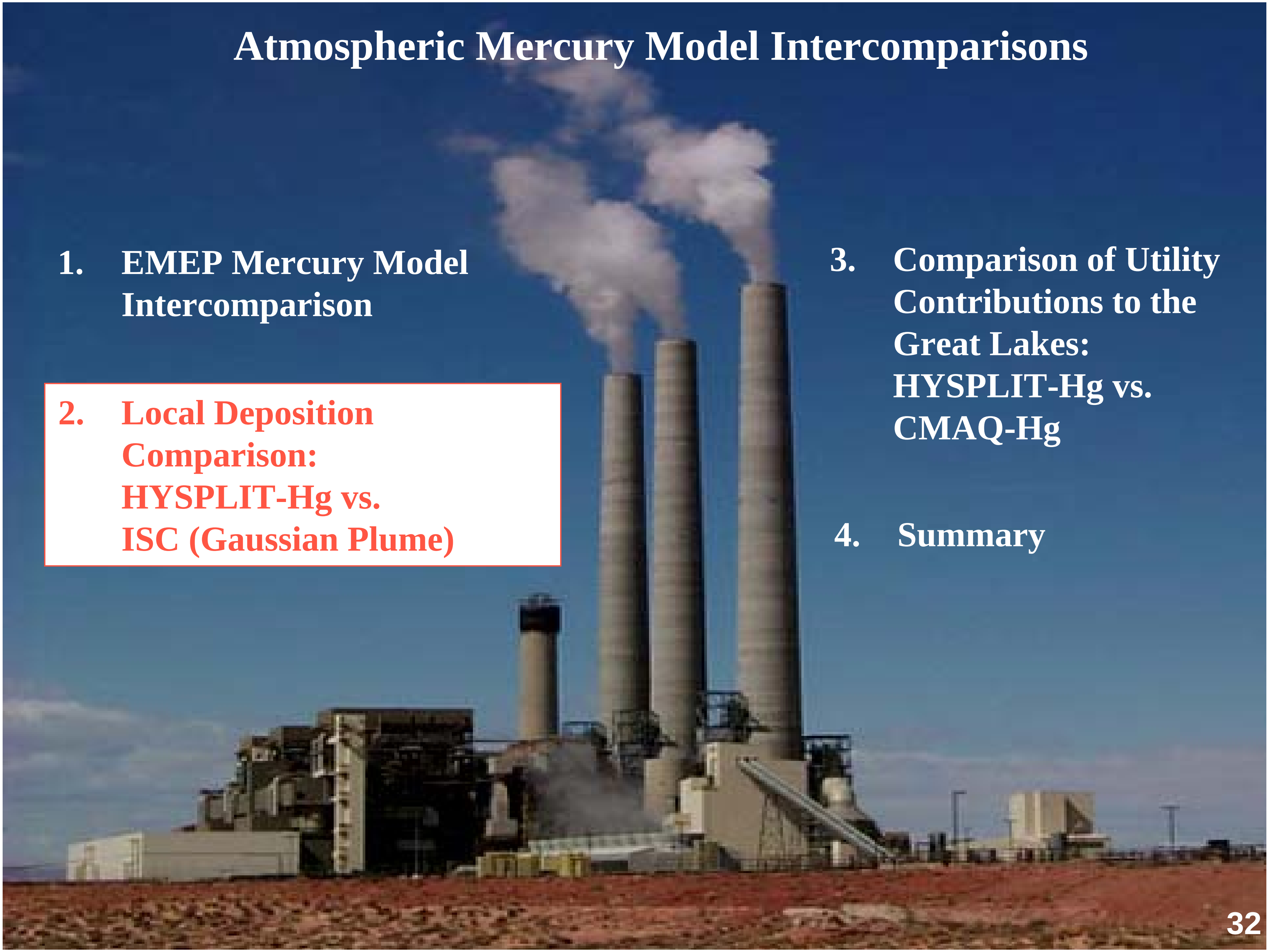


Items of Hg atmospheric balances for the countries in 1999, t/yr [average modeled result *(with ranges in parentheses)*]

Item	The UK	Italy	Poland
Total deposition	**3.5** *(3.1-4.2)*	**4.7** *(3.2-6.6)*	**11.8** *(9.6-13.1)*
Dep. from own emissions	**1.3** *(0.8-1.6)*	**1.3** *(0.6-1.9)*	**7.4** *(4.8-9.1)*
Dep. from European emissions	**0.3** *(0.2-0.6)*	**0.8** *(0.5-1.3)*	**2.1** *(1.4-2.6)*
Outflow	**7.3** *(7.0-7.8)*	**8.4** *(7.9-9.2)*	**18.2** *(16–21)*

Stage III Publication:

2005 Ryaboshapko, A., Artz, R., Bullock, R., Christensen, J., Cohen, M., Draxler, R., Ilyin, I., Munthe, J., Pacyna, J., Petersen, G., Syrakov, D., Travnikov, O. *Intercomparison Study of Numerical Models for Long Range Atmospheric Transport of Mercury*. ***Stage III. Comparison of Modelling Results with Long-Term Observations and Comparison of Calculated Items of Regional Balances***. Meteorological Synthesizing Centre – East, Moscow, Russia.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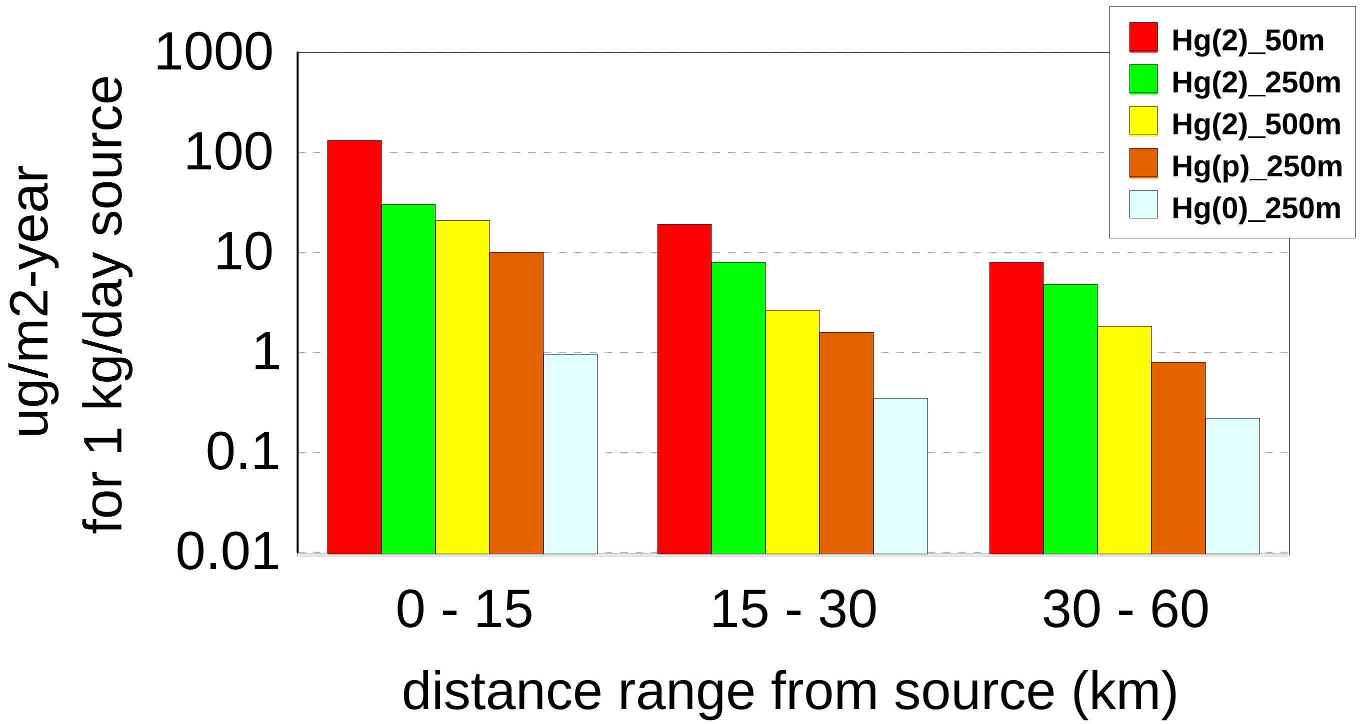
32

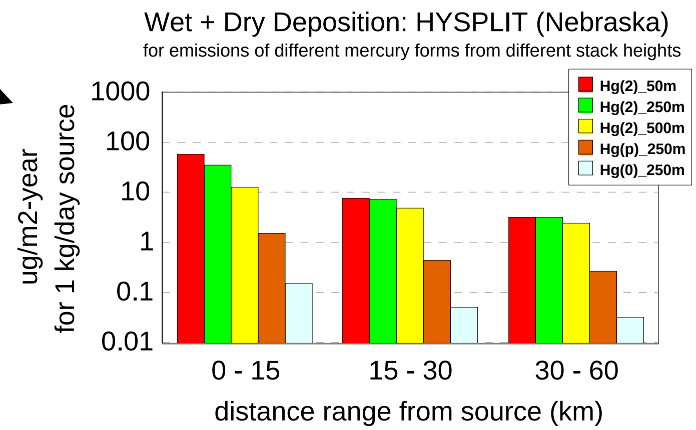
Wet + Dry Deposition: ISC (Kansas City)

for emissions of different mercury forms from different stack heights



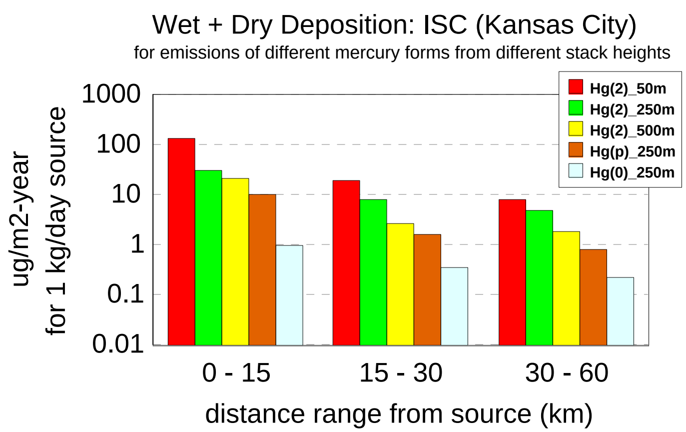
Calculated from data used to produce Appendix A of USEPA (2005): Clean Air Mercury Rule (CAMR) Technical Support Document: Methodology Used to Generate Deposition, Fish Tissue Methylmercury Concentrations, and Exposure for Determining Effectiveness of Utility Emissions Controls: Analysis of Mercury from Electricity Generating Units

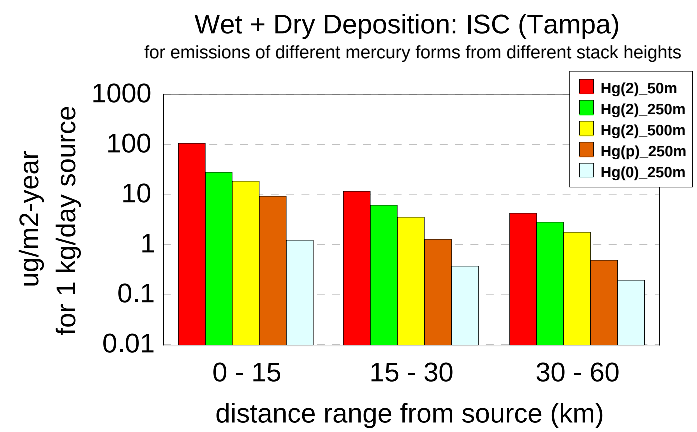
HYSPLIT 1996

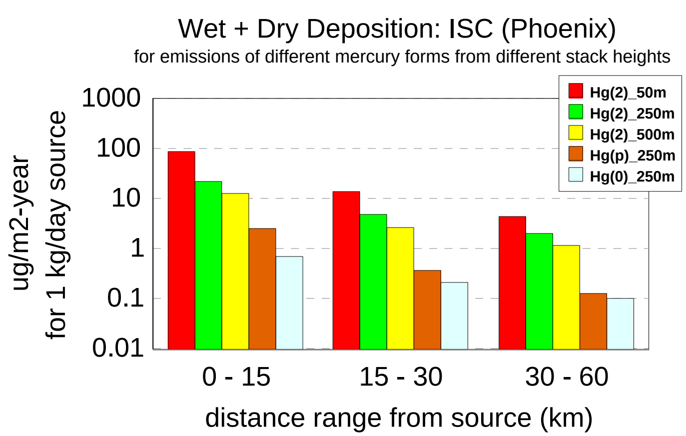


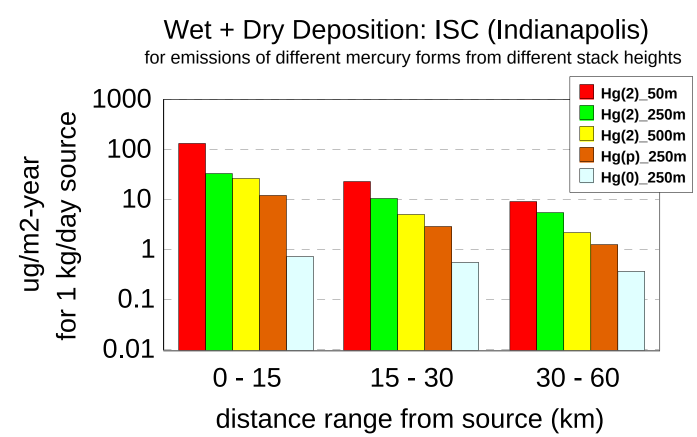
Different Time Periods and Locations, but Similar Results

ISC: 1990-1994

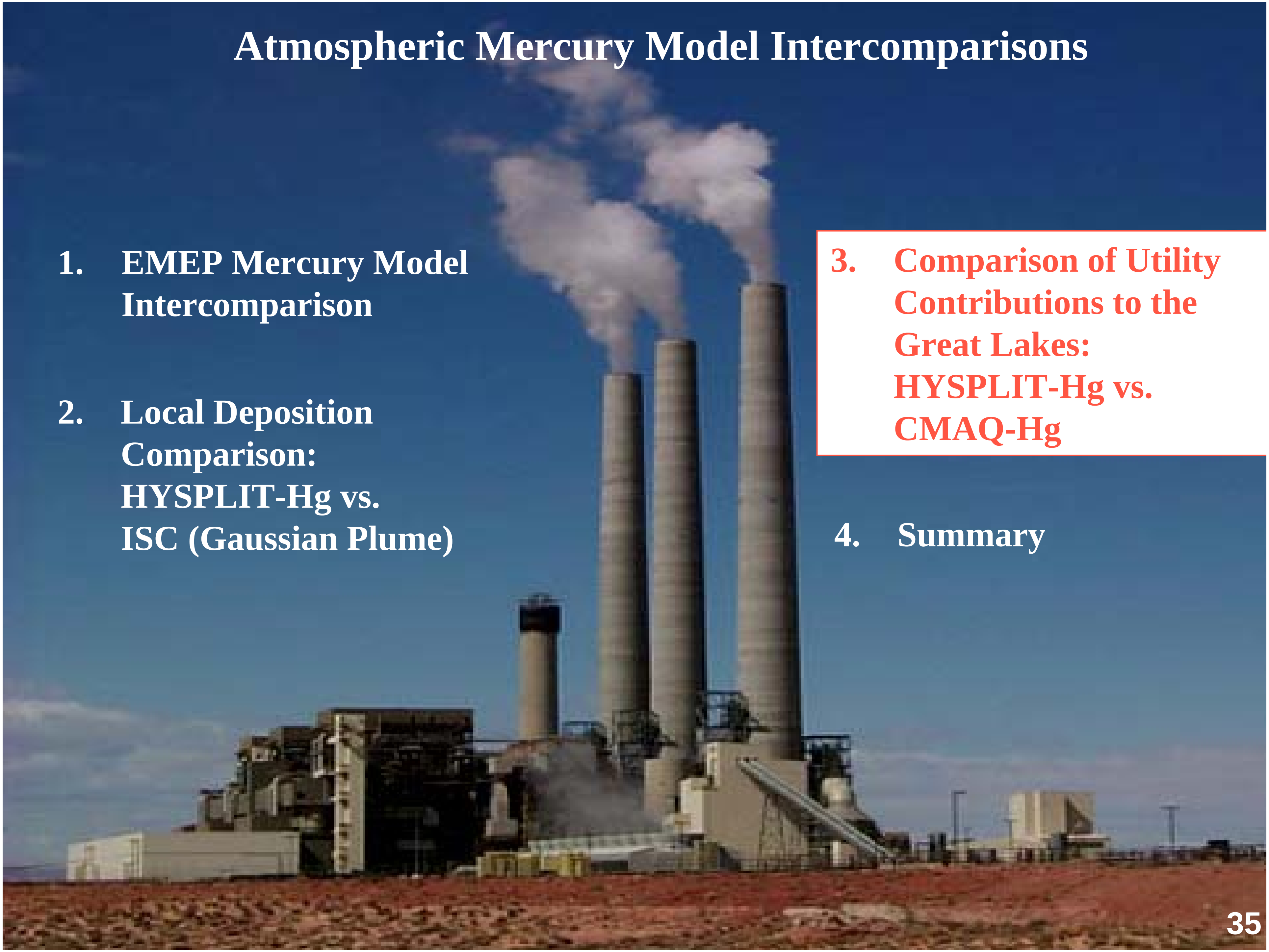








Atmospheric Mercury Model Intercomparisons



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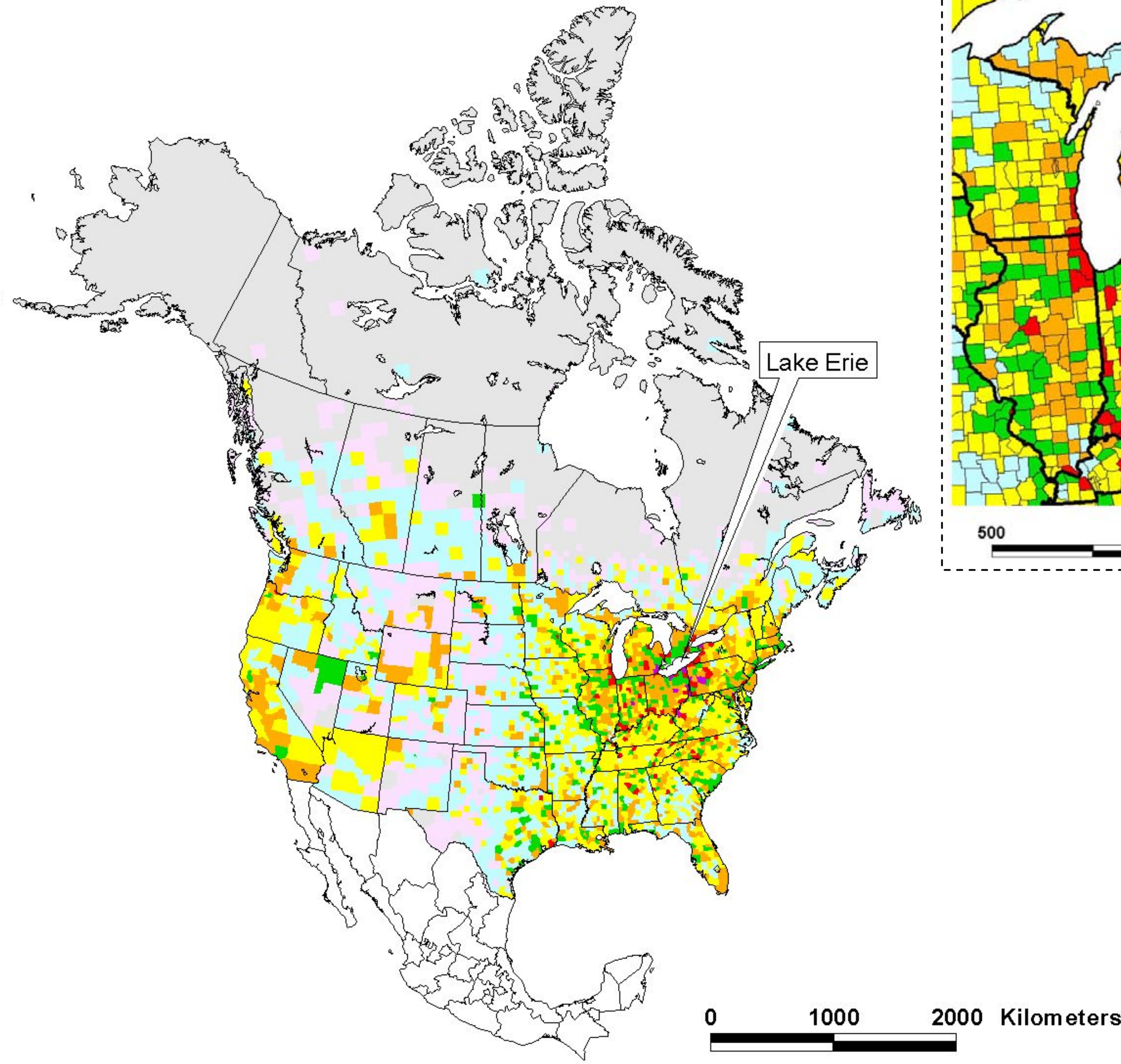
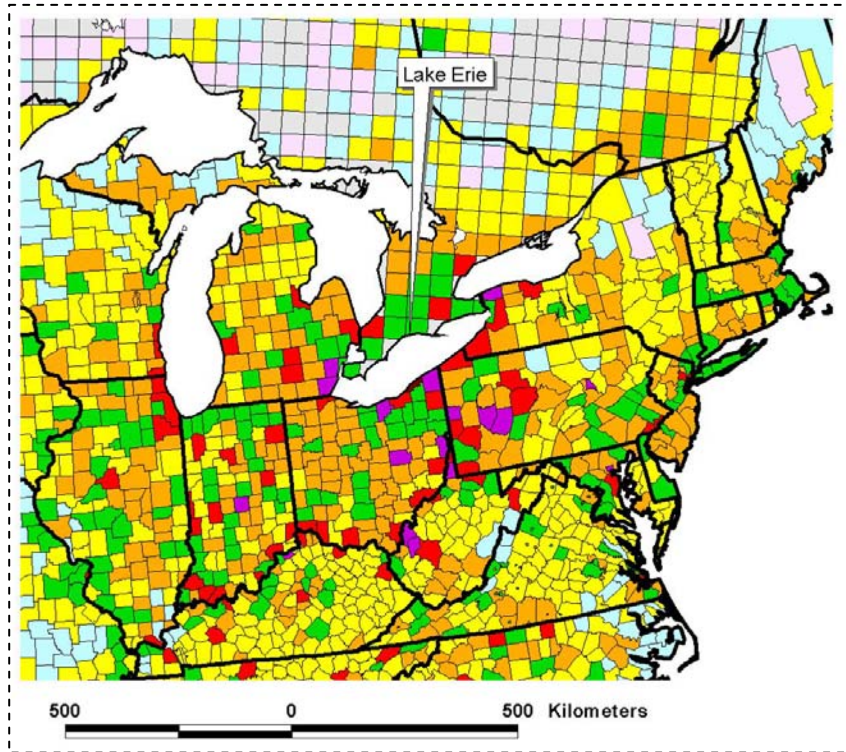
4. **Summary**

HYSPLIT-Hg results for Lake Erie (1999)

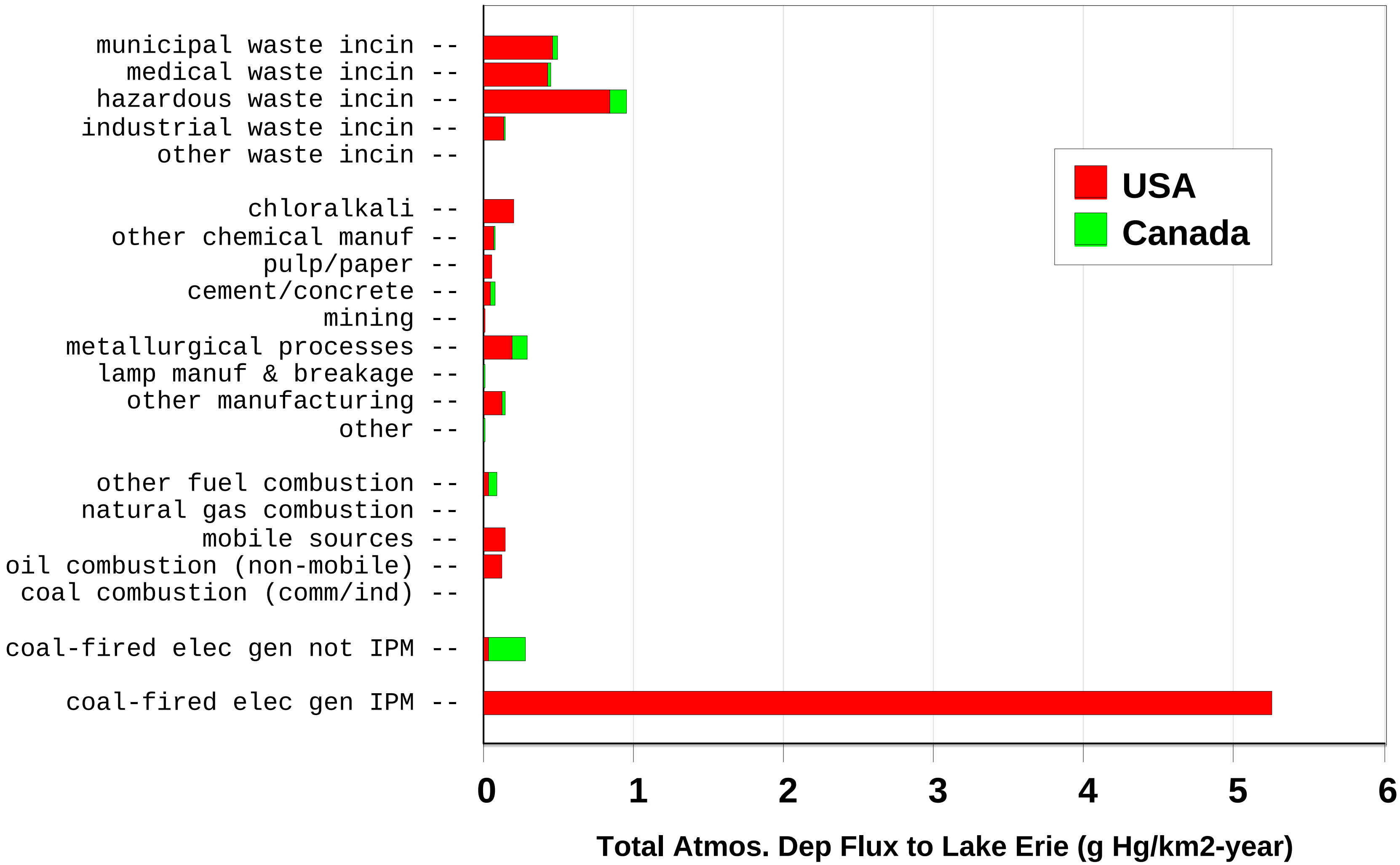




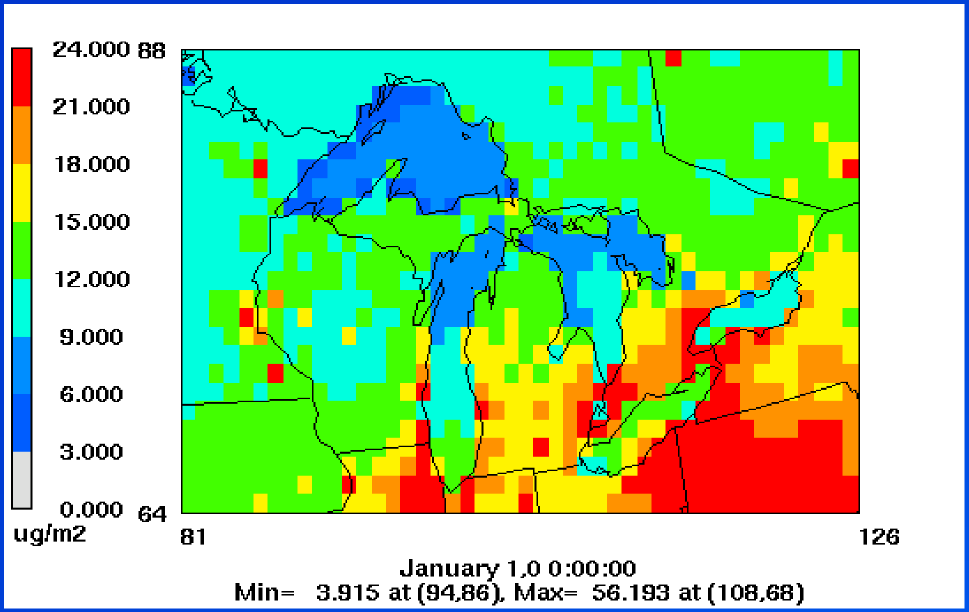
Deposition Contribution of Source Area to Receptor (ug deposited / year per km^2 of receptor area) per (km^2 of source area)

- 0 - 0.0001
- 0.0001 - 0.001
- 0.001 - 0.01
- 0.01 - 0.1
- 0.1 - 1
- 1 - 10
- 10 - 100
- 100 - 1,000
- 1,000 - 10,000
- 10,000 - 100,000

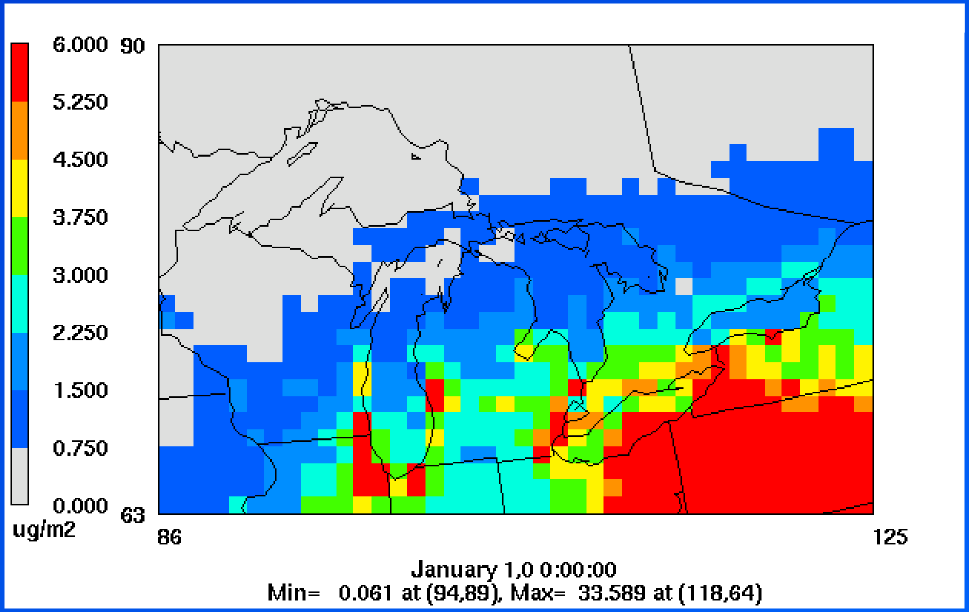
HYSPLIT-Hg results for Lake Erie (1999)



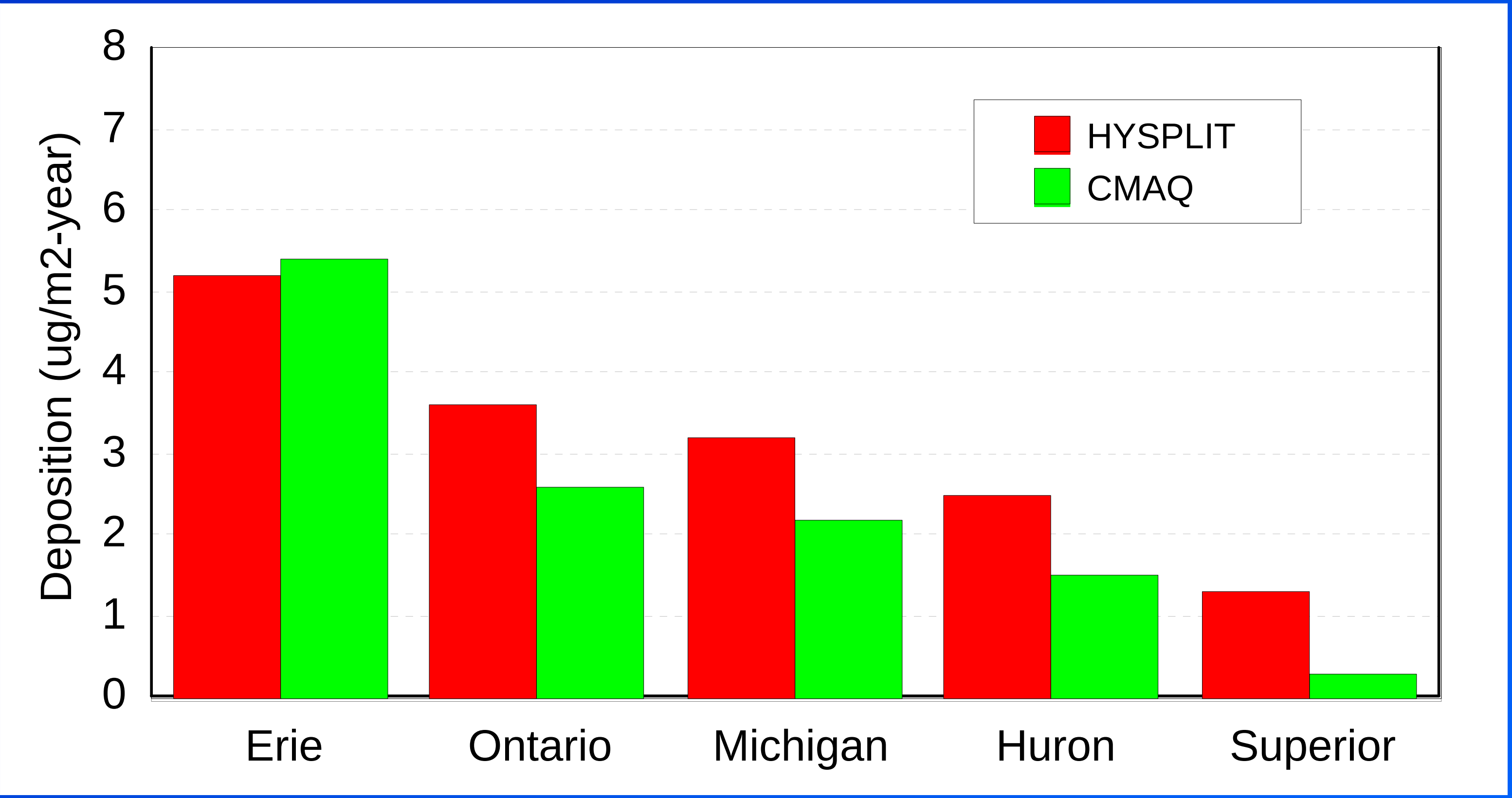
CMAQ-Hg results from EPA analysis performed for the Clean Air Mercury Rule



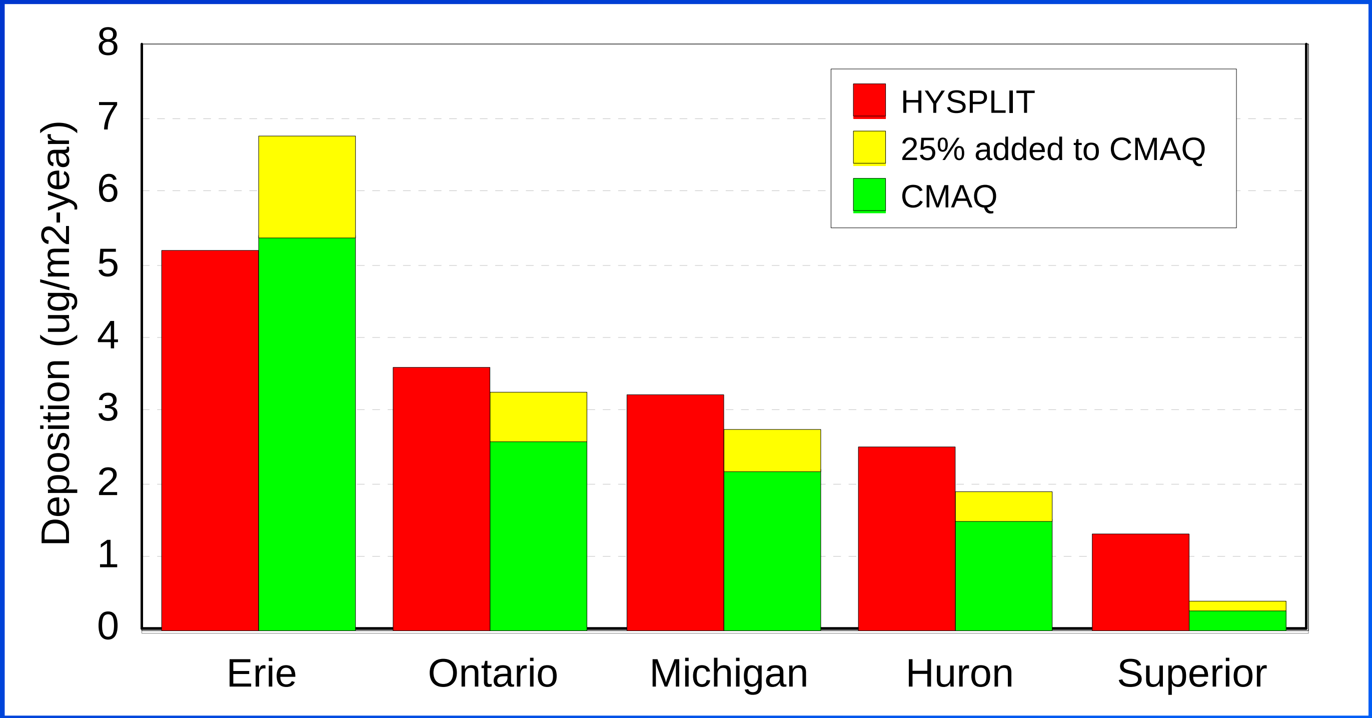
Modeled Mercury Deposition in the Great Lakes Region from all sources during 2001



Modeled Mercury Deposition in the Great Lakes Region attributable to U.S. coal-fired power plants during 2001

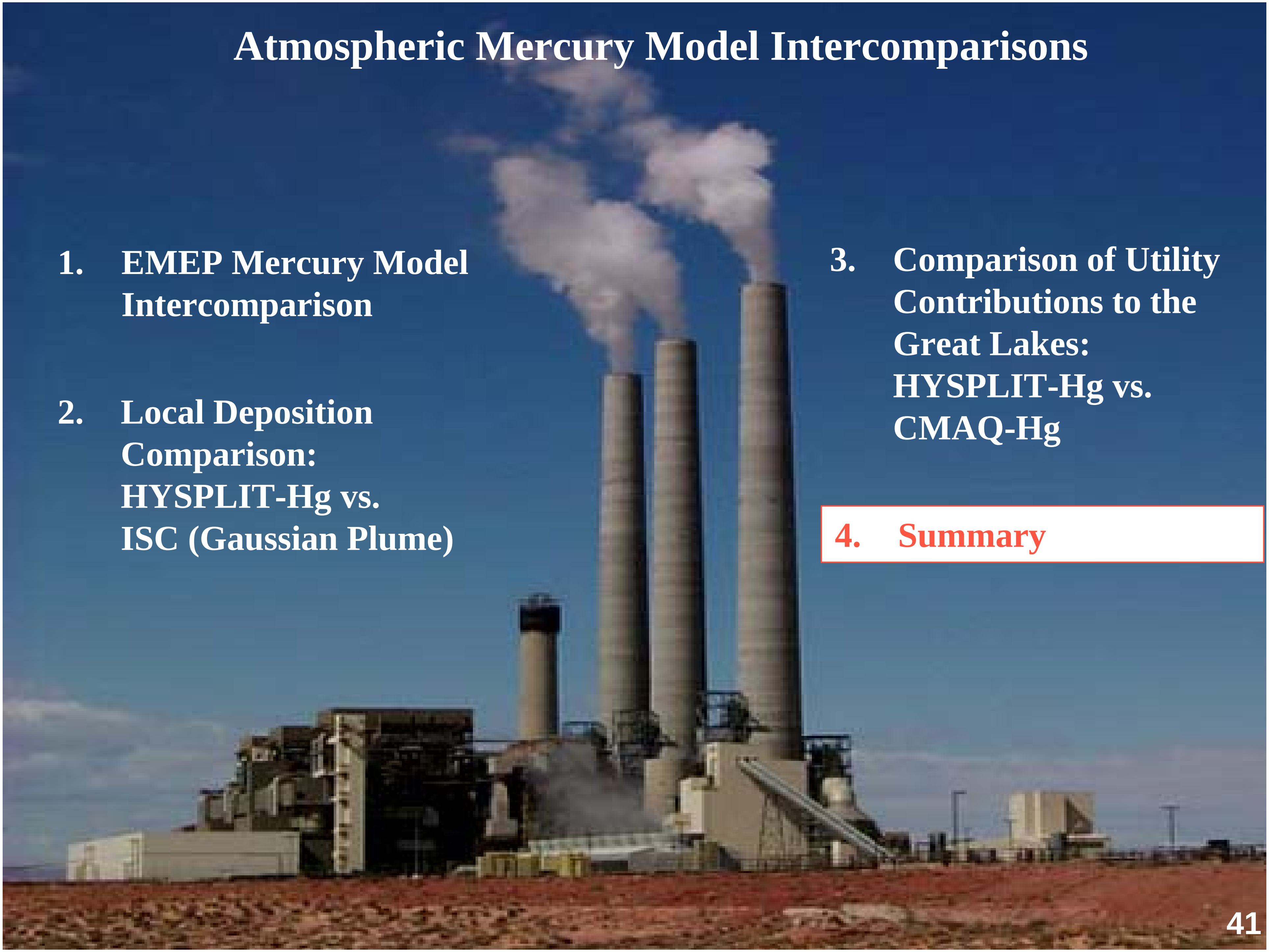


Model-estimated U.S. utility atmospheric mercury deposition contribution to the Great Lakes: HYSPLIT-Hg (1996 meteorology, 1999 emissions) vs. CMAQ-HG (2001 meteorology, 2001 emissions).



- Model-estimated U.S. utility atmospheric mercury deposition contribution to the Great Lakes: HYSPLIT-Hg (1996 meteorology, 1999 emissions) vs. CMAQ-Hg (2001 meteorology, 2001 emissions).

- This figure also shows an added component of the CMAQ-Hg estimates -- corresponding to 30% of the CMAQ-Hg results – in an attempt to adjust the CMAQ-Hg results to account for the deposition underprediction found in the CMAQ-Hg model evaluation.



Atmospheric Mercury Model Intercomparisons

1. EMEP Mercury Model Intercomparison

2. Local Deposition Comparison: HYSPLIT-Hg vs. ISC (Gaussian Plume)

3. Comparison of Utility Contributions to the Great Lakes: HYSPLIT-Hg vs. CMAQ-Hg

4. Summary

Summary of Model Intercomparisons

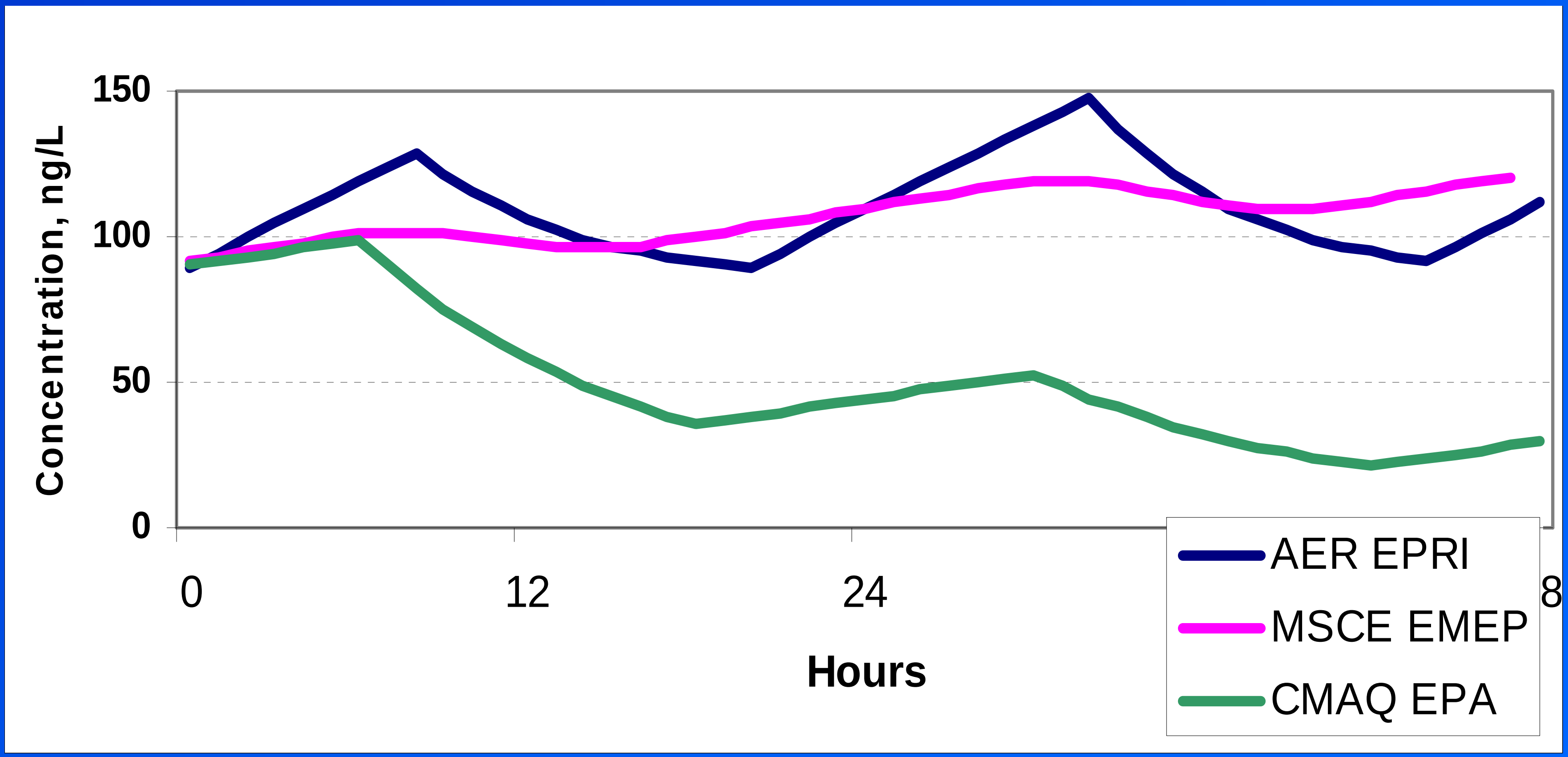
- **Extremely useful for improving models**
- **Opportunity to work together and pool resources**
  (e.g., everyone doesn't have to create their own inventory or assemble monitoring data for evaluation)
- **Funding is a problem… most studies do not fund the individual participants….**
- **10% of the work is doing the initial modeling analysis;**
- **90% of the work is trying to figure out why the models are different – but we rarely have the resources to do much of this**

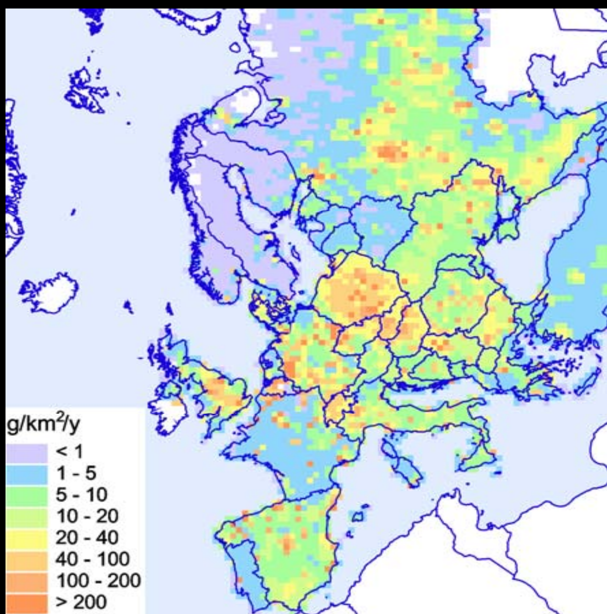
Thanks!

Extra Slides

Effect of Different Assumptions Regarding Hg(p) Solubility

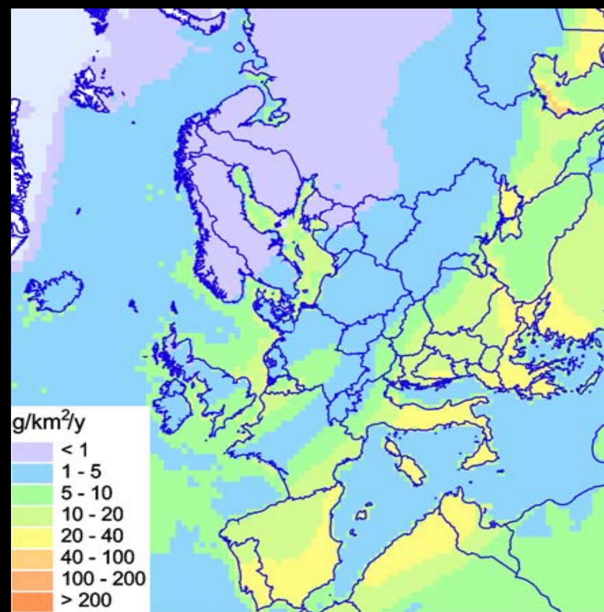
AER/EPRI 0%; MSCE-EMEP 50%; CMAQ-EPA 100%





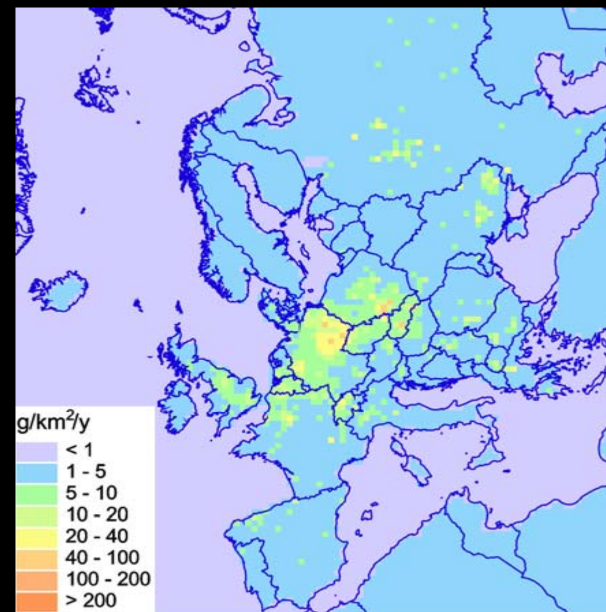
2000 European anthropogenic Hg emissions

240 t/yr



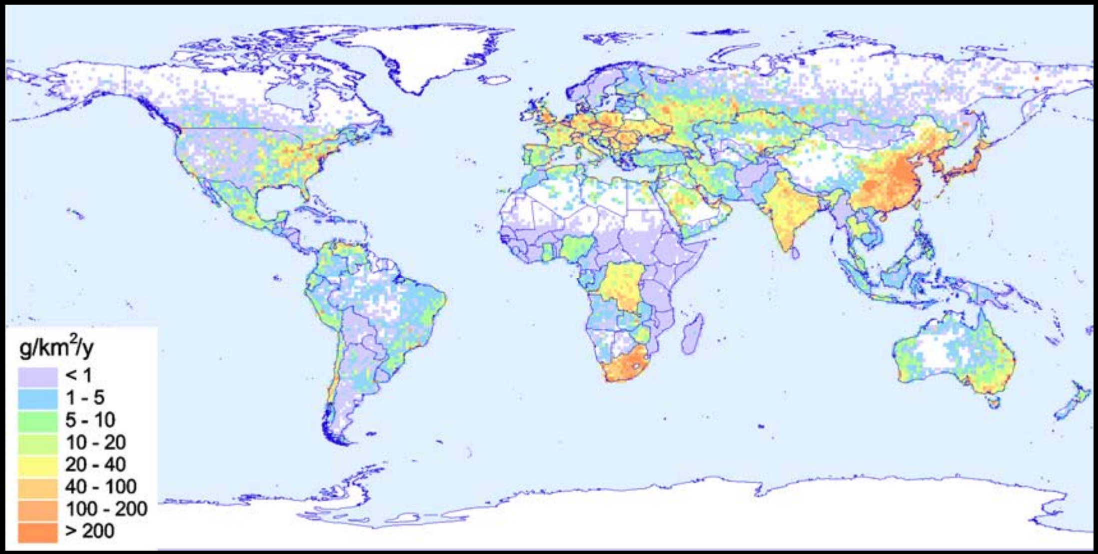
European natural Hg emissions

180 t/yr

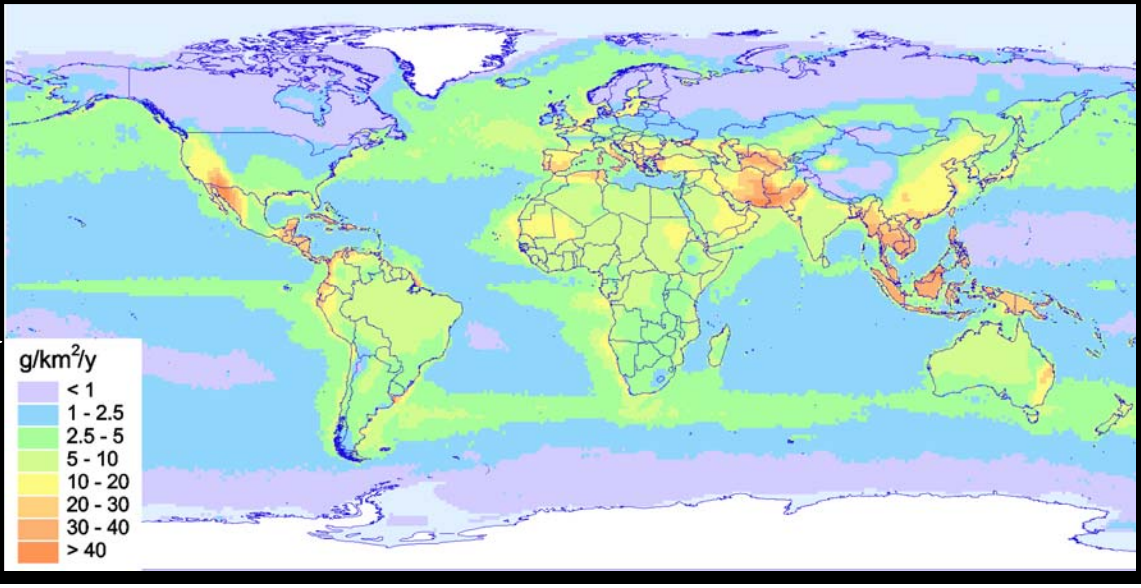


European anthropogenic Hg re-emissions

50 t/yr

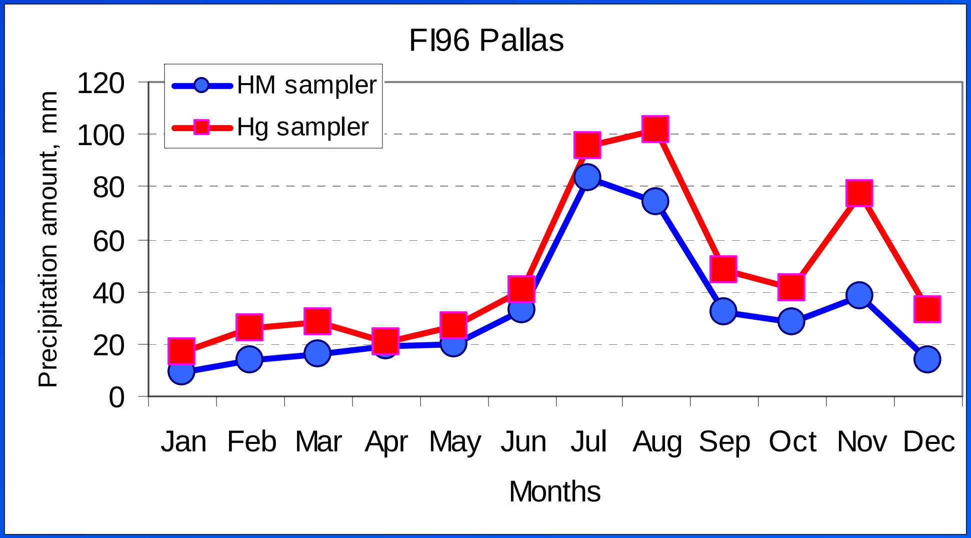


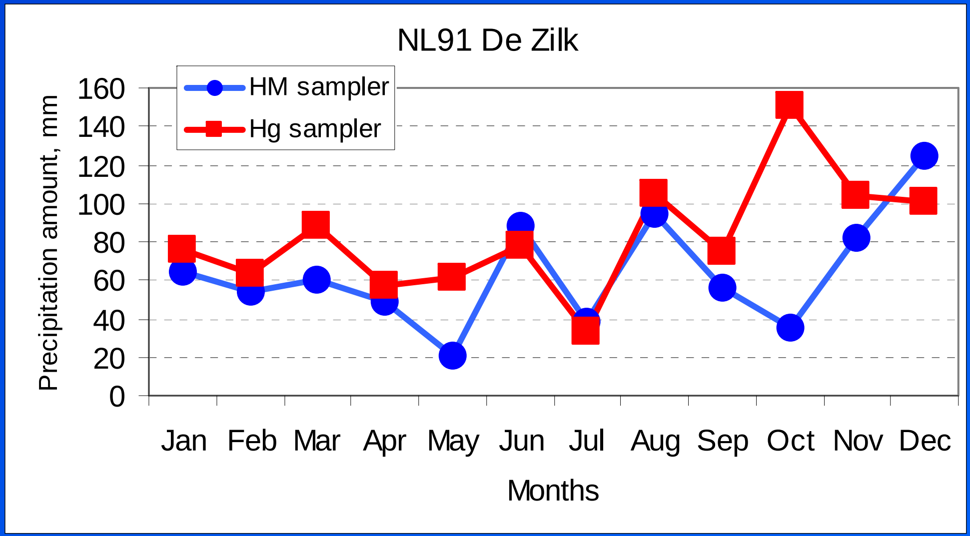
1995 global anthropogenic Hg emissions 1900 t / yr

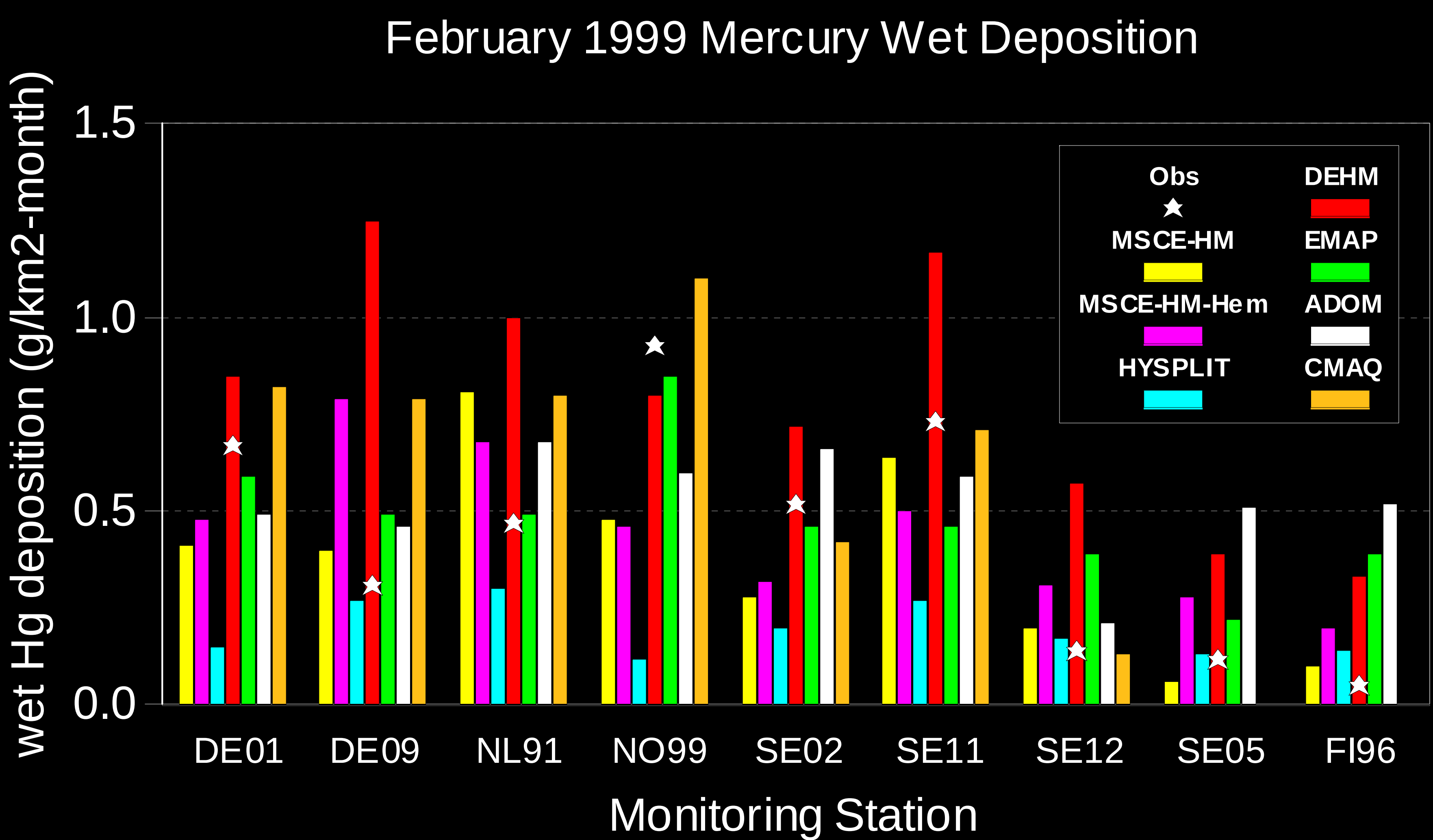


global natural Hg emissions 1800 t / yr

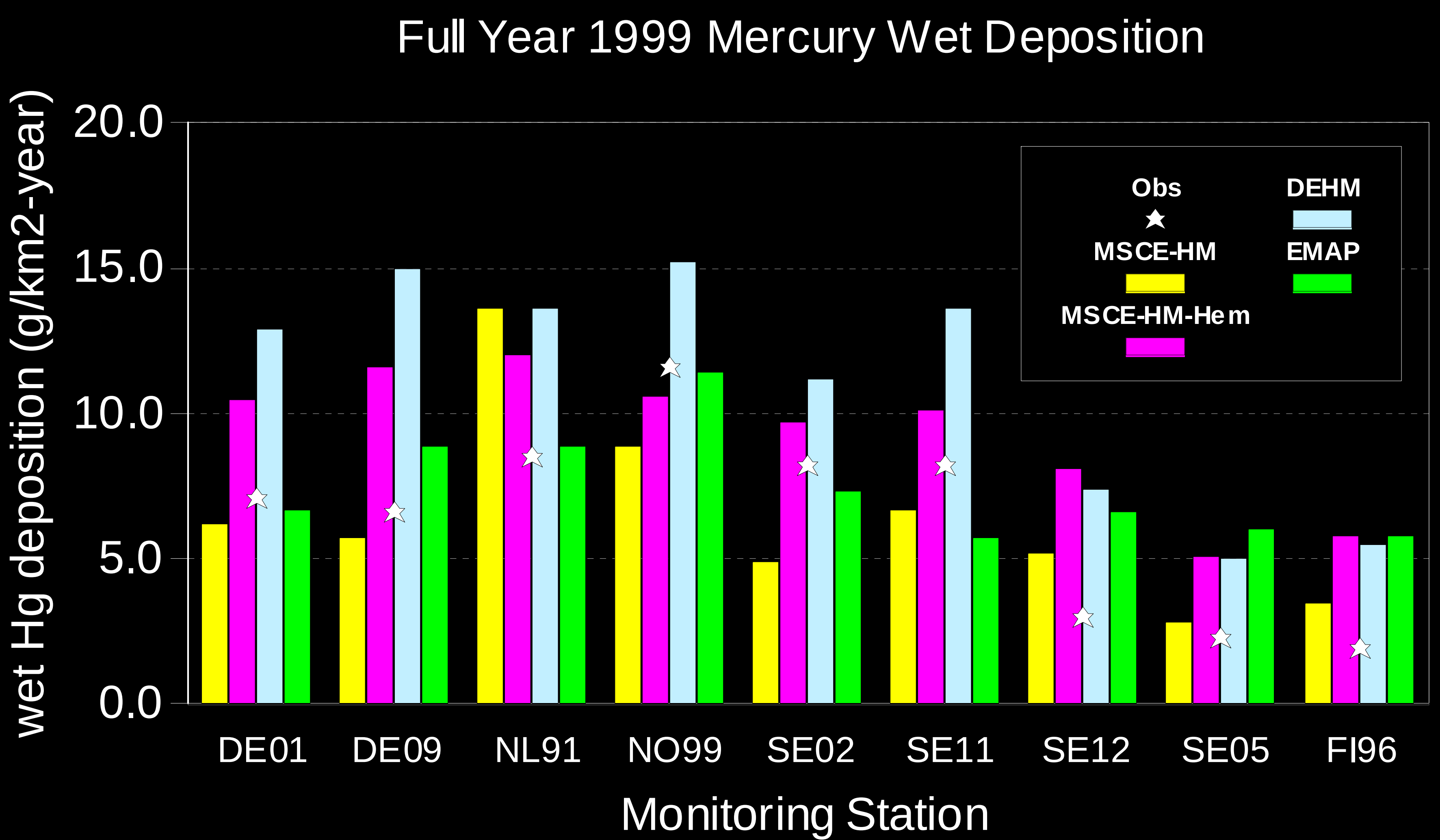
There are uncertainties in measurements -- even of precipitation amount…



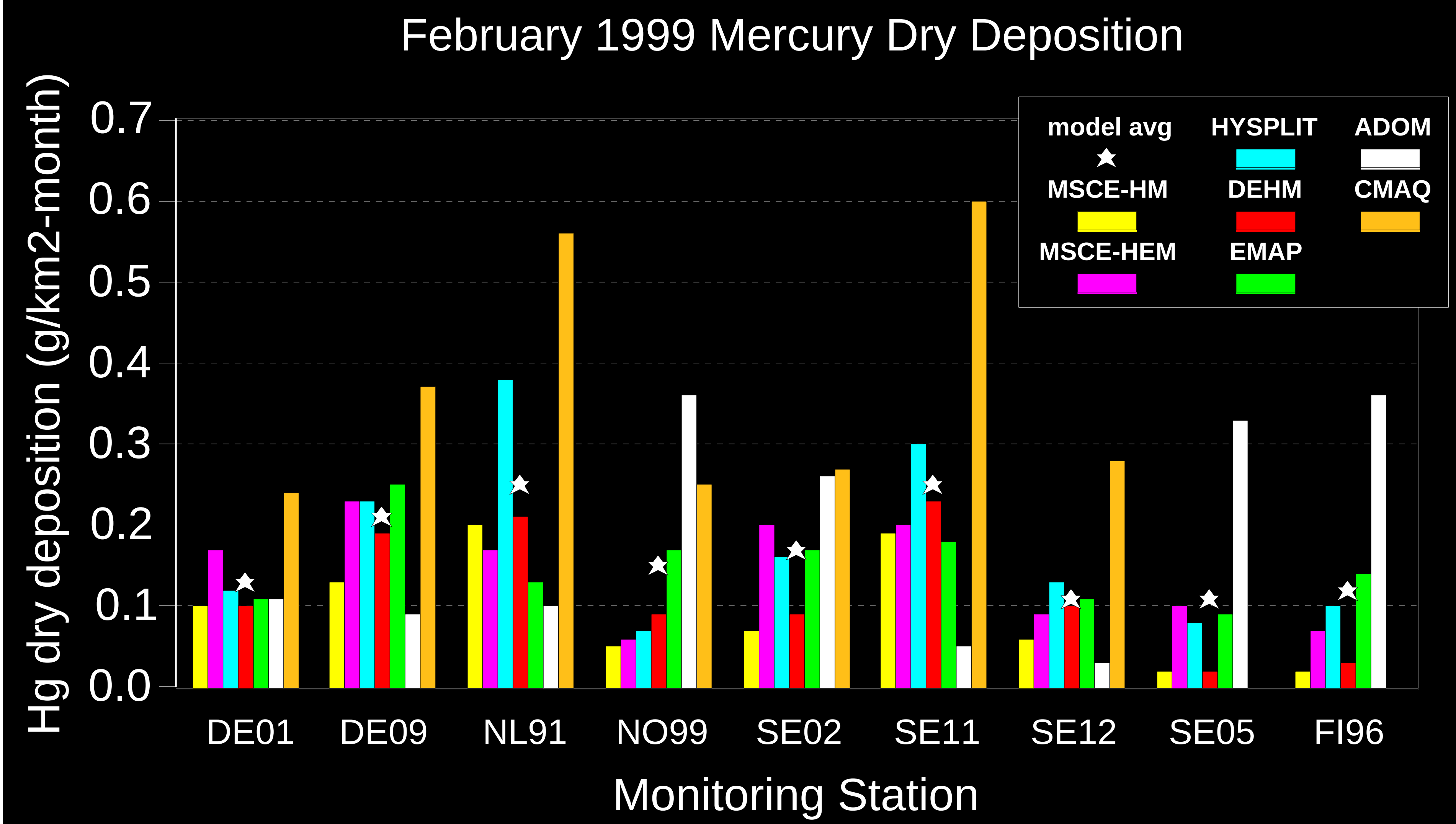


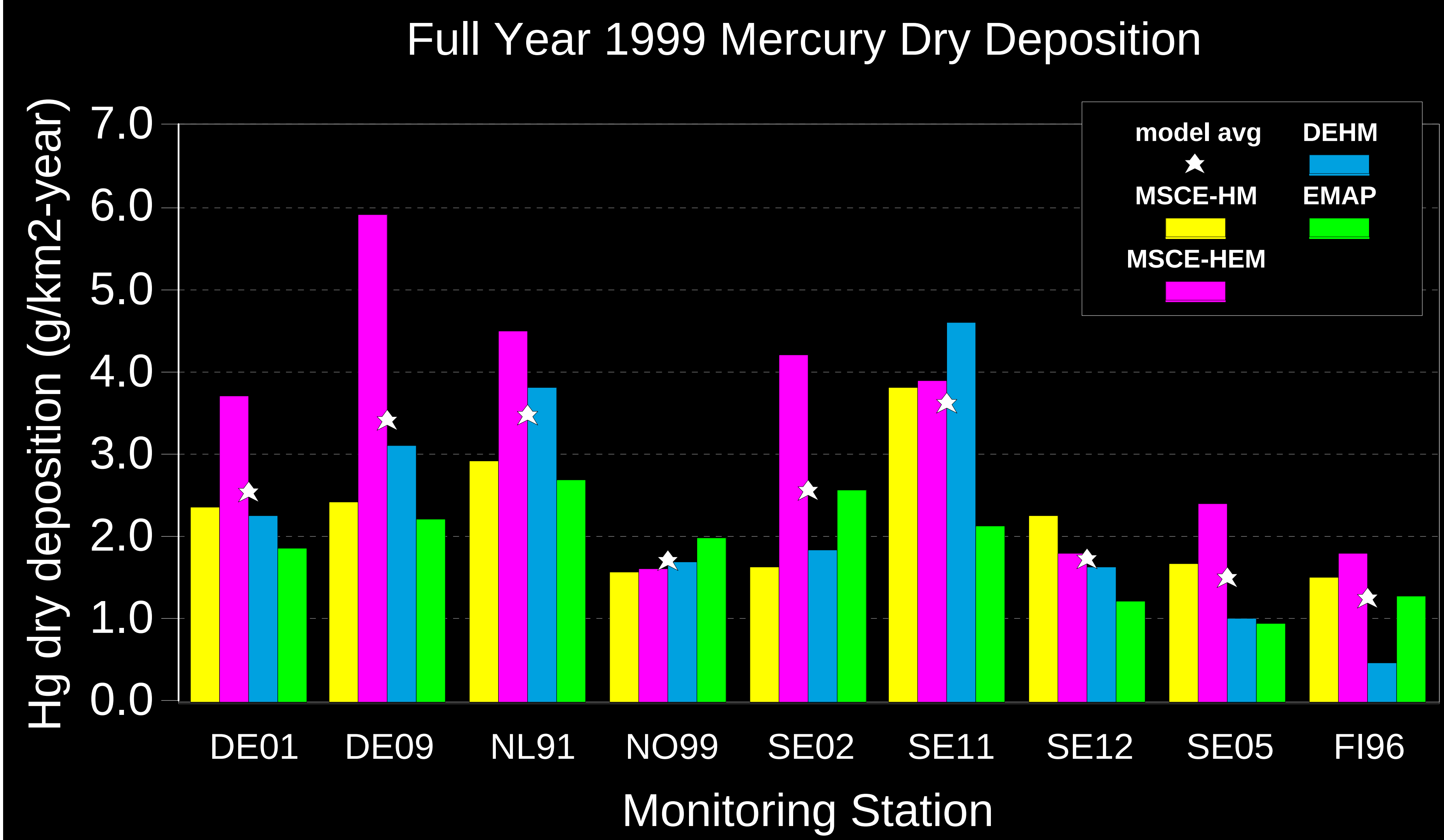


February 1999 Mercury Wet Deposition
wet Hg deposition (g/km2-month)
1.5
1.0
0.5
0.0
Obs
DEHM
MSCE-HM
EMAP
MSCE-HM-Hem
ADOM
HYSPLIT
CMAQ
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station



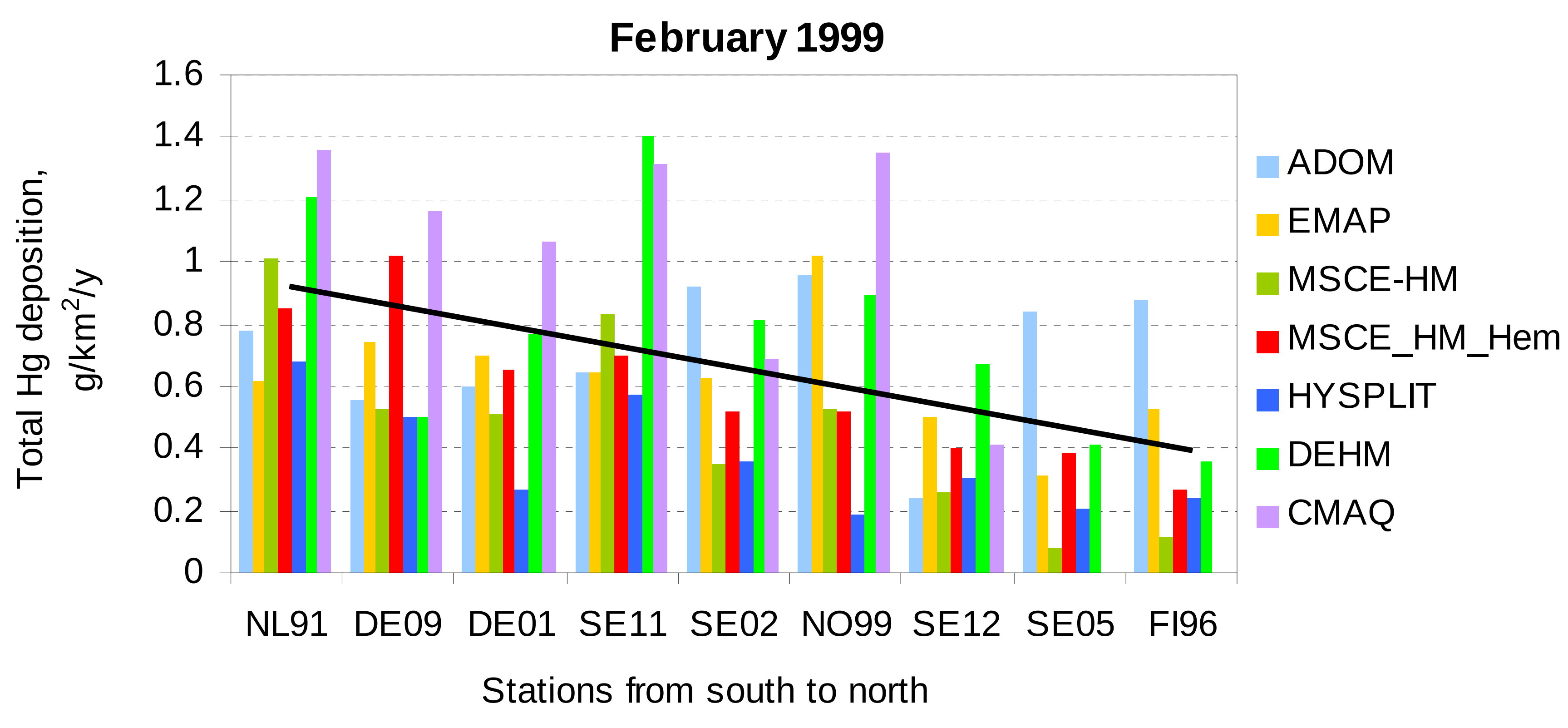
Full Year 1999 Mercury Wet Deposition
wet Hg deposition (g/km2-year)
20.0
15.0
10.0
5.0
0.0
Obs
DEHM
MSCE-HM
EMAP
MSCE-HM-Hem
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station


February 1999 Mercury Dry Deposition
Hg dry deposition (g/km2-month)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
model avg
HYSPLIT
ADOM
MSCE-HM
DEHM
CMAQ
MSCE-HEM
EMAP
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station

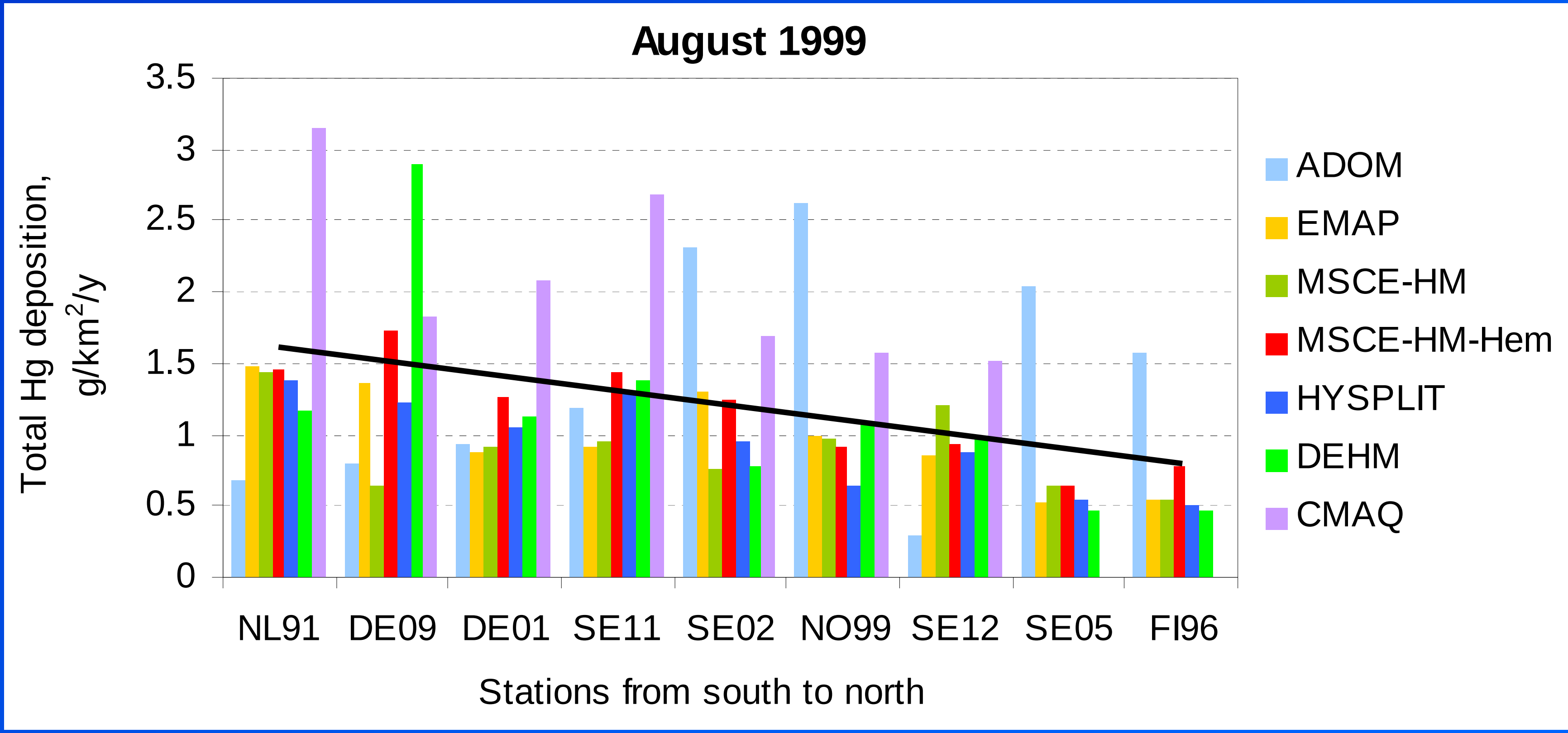

Full Year 1999 Mercury Dry Deposition
Hg dry deposition (g/km2-year)
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0.0
model avg
DEHM
MSCE-HM
EMAP
MSCE-HEM
DE01
DE09
NL91
NO99
SE02
SE11
SE12
SE05
FI96
Monitoring Station

- **In the following, the total model-predicted deposition *(= wet + dry)* is compared**

Total Modeled Hg Deposition (wet + dry)

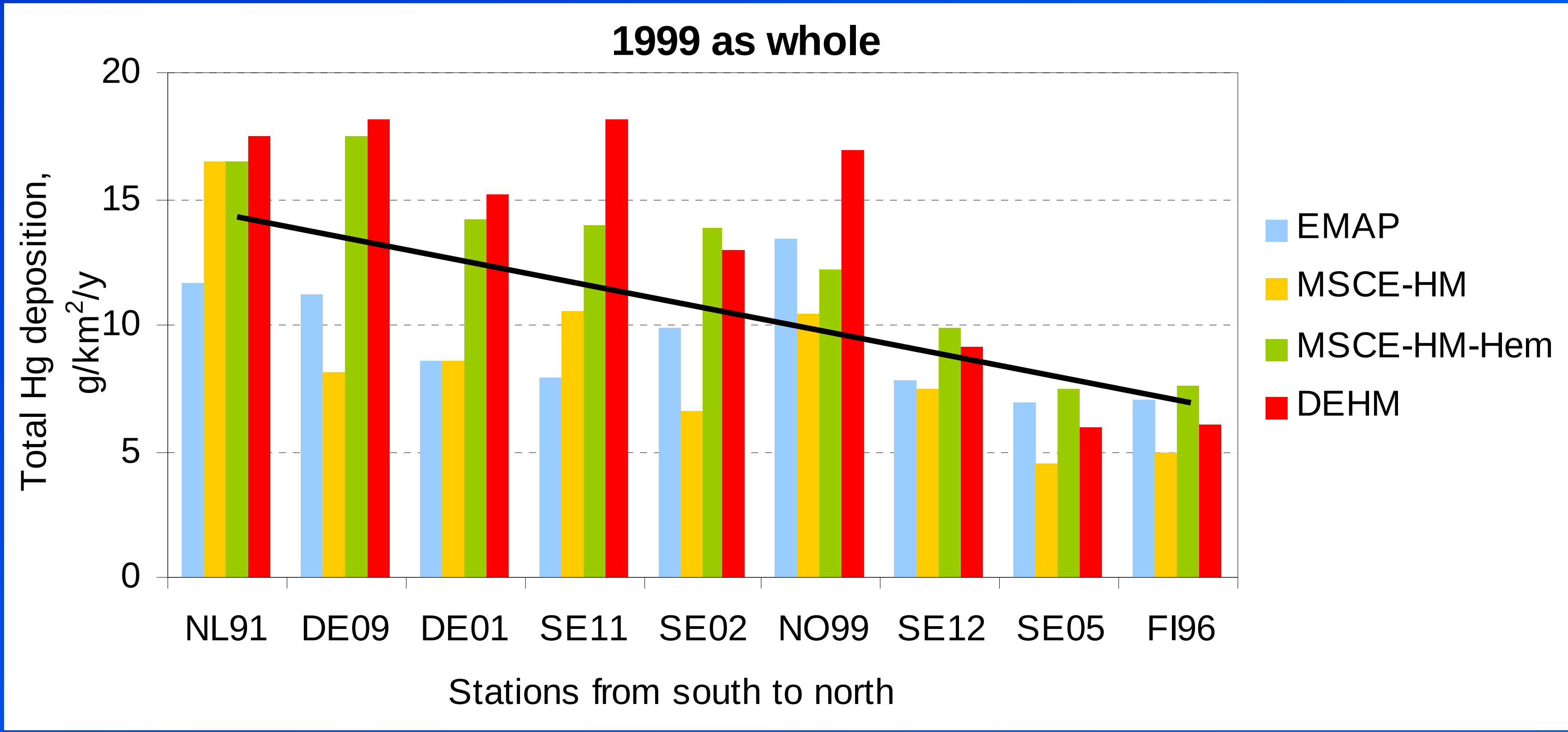


Total Modeled Hg Deposition (wet + dry)



Note: ADOM was not run for August, so for this graph, ADOM results for July were used

Total Modeled Hg Deposition (wet + dry)



EMEP model results in relation to the other models

Range	Deposition over polluted area in Feb 1999, g/km^2		Total deposition over the countries in Feb 1999, kg		
	Wet	Dry	The UK	Italy	Poland
Minimum	**0.24**	**0.10**	**76**	**143**	**300**
MSCE-HM	**0.54**	**0.16**	**235**	**261**	**1070**
MSCE-HM-Hem	**0.65**	**0.19**	**170**	**164**	**730**
Maximum	**1.03**	**0.39**	**240**	**334**	**1190**

Conclusions: Uncertainties in Mercury Modeling

- Elemental Hg in air - factor of **1.2**
- Particulate Hg in air - factor of **1.5**
- Oxidized gaseous Hg in air - factor of **5**
- Total Hg in precipitation - factor of **1.5**
- Wet deposition - factor of **2.0**
- Dry deposition - factor of **2.5**
- Balances for countries - factor of **2**