

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

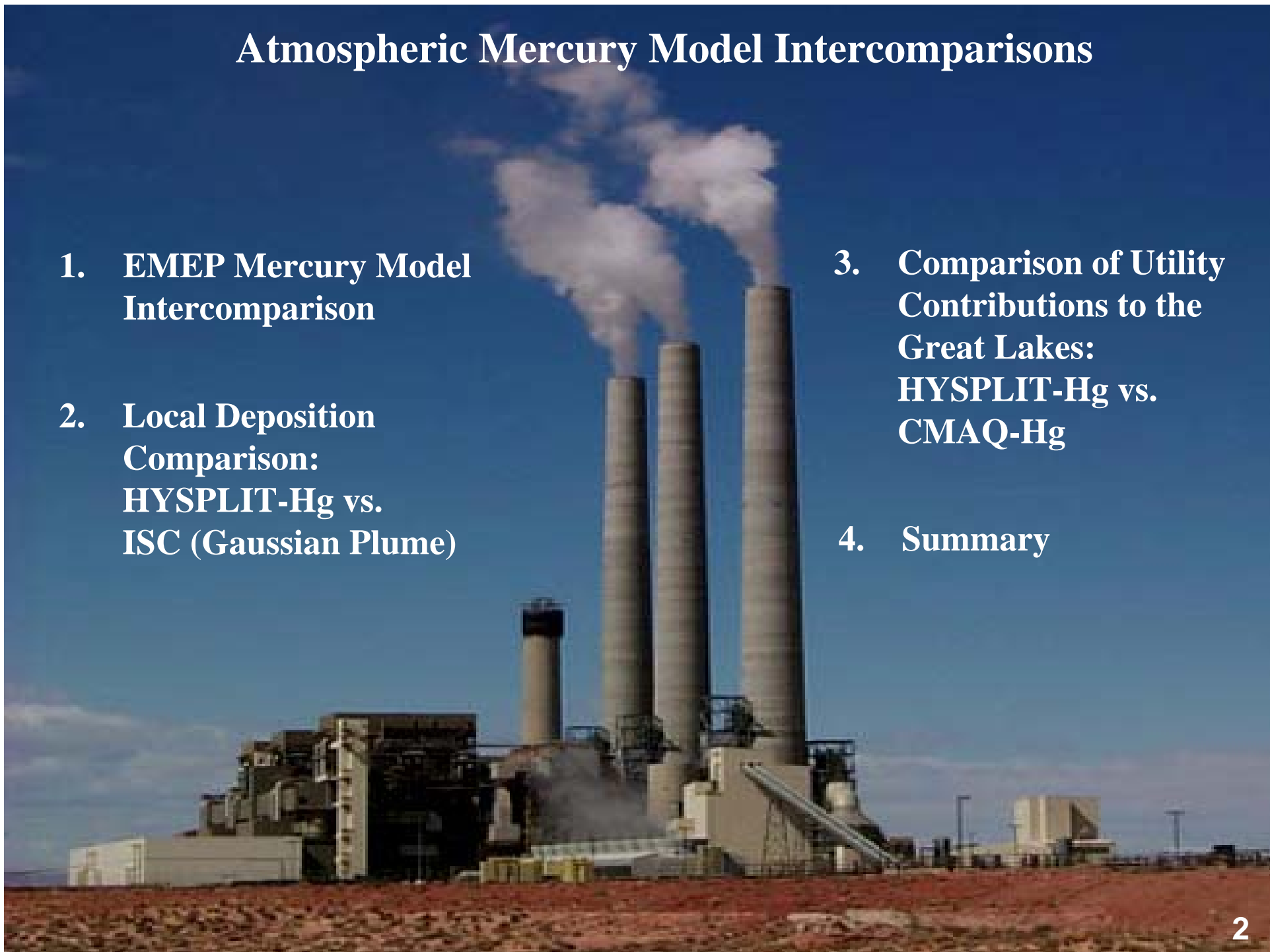
Atmospheric Mercury Model Intercomparisons

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



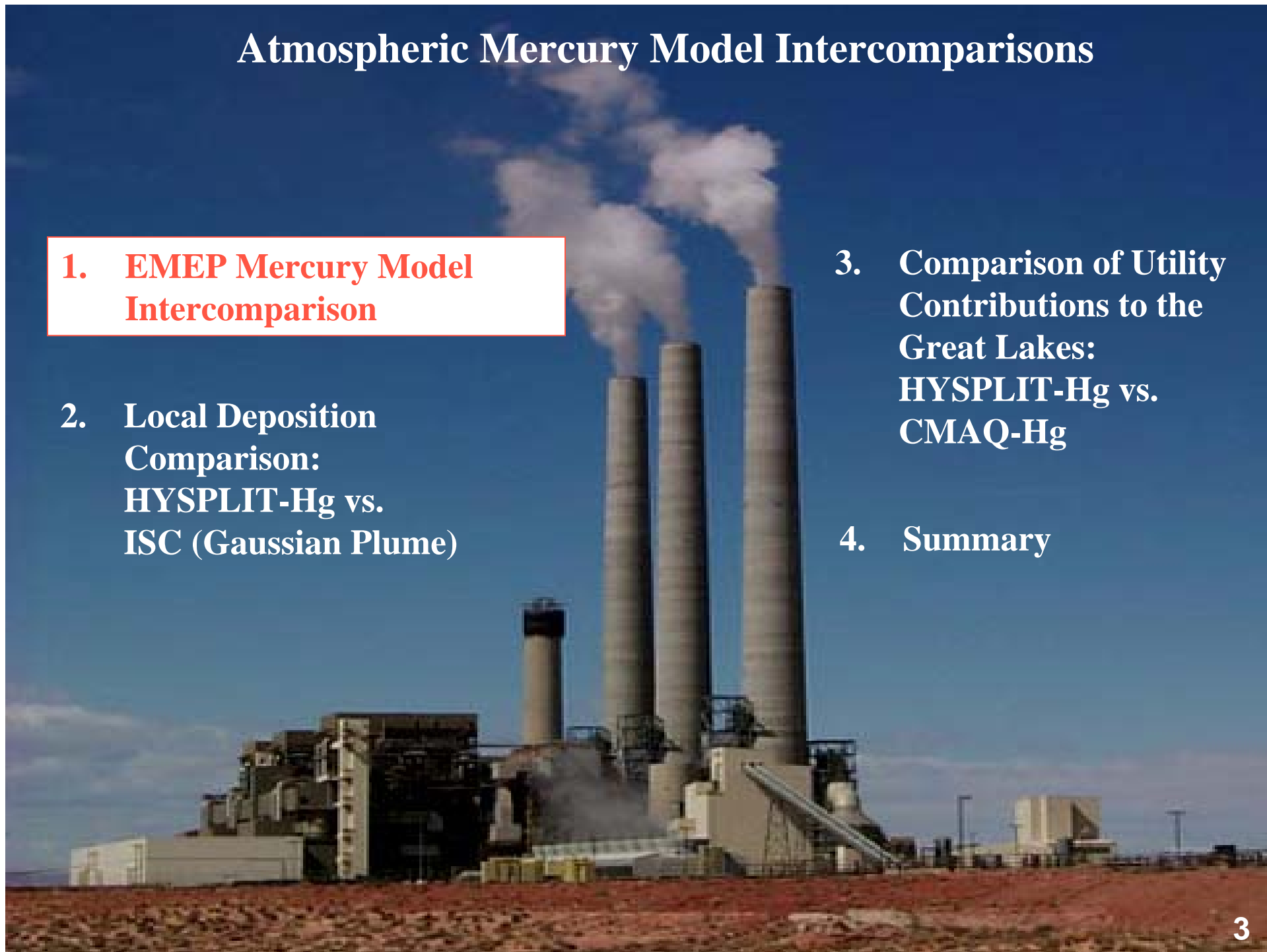
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Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

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**

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
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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**

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

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Participating Models

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

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


Intercomparison Conducted in 3 Stages

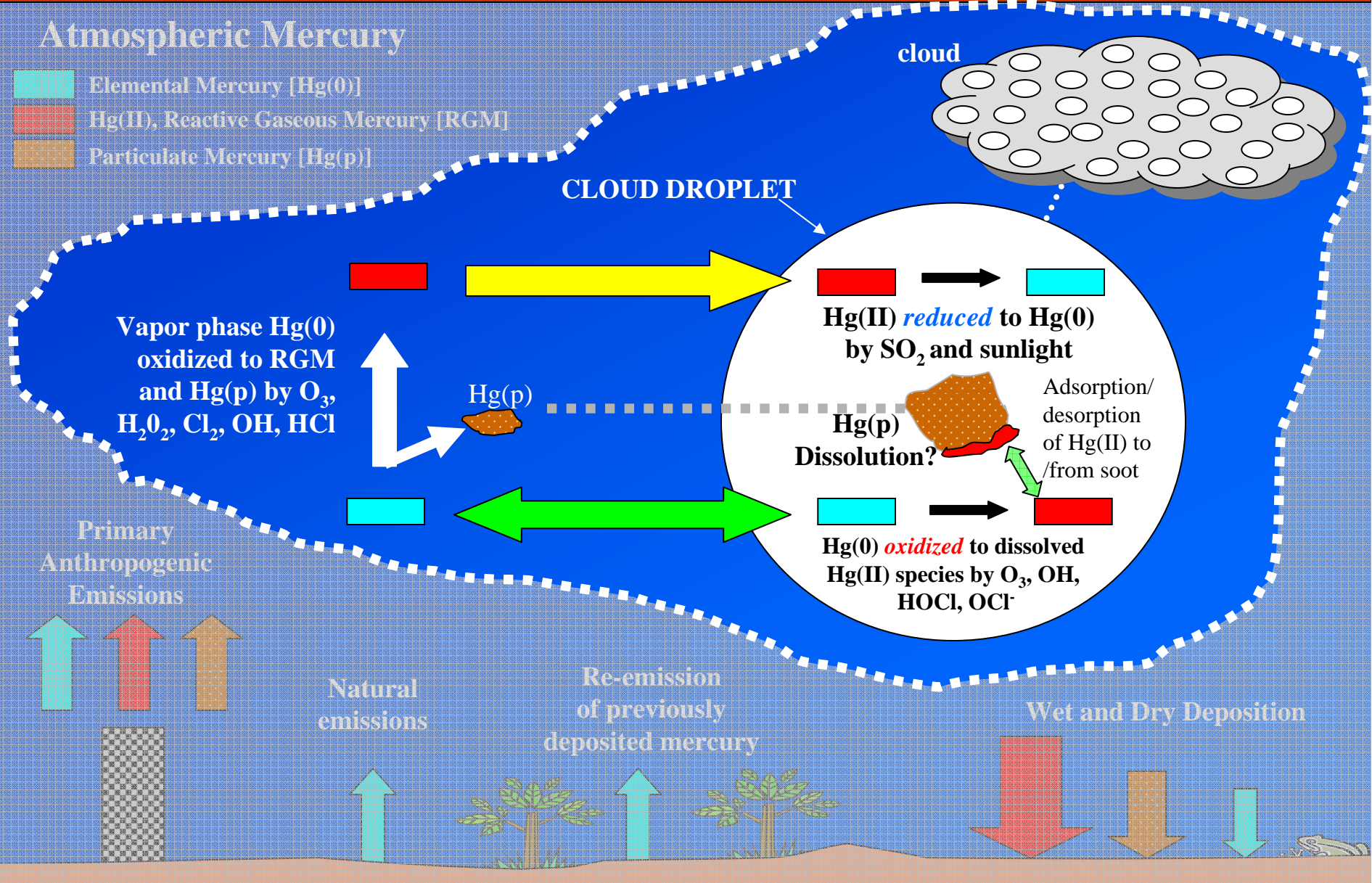
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Atmospheric Mercury

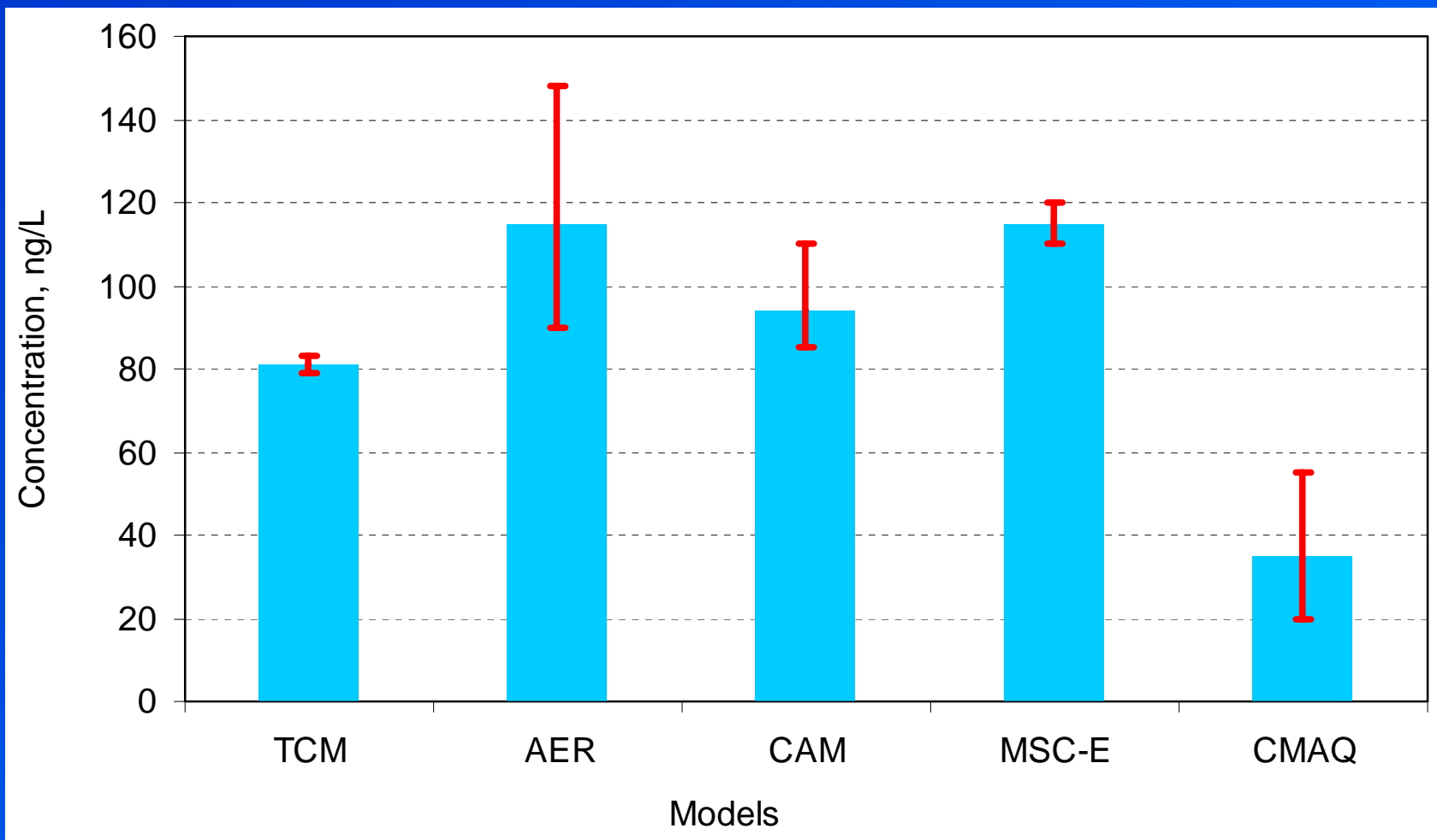
-  Elemental Mercury [Hg(0)]
-  Hg(II), Reactive Gaseous Mercury [RGM]
-  Particulate Mercury [Hg(p)]



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Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
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Variation of Hg concentrations (ng/L)



Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

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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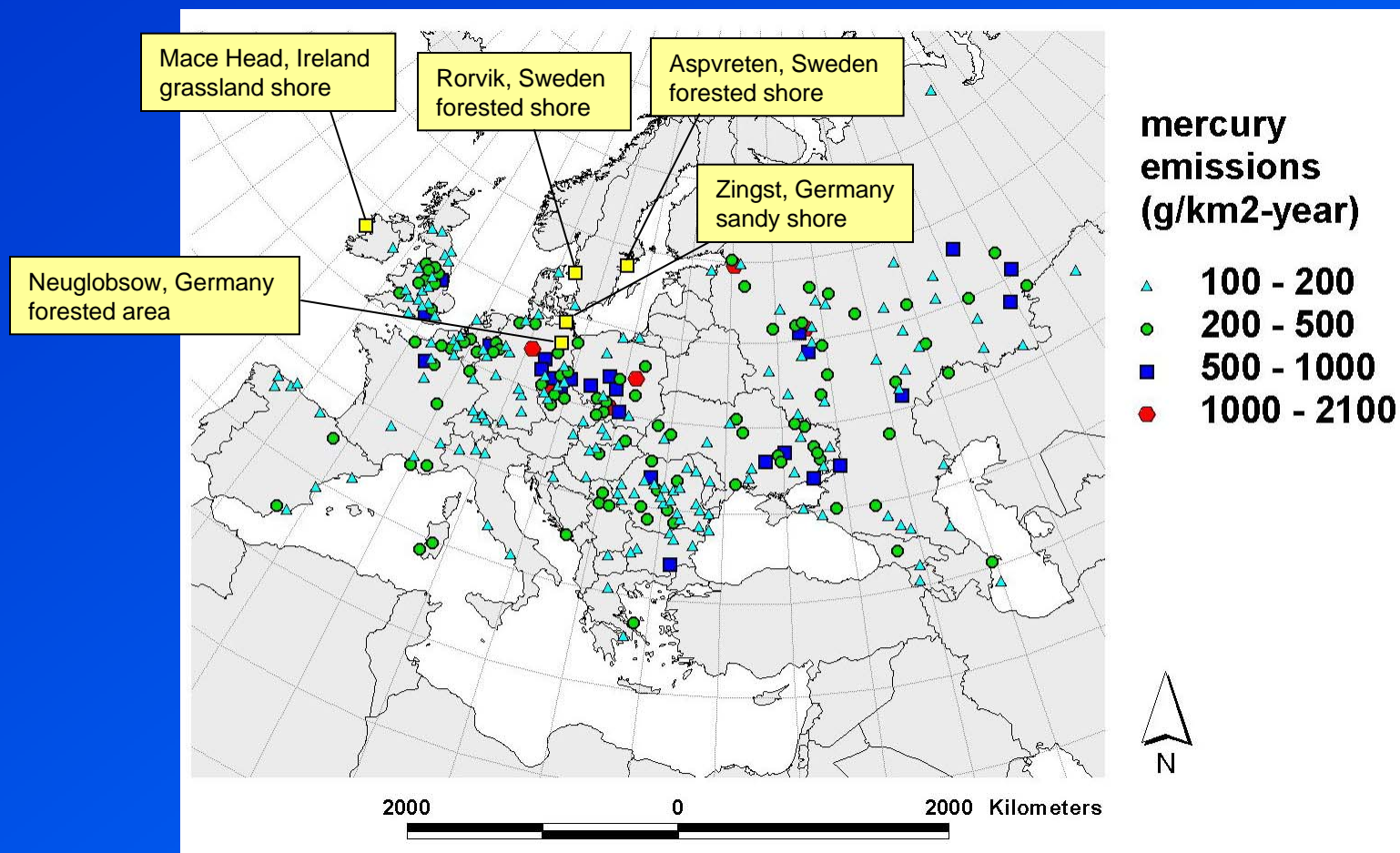
Model	CMAQ-Hg	ADOM	HYSPLIT	EMAP	GRAHM	DEHM	MSCE-Hg
Model type	Eulerian	Eulerian	Lagrangian	Eulerian	Eulerian	Eulerian	Eulerian
Scale/ Domain	regional/ Central and Northern Europe	regional/ Central Europe	regional/ EMEP	regional/ EMEP	global	Hemispheric	regional/ EMEP
Source of meteorological data	ECMWF 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	1° x 1°	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 ₂ O ₂ , Cl ₂ , OH [•]	O ₃	O ₃ , H ₂ O ₂ , Cl ₂ , HCl	O ₃ , OH [•]	O ₃	O ₃	O ₃ (f)
Liquid-phase oxidation agents	O ₃ , OH [•] , HOCl, OCl ⁻	O ₃	O ₃ , OH [•] , 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

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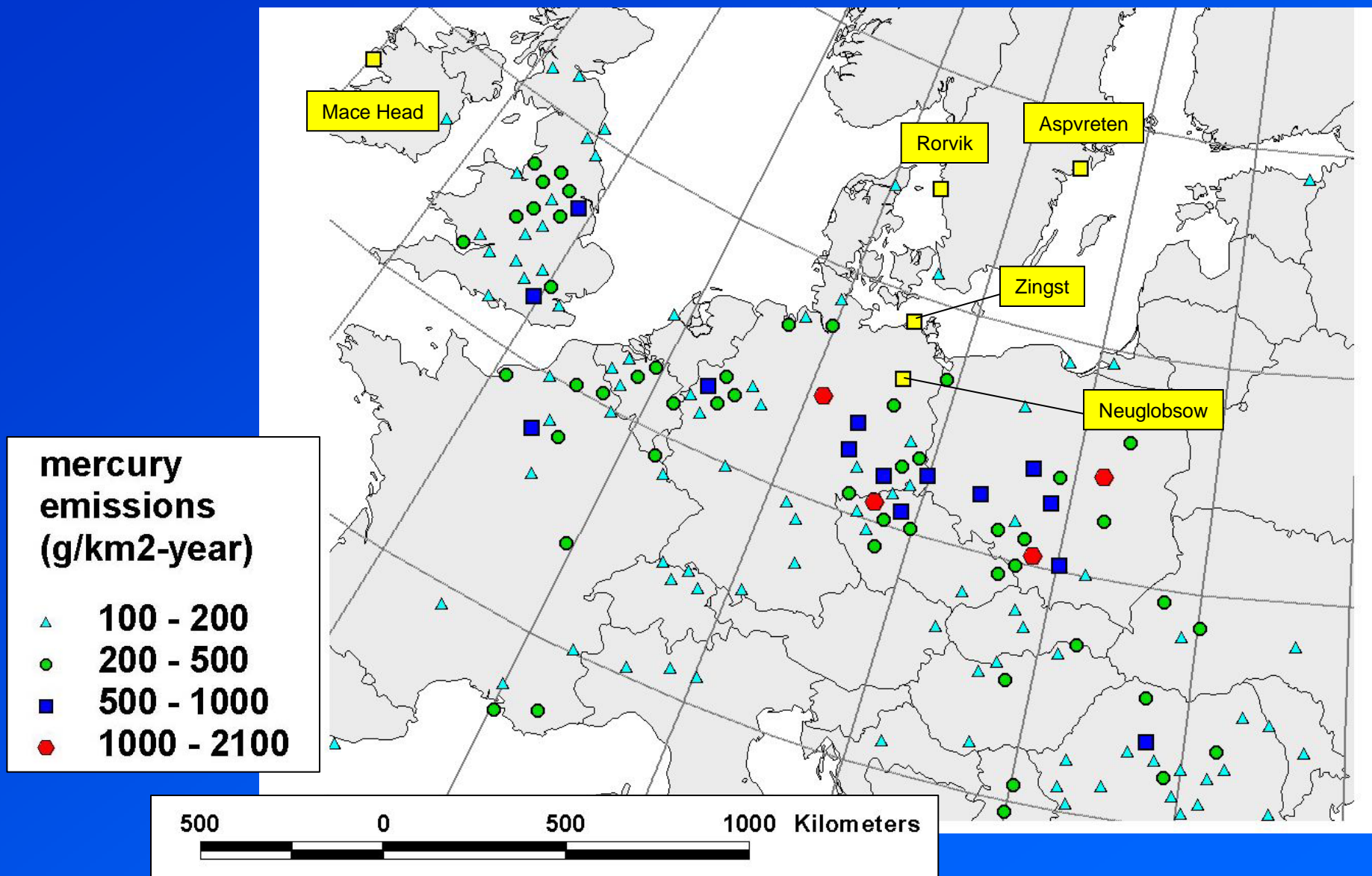
Anthropogenic Mercury Emissions Inventory and Monitoring Sites for Phase II

(note: only showing largest emitting grid cells)



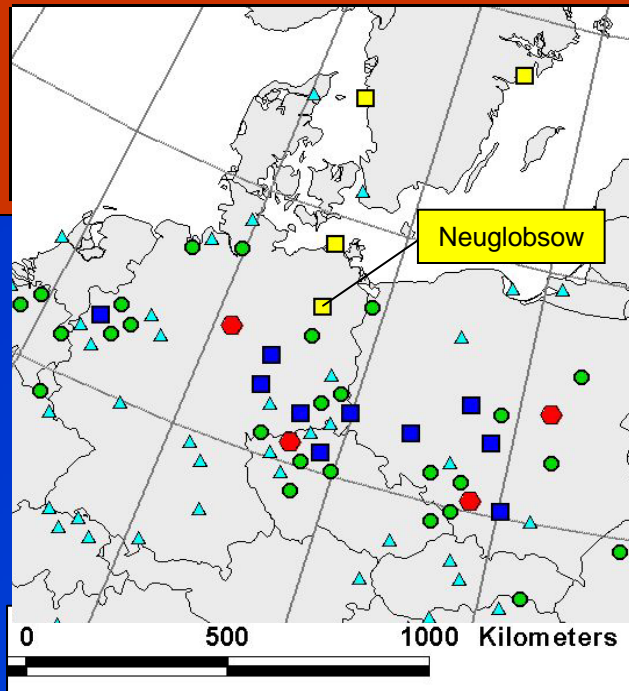
EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury

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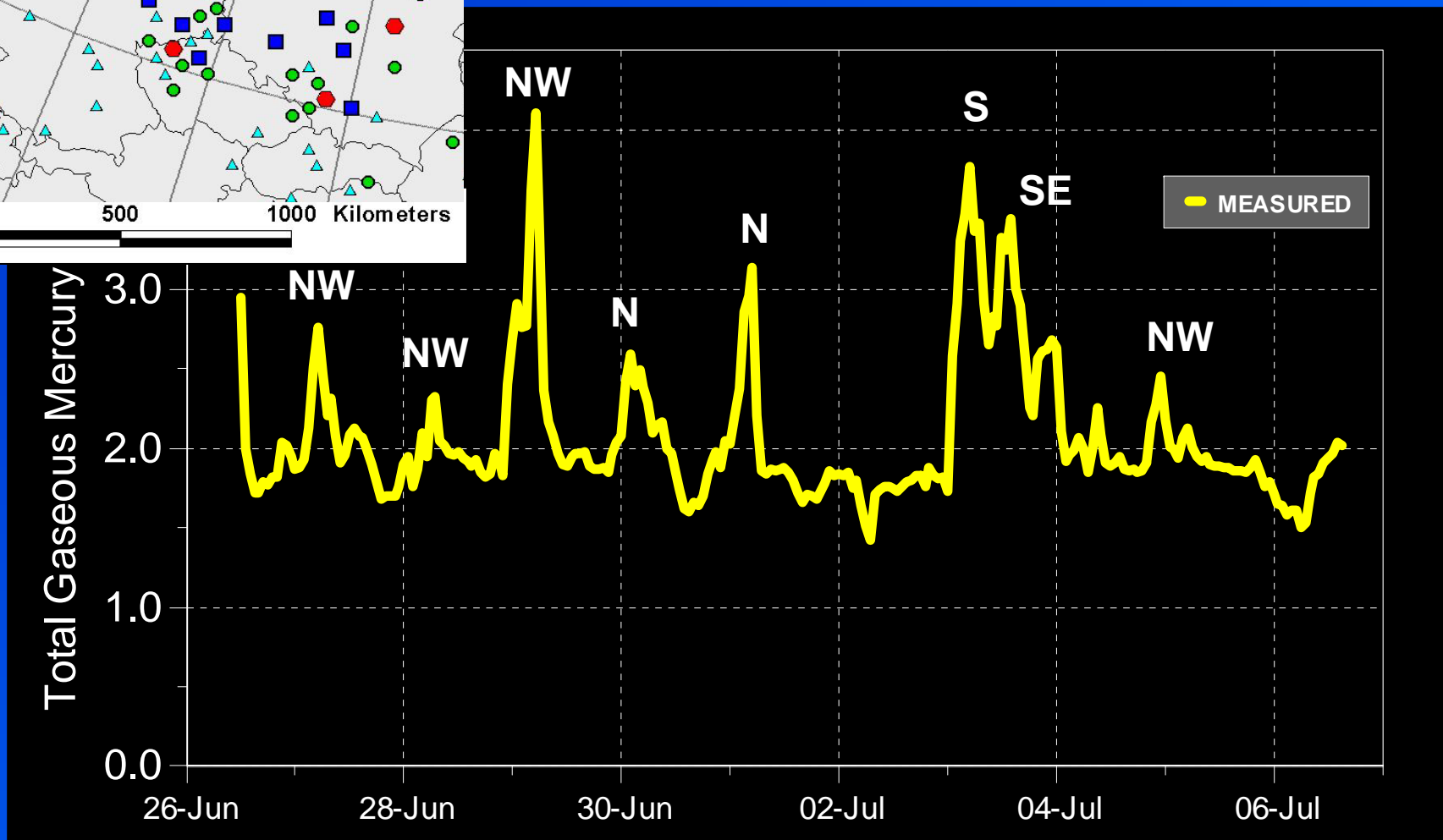


of Numerical Models for Long-Range Atmospheric Transport of Mercury

Stage II		Stage III			Conclu- sions
Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	



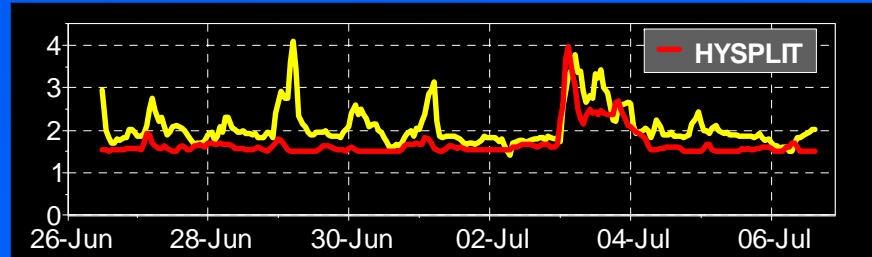
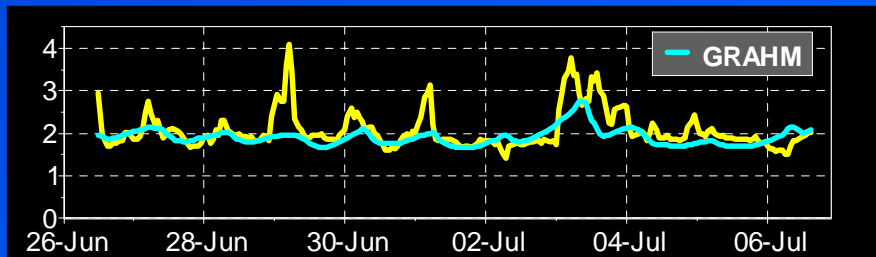
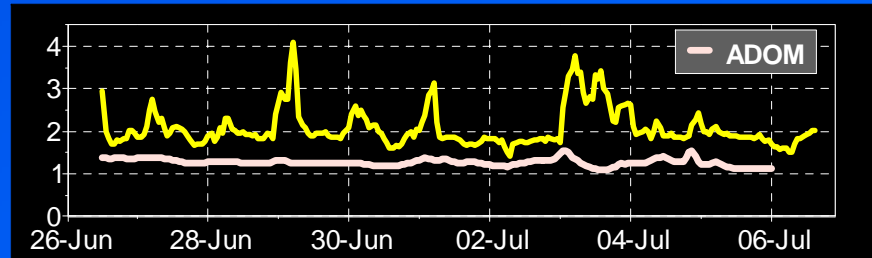
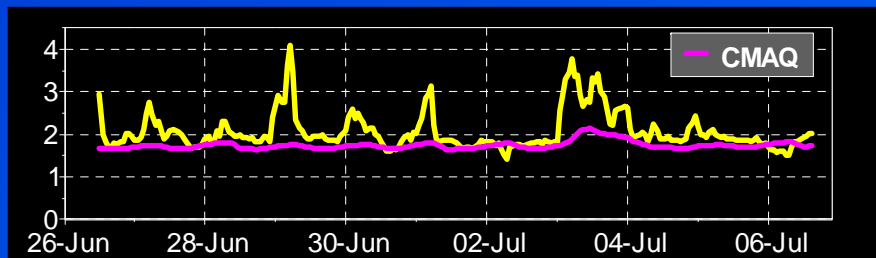
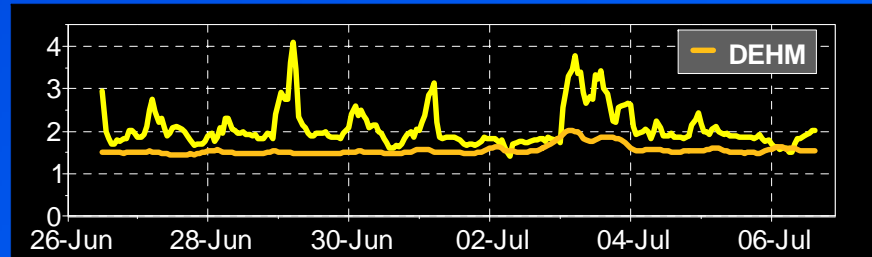
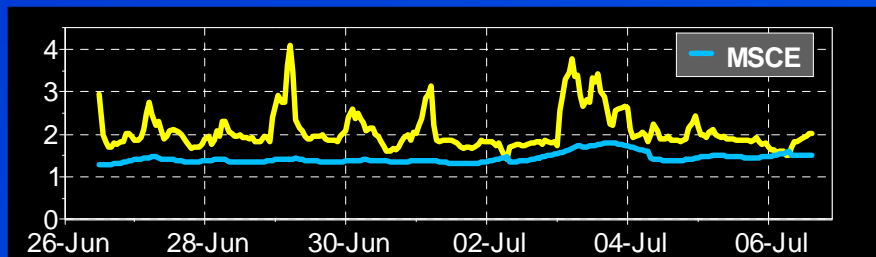
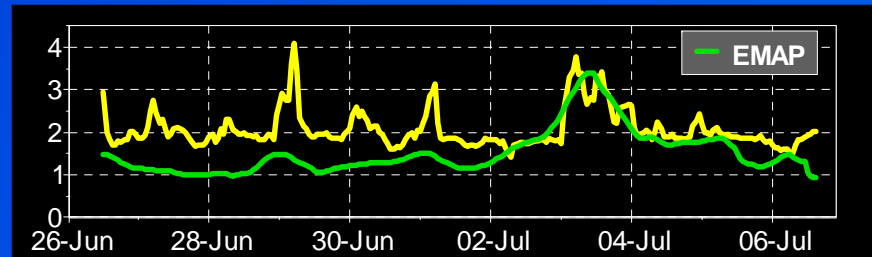
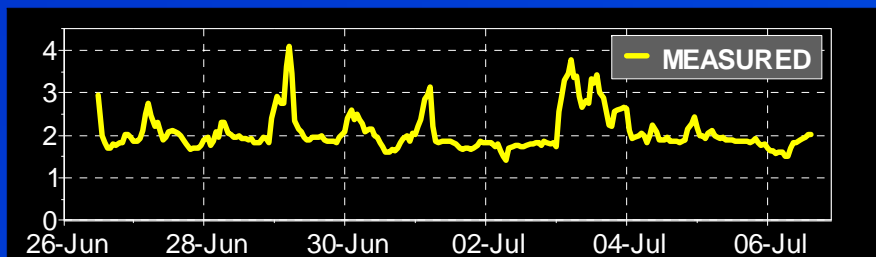
Mercury at Neuglobsow: June 26 – July 6, 1995



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

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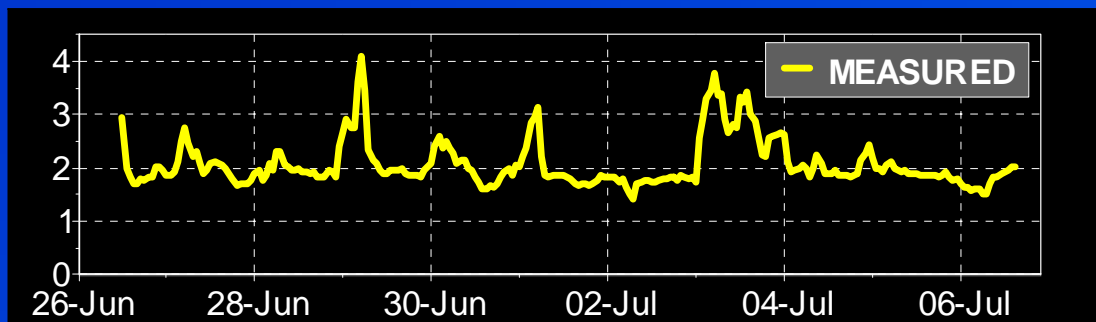
Total Gaseous Mercury (ng/m³) at Neuglobsow: June 26 – July 6, 1995



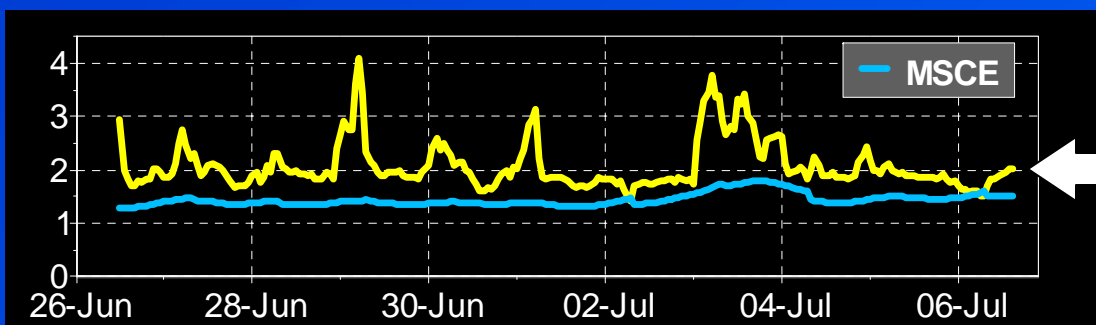
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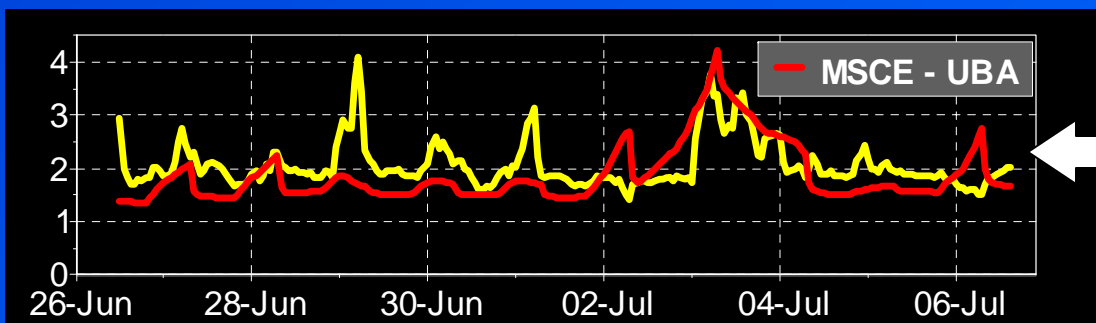
Total Gaseous Mercury (ng/m³) at Neuglobsow: June 26 – July 6, 1995



The emissions inventory is a critical input to the models...



Using default emissions inventory

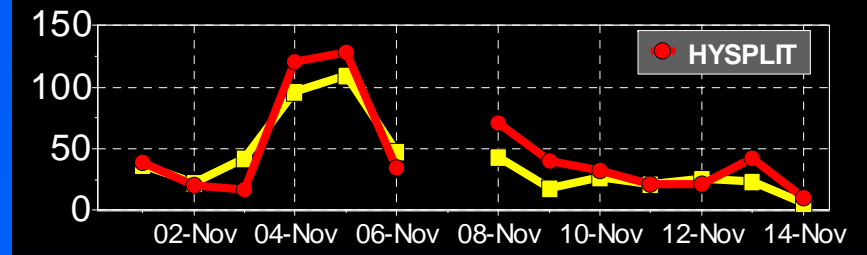
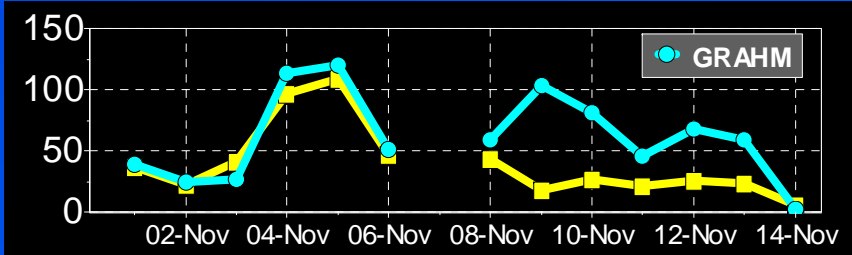
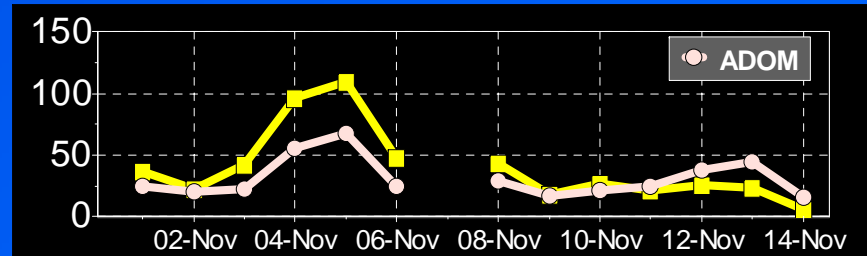
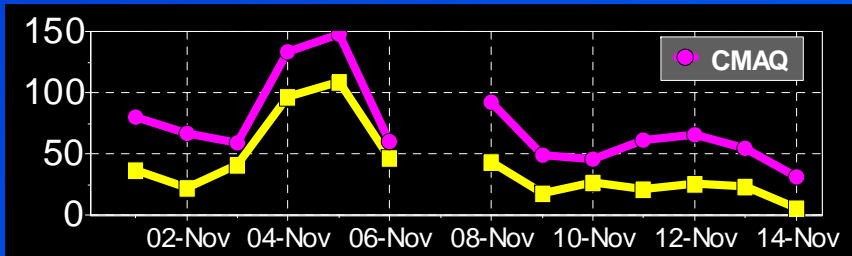
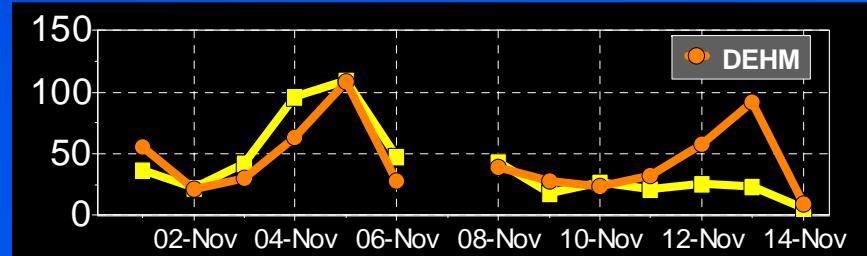
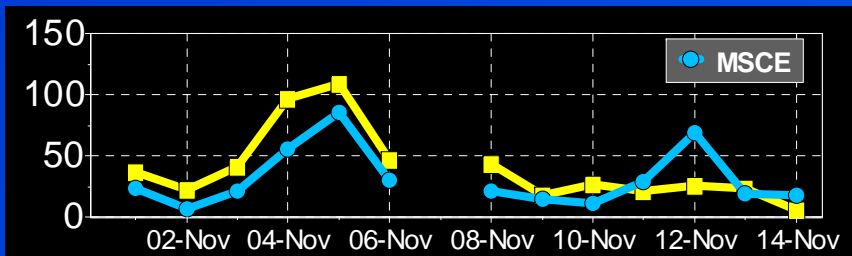
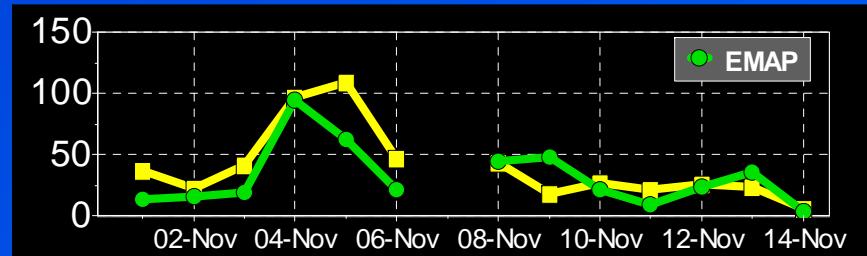
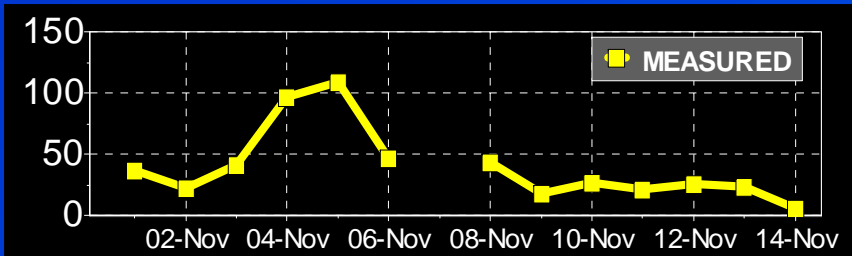


Using alternative emissions inventory

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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

Total *Particulate* Mercury (pg/m³) at Neuglobsow, Nov 1-14, 1999



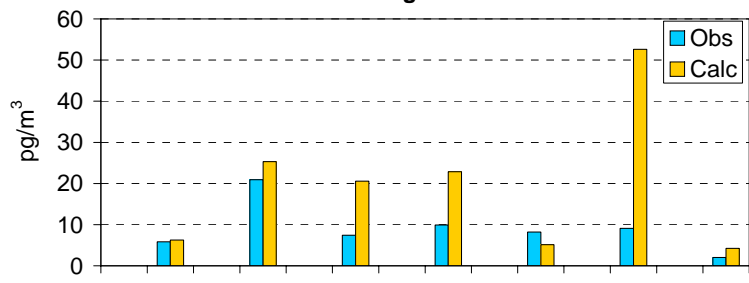
EMEP Intercomparison Study of Numerical Models for

Intro- duction	Stage I	Stage II		
	Chemistry	Hg ⁰	Hg(p)	RGM

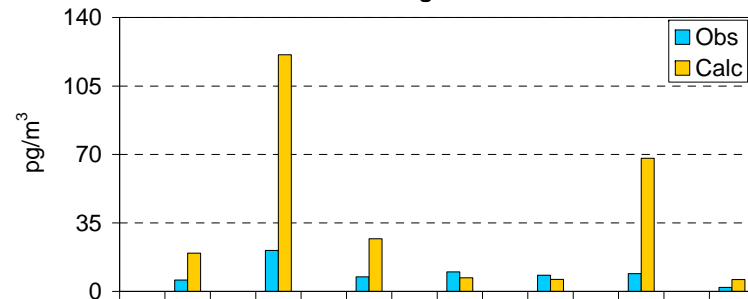
conclu-
ions

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

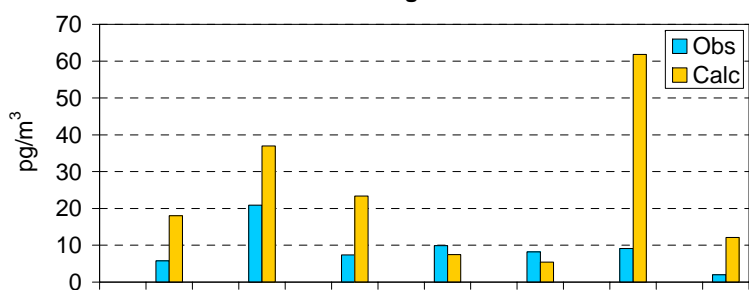
MSCE Neuglobsow RGM



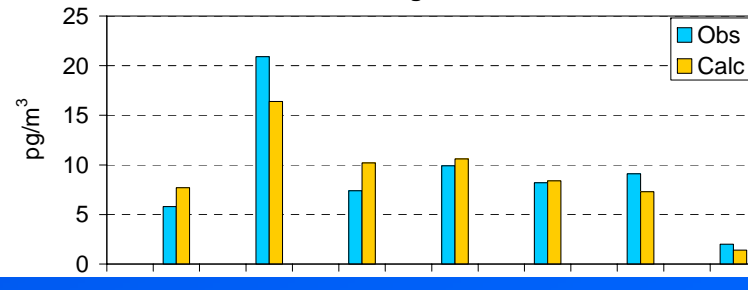
GRAHM Neuglobsow RGM



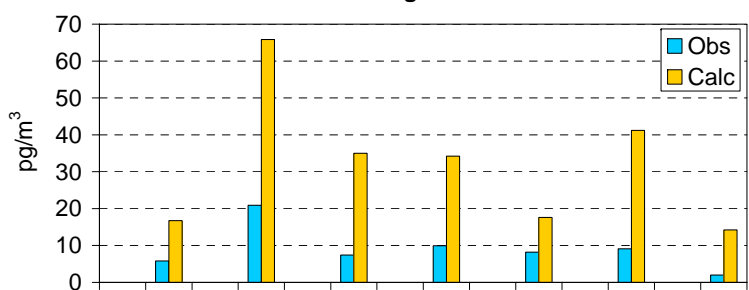
ADOM Neuglobsow RGM



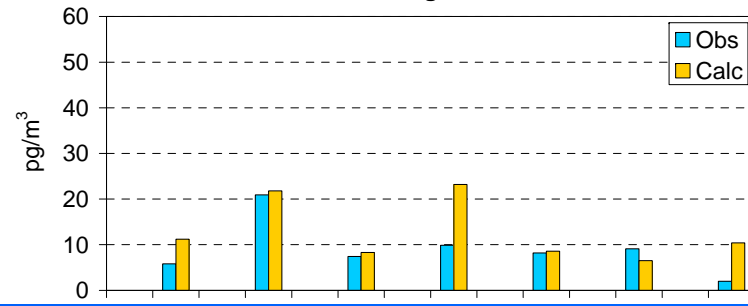
EMAP Neuglobsow RGM



CMAQ Neuglobsow RGM



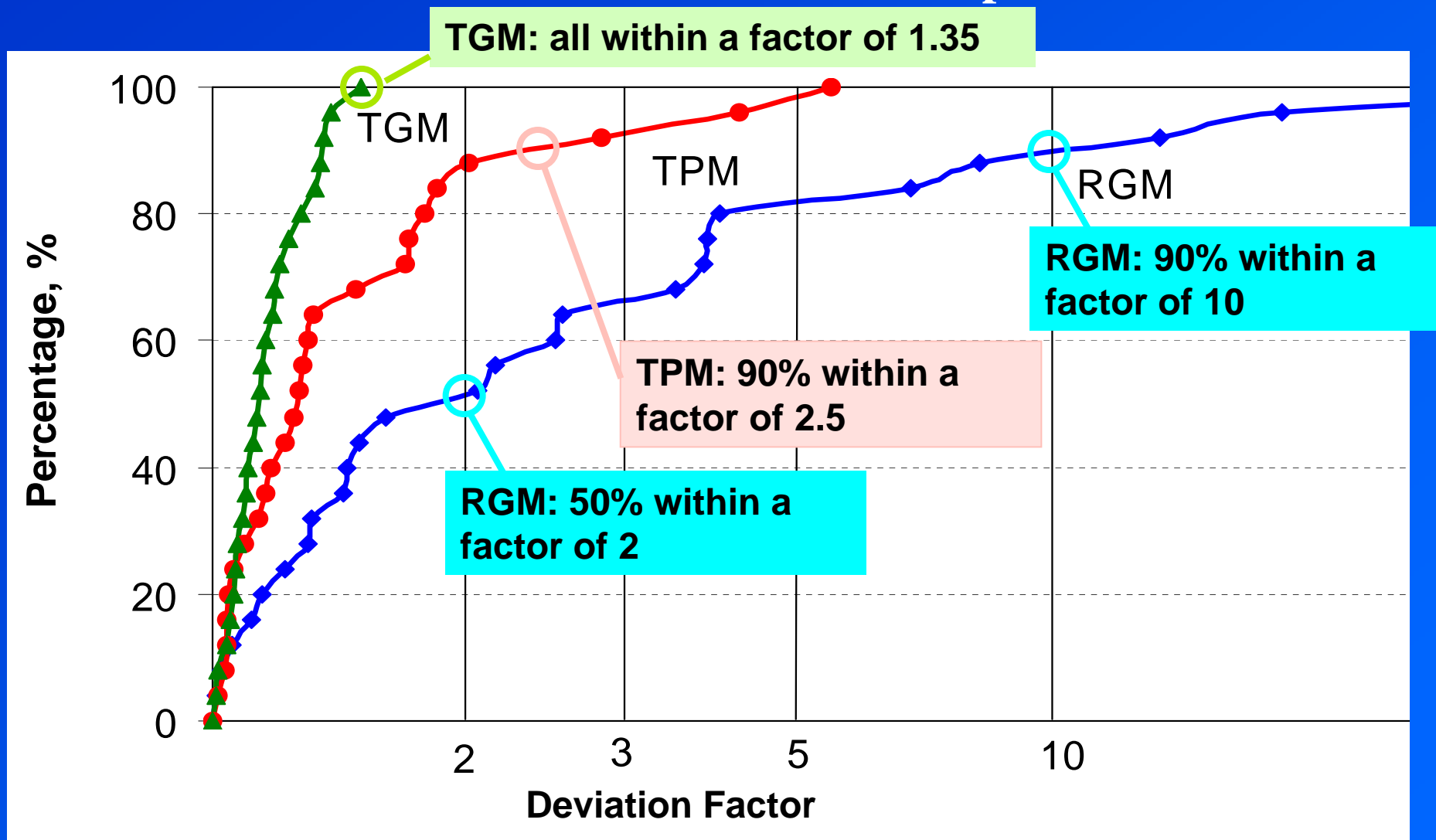
HYSPLIT Neuglobsow RGM



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Overall Phase II statistics for 2-week episode means



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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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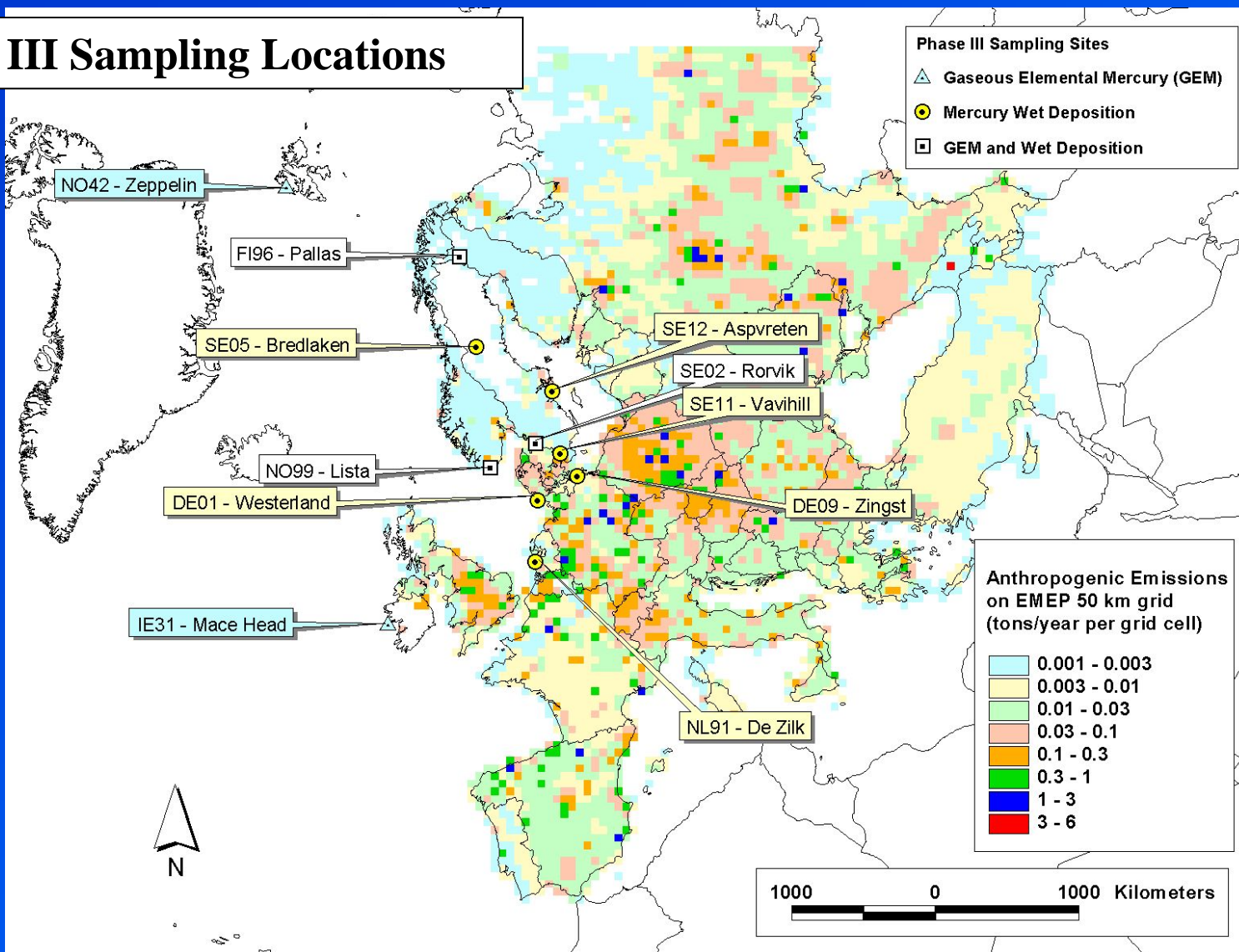
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Phase III Sampling Locations



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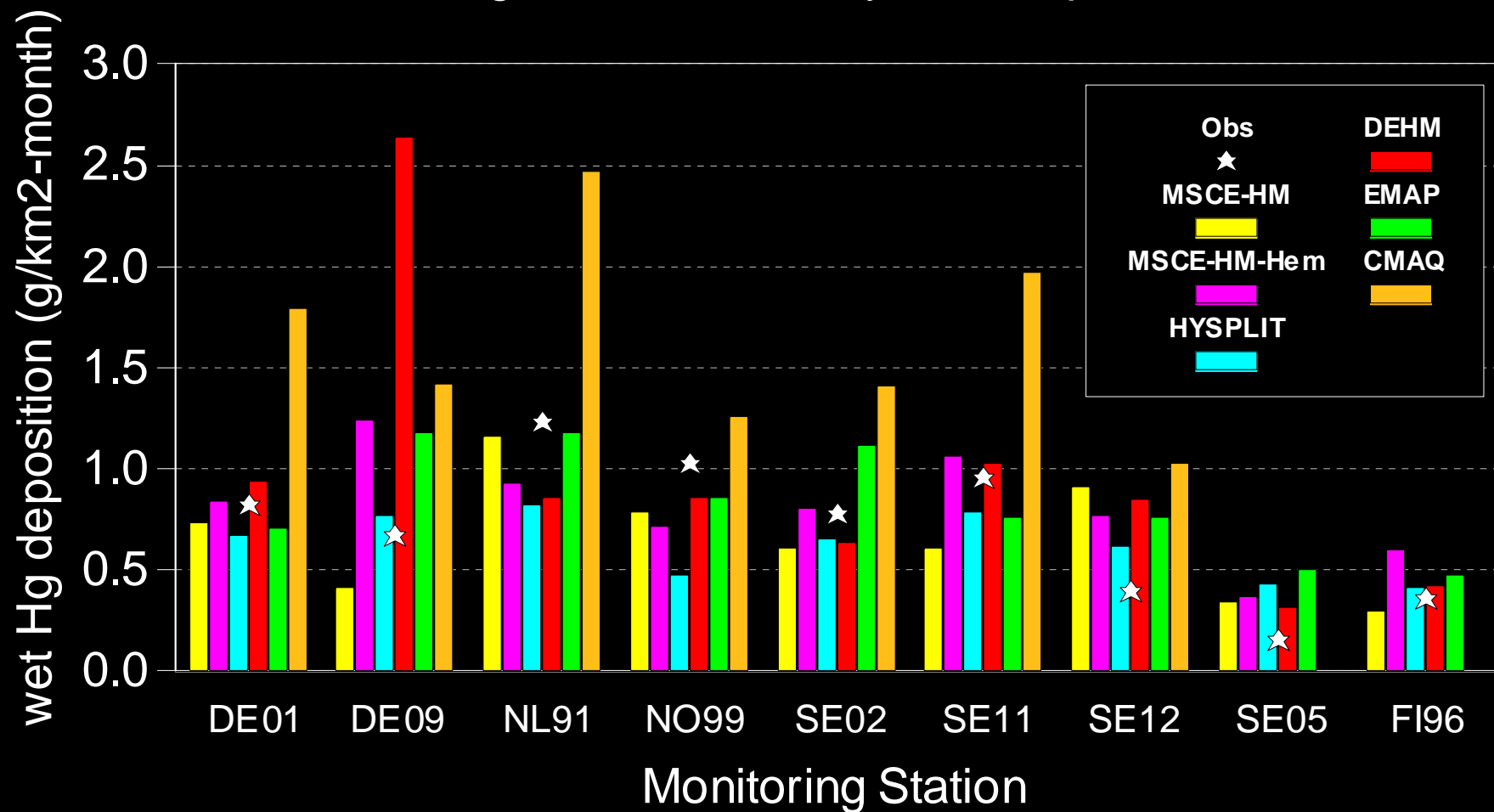
Due to resource constraints, not all models simulated the entire year 1999...

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

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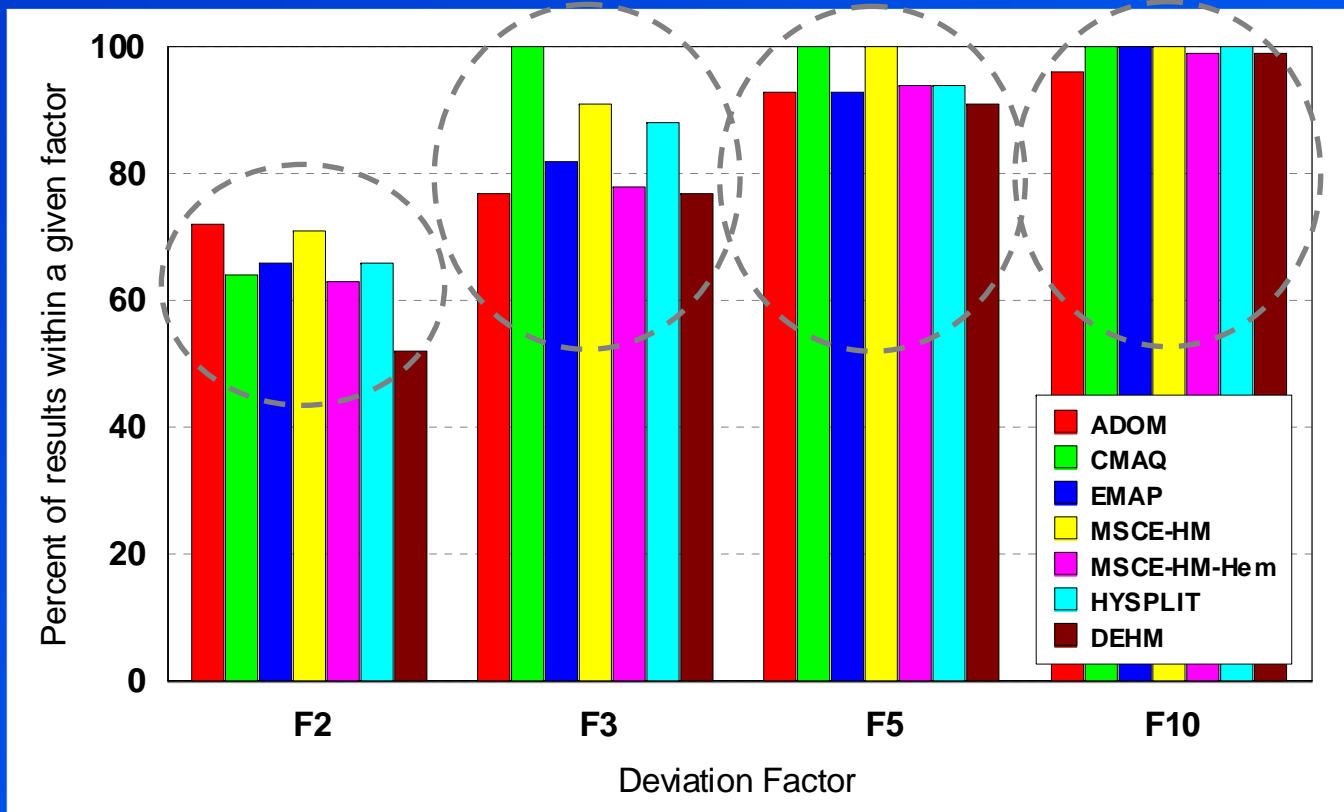
August 1999 Mercury Wet Deposition



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

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~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

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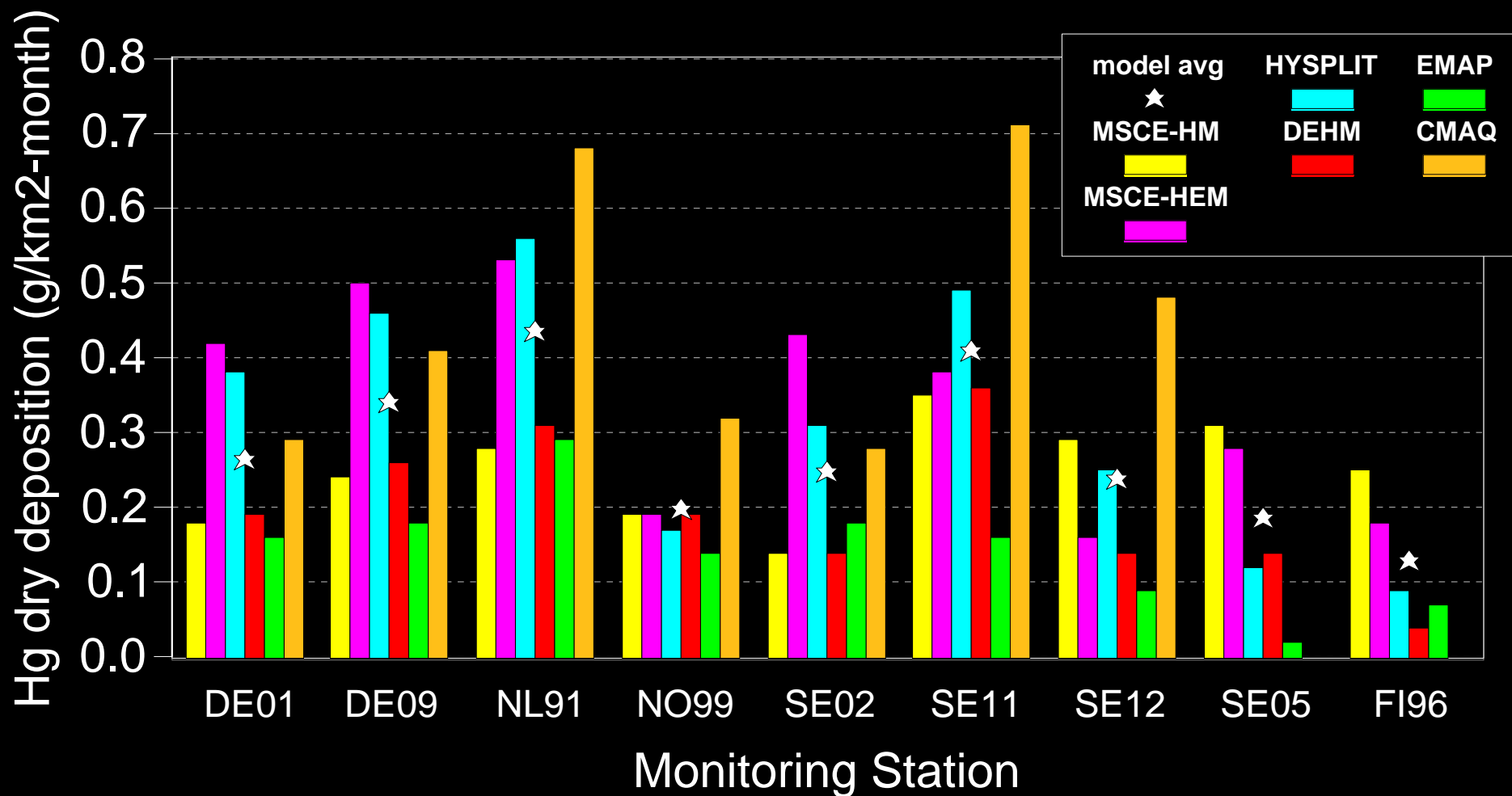
Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
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- For *dry deposition*, there are no measurement results to compare the models against;
- However, the models can be compared against *each other...*

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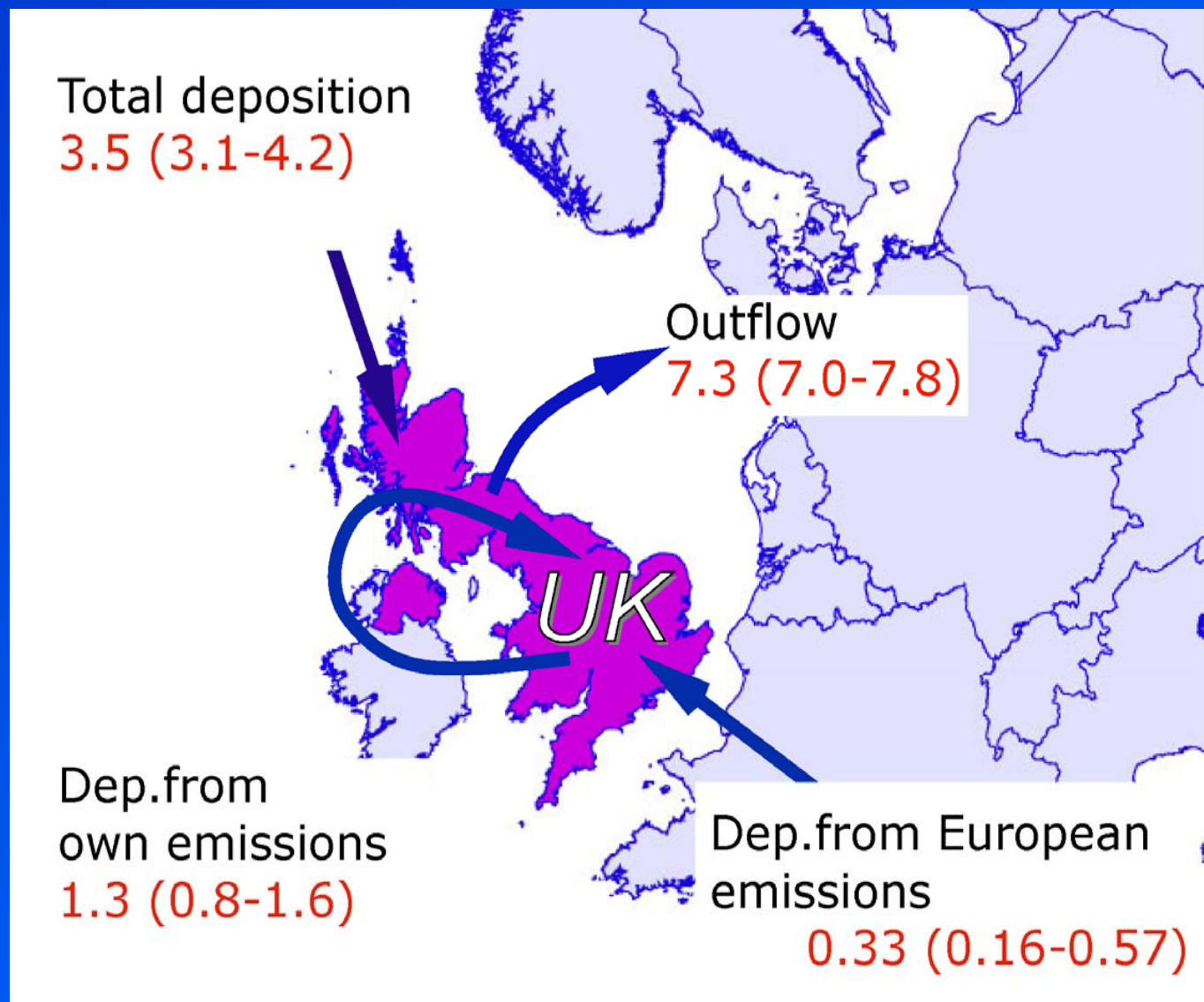
August 1999 Mercury Dry Deposition



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

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



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

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**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 <i>(3.1-4.2)</i>	4.7 <i>(3.2-6.6)</i>	11.8 <i>(9.6-13.1)</i>
Dep. from own emissions	1.3 <i>(0.8-1.6)</i>	1.3 <i>(0.6-1.9)</i>	7.4 <i>(4.8-9.1)</i>
Dep. from European emissions	0.3 <i>(0.2-0.6)</i>	0.8 <i>(0.5-1.3)</i>	2.1 <i>(1.4-2.6)</i>
Outflow	7.3 <i>(7.0-7.8)</i>	8.4 <i>(7.9-9.2)</i>	18.2 <i>(16-21)</i>

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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

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.

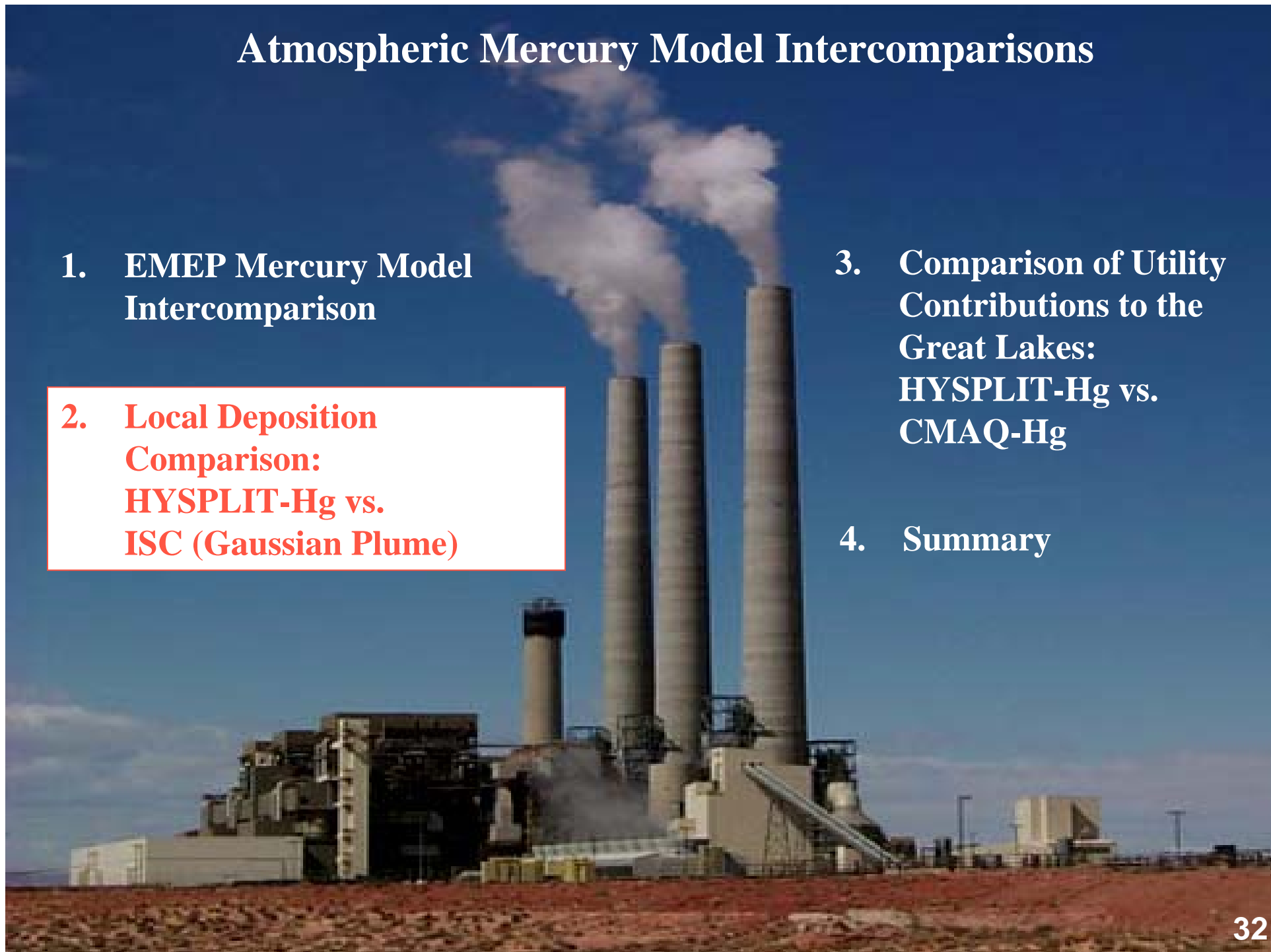
Atmospheric Mercury Model Intercomparisons

1. EMEP Mercury Model Intercomparison

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

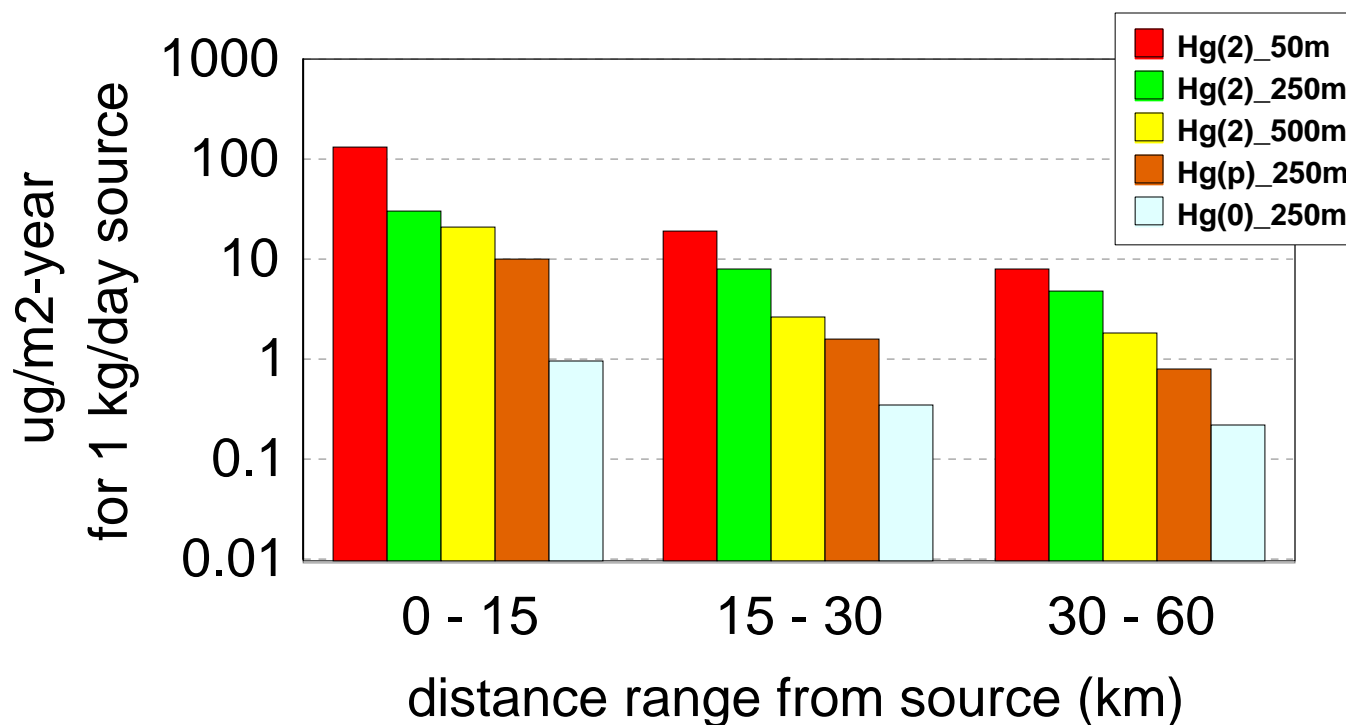
3. Comparison of Utility Contributions to the Great Lakes:
HYSPLIT-Hg vs.
CMAQ-Hg

4. Summary



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

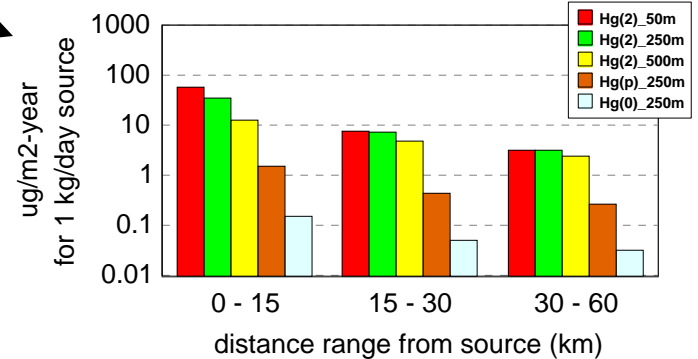
HYSPLIT 1996



ISC: 1990-1994

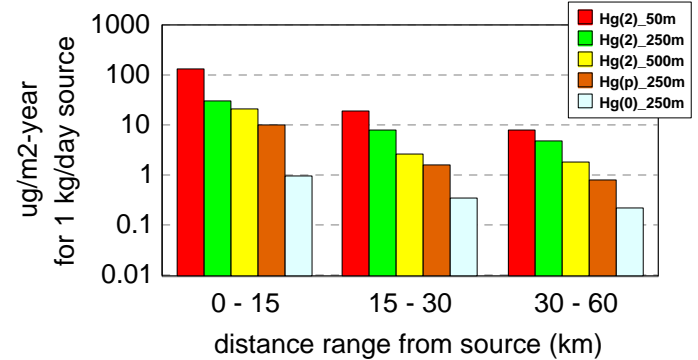


Wet + Dry Deposition: HYSPLIT (Nebraska)
for emissions of different mercury forms from different stack heights

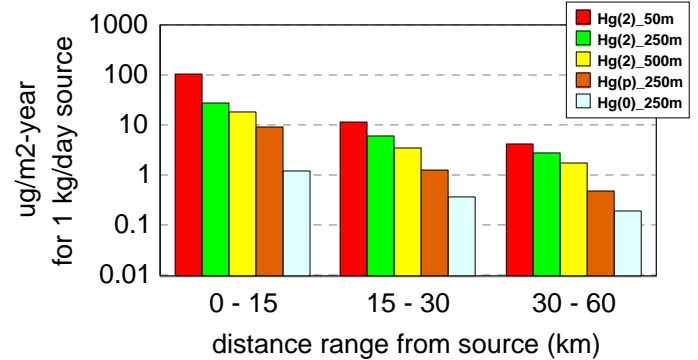


Different Time Periods and Locations, but Similar Results

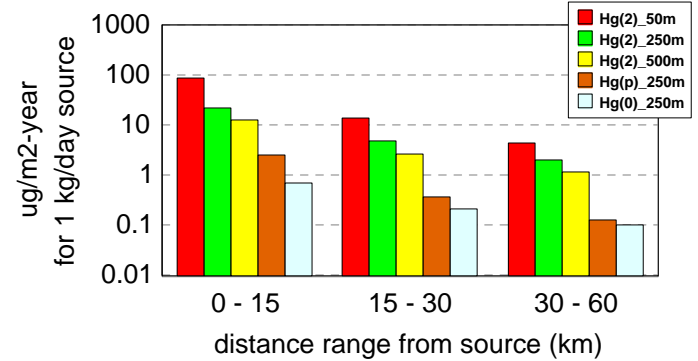
Wet + Dry Deposition: ISC (Kansas City)
for emissions of different mercury forms from different stack heights



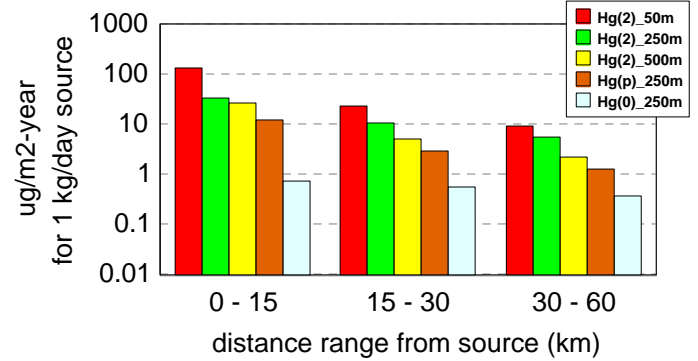
Wet + Dry Deposition: ISC (Tampa)
for emissions of different mercury forms from different stack heights



Wet + Dry Deposition: ISC (Phoenix)
for emissions of different mercury forms from different stack heights



Wet + Dry Deposition: ISC (Indianapolis)
for emissions of different mercury forms from different stack heights



Atmospheric Mercury Model Intercomparisons

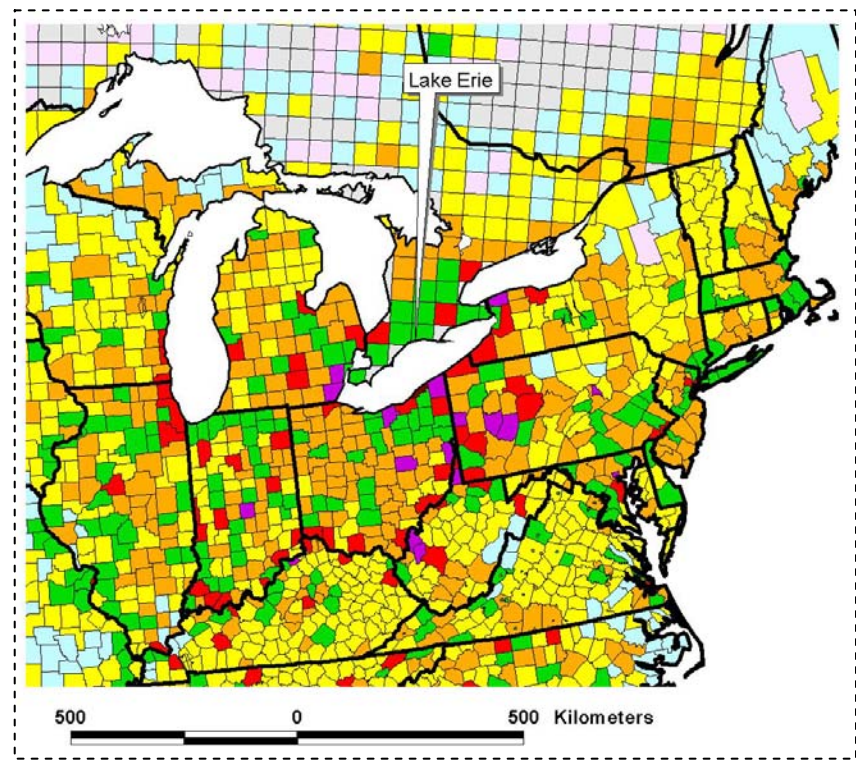
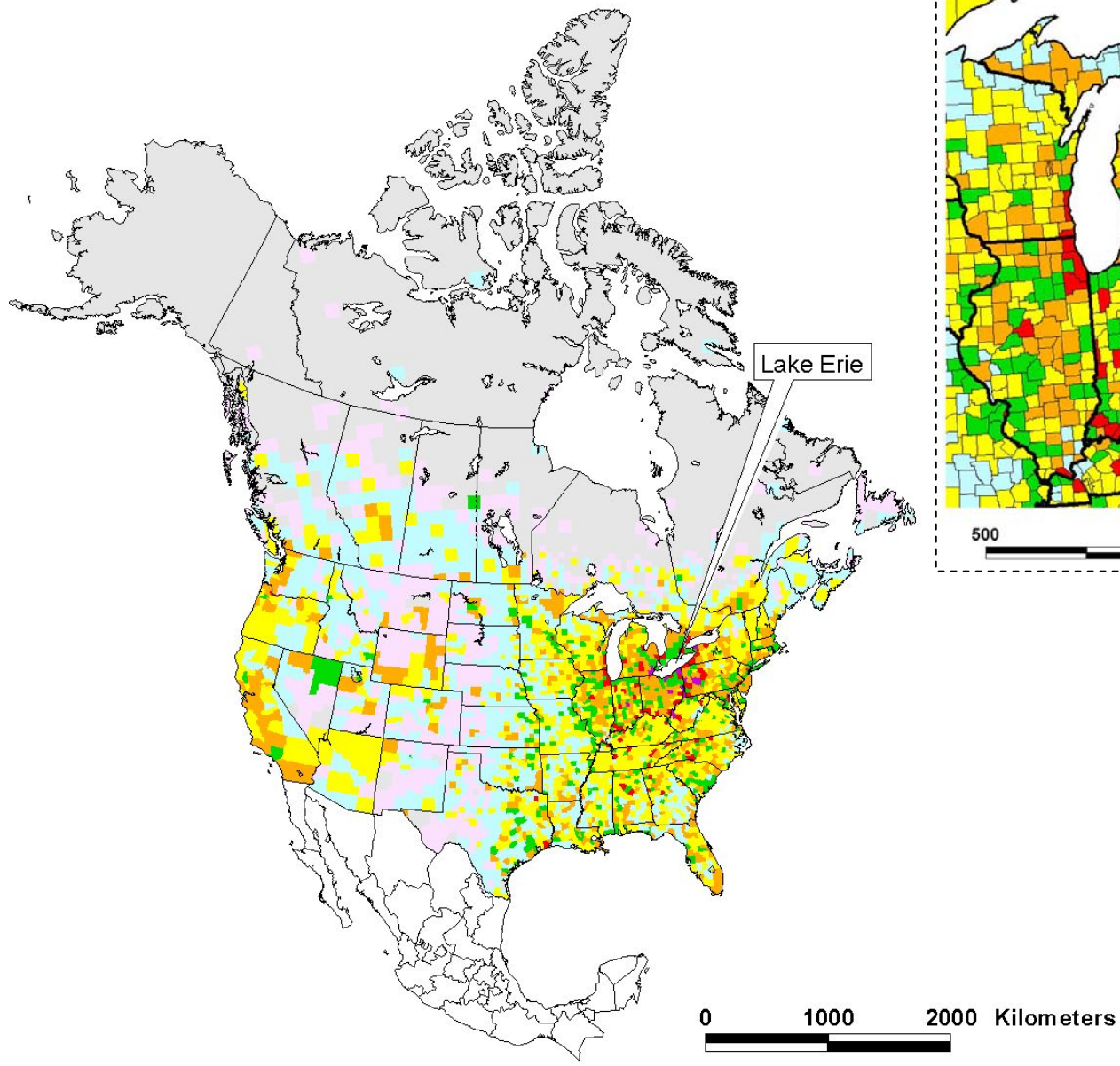
1. EMEP Mercury Model Intercomparison

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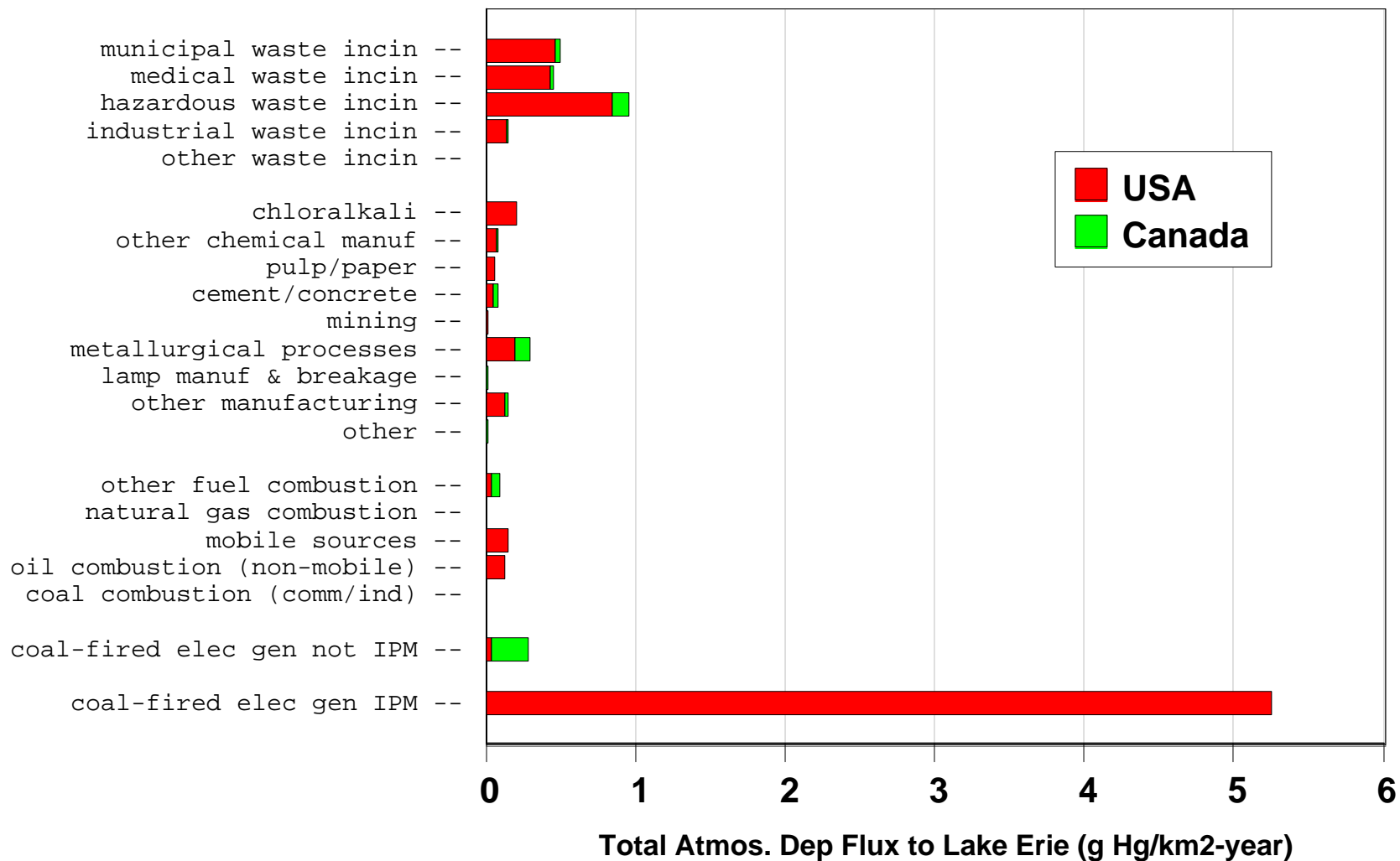
HYSPLIT-Hg results for Lake Erie (1999)



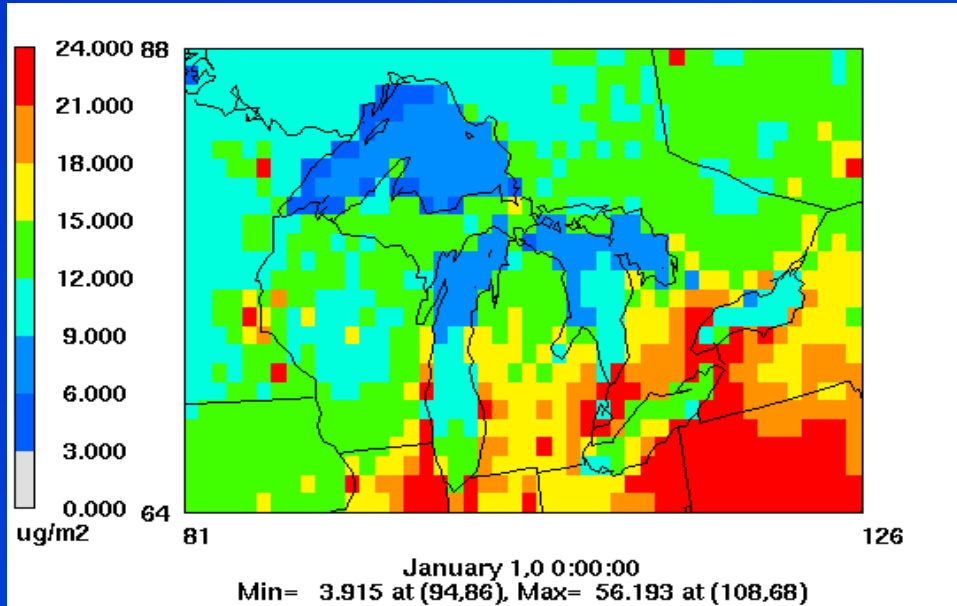
Deposition Contribution of Source Area to Receptor (ug deposited / year per km² of receptor area) per (km² of source area)

- 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

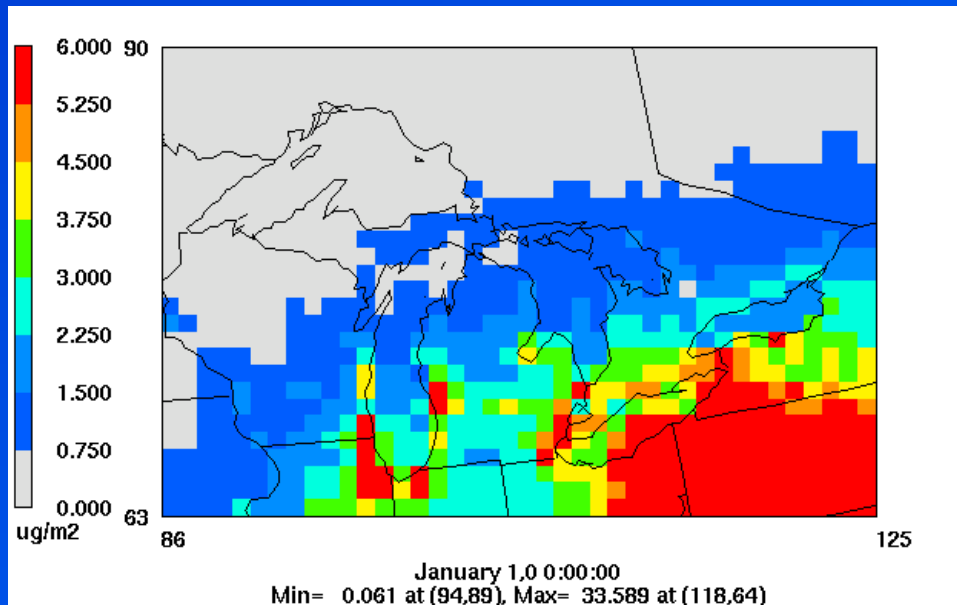
HYSPLIT-Hg results for Lake Erie (1999)



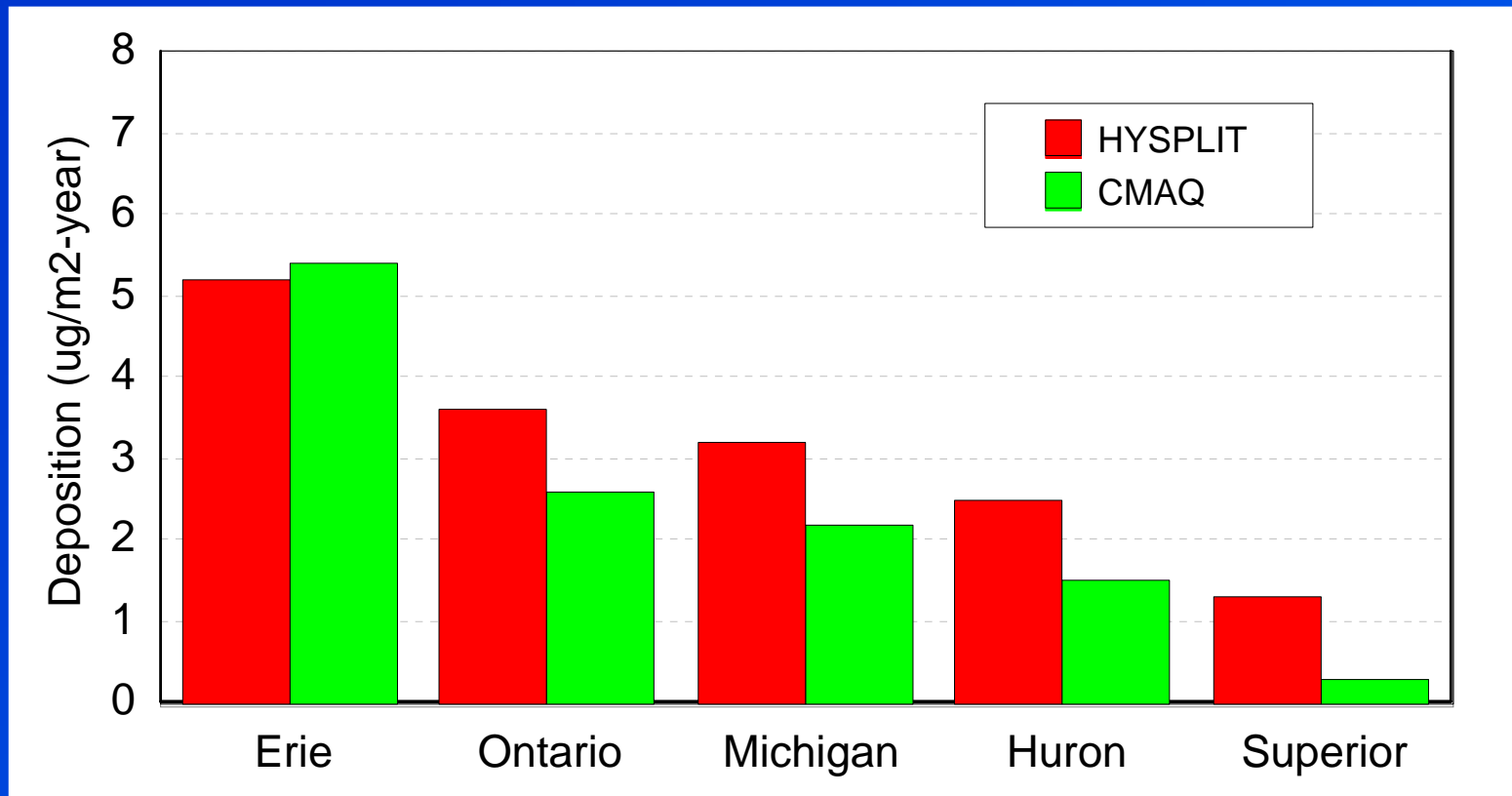
CMAQ-Hg results from EPA analysis performed for the Clean Air Mercury Rule



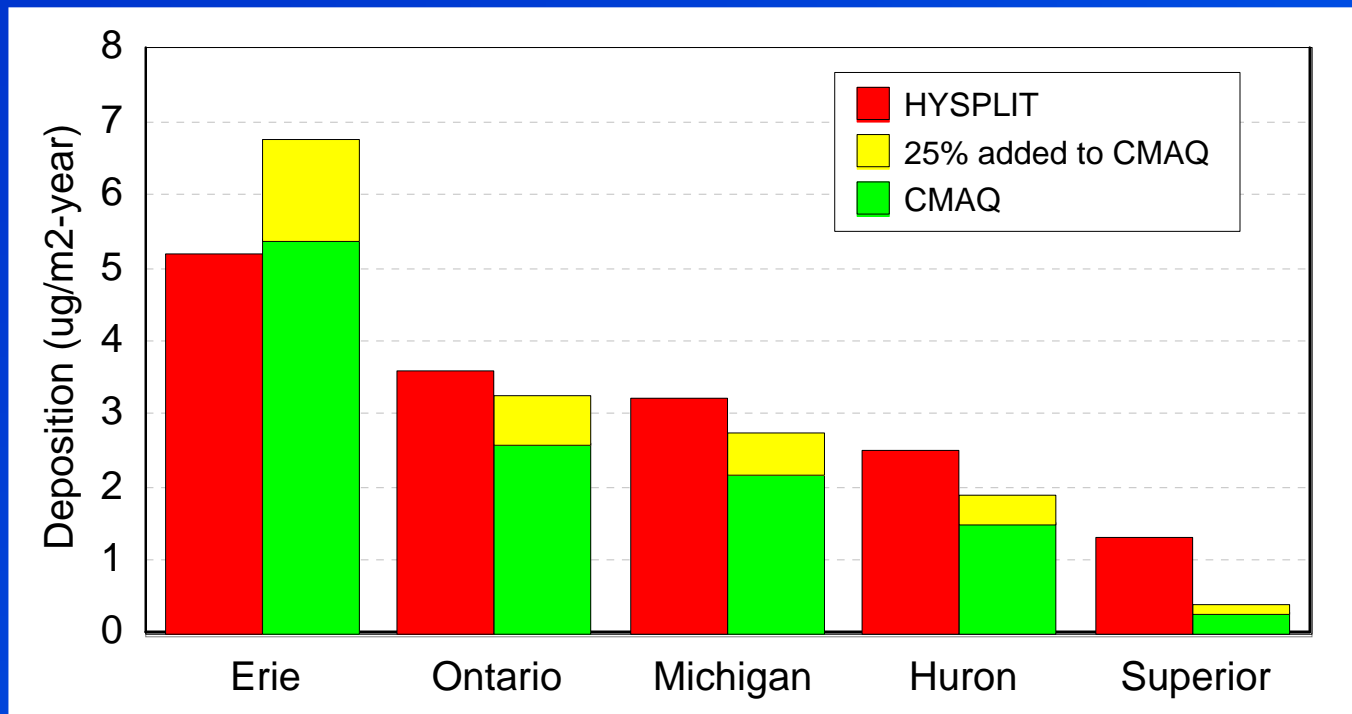
**Modeled Mercury
Deposition in the
Great Lakes Region
from all sources
during 2001**



**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.

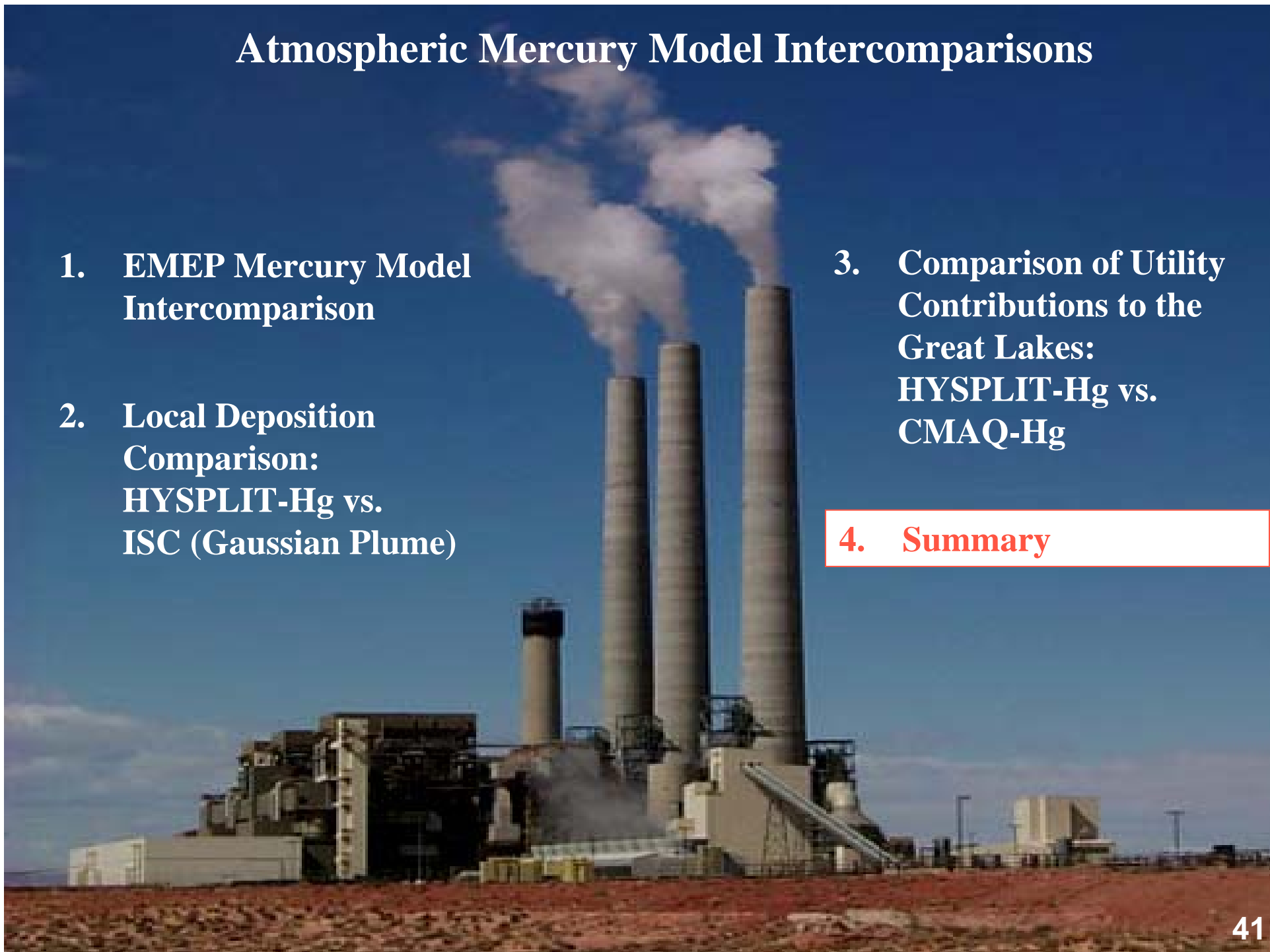
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4. Summary



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!

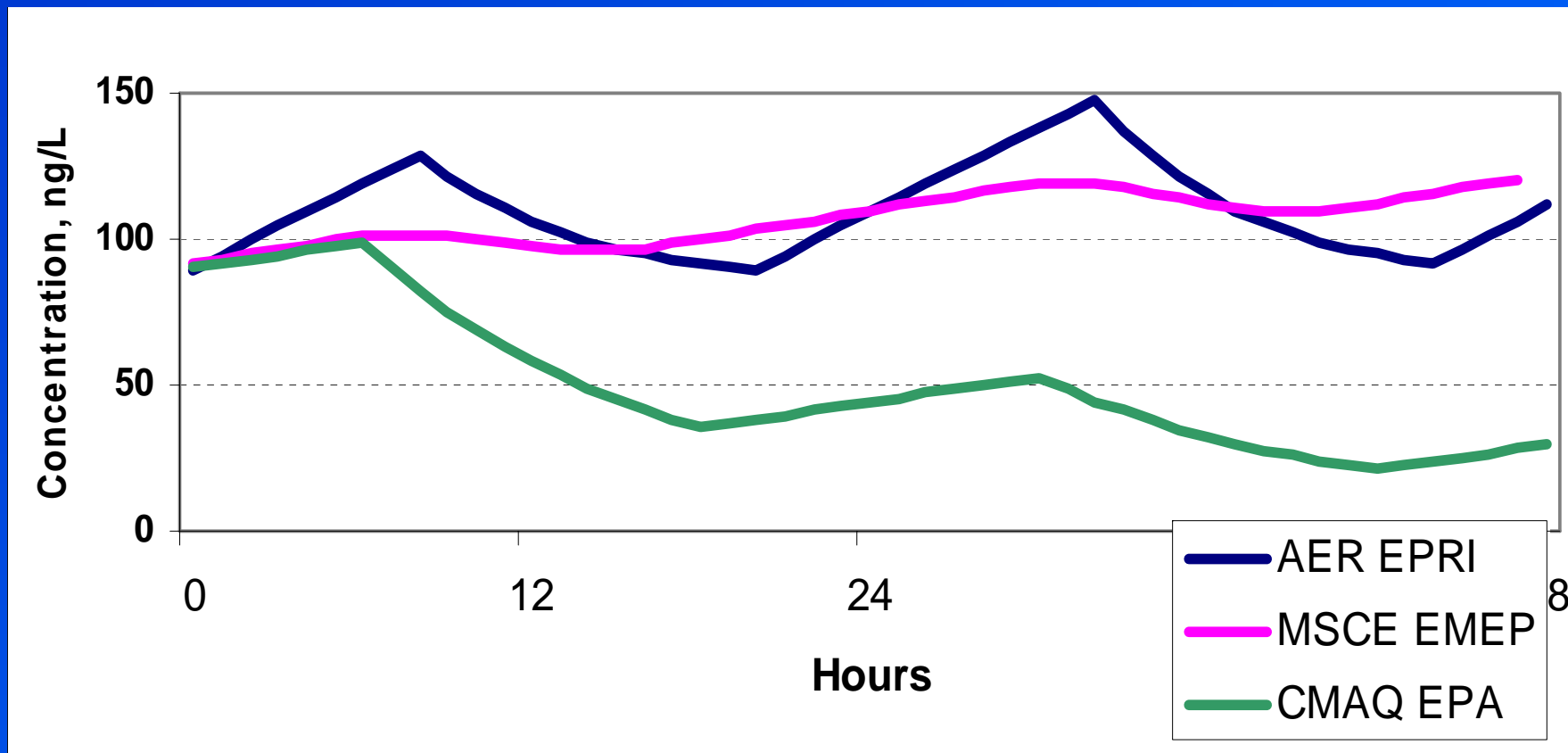
Extra Slides

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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

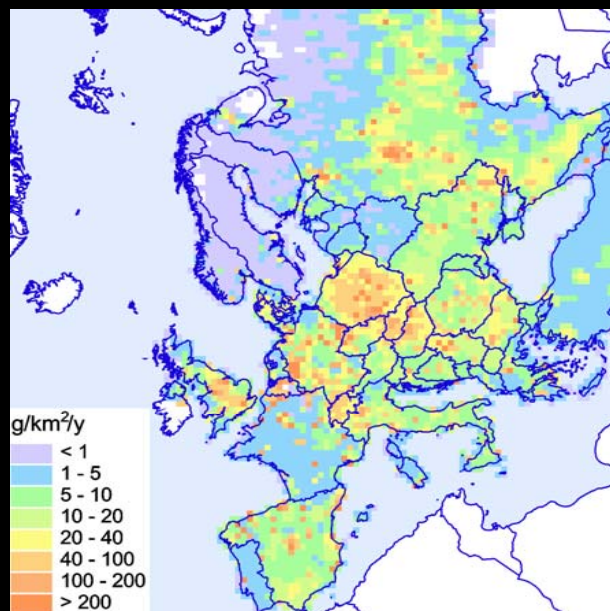
Effect of Different Assumptions Regarding Hg(p) Solubility

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



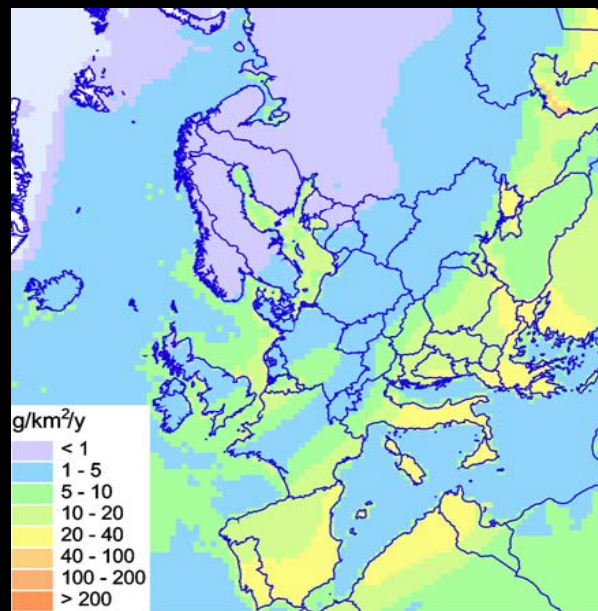
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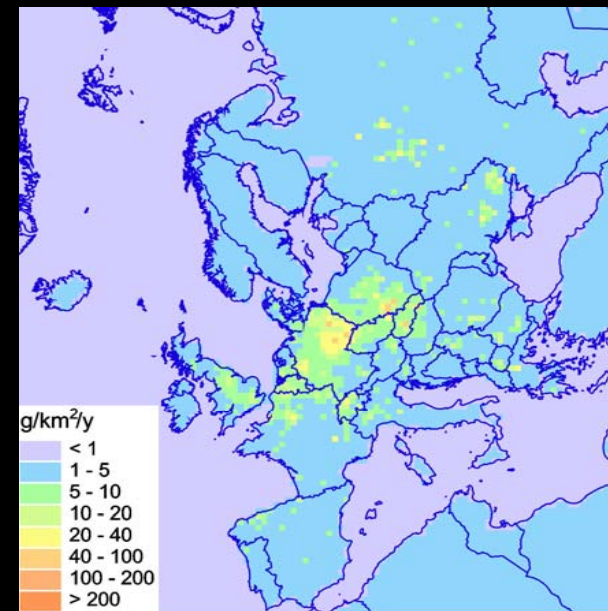
2000 European anthropogenic Hg emissions

240 t/yr



European natural Hg emissions

180 t/yr

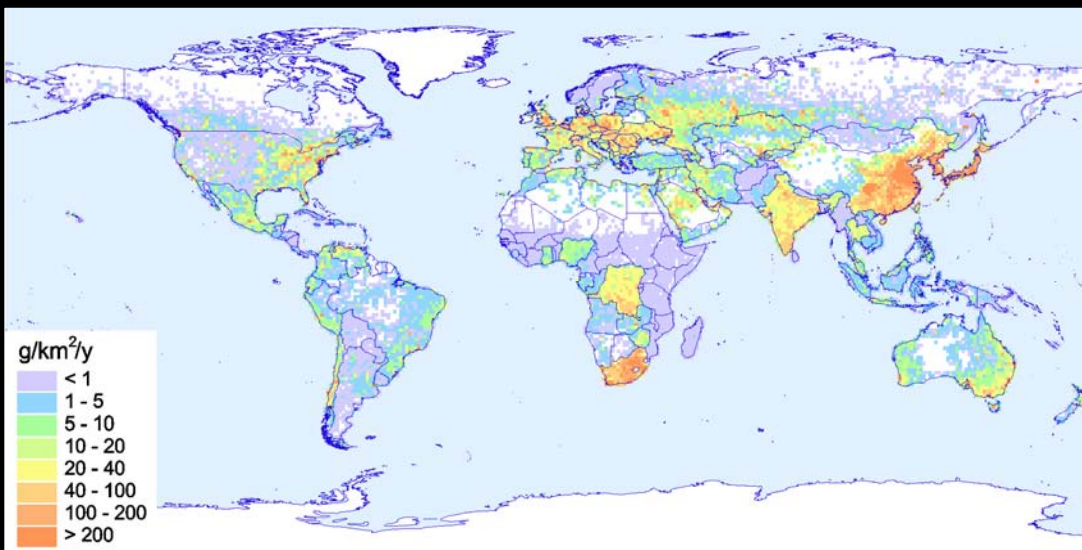


European anthropogenic Hg re-emissions

50 t/yr

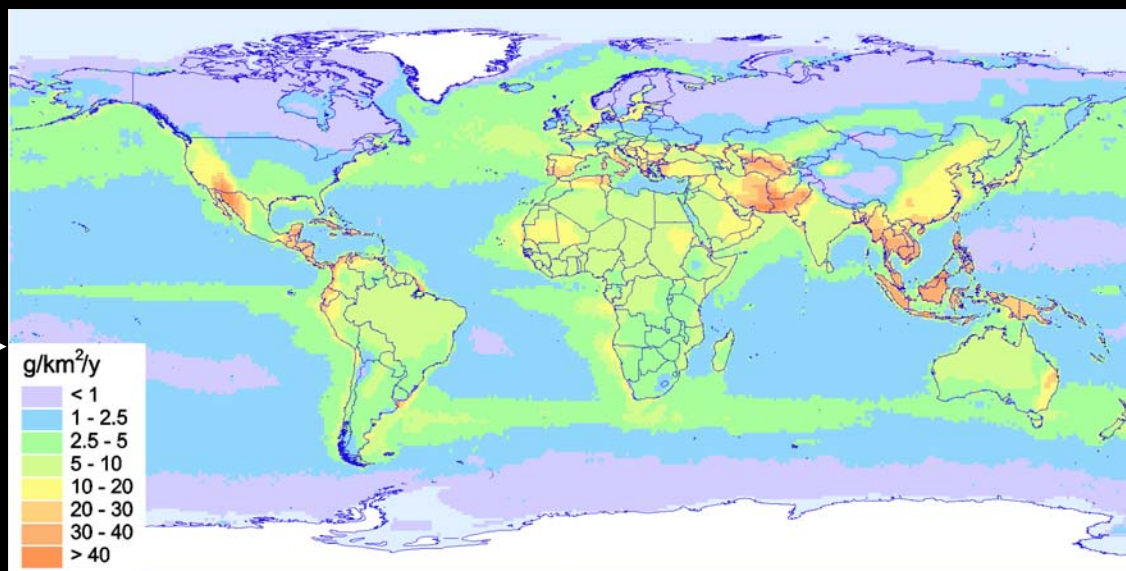
EMEP Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
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← 1995 global anthropogenic Hg emissions 1900 t/yr

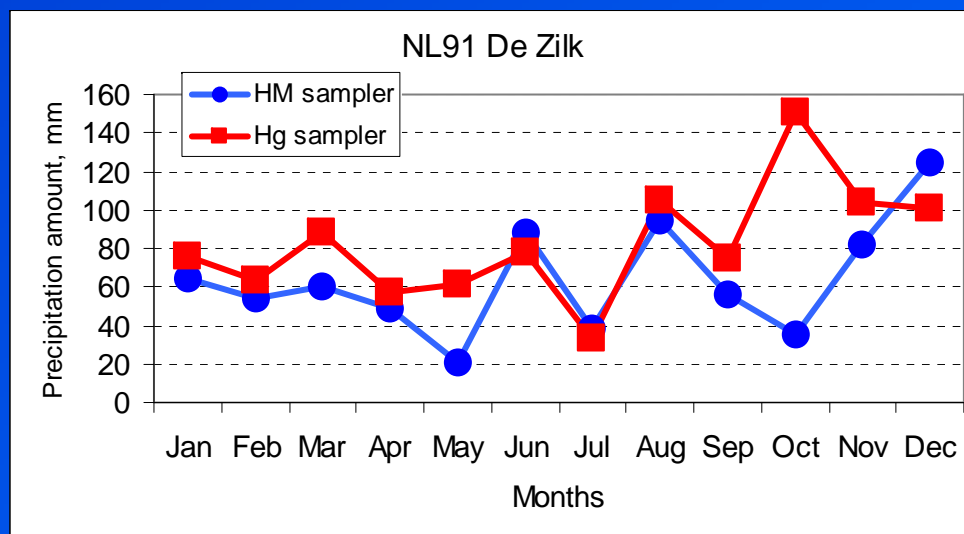
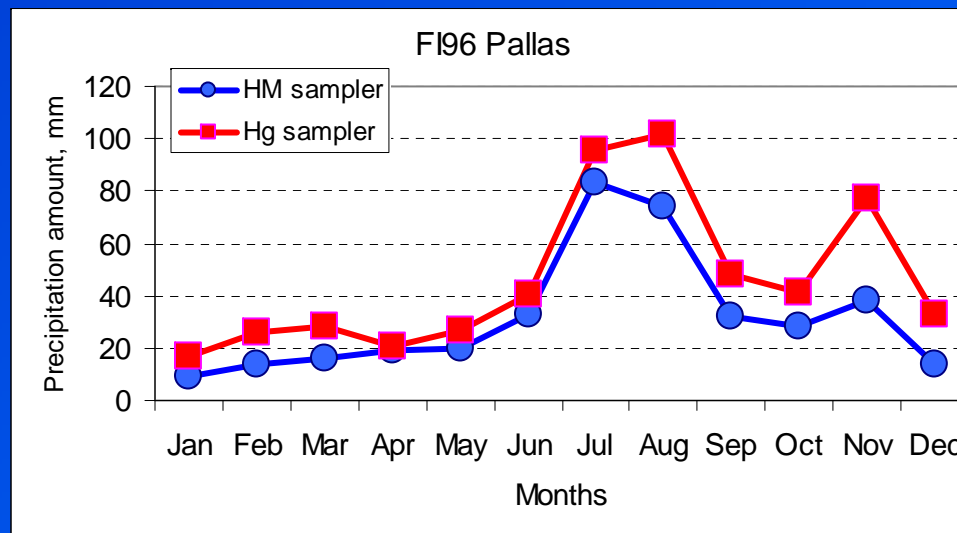
global natural Hg emissions 1800 t/yr →



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

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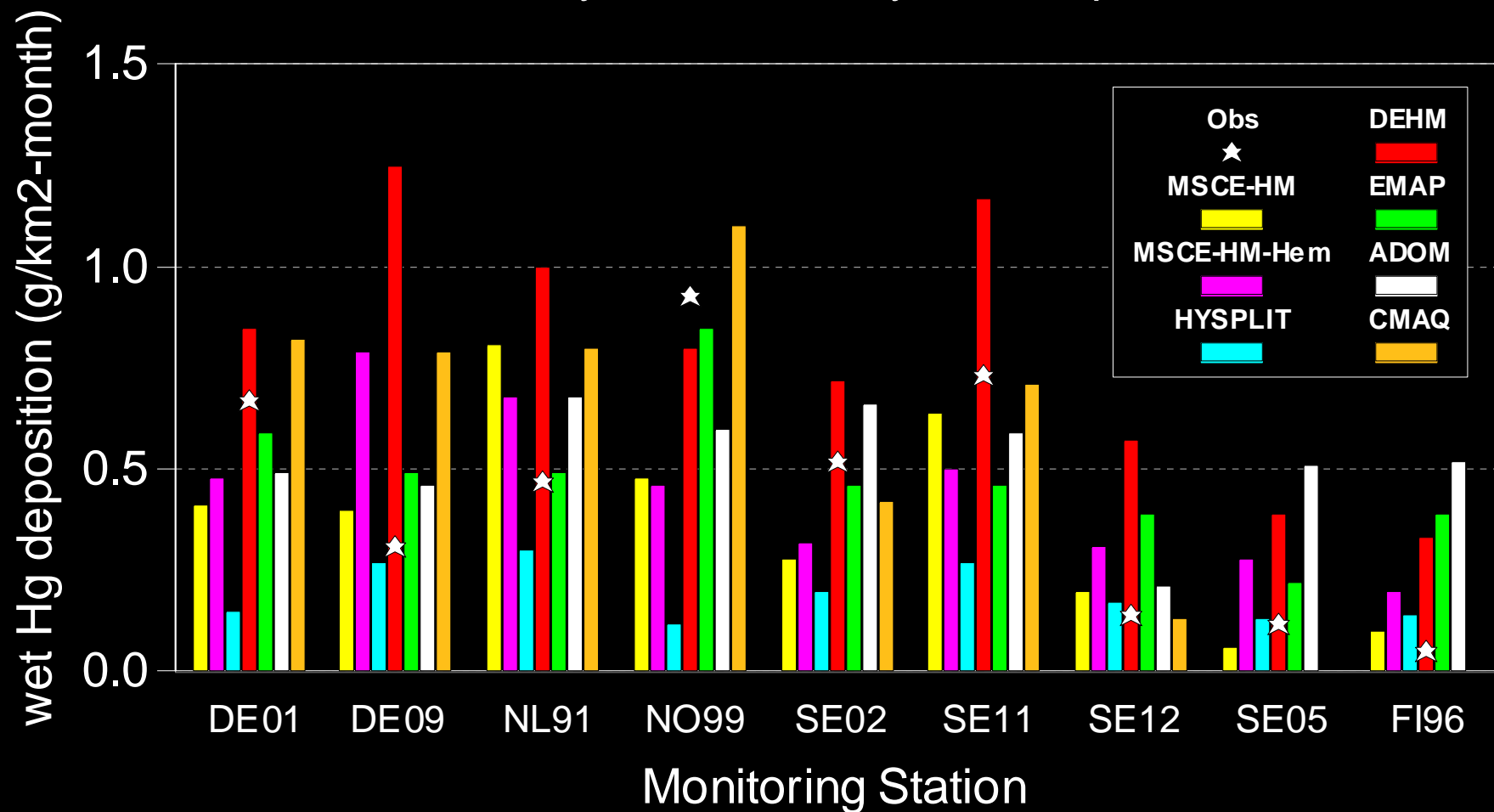
There are uncertainties in measurements -- even of precipitation amount...



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

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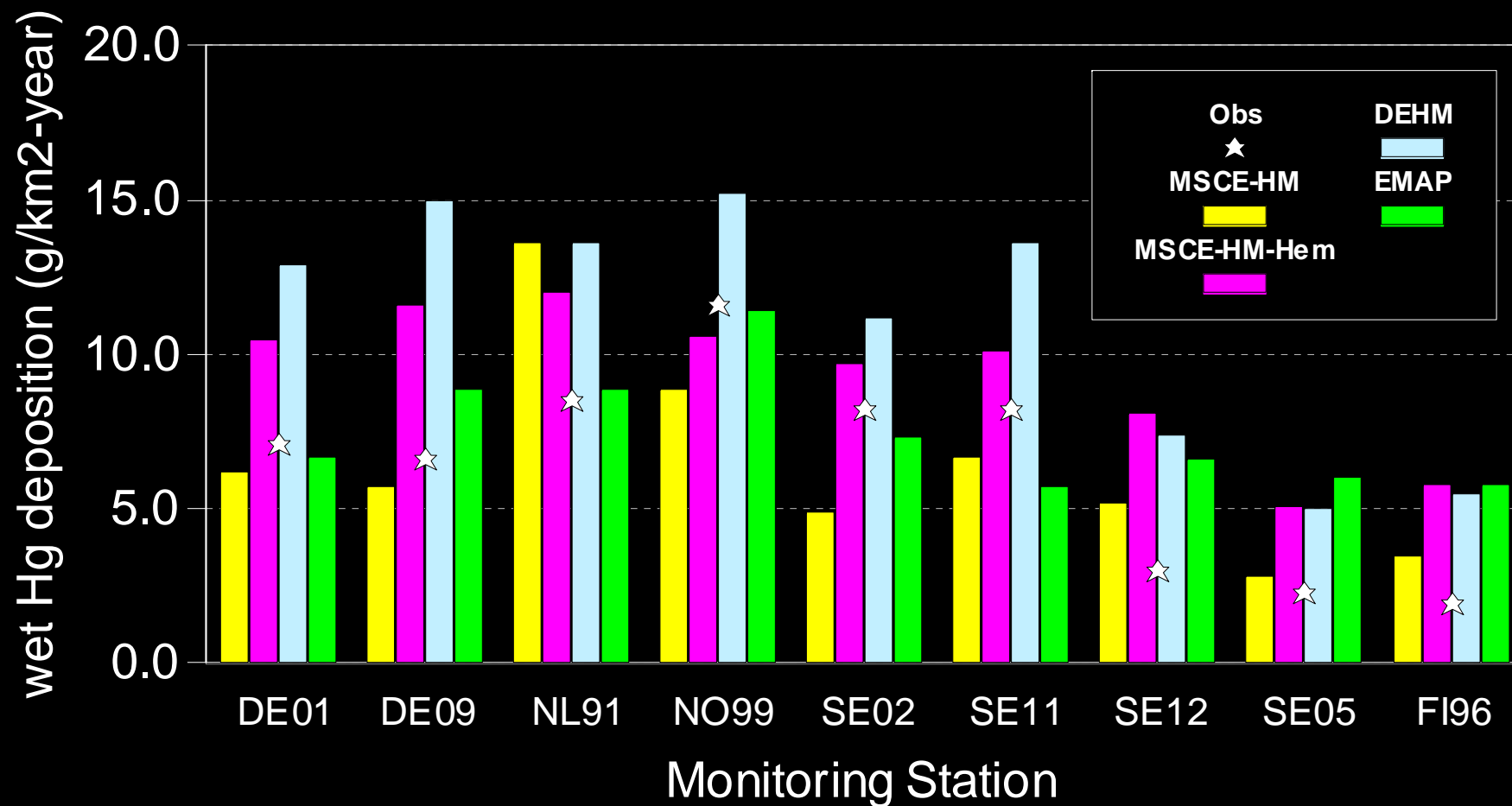
February 1999 Mercury Wet Deposition



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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

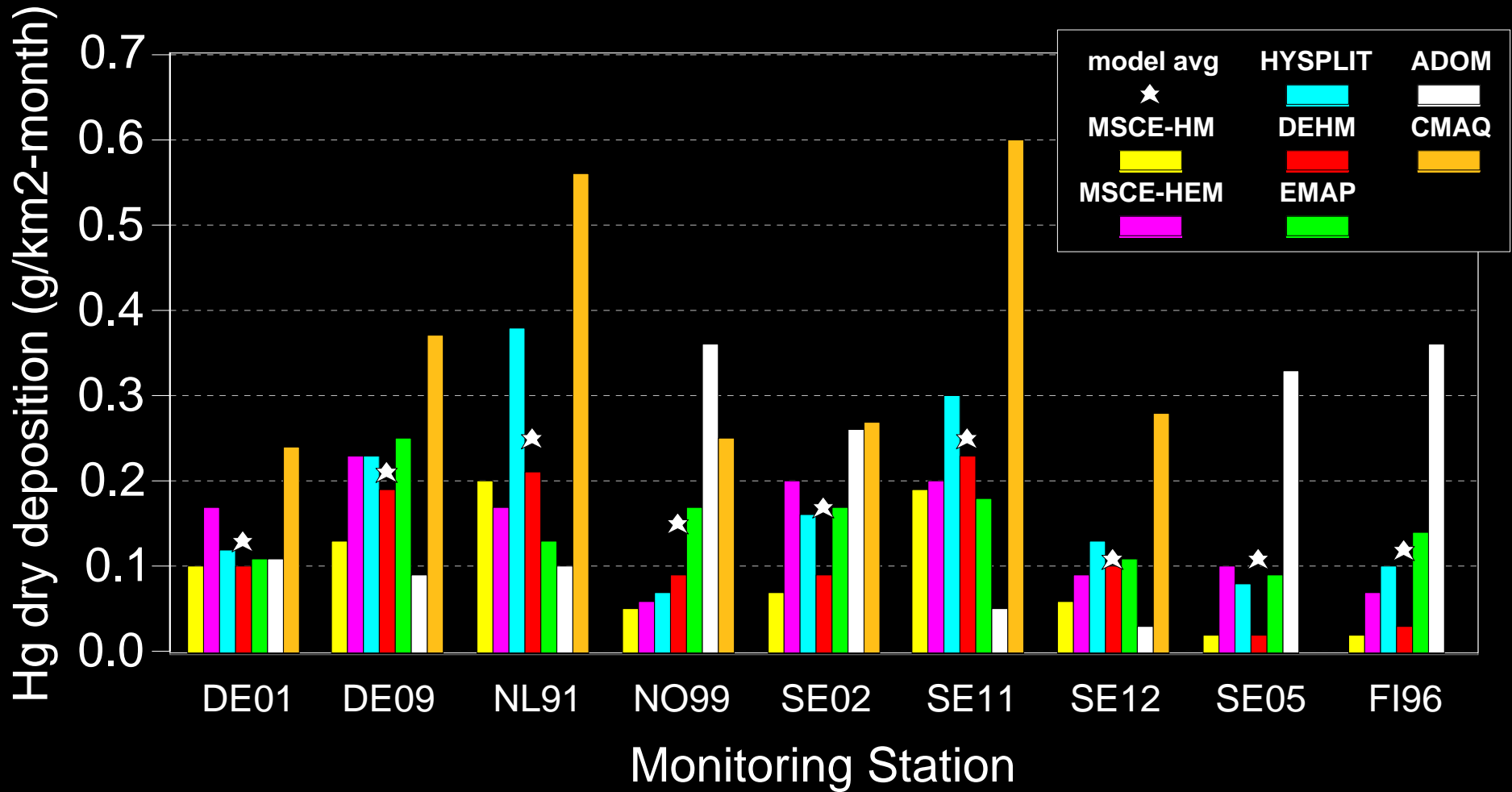
Full Year 1999 Mercury Wet Deposition



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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

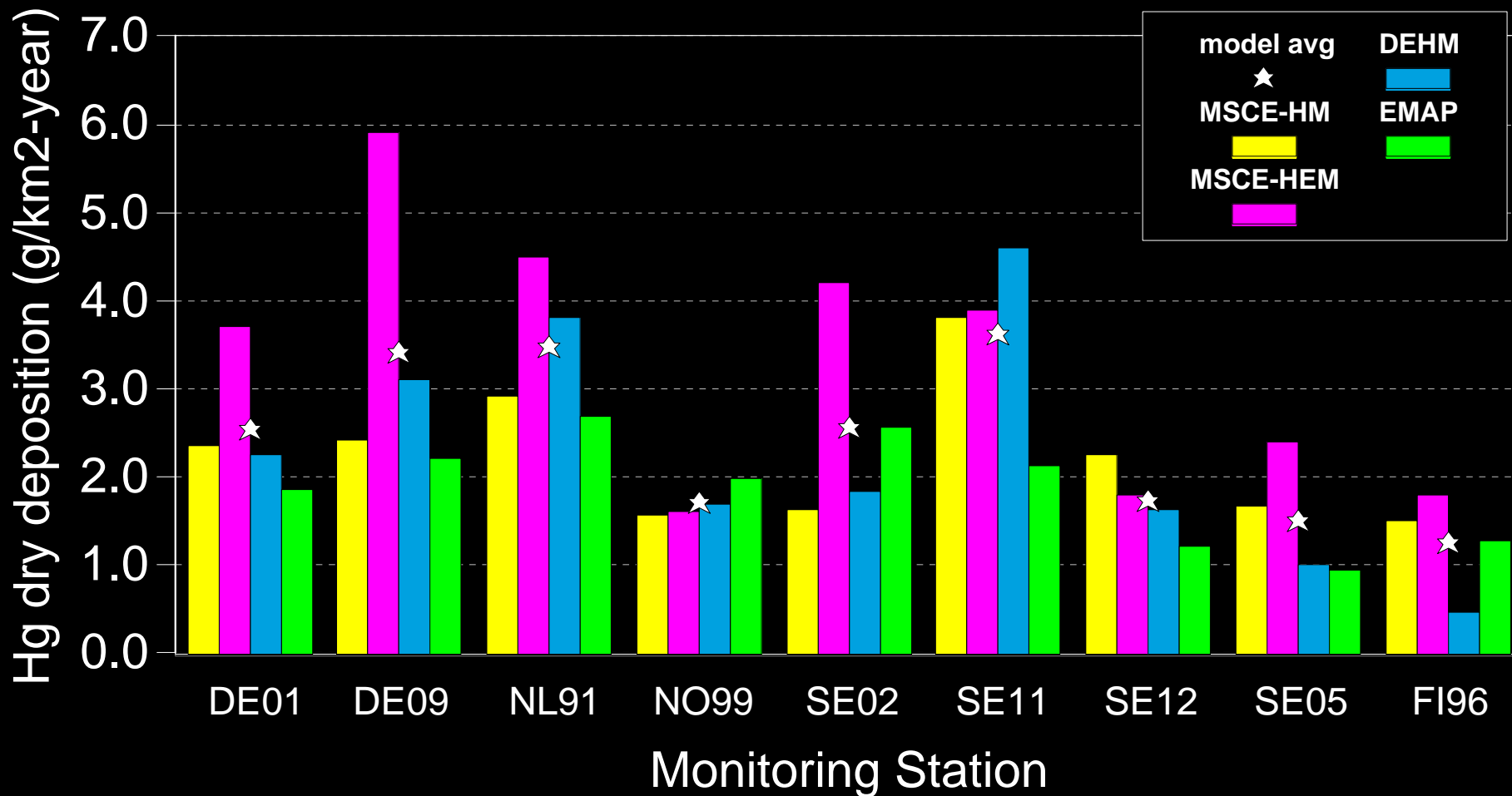
February 1999 Mercury Dry Deposition



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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

Full Year 1999 Mercury Dry Deposition



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

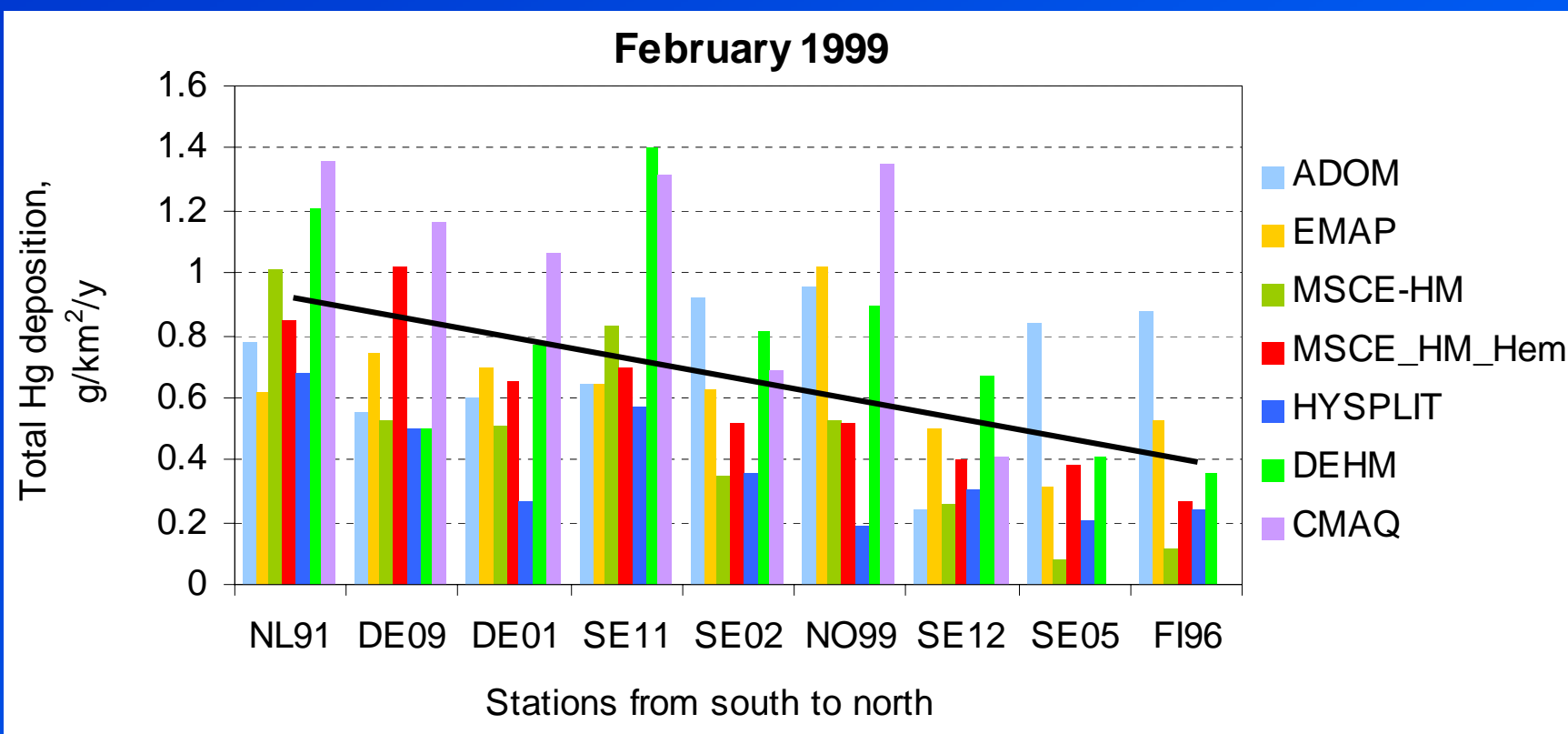
Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

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

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

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	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

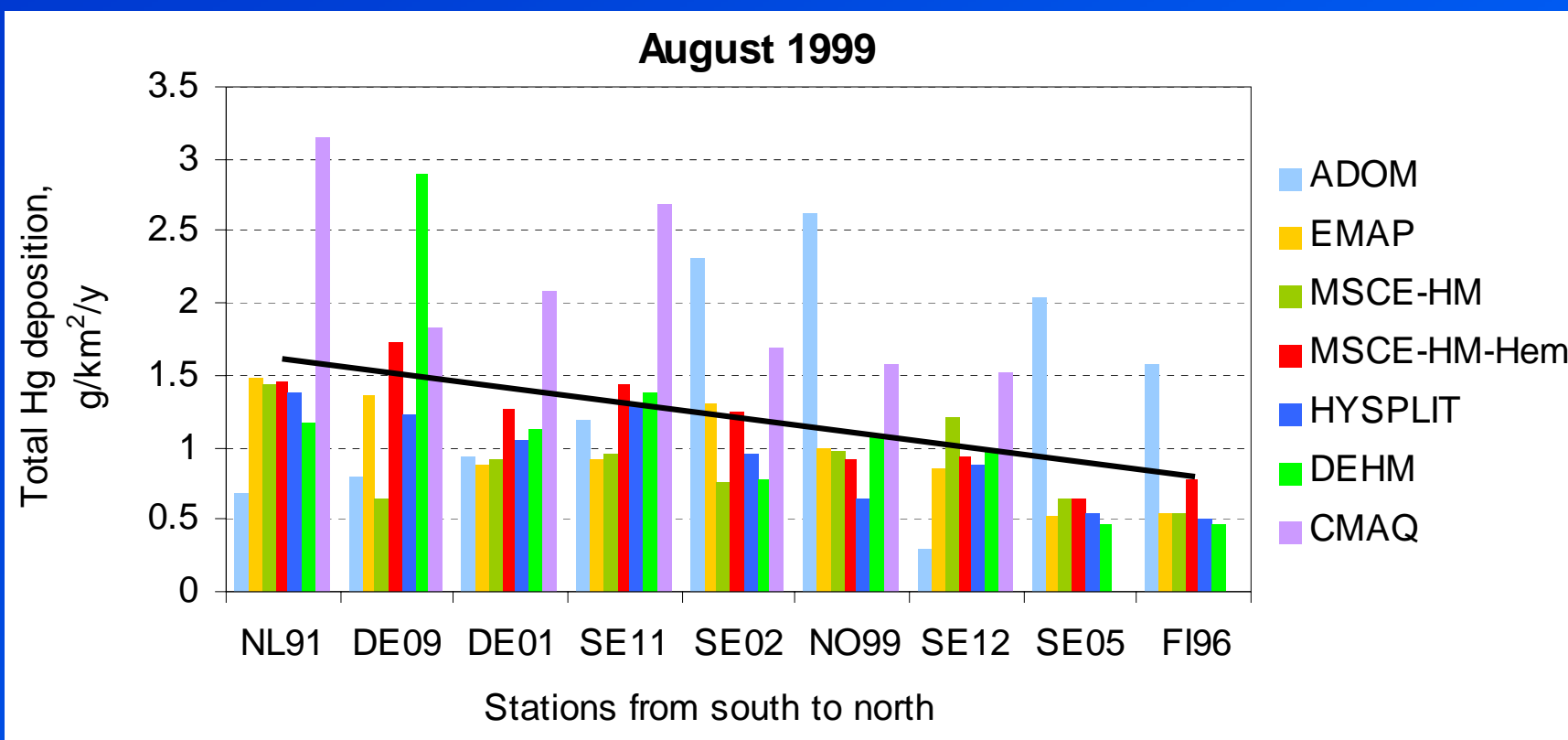
Total Modeled Hg Deposition (wet + dry)



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

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	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

Total Modeled Hg Deposition (wet + dry)

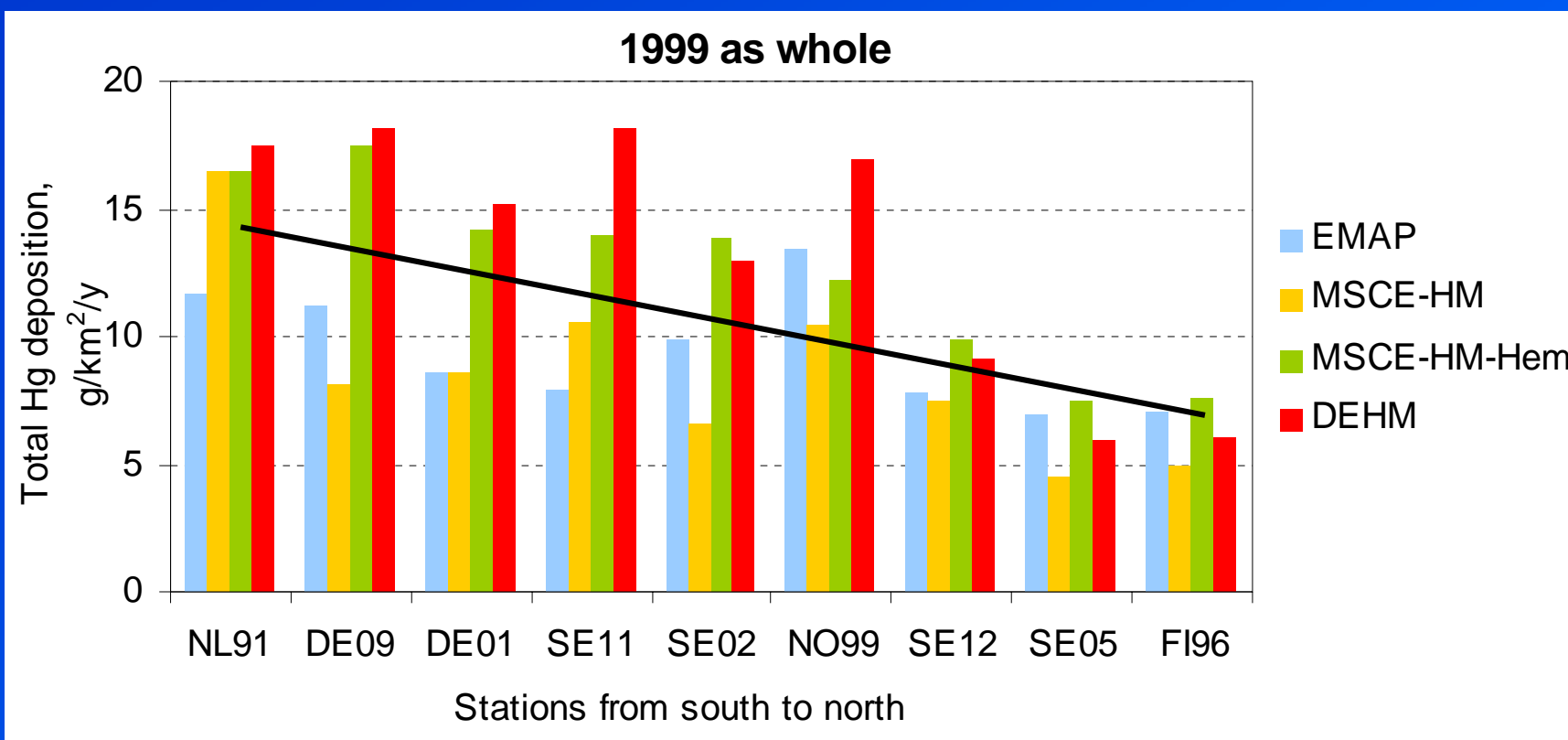


Note: ADOM was not run for August, so for this graph, ADOM results for July were used

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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
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Total Modeled Hg Deposition (wet + dry)



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	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

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

Intro- duction	Stage I	Stage II			Stage III			Conclu- sions
	Chemistry	Hg ⁰	Hg(p)	RGM	Wet Dep	Dry Dep	Budgets	

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**