

THE AUTHORS (THE MODELS):

Richard S. ARTZ (HYSPLIT)

Deputy Director
NOAA Air Resources Laboratory
1315 East West Highway, SSMC3, R/ARL,
Room 3316, Silver Spring MD 20910
USA
tel: 301 713 09 72; fax: 301 713 01 19
e-mail: richard.artz@noaa.gov

Jesper CHRISTENSEN (DEHM)

National Environmental Research Institute
Department of Atmospheric Environment
P.O.Box 358, DK-Roskilde
DENMARK
tel: 45 46 30 11 75; fax: 45 46 30 11 14
e-mail: jc@dmu.dk

Ashu DASTOOR (GRAHM)

Modelling and Integration Research Division, Air
Quality Research Branch, Meteorological Service of
Canada, Environment Canada
2121 Trans-Canada Highway, 5th Floor
Dorval, H9P 1J3 Quebec
CANADA
tel: 514 421 4766; fax: 514 421 2106
e-mail: ashu.dastoor@ec.gc.ca

Roland R. DRAXLER (HYSPLIT)

NOAA Air Research Laboratory
1315 East West Highway SSMC 3, Room 3316,
R/ARL Silver Spring, MD 20910
USA
tel: 301 713 02 95 x117; fax: 301 713 01 19
e-mail: roland.draxler@noaa.gov

Iliia ILYIN (MSCE-HM)

Meteorological Synthesizing Center- East
Ul. Arhitektor Vlasov, 51,
Moscow 117393
RUSSIA
tel: +7 095 128 96 21; fax: +7 095 125 24 09
e-mail: ilia.ilyin@msceast.org

Gerhard PETERSEN (ADOM)

GKSS - Research Centre
Institute of Hydrophysics
Max-Planck-Strasse 1, D-21502 Geesthacht
GERMANY
tel: 49 41 52 87 18 47; fax: 49 41 52 87 18 88
e-mail: petersen@gkss.de

Dimiter SYRAKOV (EMAP)

National Institute of Meteorology and Hydrology
Tzarigradsko chaussee 66, 1784 Sofia
BULGARIA
tel: 3592 975 39 86; fax: 3592 988 03 80
e-mail: Dimiter.Syrakov@meteo.bg

Russell O. BULLOCK (CMAQ)

U.S. Department of Commerce,
NOAA, Air Resources Laboratory (on assignment to
the U.S. EPA Office of Research and Development),
Mail Drop 80, Research Triangle Park NC 27711
USA
tel: 919 541 13 49; fax: 919 541 13 79
e-mail: bro@hpcc.epa.gov

Mark COHEN (HYSPLIT)

NOAA Air Research Laboratory
1315 East West Highway SSMC 3, Room 3316,
R/ARL Silver Spring, MD 20910
USA
tel: 301 713 0295 x122; fax: 301 713 0119
e-mail: mark.cohen@noaa.gov

Didier DAVIGNON (GRAHM)

Modelling and Integration Research Division, Air
Quality Research Branch, Meteorological Service of
Canada, Environment Canada
2121 Trans Canada Highway, 5th Floor
Dorval, Quebec H9P 1J3
CANADA
tel: 514 421 7242; fax: 514 421 2106
e-mail: didier.davignon@ec.gc.ca

Ralf EBINGHAUS (Measurements)

GKSS - Research Centre
Centre Institute of Coastal Research
Max-Planck-Strasse 1, D-21502 Geesthacht
GERMANY
tel: 49 41 52 87 18 47; fax: 49 41 52 87 18 88
e-mail: petersen@gkss.de

John MUNTHE (Measurements)

Swedish Environmental Research Institute
Dagjamningsgatan 1, P.O. Box 47086
S-40758 Goteborg
SWEDEN
tel: 46 31 725 62 00; fax: 46 31 725 62 90
e-mail: john.munthe@ivl.se

Alexey RYABOSHAPKO (MSCE-HM)

Meteorological Synthesizing Center- East
Ul. Arhitektor Vlasov, 51,
Moscow 117393
RUSSIA
tel: +7 095 128 96 21; fax: +7 095 125 24 09
e-mail: alexey.ryaboshapko@msceast.org

CONTENTS

Introduction	4
1 Program of the second stage and working plan	6
2 Input data	7
3 Participating models	12
4 The first results of the comparison	14
5 Current plans for further work	18
References	19

INTRODUCTION

Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on Heavy Metals (signed currently by 36 countries) determined the main task for the EMEP in this field: "EMEP shall, using appropriate models, ..., provide to the Executive Body for the Convention calculations of transboundary fluxes and depositions of heavy metals within the geographical scope of EMEP". According to the Protocol, Meteorological Synthesizing Centre "East" (MSC-E) of EMEP has a responsibility to perform model calculations of transboundary transport and deposition of heavy metals over Europe. The Steering Body of EMEP at its 18th session decided (EB.AIR/GE.1/24, 1994) that MSC-E had to organize an intercomparison study of atmospheric long-range transport models for lead, cadmium and mercury. Such a study is considered by the Steering Body to be one of the essential prerequisites for development and application of operational models for heavy metals.

The mercury model intercomparison study was launched in 1999. The mercury intercomparison study is focused on:

- an evaluation of parameterizations of the main physical-chemical processes of mercury transformations in the gaseous and the liquid phase;
- a comparison of modeling results with measurements obtained from both short-term campaigns and from the EMEP monitoring network and other international and national programs;
- a comparison of the main features of long-range transport of different mercury forms.

The program of the mercury model intercomparison study envisages four stages:

Stage I. Comparison of modules for physico-chemical transformations of mercury species in a cloud/fog environment with prescribed initial mercury concentrations in ambient air and other physical and chemical parameters relevant for atmospheric mercury transformations.

Stage II. Comparison of model results with observations during 1-2 weeks episodes. Hourly and daily averages and event based averages of mercury concentrations in air, obtained from the joint Swedish/Canadian/German field campaign TRANSECT 1995 and from the European Union Environment & Climate project Mercury Species Over Europe (MOE-1999) will be used.

Stage III. Comparison of model results with observed monthly and annual means of mercury concentrations in air and precipitation and deposition fluxes available from European monitoring stations in 1998.

Stage IV. Comparison of model predicted atmospheric budgets of mercury species in the entire EMEP domain and for selected European countries (UK, Poland and Italy), including dry and wet deposition within and outside the area of the countries.

The first stage of the intercomparison study was started in 1999 and finished in 2001. In addition to the MSC-East four scientific groups from Germany, Sweden and the USA took part in the study. The results were presented in a MSC-East technical report (MSC-E 2/2001) and will be published in "Atmospheric Environment" journal (accepted in May 2002).

The second stage started in 2001 is focused on comparison of modeled and observational results. The observations were performed at five measurement sites in Europe during two short-term campaigns in 1995 and 1999. In the first case main attention was paid to the mercury elemental form. In 1999 reactive gaseous and aerosol mercury were measured in addition to the elemental form. To discuss the program of the second stage a workshop was organized in Moscow in February 2002. The participants discussed the volume of calculations, methods of their statistical processing, and formats of reporting data. All details were agreed and accepted as a working plan.

Seven scientific groups involved in atmospheric mercury modeling participated in the second stage. They represent all the most advanced scientific and operational mercury models of regional and global types known by the moment. They are:

- GKSS-Forschungszentrum Geesthacht GmbH (Germany), the European mercury version of the Acid Deposition and Oxidants Model (ADOM).
- U.S. Environmental Protection Agency (USA), the Community Multi-Scale Air Quality (CMAQ) model.
- Environment Canada (Canada), Global/Regional Atmospheric Heavy Metals Model (GRAHM).
- National Ocean and Atmosphere Administration (USA), Hybrid Single Particle Lagrangian Integrated Trajectory model, version 4 (HYSPLIT_4)
- National Institute of Meteorology and Hydrology (Bulgaria), Eluarian Model for Air Pollution (EMAP)
- National Environmental Research Institute (Denmark), Danish Eluarian Hemispheric Model (DEHM)
- Meteorological Synthesizing Centre-East (Russia), MSCE Heavy Metal model (MSCE-HM).

The second stage of the mercury model intercomparison campaign should help to answer two very important questions:

- can the current atmospheric mercury models of regional and global levels reproduce mean values and short-term (hours) peaks of total gaseous mercury concentrations observed in Central Europe at monitoring stations?
- can the models catch mean values and variations of concentration of particulate mercury and reactive gaseous mercury compounds?

1. PROGRAM OF THE SECOND STAGE AND WORKING PLAN

The program of the second stage of the Hg model intercomparison study was preliminary agreed by a workshop held in Moscow in February 2001. The second stage is devoted to modelling the short-term episodes of mercury atmospheric transport and deposition in Northern Europe. Two experimental measuring campaigns are used as the basis of the second stage. Both of them were carried out at four monitoring stations: Neuglobsow (Germany), Zinst (Germany), Rörvik (Sweden) and Aspvreten (Sweden). In June-July 1995 the observations were done in the framework of German-Swedish TRANSECT experiment when mercury in air was simultaneously measured at 2 German and 2 Swedish monitoring stations situated along 800 km line. The measurement results used for the second episode were obtained in the framework of a project "Mercury over Europe" (November 1999). Besides, the data of continuous measurements at Mace Head station (Ireland) are also used, mainly for the hemispheric and global models.

Modelling of the 1st episode covering fortnight period of summer-95 is focussed on total gaseous mercury concentrations (TGM) as the main parameter for comparison with the observations. Modelling of the 2nd episode is focussed on particulate (TPM) and gaseous oxidized mercury (RGM) concentrations as the main parameters for comparison with the observations.

Because it was agreed to focus on five exact geographical points the size and coverage of modelling domain do not play any role. Therefore, each participation team is free to use its own domain. Some models are of regional character and adopt the EMEP modelling domain with 50*50 km resolution. The other models are global with coarser resolution.

Two main statistical parameters were agreed to be used for quantitative characterization of the comparison: correlation coefficient (measurements against calculations) and fractional bias. A separate task is to check how this or that model can follow some short-term peaks in the measurements. This characteristic of the models should be given on qualitative base (the coincidence or convergence for periods of each peak).

The workshop accepted a timetable of preparation of the stage final report and a possible publication in scientific literature. The timetable is presented in Table 1.

Table 1. *Timetable for the second stage fulfilment*

Actions	Time period
Preparation for calculations in full agreed volume	February – March, 2002
Calculations	April, 2002
Sending the results in agreed formats to MSC-East	May, 2002
Processing of the results	May – June, 2002
Preparation of a draft of a joint report	June – September, 2002
Preparation of the final version of the report	October – December, 2002
Preparation of the first version of a joint scientific article	January – April, 2003
Submission of the manuscript to a journal	April – June, 2003

2. INPUT DATA

Measurements

The measurements of concentrations of different mercury species were carried out at two German, two Swedish and one Irish monitoring stations. Locations of the stations and characteristics of surrounding areas are presented in Table 2. The data of the table show that Neuglobsow station is situated in north-eastern part of Germany and can be under influence of industrial emission sources of Central Europe. Zingst station is on the shore of the Baltic Sea where no local emission sources are situated. However, the station can also be influenced by the Central European sources. Swedish stations are more remote and reflect air pollution levels over relatively clean Scandinavia. Station “Mace Head” is located in the most western part of Europe. It can characterise air composition over the North Atlantic.

Table 2. *Locations and characteristics of the monitoring stations*

Station/country	Coordinates		Elevation, m	Characteristic of surrounding area
	Lat	Lon		
Neuglobsow, Germany	53.2N	13.0E	62	Forested area
Zingst, Germany	54.4N	12.7E	1	Sandy sea shore of the Baltic
Rörvik, Sweden	57.4N	11.9E	10	Western forested shore of Sweden
Aspvreten, Sweden	58.8N	17.4E	20	Eastern forested shore of Sweden
Mace Head, Ireland	53.3N	9.9W	20	Atlantic shore, grassland

Two measurement campaigns were organised to assess spatial distribution and temporal variations of mercury species over Germany and Sweden. Sampling and analytical methodology as well as the results were described in detail in [Schmolke *et al.*, 1999; Ebinghaus *et al.*, 2002]. The accuracy of TGM measurement was rather high. On the base of comparison of different measuring techniques at the same time and at the same place [Ebinghaus *et al.*, 1999] it is possible to estimate that possible error did not exceed $\pm 15\%$. Uncertainty of RGM and TPM measurement data was much higher – up to an order of magnitude.

The first campaign (the first episode of the model intercomparison study) was in summer 1995. Total gaseous mercury concentrations were measured simultaneously at German and Swedish stations. The second campaign (episode) was fulfilled in November 1999. Three mercury forms – total gaseous mercury (TGM), reactive gaseous mercury (RGM) and total particulate mercury (TPM) - were measured in this case at all 5 stations.

The variability of the TGM concentration is demonstrated in Figure 1 (the first episode) drawn on the base of hourly means. One can see from the figures that German sites are characterised by short-term but high peaks up to 4 ng/m^3 . Such peaks are much higher than the mean values and obviously exceed possible measurement errors. The variability of the data of Swedish stations is much lower.

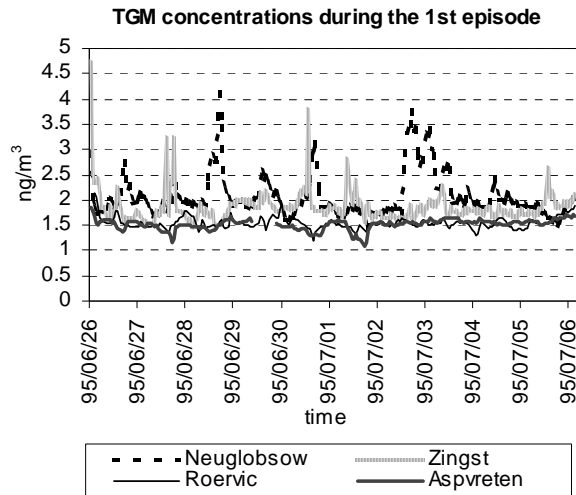


Figure 1. Observed TGM concentrations at the monitoring stations during the 1-st episode

During the second episode several samples of RGM and TPM were collected at each monitoring station.

The results of RGM measurements are presented in Figures 2 and 3 for Neuglobsow and Mace Head as an example. Surprisingly, the highest concentrations (up to 25 pg/m^3) were measured at Mace Head station, which can be considered as background one. At Neuglobsow mean concentration value is on the level of 10 pg/m^3 and lower at the other stations.

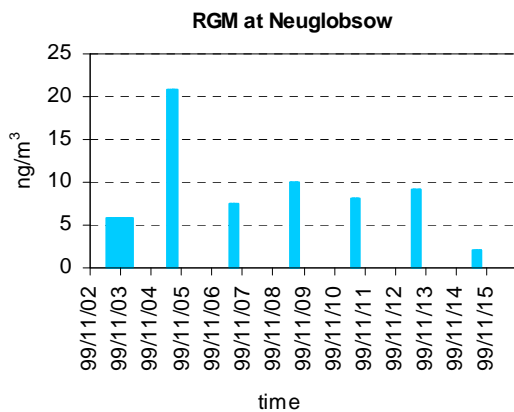


Figure 2. Measured RGM concentrations at Neuglobsow station

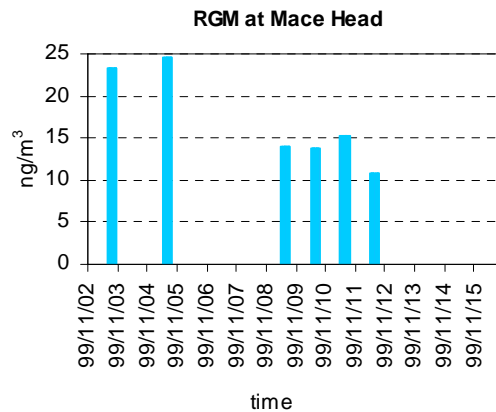


Figure 3. Measured RGM concentrations at Mace Head station

The results of TPM concentration measurements demonstrate a very strong difference between the stations. The highest values are characteristic of Neuglobsow where the TPM concentrations can exceed 100 pg/m^3 . Northward the concentration values drop. At Aspveten they are below 20 pg/m^3 during the whole period of the experiment. The

concentrations at Mace Head station are relatively low and vary between practically zero and 30 pg/m^3 . Analysing the figures one can notice a practically synchronous peak at Newglobosow, Zingst and Roervik during November 5-7 (see Figs. 4 and 5 as an example).

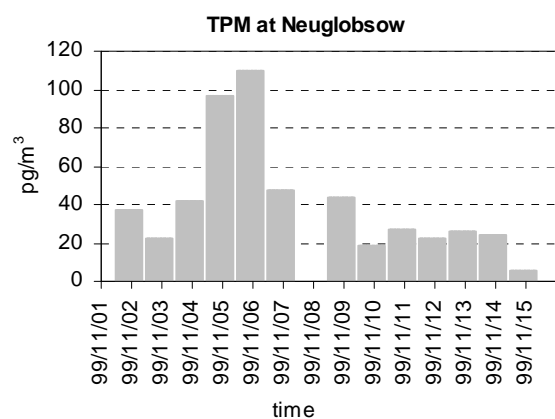


Figure 4. Measured TPM concentrations at Neuglobsow station

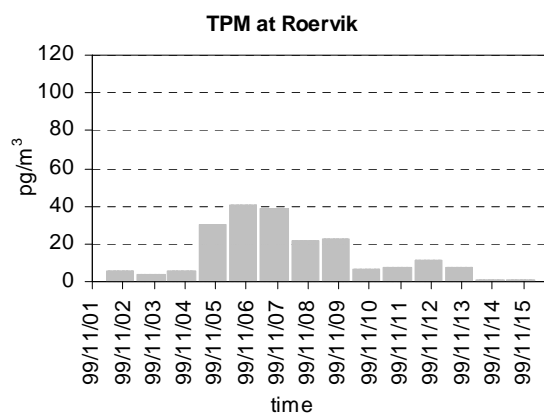


Figure 5. Measured TPM concentrations at Roervik station

Emission data

Mercury content in the atmosphere is determined by all kinds of emissions – direct anthropogenic, purely natural and secondary anthropogenic re-emission. In Central Europe the main contribution is made by the anthropogenic emissions. Hence, it was decided to use in the regional models only direct anthropogenic emissions. It is believed that such underestimation of the emission cannot effect seriously on the calculated values. There are some reasons of disdain of natural emission and re-emission for the area of the measurements. First, the mercury geochemical belt is far from the monitoring stations. Second, possible strong natural emission in Gulf Stream zone cannot influence strongly also because of remote location. The data for the re-emission are still very unreliable. At the same time remote natural emission and re-emission outside the domain are taken into account by adopted concentration values on the domain borders (boundary conditions). For the models of global type all kinds of mercury emissions should be considered.

For 1995 anthropogenic emission data suggested by *J.Pacyna and E.Pacyna* [2002] are used. As to 1999 episode, no corrections are made for the 1995 anthropogenic emission data. The data consist of mercury emissions from individual point sources within Europe and national total emission values for area sources in European countries. All the emission height data are divided into three vertical layers: 0-56 m, 56-136 m and (136 - 251 m). All the area sources have the same mercury speciation ratio: Hg^0 - 80%, RGM - 15%, and TPM -

5%. As for the point sources, each of them has its individual mercury speciation ratio. In this case Hg^0 contribution can vary from 20% (waste disposal) to 80% (cement production).

It should be mentioned that both total mercury emission value and configuration of mercury emission field changed drastically in the beginning of the 90s. Earlier the main emission sources were located in eastern part of Germany. The emission distributions for 1990 [Berdowski *et al.*, 1997] and for 1995 [Pacyna and Pacyna, 2002] over EMEP region are presented in Figure 6 (a and b). Comparison of Figures 6a and 6b shows that the emission generally dropped during 5 years and that the “hot spot” in Eastern Germany practically disappeared. This last circumstance can be very important for modelling the mercury concentrations at German monitoring stations.

It was agreed that the global modelling groups (models GRAHM and DEHM) should use their own emission data on the hemispheric/global scale. These data based mainly on estimates provided by *E.Pacyna and J.Pacyna* [2002].

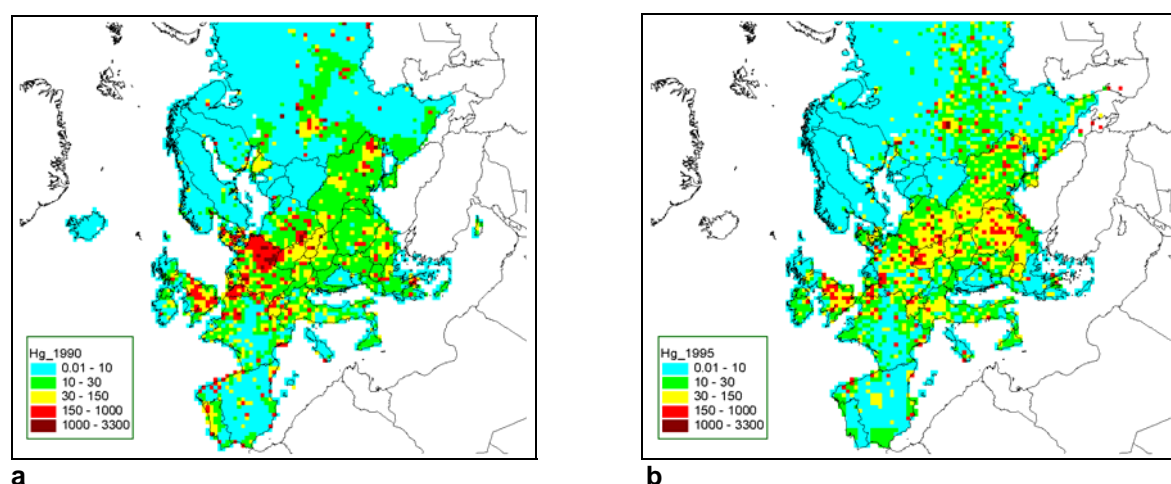


Figure 6. The mercury emission field in EMEP region for 1990 - **a** and 1995 - **b**, $g/km^2/y$

Analysis of air mass transport

Pilot analysis of air mass transport during the both episodes has been provided by MSC-E using the HYSPLIT model available through the Internet (www.arl.noaa.gov/ready.html). Such an analysis helps to distinguish periods when trajectories of air masses were connected with areas of known anthropogenic sources. In these cases one can assume elevated concentration values measured at the monitoring stations. On regional level airborne transport of polluted air masses takes place as usual within the first hundred meters of the lowest atmosphere. Hence, the trajectories were constructed for three vertical layers – 100 m, 500 m and 1000 m.

Figures 7 (a and b) present two meteorological situations. In the first case (Fig. 7.a) air masses were transported to Neuglobsow station from north-eastern Atlantic. It could be hardly suppose any elevated concentrations of any mercury form. In 24 hours the meteorological situation changed, and air masses were transported to Neuglobsow from heavy industrialized areas of Germany, Poland and Czech Republic. Indeed, the second case is characterized by obvious peak of mercury concentration (see Fig. 1).

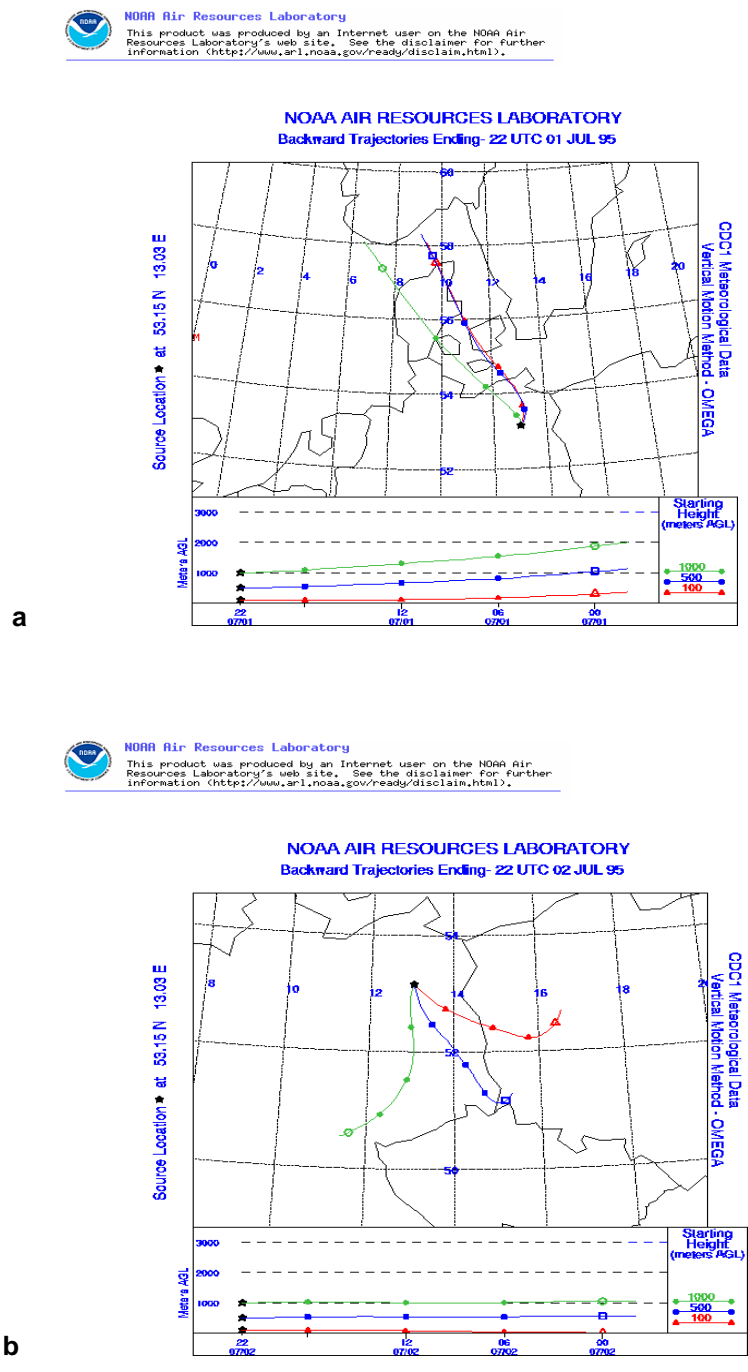


Figure 7. Air mass back trajectories for Neuglobsow station at 3 vertical levels: **a** – air masses came to the station at 22:00 July 02, 1995; **b** – air masses came to the station at 22:00 July 03, 1995

3. THE PARTICIPATING MODELS

The modelling efforts described in the subsequent paragraphs have been made to simulate the atmospheric transport and fate of mercury and to derive model predicted time series of mercury concentrations in ambient air to be compared with observations. The participating models comprise a regional Lagrangian formulation as well as Eulerian approaches on regional, hemispheric and global scales employing extensive gas- and aqueous phase chemical mechanisms and explicitly tracking numerous species concentrations. Lagrangian models are usually formulated under assumptions of simplified turbulent diffusion, no convergent flows and no wind shear. In these approaches only first-order chemical reactions can be treated rigorously. However, the Lagrangian approach avoids many of the computational complexities associated with the simultaneous solution of many differential equations; this generally results in requiring significantly less computational resources and can facilitate an understanding of problems that do not require descriptions of interactive non-linear processes. Further progress in understanding the atmospheric cycling of mercury has emphasised the need for direct modelling of the complex physico-chemical transformations of atmospheric mercury species by comprehensive Eulerian models. These approaches employ extensive gas- and aqueous phase chemical mechanisms and explicitly track numerous species concentrations. Also, a more detailed numerical formulation of physical and chemical processes occurring within and below precipitating and non-precipitating clouds is included. Typically, these models contain modules designed to calculate explicitly the chemical interactions that move gas-phase species into and among the various aqueous phases within clouds as well as calculate the aqueous-phase chemical transformations that occur within cloud- and precipitation droplets. The main properties of the participating models are presented in Table 3.

Table 3. *Main properties of the participating models*

Model	Type	Scale/domain	Model top height, m	Resolution	Boundary concentrations ^(a)			Oxidation agents	Reduction agents
					Hg(0) ₃ , ng/m ³	RGM ₃ , pg/m ³	TPM ₃ , pg/m ³		
CMAQ-Hg	Eulerian	Central–Northern Europe	about 15000	36 x 36 km	1.7	17	17	O ₃ , Cl ₂ , H ₂ O ₂ , OH	SO ₃ ^{•-} , HO ₂
ADOM	Eulerian	Regional, Central Europe	10000	55 x 55 km	1.5	2	20	O ₃	SO ₃ ^{•-}
HYSPLIT	Lagrangian	Central-Northern Europe	15000	36 x 36 km, 108 x 108 km ^(b)	(c)	(c)	(c)	O ₃ , OH, H ₂ O ₂ , Cl ₂ , HCl	SO ₃ ^{•-} , HO ₂ ,
EMAP	Eulerian	Regional/EMEP	5000, 8 layers	50 x 50 km	1.5	10	10	O ₃	SO ₃ ^{•-}
GRAHM	Eulerian	Global	about 30000	1 x 1 degree	No	No	No	O ₃	SO ₃ ^{•-}
DEHM	Eulerian	Hemispheric	15000	50 x 50 km 150 x 150 km ^(d)	1.5	0	0	O ₃	SO ₃ ^{•-}
MSCE-HM	Eulerian	Regional/EMEP	3900	50 x 50 km	1.6-1.7 ^(e)	0	20	O ₃ , OH	SO ₃ ^{•-} , HO ₂

^(a) – under normal conditions;

^(b) – for mother domain and outside the domain, correspondingly;

^(c) – cannot be specified for Lagrangian model in an explicit form;

^(d) – for EMEP domain and outside the domain, correspondingly;

^(e) – depending on a boundary.

4. THE FIRST RESULTS OF THE COMPARISON

By the moment the main part of the modelling results are under processing. However, preliminary analysis allows to see some similarities between the models, some discrepancies for all the models, some successful agreements with the observations. A comprehensive analysis of all cases will be done late in the final report for the second stage of mercury model intercomparison.

Basic statistical parameters for modelled TGM against observed TGM during the first episode are showed in Table 4. First of all, the statistical results demonstrate that all models are within $\pm 40\%$ of the observed values. There is an obvious regularity: all models underestimate the TGM concentrations at German stations and a little bit overestimate them at Swedish stations. Standard deviation values as a measure of variability show that practically in all cases the observational values vary in more broad range than the modelled ones. Some models demonstrate rather high correlation with the observations. However, no one model can follow the sharp peaks in observational series. Figure 8 presents the CMAQ modelling data (the best example) against the observational line. One can see that the modelled and observed peaks clash practically in time, however, in all cases the model underestimate the amplitude of the peaks.

Table 4. Statistical parameters for TGM concentrations during the first episode, ng/m^3

Station	Parameter	Obs	MSCE	CMAQ	EMAP	ADOM	DEHM	GRAHM	HYSPLIT
Neuglobsow	Ar.M.	2.10	1.67	1.75	1.55	1.28	1.56	2.08	
	Median	1.93	1.74	1.72	1.36	1.27	1.52	1.93	
	SD	0.45	0.30	0.10	0.55	0.08	0.12	0.17	
	Max	4.11	2.20	2.14	3.41	1.56	2.03	4.11	
	Min	1.42	0.87	1.64	0.98	1.10	1.45	1.42	
	Cor.Co.		0.07	0.60	0.55	0.07	0.55	0.57	
Zingst	Ar.M.	1.83	1.67	1.72	1.32	1.28	1.51	1.85	
	Median	1.78	1.71	1.72	1.21	1.28	1.49	1.78	
	SD	0.27	0.23	0.03	0.36	0.08	0.06	0.17	
	Max	3.79	2.12	1.80	2.25	1.58	1.71	4.73	
	Min	1.45	1.06	1.68	0.86	1.09	1.45	1.45	
	Cor.Co.		0.02	0.08	-0.12	0.04	0.06	0.08	
Roervik	Ar.M.	1.55	1.76	1.70	1.12	1.29	1.49	1.55	
	Median	1.53	1.77	1.70	1.10	1.28	1.48	1.53	
	SD	0.12	0.14	0.02	0.12	0.10	0.04	0.14	
	Max	2.14	2.21	1.80	1.64	1.58	1.65	2.14	
	Min	1.20	1.50	1.65	0.87	1.06	1.44	1.20	
	Cor.Co.		-0.08	0.26	0.17	-0.01	0.15	0.36	
Aspvreten	Ar.M.	1.52	1.65	1.69	1.19	1.26	1.49	1.52	
	Median	1.54	1.68	1.69	1.17	1.25	1.49	1.54	
	SD	0.11	0.18	0.03	0.08	0.09	0.03	0.13	
	Max	1.86	2.02	1.75	1.50	1.52	1.58	1.72	
	Min	1.10	1.12	1.62	1.08	1.09	1.44	1.10	
	Cor.Co.		0.16	-0.03	0.10	0.16	0.21	0.30	

The same statistical parameters for modelled TGM against observed TGM during the second episode are showed in Table 5. One can see that in this case the models do not underestimate the observations. A possible explanation of this fact is that real mercury emission was reduced between 1995 and 1999, while the models used the emission data of 1995 calculating concentrations for 1999. The models, which use MM5 (CMAQ, DEHM, HYSPLIT) meteorological processor, demonstrate better correlation coefficients, however, like for the first episode no one model can follow all observational peaks.

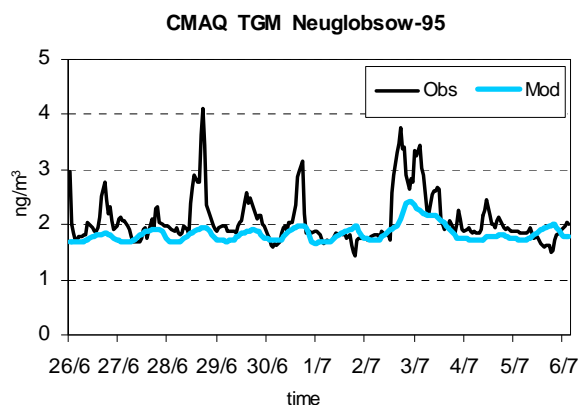


Figure 8. Comparison of observations of TGM with the results obtained by CMAQ model for the first episode

Table 5. Statistical parameters for TGM concentrations during the second episode, ng/m^3

Station	Parameter	Obs	MSCE	CMAQ	EMAP	ADOM	DEHM	GRAHM	HYSPLIT
Neuglobsow	Ar.M.	2.02	2.32	2.27	1.35		1.79	2.05	
	Median	1.89	2.32	2.25	1.22		1.79	2.01	
	SD	0.46	0.32	0.27	0.56		0.19	0.26	
	Max	4.07	3.23	2.91	3.76		2.18	2.93	
	Min	1.24	1.68	1.79	0.89		1.46	1.66	
	Cor.Co.		0.11	0.51	0.29		0.56	0.39	
Zingst	Ar.M.	1.58	2.18	2.09	1.17		1.65	1.91	
	Median	1.48	2.15	2.06	1.11		1.61	1.84	
	SD	0.35	0.23	0.27	0.36		0.16	0.24	
	Max	2.75	2.78	2.71	2.50		2.10	2.56	
	Min	1.17	1.75	1.72	0.75		1.44	1.61	
	Cor.Co.		0.01	0.67	0.38		0.69	0.53	

The modellers faced the most serious difficulties trying to simulate RGM concentrations. Sometimes the results obtained look like quite reasonable. For example, the results of GRAHM model for Roervik station demonstrate a good agreement with the observations (see Figure 9.a). In this case mean modelled value for the whole episode is only 40% lower than the observed value. At the same time this model underpredicts the observations at Mace Head station one-two orders of magnitude (see Figure 9.b). At the same time the model satisfactorily predicts TPM at this station (see Figure 9.c). This fact demonstrates that the model is capable of simulating air transport dynamics, and possible reasons of disagreement for RGM (Figure 9.b) is in treatment of RGM behaviour.

As a whole the difference on the level of one order of magnitude is characteristic of all participating models for RGM. It means that our knowledge on RGM composition and physical-chemical properties is very poor. Hence, parameterisation of the removal processes for RGM in current models cannot be satisfactorily accurate. Obvious underestimation of RGM concentrations by both global models (GRAHM and DEHM) at Mace Head station suggests that some significant natural sources of this mercury form can exist in the oceanic atmosphere.

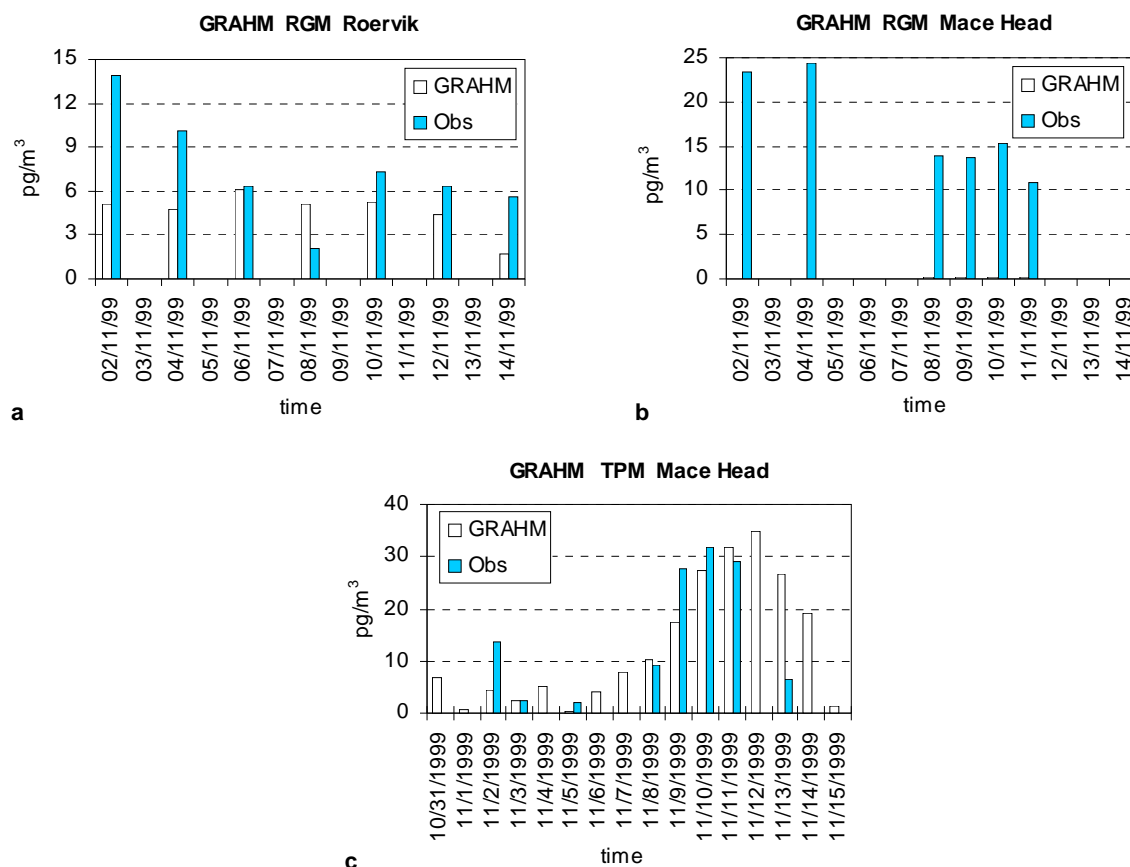


Figure 9. Observed and modelled concentrations by GRAHM model: **a** - RGM at Roervik; **b** - RGM at Mace Head; **c** - TPM at Mace Head

All models revealed a good capacity to simulate TPM concentrations and their temporal variations. It is characteristic of both regional and global/hemispheric models. Mean values for the whole second episode and correlation coefficients are showed in Table 6. One can see that all modelling mean values have the same order of magnitude as the results of the observations. Very often the difference does not exceed few tens of per-cent. As one can suppose a priori the highest values are typical for Neuglobsow station in the vicinity of main European industrial sources. In most cases the models demonstrate very high correlation between modelling and observational results.

Table 6. Mean TPM concentrations and correlation coefficients (in brackets) for the second episode

Station Mod or Obs	Neuglobsow	Zingst	Roervik	Aspvreten	Mace Head
Observations	40	33	15	10	14
CMAQ	77 (0.94)	65 (0.84)	49 (0.88)	50 (0.72)	-
EMAP	32 (0.78)	26 (0.70)	15 (0.78)	10 (0.53)	-
ADOM					
HISPLIT					
MSCE-HM	31 (0.70)	34 (0.60)	15 (-0.23)	12 (-0.14)	15 (0.82)
DEHM	33 (0.88)	26 (0.81)	17 (0.86)	15 (0.73)	18 (0.80)
GRAHM	62 (0.67)	43 (0.79)	40 (0.86)	52 (0.59)	14 (0.73)

For the most polluted station Neuglobsow all models caught an observed TPM concentration peak which took place when polluted air masses came from known source areas between the 3rd and the 5th November, 1999. This is demonstrated by Figure 10 where the results of MSCE-HM (Fig. 10.a) and DEHM (Fig. 10.b) are compared with the observations. It is important to stress that the models are very different: MSCE-HM is regional model while DEHM operates with the whole Northern Hemisphere. Nevertheless, the results of calculations are very similar.

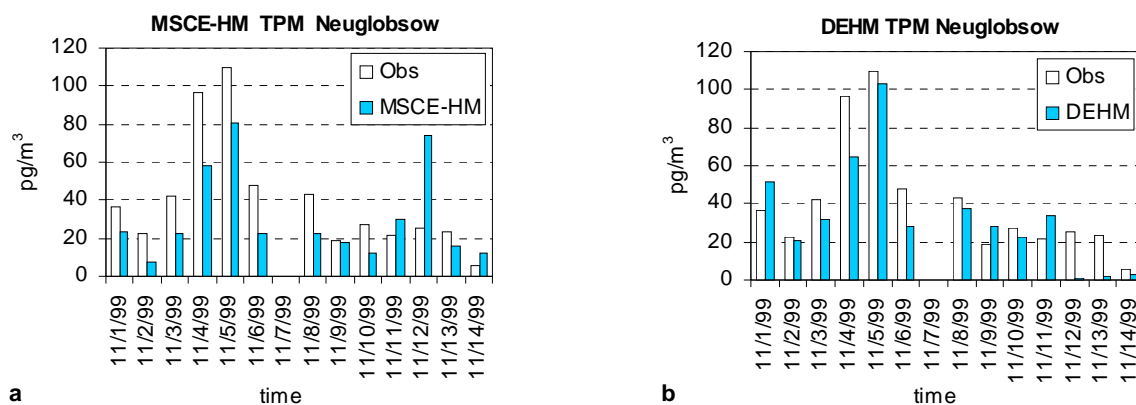


Figure 10. Comparison of observed TPM concentrations at Neuglobsow stations with calculates: **a** - MSCE-HM regional model; **b** – DEHM hemispheric model.

5. CURRENT PLANS FOR FURTHER WORK

To discuss the current results and to outline steps of further work a visit of Dr. Gerhard Petersen to MSC-East was organised from 2 to 5 July, 2002. First of all, the results of calculations of TGM were considered. Revealed incapacity of all models to follow the sharp observational peaks suggests that the emission field used in the calculations can underestimate or even ignore some strong point emission sources on the territories of Germany, Poland and Czech Republic. In this connection, a plan is accepted to use another emission field (produced in the framework of UBA project for 1990) for ADOM and MSCE-HM models. This work will be additionally done in GKSS and MSC-East.

Very contradictive results obtained by all models for RGM caused most likely by uncertainties in model parameterisation for this mercury form. Nothing is known about any natural sources of RGM. Within the next few months some consultations should be provided with chemists who can have any experience in this field and can help to understand the obtained modelling results.

In accordance with the commonly accepted working plan the calculation results of all models should be processed in the same manner by the end of September 2002. After that a draft of technical report for EMEP and other institutions will be prepared. At this phase of the work it would be desirable to convene a meeting of participating scientific groups to consider the results and to discuss plans for fulfilment of the 3rd and the 4th studies of mercury model intercomparisons. Besides, taking into account interest of scientific community to the model comparisons, it is very important to present the results of the comparison campaign in the form of a scientific article.

REFERENCES

- Berdowski J.J.M., Baas J., Bloos J.P.J., Visschedijk A.J.H. and P.Y.J. Zandveld [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Science, Energy Research and Process Innovation, UBA-FB report 104 02 672/03, Apeldoorn, 239 p.
- Ebinghaus R., Tripathi R.M., Walischläger D. and S.E. Lindberg [1999] Natural and Anthropogenic Mercury Sources and Their Impact on the Air-Surface Exchange of Mercury on Regional and Global Scales. In: R. Ebinghaus, R.R. Turner, L.D. de Lacerda, O.Vasiliev, and W.Salomons (Eds.), *Mercury Contaminated Sites. Characterization, Risk Assessment and Remediation*. Springer, pp. 3-50.
- Ebinghaus R., Kock H.H., Coggins A.M., Spain T.G., Jennings S.G. and Ch. Temme (2002: Long-term measurements of atmospheric mercury at Mace Head, Irish west coast between 1995 and 2001, submitted to *Atmospheric Environment*.
- Schmolke S., Schroeder W.H., Munthe J., Kock H.H., Schneeberger D. and R. Ebinghaus [1999] Simultaneous measurements of Total Gaseous Mercury at four sites on a 800 km transect: Spatial distribution and short time variability of Total Gaseous Mercury over Central Europe, *Atmospheric Environment*, v.33, pp.1725 - 1733.
- Pacyna E.G. and J.M. Pacyna [2002] Global emission of mercury from anthropogenic sources in 1995. *Water, Air and Soil Pollution*, v. 137, No. 1, pp.149-165.