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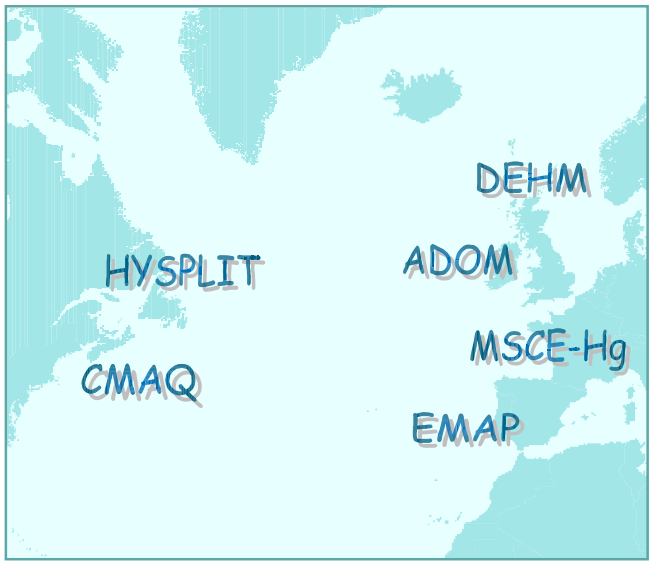
Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury

Stage III. Comparison of modelling results with long-term observations and comparison of calculated items of regional balances

Technical Report 1/2005



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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

Alexey Ryaboshapko, Richard Artz, Russell Bullock, Jesper Christensen, Mark Cohen, Roland Draxler, Ilia Ilyin, John Munthe, Jozef Pacyna, Gerhard Petersen, Dimiter Syrakov, Oleg Travnikov

Meteorological Synthesizing Centre - East

Leningradsky prospekt, 16/2, 125040 Moscow
Russia
Tel./Fax: +7 095 214 39 93
E-mail: msce@msceast.org
Internet: www.msceast.org

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	**O. Russell BULLOCK, Jr.** (**CMAQ**) U.S. Department of Commerce, NOAA, Air Resources Laboratory (on Assignment to the U.S. EPA Office of Research and Development), Mail Drop E243-03 Research Triangle Park NC 27711 USA tel: 919 5411349; fax: 919 5411379 e-mail: bro@hpcc.epa.gov
Jesper CHRISTENSEN (**DEHM**) National Environmental Research Institute Department of Atmospheric Environment P.O.Box 358 DK-Roskilde DENMARK tel: 45 46 301175; fax: 45 46 301114 e-mail: jc@dmu.dk	**Mark COHEN** (**HYSPLIT**) NOAA Air Research Laboratory 1315 East West Highway SSMC 3 Room 3316, R/ARL Silver Spring MD 20910 USA tel: 301-7130295 x122; fax: 301-7130119 e-mail: mark.cohen@noaa.gov
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	**Ilia ILYIN** (**MSCE-HM-regional**) Meteorological Synthesizing Center “East” Leningradsky prospekt, 16/2 Moscow 125040 RUSSIA tel./fax: +7 095 214 39 93 e-mail: ilia.ilyin@msceast.org
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	**Jozef PACYNA (Emission)** Norwegian Institute for Air Research NILU P.O.Box 100 2007 Kjeller NORWAY tel. 47 63 89 81 55; fax 47 63 89 80 50 jozef.pacyna@nilu.no
Gerhard PETERSEN (**ADOM**) GKSS - Research Centre, Institute of Hydrophysics Max-Planck-Strasse 1 D-21502 Geesthacht GERMANY tel: 49 41 52871847; fax: 49 41 52871888 e-mail: Petersen@gkss.de	**Alexey RYABOSHAPKO** (**MSCE**) Meteorological Synthesizing Center “East” Leningradsky prospekt, 16/2 Moscow 125040 RUSSIA tel./fax: +7 095 214 39 93 e-mail: alexey.ryaboshapko@msceast.org
Dimiter SYRAKOV (**EMAP**) National Institute of Meteorology and Hydrology Tzarigradsko chaussee 66 1784 Sofia BULGARIA tel: 3592 9753986; fax: 3592 9880380 e-mail: Dimiter.Syrakov@meteo.bg	**Oleg TRAVNIKOV** (**MSCE-HM-hemispheric**) Meteorological Synthesizing Center “East” Leningradsky prospekt, 16/2 Moscow 125040 RUSSIA tel./fax: +7 095 214 39 93 e-mail: oleg.travnikov@msceast.org

CONTENT

1. INTRODUCTION

Recently it has been widely recognized that mercury is a hazardous heavy metal, which possesses unique characteristics in the environment. It enters the atmosphere from both anthropogenic and natural sources. It migrates through the environment in different physical-chemical states. It is chemically and biochemically active in soils, water bodies and in the atmosphere. Finally, mercury in the forms of its organic compounds can be accumulated it trophic chains. In these forms it is the most dangerous for human and animals whose diet is connected with fish and other fresh water or oceanic food.

Taking into account high jeopardy of mercury and its high mobility in the environment the 1979 Convention on Long-Range Transboundary Air Pollution included this metal into its consideration. The Protocol on Heavy Metals, which came into forth on December 2004, determines mercury as on of three metals of the highest priority. A number of investigations have shown that anthropogenic activity of recent centuries led to long-term accumulation of mercury in all main geospheres – the atmosphere, the pedosphere (soils), and the Ocean. To assess global biogeochemical aspects of mercury environmental cycle and pollution UNEP launched the “Global Mercury Assessment” project.

Complicated mercury behaviour in the environment and diversity of its form make study of this metal especially difficult. In such cases numerical modelling becomes a powerful tool of investigations. One should also bear in mind that routine monitoring of mercury is very expensive, and therefore density of monitoring networks cannot be high. All these facts were recognized by a number of researchers and decision-makers. The Protocol on Heavy Metals considers the modelling approach as a basis for assessment of mercury transboundary pollution in Europe. It determined the main task for the European Monitoring and Evaluation Program (EMEP) in this field as follows: “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”. 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.

Many numerical models of different types have been developed to evaluate mercury atmospheric transport and deposition on local, regional and global levels. They are widely used as purely scientific instruments or as applied methods to solve problems of local or national levels. It is understandable that the models can differ from each other depending on their complexities and their tasks. Natural questions arise: Can the models produce reliable results? How far are the modelling results from the available observations? To what extent do different models agree with each other?

Recognizing the importance of these questions 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 heavy metals. Such a study is considered by the Steering Body to be one of the essential prerequisites for development and application of EMEP operational models. Intercomparison studies have already been completed for lead [*Sofiev et al*., 1996] and for cadmium [*Gusev et al*., 2000]. Besides, model intercomparison exercises were initiated by World Meteorological Organization for sulphur compounds on regional and global scale [*Rasch et al*., 2000; *Barrie et al.,* 2001].

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

- an evaluation of parameterisations of the main physical-chemical processes of mercury transformations in the gaseous and the liquid phase;
- a comparison of modelling 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.

Taking into account significant complexity of mercury models, necessity to consider main modelling processes separately it was decided to divide the program of the mercury model intercomparison study into three 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) have been 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 1999. 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 from sources within and outside the area of the countries.

The first stage of the intercomparison study was started in 1999 and finished in 2001. Five scientific groups from Germany, Sweden, the USA and MSC-East took part in the study. The results were presented in a MSC-East technical report [*Ryaboshapko et al.,* 2001] and published in scientific literature [*Ryaboshapko et al*., 2002a].

The second stage (started in 2001) is focused on the comparison of modelled 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 mercury elemental form. In 1999 reactive gaseous and aerosol mercury were measured in addition to the elemental form. Seven scientific groups involved in atmospheric mercury modelling participated in the second stage. They represent the most advanced scientific and operational mercury models of regional and global types. The results of the second stage were analysed and discussed in MSC-East reports [*Ryaboshapko et al.,* 2002b; 2003]. A corresponding scientific article is under preparation.

The last stage of the mercury model comparison combined performances of seven models of atmospheric mercury transport and deposition of regional and global levels. 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.
- National Ocean and Atmosphere Administration (USA), Hybrid Single Particle Lagrangian Integrated Trajectory model, version 4 for mercury (HYSPLIT)

- National Institute of Meteorology and Hydrology (Bulgaria), Eulerian Model for Air Pollution (EMAP)
- National Environmental Research Institute (Denmark), Danish Eulerian Hemispheric Model (DEHM)
- EMEP Meteorological Synthesizing Centre-East, MSCE Heavy Metal model, Regional version (MSCE-HM)
- EMEP Meteorological Synthesizing Centre-East, MSCE Heavy Metal model, Global version (MSCE-HM-Hem)

At this stage the EMEP / MSC-East was presented by two model versions: regional one – for EMEP domain, and global one – for the Northern Hemisphere.

The program of the third stage of the study met to the greatest degree the requirements of EMEP. It was focused on answering two important questions:

- To what extent can the current atmospheric mercury models of regional and global levels reproduce annual and monthly mean values of mercury concentrations and depositions observed at monitoring stations in Central Europe?
- What is accuracy of model assessment of items of mercury atmospheric balances for individual European countries?

To discuss the program of the third stage, the volume of calculations, methods of their statistical processing, and formats of reporting data two workshops was organised in Moscow in April 2003 and in April 2004 (MSC-East). All details were agreed and accepted as a working plan. Some first results of the last stage and the study as a whole were presented by the participants at the International Conference “Mercury as a Global Pollutant” (Slovenia, Ljubljana, 2004).

2. PROGRAM OF THE THIRD STAGE

The third stage of the mercury model intercomparison study aims to provide comparison of model results with monthly and annual mean measurements in 1999 and comparison of model predicted deposition budgets for selected European countries (UK, Poland, Italy). The participants of the workshop in Moscow (April 15-16, 2003) discussed and agreed the following program of the stage.

All the models, both regional and global ones consider Europe as the main modelling domain. The reference year is 1999.

The calculating parameters are:

- GEM in air (ng/m^3)
- Hg concentration in precipitation (ng/L)
- Precipitation amount (mm/month)
- Hg wet deposition (g/km^2/month)
- Hg dry deposition (g/km^2/month)
- Hg total deposition (g/km^2/month for a given geographical point or g/y for a given country).

Temporal resolution of the calculating parameters is 1 month for each selected monitoring station. Total depositions over selected countries are presented as monthly and annual values. The modellers are free to calculate the parameters for the whole year (12 months) or for some selected months. In the last case winter and summer months are chosen for the calculations.

Common subsidiary input parameters are:

- sulphur dioxide field with 50x50 km resolution (monthly mean values);
- ozone field with 50x50 km resolution (monthly mean values);
- soot field with 50x50 km resolution (monthly mean values);
- chlorine in the gas phase – fixed value of 100 ppt – within the lowest hundred meters over the ocean at night; zero above 100 m over the ocean at night; zero - during daytime; zero - over the continents;
- fixed value of pH of cloud water equal to 4.5;
- fixed value of chloride concentration in cloud water equal to 2.5 mg/l or $7x10^{-5}$ M;
- OH radical in cloud water - midday (maximum) concentration of OH radical – 10^{-12} M; at night-time the radical concentration is zero; sinusoidal trend during daytime.

Subsidiary input parameters to be used individually are:

- meteorological data sets;
- global concentration fields of SO_2, CO, NO_x, VOC_s, soot;
- boundary conditions (concentrations of modelled species at domain boundaries) for the regional models;
- initial concentration values for global (hemispheric) models.

The following monitoring stations are used for comparison of gaseous elemental mercury concentrations in air:

- Pallas, FI96, Finland
- Mace Head, IE31, Ireland
- Zeppelinfjell, NO42, Norway
- Lista, NO99, Norway
- Rörvik, SE02, Sweden
- Storhöfdi, IS91, Iceland

The following monitoring stations are used for mercury concentrations in precipitation (total mercury):

- Westerland, DE01, Germany
- Zingst, DE09, Germany
- Pallas, FI96, Finland
- Rörvik, SE02, Sweden
- Lista, NO99, Norway
- Bredkälen, SE05, Sweden
- Vavihill, SE11, Sweden
- Aspvreten, SE12, Sweden

The following sets of emission data are commonly used:

- anthropogenic Hg^0, Hg^{+2} and HgP emission fields (50x50km) within EMEP domain for 2000, three vertical layers;
- global anthropogenic Hg^0, Hg^{+2} and HgP emission fields (1x1 degree) for 1995 in accordance with (*Pacyna and Pacyna*, 2002), one vertical layer;
- natural Hg^0 emissions from the land and the sea surface within EMEP domain, spatial resolution 50x50 km (prepared by MSC-E);
- global natural emission of Hg^0 from the continents and the World ocean, spatial resolution 1x1 degree (prepared by MSC-E);
- re-emission from land within EMEP region, spatial resolution 50x50 km (prepared by MSC-E).

The emission values are given as annual mean values without any seasonal variations.

For quantitative characterization of the comparison the following parameters are used:

- arithmetic mean (normal) value (AM);
- coefficient of correlation between measurements and calculations (CC);
- relative bias (RB);
- deviation factor – “higher to smaller” ratio of observational and modelled values for a given pair;
- the highest deviation - maximum “higher to smaller” ratio of observed and modelled values within a given series of pairs of results (MD);
- factor 2 coverage (for oxidized mercury forms) or 1.2 coverage (for elemental mercury) – per cent of results within the corresponding factor for a given series of the data (F2 or F1.2);

Mercury depositions over selected countries are calculated in accordance with three emission scenarios:

- depositions caused by national anthropogenic emission of a given country (NAS);
- depositions caused by all anthropogenic European sources without sources of a given country (EAS);
- depositions caused by all possible sources including anthropogenic, natural and re-emission (APS); in this case the regional models take into account global sources by prescribing boundary conditions.

3. THE MEASUREMENT DATA

There were only a limited number of monitoring sites where mercury concentrations were measured in 1999 in the framework of EMEP monitoring program. At five sites gaseous elemental mercury was measured in air. Sampling of precipitation to measure wet deposition of mercury was performed at nine EMEP sites. The locations of the measurement sites are given in Fig. 3.1 and Table 3.1. Figure also shows that practically all the stations are located northward from the main European mercury anthropogenic sources. Roughly they are ranged in meridian direction from 52°N to 79°N.

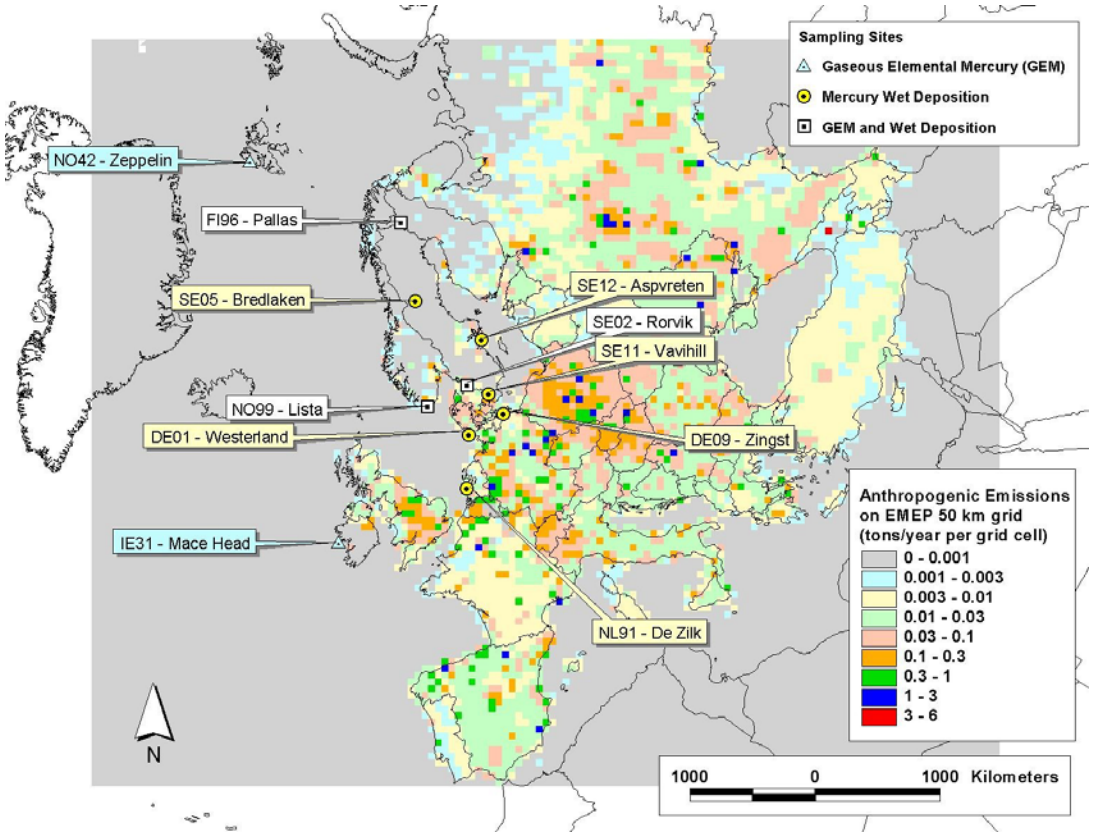


Fig. 3.1. *Locations of the mercury monitoring sites and distribution of mercury anthropogenic emissions over EMEP domain*

The monitoring sites can be divided into 3 groups. The sites of Germany (DE01, DE09) and The Netherlands can be considered as “polluted” since they are located relatively close to strong anthropogenic source areas. The second group is the “regional” sites located in southern Scandinavia (NO99, SE02, SE11, SE12) where anthropogenic effects caused by European anthropogenic sources are noticeable. The rest are “background” sites located in the Arctic and in the Atlantic.

Table 3.1. *Locations of the monitoring sites*

Station name	EMEP code	Country	Latitude	Longitude	Height, m
Gaseous elemental mercury concentrations in air					
Pallas	FI96	Finland	67° 58´ N	24° 07´ E	566
Mace Head	IE31	Ireland	53° 19´ N	10° 17´ W	5
Zeppelin	NO42	Norway	78° 54´ N	11° 53´ E	474
Lista	NO99	Norway	58° 06´ N	06° 34´ E	13
Rörvik	SE02	Sweden	57° 25´ N	11° 56´ E	10
Total mercury concentrations in precipitation (type of sampler)					
Westerland (wet only)	DE01	Germany	54° 55´ N	08° 18´ E	12
Zingst (wet only)	DE09	Germany	54° 26 N	12° 44´ E	1
Rörvik (bulk)	SE02	Sweden	57° 25´ N	11° 56´ E	10
Bredkälen (bulk)	SE05	Sweden	63° 51´ N	15° 20´ E	404
Vavihill (bulk)	SE11	Sweden	56° 01´ N	13° 09´ E	172
Aspvreten (bulk)	SE12	Sweden	58° 48´ N	17° 23´ E	20
De Zilk (wet only)	NL91	Netherlands	52° 18´ N	04° 30´ E	4
Lista (bulk)	NO99	Norway	58° 06´ N	06° 34´ E	13
Pallas (bulk)	FI96	Finland	67° 58´ N	24° 07´ E	566

Reported values of monthly mean concentrations of elemental gaseous mercury (GEM) in 1999 are presented in Table 3.2 and in Fig. 3.2 [*Ilyin et al*., 2001]. It can be noted that GEM concentrations are generally at their minimum during the warm season.

Table 3.2. *Monthly mean concentrations of gaseous elemental mercury (GEM) in 1999, ng/m^3*

Station: code, name, country	Method	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Year
FI96 Pallas Finland	manual	1.50	1.60	1.85	1.60	1.40	1.30	1.23	1.33	1.10	1.30	1.38	1.50	1.42
IE31 Mace Head, Ireland	automate	1.83	1.70	1.85	1.64	1.60	1.69	1.57	1.51	1.54	1.90	1.96	2.10	1.74
NO42 Zeppelin, Norway	automate	2.13	2.93*	1.83	2.60*	1.60	nd	nd	nd	1.75	1.20	nd	1.65	1.69**
NO99 Lista Norway	automate	1.80	1.70	2.10	1.90	1.60	nd	nd	nd	2.60*	1.50	1.80	1.70	1.76**
SE02 Rörvik, Sweden	manual	1.35	1.33	1.43	1.47	nd	1.35	1.35	1.44	1.62	1.30	1.48	1.28	1.40

nd – no data; * - outliers; ** - outliers have been removed;

Manual = Manual sampling on gold trap, analysis via dual amalgamation Cold Vapour Atomic Fluorescense Spectrometry (CVAFS). Automated = Tekran instrument based on automated gold trap sampling and CVAFS detection.

From Fig. 3.2 it is clear that the values measured at the Norwegian sites NO42 and NO99 are very variable in comparison to the data from the other stations. At the Arctic site Zeppelin the mean concentration in February was extremely high - about 3 ng/m^3. This places some doubts on the representativity and quality of the measurements. After consulting with EMEP CCC, it was decided that these values should be regarded as outliers and to be removed them from database for the comparison [*Torunn Berg*, private communication]. The GEM concentrations measured at SE02 stations seem to be relatively low, but there are no grounds to remove them from the database.

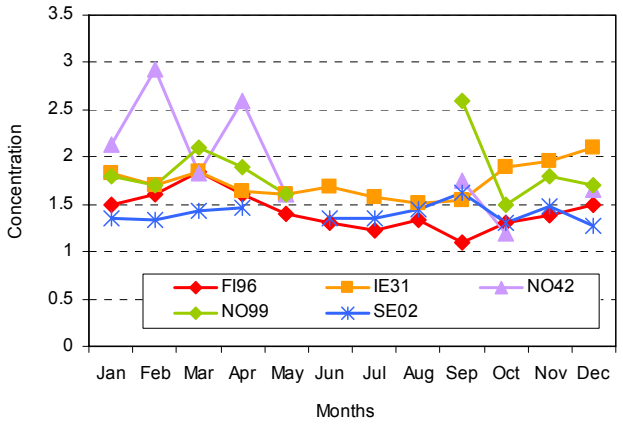


Fig. 3.2. *Measured values of monthly mean concentrations of gaseous elemental mercury (GEM) at EMEP sites in 1999, ng/m^3*

There is some uncertainty regarding terminology used for air mercury measurements and modelling. Most sampling methods for airborne gaseous mercury include a filtering step to remove particles. Thus, the sampled mercury should represent only gaseous mercury forms (typically called as “Total Gaseous Mercury” or TGM). Gaseous forms are presented mainly by elemental mercury and by a small amount of oxidised gas phase mercury species such as $HgCl_2$ (usually denoted "Reactive Gaseous Mercury" or RGM). The automated Tekran instrument can also be equipped with a speciation unit, which, in addition to a filter for particle collection, also contains a denuder for sampling of RGM. In most cases, the absolute difference between GEM and TGM is very small and for the purposes of this intercomparison, negligible. All the models calculate GEM and RGM separately. Hence, in this report the term Gaseous Elemental Mercury (GEM) is used to more accurately denote both the measurement and calculation data.

Mercury concentrations in precipitation were measured only in Northern Europe – at one site in the Netherlands, at two sites in northern Germany and at six Scandinavian sites. All measurement results as mean monthly concentrations are presented in Table 3.3 and Fig. 3.3.

Table 3.3. *Monthly mean concentrations of mercury in precipitation at EMEP monitoring sites in 1999, ng/L*

Month	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	16.7	12.0	8.4	8.8	7.3	13.0	4.5	4.8	2.4
February	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
March	6.7	8.3	11.9	9.1	11.4	20.5	11.7	4.5	4.9
April	12.2	14.7	13.9	11.8	9.0	13.2	7.2	3.8	5.8
May	9.2	19.8	23.0	17.5	18.7	9.7	9.0	6.6	4.5
June	35.8*	11.7	12.6	ND	11.1	9.9	ND	3.6	7.1
July	60.6*	13.0	16.6	8.5	12.7	11.6	36.9*	6.6	7.6
August	10.6	8.5	13.1	14.1	6.4	6.9	10.3	3.0	4.9
September	6.9	11.8	13.4	14.3	9.1	12.0	7.8	5.4	3.6
October	7.5	18.4	10.3	6.5	6.8	8.2	16.1	2.0	3.2
November	6.4	15.6	8.1	14.1	13.8	11.9	16.6	3.4	2.3
December	6.6	7.6	7.1	6.1	ND	9.3	7.6	4.3	3.3
Annual (without outliers)	9.5	12.7	12.3	10.8	10.5	11.4	9.9	4.2	4.4

ND – No Data; * - outliers

Fig. 3.3 demonstrates that two values from German site DE01 and one value from Swedish site SE12 exceed the limits of typical variations. It can be assumed that these values are non-representative and should be considered as outliers. In the SE12 case, the value associated with a very low monthly precipitation (3 mm). Sampling or analytical problems may also be a cause (*Torunn Berg*, private communication). Removing the outliers from our consideration shows (Fig. 3.4) that the bulk of the measurement data is within the range from 2 to 20 ng/L.

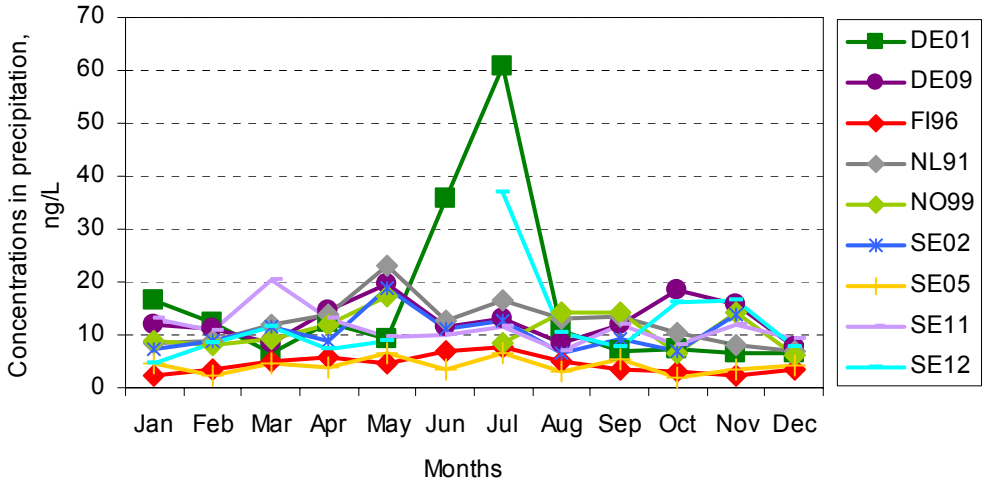


Fig. 3.3. *Monthly mean concentrations of mercury in precipitation at EMEP monitoring sites in 1999, ng/L*

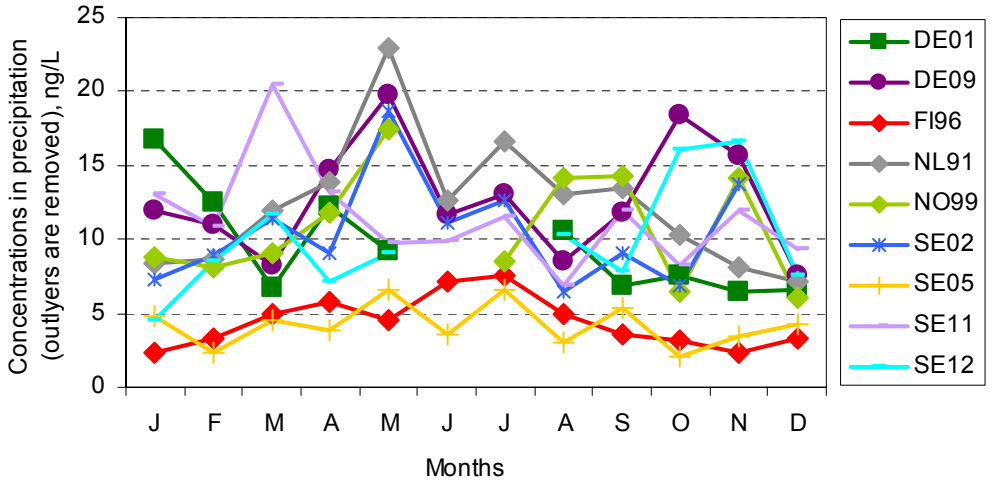


Fig. 3.4. *Monthly mean concentrations of mercury in precipitation at EMEP monitoring sites in 1999 without outliers, ng/L*

The northern location of the monitoring sites restricts considerably possibilities to reveal any spatial trends at different distances from the main European mercury sources. Nevertheless, annual values (without outliers) display higher concentrations at the polluted sites of Germany and The Netherlands

while the minimum concentrations are characteristic for the most northern “background” sites SE05 and FI96. These stations demonstrate a marked seasonal trend of Hg concentrations in precipitation with maximum in the warm season. The data from the other stations reveal no obvious seasonal trend.

Another essential parameter to estimate mercury wet deposition is precipitation amount. The monthly precipitation amounts measured at the considered stations are presented in Table 3.4. It also should be mentioned that the measured annual precipitation amount at site SE12 (340 mm) seems to be atypically low for Central Sweden and possibly underestimated.

Table 3.4. *Precipitation amounts* observed at the EMEP sites in 1999, mm*

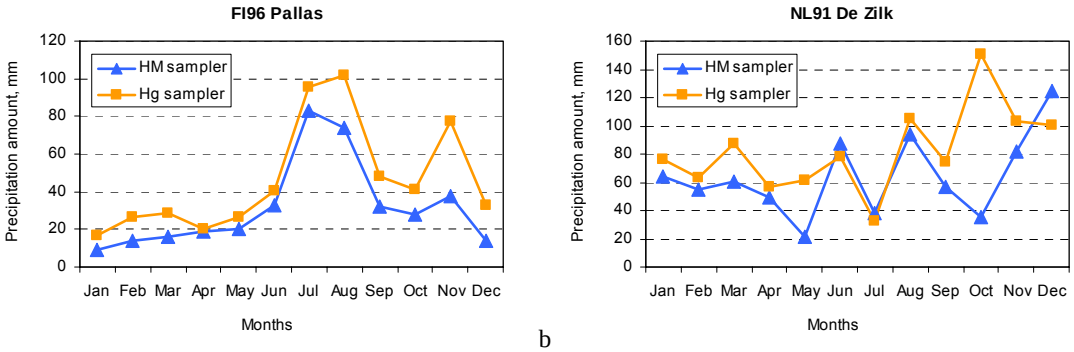
Month	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	42.3	40.4	64.5	122.7	69.8	74.4	30.8	43.7	9
February	53.8	28.5	54.5	115.1	58.3	67.3	17.0	49.1	14
March	79.6	64.0	60.3	157.8	63.5	22.8	26.0	28.3	16
April	16.6	35.7	49.2	67.0	54.3	42.6	13.9	42.2	19
May	27.3	22.1	21.0	46.9	35.5	56.4	16.3	27.3	20
June	79.3	92.1	87.8	172.9	146.7	87.5	75.0	80.8	33
July	41.3	32.0	38.3	64.2	62.7	46.0	3.0	63.5	83
August	77.3	78.3	94.2	73.0	121.4	138.9	39.2	54.9	74
September	101.7	15.5	56.3	92.7	68.2	64.2	34.5	57.2	32
October	106.6	43.6	35.2	165.0	87.3	53.6	15.4	61.1	28
November	45.4	29.1	82.2	54.8	12.0	19.2	23.2	29.2	38
December	140.6	99.3	121.2	141.1	87.6	120.5	45.2	28.4	14
Annual	812	581	765	1273	867	793	340	566	380

* Note:The amounts for DE01, DE09, NO99, SE02, SE05, SE11 and SE12 are taken from CCC/EMEP Report 3/2001; Dr. W.Aas (*CCC*, private communication) thinks that these values are closer to real precipitation amount; The precipitation amounts for NL91 and FI96 were provided by CCC/EMEP additionally.

There are two problems, which can seriously complicate the procedure of comparison of the measured values and the modelling results for mercury wet deposition. The first one is connected with the fact that the sampling of precipitation was provided by two types of the instrument – bulk sampler and wet-only sampler. In bulk samplers, dry deposition may theoretically contribute to the amount of mercury sampled by deposition in the funnel (as a rule, made of very chemically inert and smooth material) whereas wet-only samplers exclude any deposition during dry periods. The experience obtained for acidifying compounds demonstrated that for remote sites of Northern Europe there was no statistically reliable difference between wet-only samplers and bulk samples [*Söderlund*, 1982; *Brukhanov et al*., 1991; *Jorander and Pedersen*, 1992; *Steadman et al*., 1990; *Granat,* Stockholm University, private communication]. *A.Iverfeldt and J.Munthe* [1993] did not find any statistically significant difference between “bulk” and “wet-only” sampling methods for mercury. Their study also suggests that there is no difference between weekly, bi-weekly and monthly sampling protocol when bulk sampler was used and integrating the results over one month. Taking these literature data in mind we can just ignore dry deposition contribution into composition of precipitation samples.

The second problem seems to be even more dramatic. The EMEP manual for sampling and analysing (*EMEP/CCC*, 1995) prescribes strictly to only use precipitation amount data obtained by a standard meteorological precipitation gauge. In practice, standardised meteorological precipitation gauges are not always available at the sampling stations and the precipitation amounts are registered

only in samplers for e.g. mercury of heavy metals. Their designs can be very different. For example, at some sites four types of samplers are used: for acidifying compounds, for heavy metals (except mercury), for mercury and for POPs. In this work the precipitation amount data obtained by mercury samplers are used (as they have been published in CCC/EMEP reports). However, this can introduce significant uncertainties when calculating deposition fluxes. Precipitation amounts collected by different samplers are presented in Fig. 3.5a,b. At sites FI96 and NL91 the precipitation amounts were measured using samplers for mercury and for other heavy metals separately. For FI96 there is high correlation (r=0.95) but the total annual amounts differ very considerably (380 mm for HMs and 556 mm for Hg). At site NL91 the correlation is poor (r=0.30), however the total difference is smaller (769 and 991 mm, accordingly). For some individual months the difference between the samplers can exceed 2 times.



a b

Fig. 3.5. *Precipitation amounts collected by sampler for heavy metals (blue) and by sampler for mercury (orange): a) – site FI96; b) – site NL91*

For the purpose of this intercomparison the precipitation amount values obtained by samplers for mercury will be used. Naturally, uncertainty in determination of precipitation amounts inevitably leads to corresponding uncertainty in determination of mercury wet deposition fluxes. It is understandable that a mercury transport / deposition model cannot produce output data better than the quality of the input information. This fact should be kept in mind when the measurement data are compared with the modelling results.

4. THE EMISSION DATA

Modern ideas on mercury cycling assume occurrence of three pathways of mercury into the atmosphere: natural emission, current anthropogenic emission and secondary anthropogenic re-emission from previously contaminated environmental compartments. Naturally, that all emission types can contribute to concentration and deposition values over Europe. There is an important difference between the emission types. It is assumed that natural emission and re-emission are presented only by gaseous elemental mercury (GEM). Industrial high-temperature sources can emit mercury in three forms: GEM, reactive gaseous mercury (RGM) and mercury in composition of particulate matter. The latter is called as usual "total particulate mercury" or TPM.

Direct anthropogenic emission over European continent for 2000 was estimated by *J.Pacyna* and his colleagues [*Pacyna et al*, 2003]. It was decided that the difference in emissions of 1999 and 2000 should be insignificant. Hence, the 2000 emission data can be applied for the modelling result comparison with the 1999 measurement data without any corrections. The data consist of mercury emissions from individual point sources within Europe and national total emission values for area sources in European countries. It was assumed that all area sources emit mercury into the lowest atmospheric layer near the ground. All the emission height data for point sources are divided into three vertical layers: below 50 m, 50-100 m and higher than 100 m. All the area sources have the same mercury speciation ratio: GEM - 80%, RGM - 15%, and TPM - 5%. As for the point sources, each of them has its individual mercury speciation ratio. It this case Hg^0 contribution can vary from 20% (waste disposal) to 80% (cement production). The emission data are estimated on annual basis believing in absence of any seasonal cycle of the anthropogenic activity.

The direct anthropogenic emission was spatially distributed over the EMEP domain with 50x50 km resolution (the EMEP grid). The emission field is shown in Fig. 4.1. All point sources were strictly attributed to the corresponding grid cells. National area sources were distributed over the grid in proportion to population density. Generally, the European direct anthropogenic emission in 2000 made up 239 t/y including 162 t/y from individual point sources (power plants, non-ferrous and ferrous foundries, cement kilns, caustic soda plants) and 77 t/y from area sources (combustion in residential heat boilers, oil combustion, waste disposal and other sources).

The intercomparison program suggests assessment of atmospheric mercury balances for three individual countries: UK, Italy and Poland. For this task the anthropogenic emission field should be obtained individually for all these countries. To do this the emissions of area sources in all boundary cells were divided between neighbouring countries in proportion to the territory share. The individual point sources were attributed to this or that country in accordance with their co-ordinates. Total national anthropogenic mercury emissions in 2000 were 8.54 t/y in the UK, 9.78 t/y in Italy and 25.6 t/y in Poland [*Pacyna et al.*, 2003].

Assessment of global distribution of anthropogenic mercury emission was done for 1995 [*Pacyna and Pacyna,* 2002]. On the global level the total anthropogenic emission changed between 1995 and 1999 insignificantly [*Pacyna et al*., 2003]. Rather high increase of the emission in South-Eastern Asia was compensated by emission reduction in developed countries. Taking this fact into account it was decided to use for the comparison study the 1995 global emission data for 1999 without any corrections. The global emission was also shared between three mercury forms, however, the data were not divided according to emission heights. Over the globe the emission was distributed with spatial resolution of 1x1 geographical degree. The map of the anthropogenic mercury emission

distribution for the globe in 1995 is presented in Fig. 4.2. *E.Pacyna and J.Pacyna* [2002] estimated the worldwide anthropogenic emission by value of 1913 t/y in 1995.

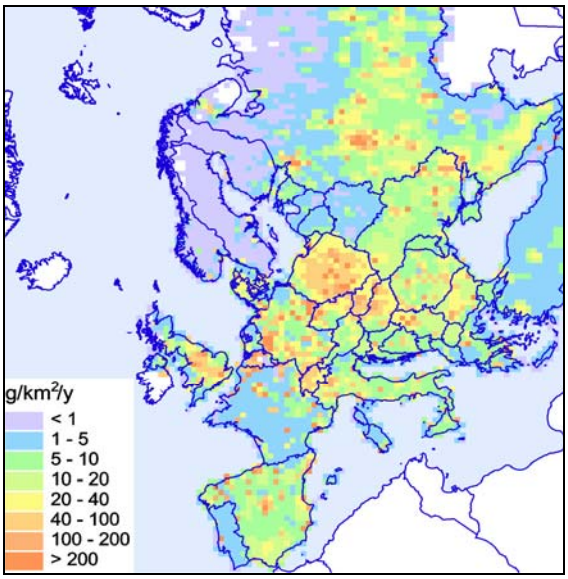


Fig. 4.1. *Spatial distribution of mercury anthropogenic emission over Europe with 50x50 km resolution in 2000, g/km²/y*

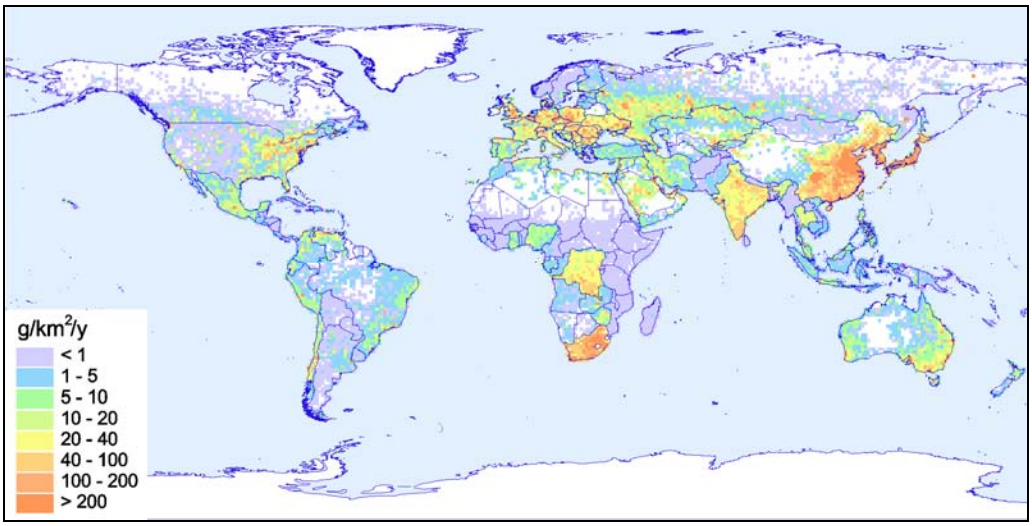


Fig. 4.2. *Spatial distribution of mercury anthropogenic emission over the globe with 1x1 degree resolution in 1995, g/km²/y*

It is well known that the oceanic surface is permanent source of elemental mercury to the atmosphere. The current global mercury emission from the oceanic surface was estimated by the value of 800 t/y [*Lamborg et al*., 2001]. Very probably, a half of this value is connected with anthropogenic re-emission from the ocean. There are some evidences that the emission intensity from water surface depends on biological activity within the surface water layer [*Kim and Fitzgerald*,

1986; *Costa and Liss*, 1999]. Taking this fact into account *O.Travnikov and A.Ryaboshapko* [2002] distributed the emission from the Ocean spatially based on spatial distribution of primary biological production of organic carbon [*Behrenfeld and Falkowski*, 1997; available through http://marine.rutgers.edu/opp].

Mercury always was presented in the surface layer of the lithosphere (soils, rocks). Accordingly natural flux of mercury to the atmosphere from continental surfaces always occurred. Here we consider natural emission as the upward flux of elemental mercury (not the net surface exchange flux of natural elemental mercury) unchangeable on geological time-scale (tens of thousand years) and caused by only natural reasons. *C.H.Lamborg et al*. [2002] estimated this mercury flux as 1000 t/y. Measurements of the intensity of this flux in different places under different conditions show that the main controlling factors are mercury content in soils/rocks and surface temperature [*Gustin et al.*, 1999]. On this basis *O.Travnikov and A.Ryaboshapko* [2002] distributed the emission from the continents. The map of spatial distribution of annual natural mercury emissions from oceanic and land surfaces over the globe is presented in Fig. 4.3. Because natural emission intensity is a function of temperature such maps were constructed separately for each month.

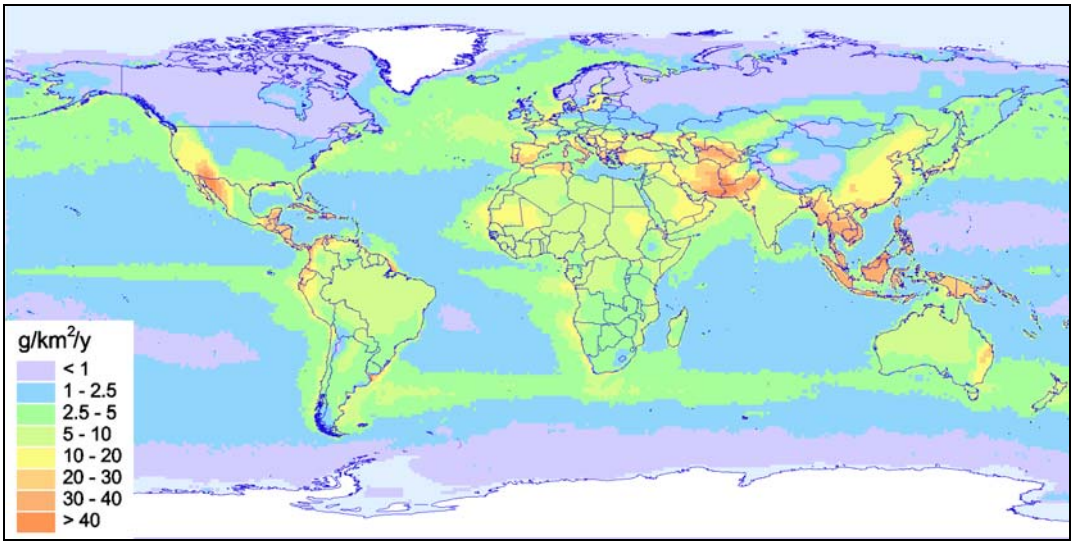


Fig. 4.3. *Spatial distribution of mercury natural emissions from oceanic and land surfaces over the globe with 1x1 degree resolution, g/km^2/y*

Natural emission in Europe is distributed unevenly. The main reason of that is occurrence of mercury geochemical anomaly in southern part of the continent where mercury content in soils can be an order of magnitude higher than in the north. Because this study is focused on comparison with European monitoring sites, the natural emission field for the EMEP domain was obtained with finer resolution – 50x50 km in the EMEP grid. Such an emission field (on annual basis) is shown in Fig. 4.4.

Intensive anthropogenic emissions of mercury over Europe and consequent depositions during at least two centuries led to mercury accumulation in European soils. In its turn elevated mercury content in soils leads to mercury re-emission to the atmosphere. *A.Ryaboshapko and I.Ilyin* [2001] estimated the current re-emission flux for Europe as 50 t/y. Assuming that re-emission intensity

should be proportional to cumulative depositions they distributed this value over Europe. The map of re-emission over Europe is shown in Fig. 4.5. One can mention that for some grid cells in Central Europe the re-emission intensity is of the same values as the current anthropogenic emission (see Fig. 4.1). It is reasonable to think that the re-emission intensity should be a function of temperature and, consequently, should have a seasonal cycle. However, in this work the intensity is accepted to be constant through the year. *C.H.Lamborg et al*. [2002] assumed that on the global level re-emission from land could account for about 30% of the current direct anthropogenic emission over the globe, however, in this work only European re-emission was taken into account.

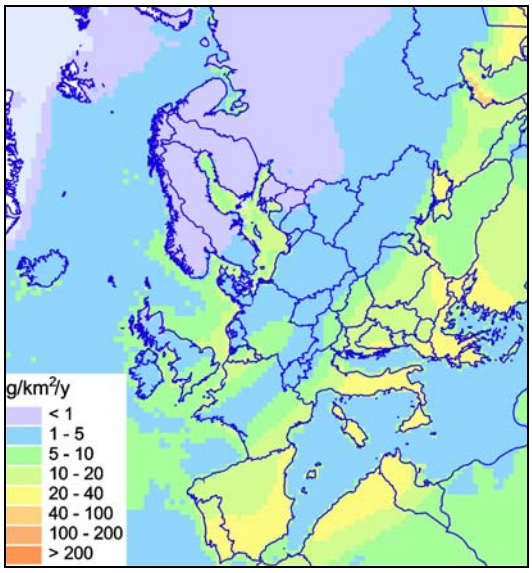


Fig. 4.4. *Spatial distribution of mercury natural emissions from oceanic and land surfaces over the EMEP domain with 50x50 km degree resolution, g/km²/y*

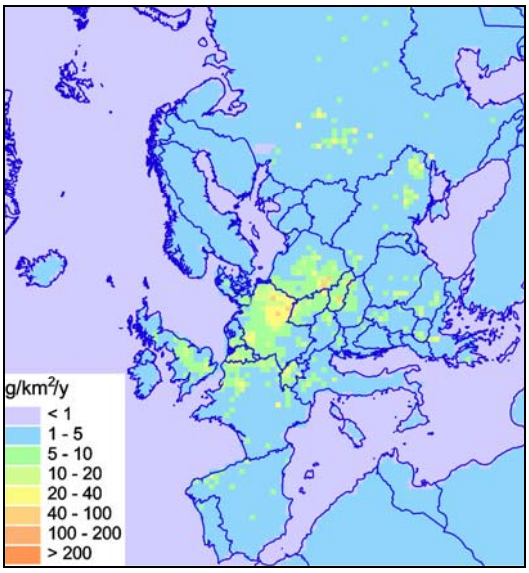


Fig. 4.5. *Spatial distribution of mercury re-emission over Europe for the end of XX century with 50x50 km degree resolution, g/km²/y*

For the calculations of mercury transport and depositions the corresponding fields of mercury emissions in different physico-chemical forms have been added. It is extremely difficult to assess degree of uncertainty of the total emission in individual grid cells. One can believe that the most accurate data are for European anthropogenic sources, which are the main contributors to the mercury emission within the area of locations of monitoring sites.

5. OTHER INPUT INFORMATION

It was agreed that all the models should use their own input meteorological data. Model ADOM is based on the meteorological data, produced by HIRLAM meteorological model, which uses ECMWF analyses. MSCE model uses data of NCEP/NCAR Re-analysis project. The same source of meteorological data is used by HYSPLIT and EMAP. CMAQ used ECMWF TOGA. Model DEHM model also based on analyses fields of ECMWF, which are processed by MM5 - Fifth Generation Pennsylvania State University / National Center for Atmospheric Research (NCAR) Mesoscale Model [*Grell et al*., 1995].

All the models use different chemical schemes of mercury transformations in the atmosphere. To avoid additional uncertainties it was decided to use the same information on concentrations of different reactants (see Chapter 2). Some input parameters should be chosen by the modellers. Subsidiary input parameters to be used individually are:

- boundary conditions (concentrations of modelled species at domain boundaries) for the regional models;
- initial concentration values for global (hemispheric) models.

6. DESCRIPTIONS OF THE PARTICIPATING MODELS

The participating models were designed to solve different problems of mercury pollution of the environment. Depending on the tasks solved the models operate with different domains. Such models like ADOM, EMAP, MSCE-HM deal with European region. CMAQ and HYSPLIT models were developed to simulate mercury transport over North American continent. The other models (DEHM and MSCE-HM-Hem) can be considered as models of global type. The participating models comprise a hemispheric Lagrangian formulation as well as Eulerian approaches; both types of models employed extensive gas- and aqueous phase chemical mechanisms and explicitly tracked numerous species concentrations. Both the Eulerian and Lagrangian formulations employ extensive gas- and aqueous phase chemical mechanisms. Both types of formulations include a detailed numerical formulation of physical and chemical processes occurring within and below precipitating and non-precipitating clouds. All participating models contain modules designed to calculate explicitly the chemical interactions that move gas-phase species into the aqueous phase 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 6.1.

Detailed descriptions of the models can be found in scientific literature, in technical reports and on the Internet. Table 6.2 presents appropriate references for the participating models.

Table 6.2. *References for detailed description of the models*

Model	**References**
CMAQ-Hg	*Bullock and Brehme* [2002]
ADOM	*Petersen et al.* [1998, 2001]
HYSPLIT	*Draxler and Hess* [1998]; *Cohen et al.* [2004]
EMAP	*Syrakov* [1995]; *BC-EMEP* [1994-1998]
DEHM	*Christensen et al.* [2004]
MSCE-HM	*Ryaboshapko et al.* [1999]; *Ilyin et al.* [2002]
MSCE-HM-Hem	*Travnikov and Ryaboshapko* [2002]

Table 6.1. *Main properties of the participating models*

Model	Type	Domain height, m	Resolution	Source of meteo data	Boundary concentrations			Oxidants		Reductants
					Hg^0, ng/m^3	RGM, pg/m^3	TPM, pg/m^3	Gas phase	Liquid phase	Liquid phase
CMAQ	Eulerian - Regional	15,000	36x36 km	ECMWF TOGA reanalysis	1.67*	18*	10*	O_3, H_2O_2, Cl_2, $OH^{\bullet}$	O_3, $OH^{\bullet}$, Cl_2	$SO_3^{=}$, hv, HO_2
ADOM	Eulerian - Regional	10,000	55x55 km	HIRLAM	1.88	0.2	20	O_3	O_3	$SO_3^{=}$
HYSPLIT	Lagrangian - Hemispheric	25,000	2.5x2.5 degree**	NCEP/NCAR reanalysis	1.2***	0	0	O_3, H_2O_2, Cl_2, HCl, $OH^{\bullet}$	O_3, $OH^{\bullet}$, Cl_2	$SO_3^{=}$
EMAP	Eulerian - Regional	5,000	50x50 km	NCEP/NCAR reanalysis	1.5	No	No	O_3, $OH^{\bullet}$	O_3	$SO_3^{=}$
DEHM	Eulerian - Hemispheric	15,000	50x50 km 150x150 km	ECMWF reanalysis	Calculated by hemispheric model			O_3	O_3	$SO_3^{=}$
MSCE-HM	Eulerian - Regional	3,900	50x50 km	NCEP/NCAR reanalysis	Calculated by hemispheric model			O_3	O_3	$SO_3^{=}$
MSCE-HM-Hem	Eulerian - Hemispheric	12,000	2.5x2.5 degree	NCEP/NCAR reanalysis	No	No	No	O_3	O_3, $OH^{\bullet}$, Cl_2	$SO_3^{=}$

* - these values for the lowest modelling atmospheric layer

** - "Resolution" has a different meaning for Lagrangian models than for Eulerian models. One measure of the resolution of a Lagrangian model is the resolution of the meteorological data used as inputs. In these simulations, the HYSPLIT model used NCEP/NCAR reanalysis data with 2.5x2.5 degree resolution.

*** - The HYSPLIT simulations were not configured to estimate the Hg0 concentration from emissions inventories alone. Reasons for this include: (a) only a 1-month "spin-up" time for each monthly simulation was used (i.e., the simulation for February 1999 was started in January 1999) and it was expected that this would lead to an underestimate of the Hg0 concentration; (b) global re-emissions were not available and were not included in the analysis – this omission was also expected to lead to an underestimate of the Hg0 concentration. The HYSPLIT results are presented both (1) without any correction for the above factors and (2) with an added background concentration of 1.2 ng/m^3, as an attempt to account for these various factors.

7. MODELS VS. OBSERVATIONS

7.1. Acid Deposition and Oxidants Model (ADOM)

ADOM was used to simulate concentrations and depositions at the site locations during two months of cold season (February and March) and two months of warm season (June and July). Accordingly, the items of mercury atmospheric balances for the selected countries were calculated for these four months.

The measured and calculated concentrations of gaseous elemental mercury (GEM) are presented in Table 7.1.1 and Fig. 7.1.1. The calculations were done only for 4 months of 1999 (two winter months and two summer months). The highest deviation of a modelled mean monthly value from a measured one is 1.35 (site FI96 Pallas in July). Relative bias for all considered sites and months is only 6%. Unfortunately, the number of the results is statistically insufficient to judge about any correlation between the observational and modelled results for the individual stations. For the totality of the results the factor of 1.2 coverage is about 64%.

Table 7.1.1. *Monthly mean observed (Obs) and modelled (Mod) concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station				
		NO99	SE02	FI96	NO42	IE31
February	Obs	1.7	1.33	1.60	ol	1.70
	Mod	**1.74**	**1.67**	**1.60**	**1.88**	**1.78**
March	Obs	2.1	1.43	1.85	1.83	1.85
	Mod	**1.65**	**1.63**	**1.57**	**1.88**	**1.70**
June	Obs	nd	1.35	1.30	nd	1.69
	Mod	**1.64**	**1.69**	**1.63**	**1.88**	**1.77**
July	Obs	nd	1.35	1.23	nd	1.57
	Mod	**1.59**	**1.57**	**1.66**	**1.88**	**1.73**

nd – no data; ol - outlier

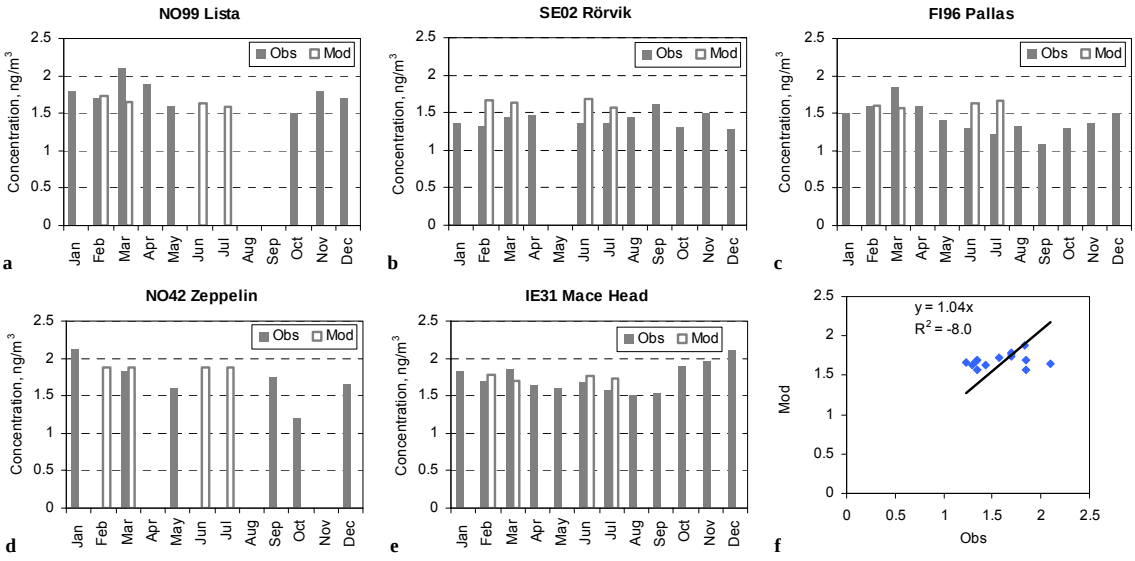


Fig. 7.1.1. *Comparison of measured and calculated by ADOM concentrations of gaseous elemental mercury (GEM) at EMEP sites*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.1.2 and Fig. 7.1.2. Variability of the data is rather high, maximum deviation of the modelled value from the observed one is 4 times (Pallas, July). However, 75% of all the results are within the factor of 2. In general, the relative bias of mean weighted concentrations (for the totality of the stations) is only 10%. The model tends to underestimate the concentration values for the stations of Central Europe and strongly overestimate the values for northern "background" stations.

It was mentioned above that the measured precipitation amounts could differ from the values used by the model as the input meteorological information. To assess uncertainty of the main modelled parameter – mercury wet deposition flux – one should have an idea about uncertainty, which is introduced by deviations between measured and forecasted precipitation amounts. Fig. 7.1.3 demonstrates that the measured and forecasted values are highly correlated (r = 0.86), however, in general the forecasted precipitation amounts are significantly higher than the values measured at the stations (a factor of 1.5). It means, that the deposition values can be correspondingly overestimated by the model. In some cases the difference exceeds 5 times.

Table 7.1.2. *Measured concentrations of Hg in precipitation at EMEP sites in 1999 (Obs) and modelled by ADOM (Mod), ng/L*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
	Mod	**3.8**	**5.5**	**7.3**	**3.9**	**7.0**	**7.3**	**4.3**	**7.0**	**13.6**
March	Obs	6.7	8.3	11.9	9.1	11.4	20.5	11.7	4.5	4.9
	Mod	**9.6**	**7.4**	**11.2**	**4.3**	**8.8**	**8.2**	**6.8**	**11.4**	**12.7**
June	Obs	ol	11.7	12.6	nd	11.1	9.9	nd	3.6	7.1
	Mod	**6.5**	**8.9**	**8.9**	**7.1**	**10.2**	**10.5**	**7.7**	**7.0**	**13.4**
July	Obs	ol	13.0	16.6	8.5	12.7	11.6	ol	6.6	7.6
	Mod	**7.5**	**12.0**	**12.2**	**11.6**	**14.0**	**10.6**	**16.9**	**12.2**	**8.6**

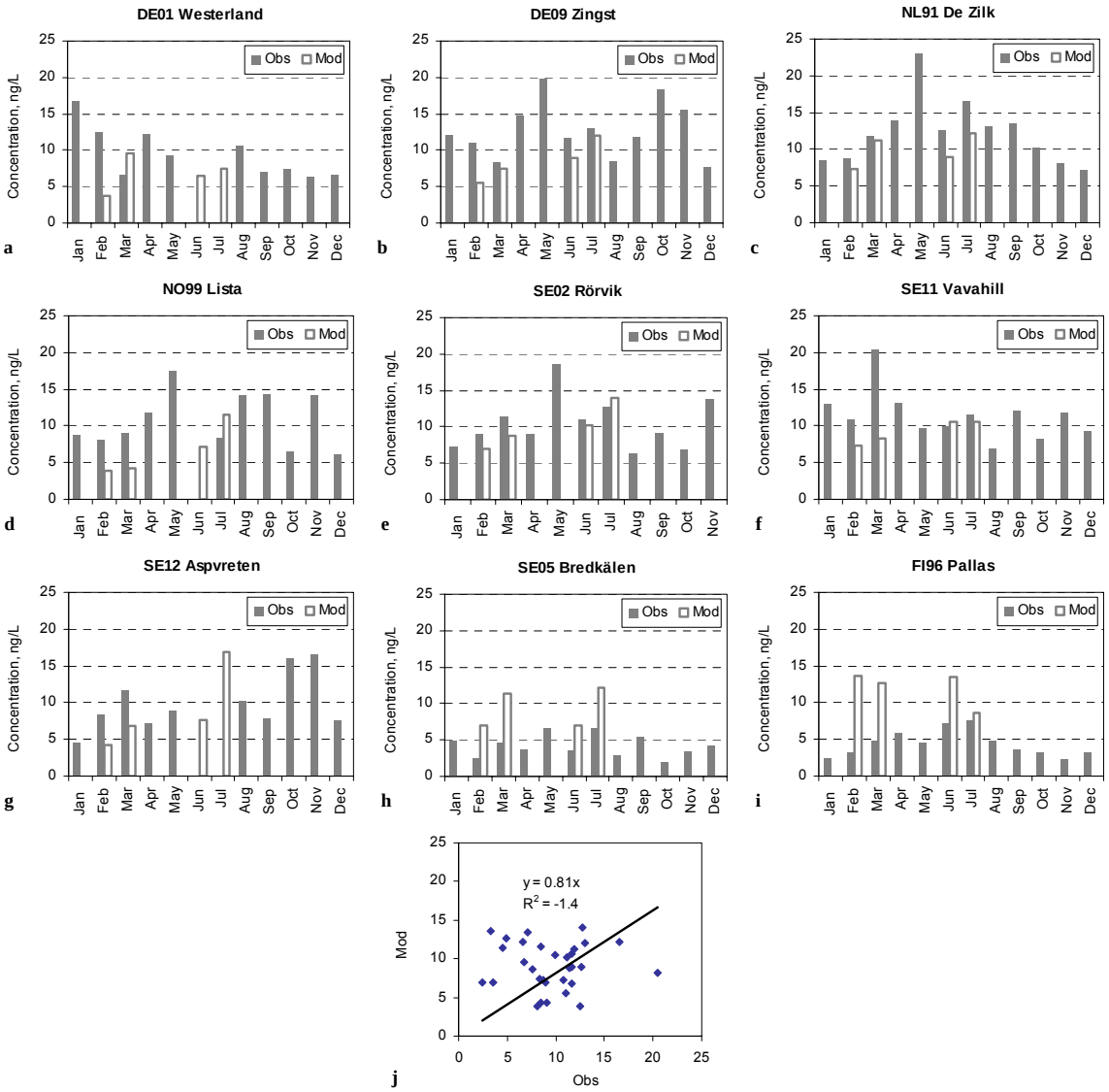


Fig. 7.1.2. *Comparison of measured and calculated by ADOM concentrations of mercury in precipitation at EMEP sites*

Table 7.1.3 and Fig. 7.1.4 presents the mercury wet deposition fluxes at the stations obtained on the basis of the observations and by the model. In many cases the model overestimates the depositions (the relative bias for the totality of the results is 40%). Main reason of such overestimation can be connected, first of all, with usage by the model of overestimated precipitation amounts (calculated by meteorological model HIRLAM). Some contribution to the overestimation can be made by high concentration values modelled for the northern stations SE05 and FI96. The maximum disagreement between the observed and modelled values can reach an order of magnitude.

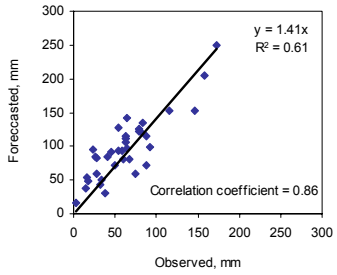


Fig. 7.1.3*. Monthly mean forecasted precipitation amounts used by ADOM against observed precipitation amounts measured at the monitoring stations, mm*

Table 7.1.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by ADOM (Mod), g/km^2*

Month	Obs / Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14	0.12	0.05
	Mod	**0.49**	**0.46**	**0.68**	**0.60**	**0.66**	**0.59**	**0.21**	**0.51**	**0.52**
March	Obs	0.53	0.53	0.72	1.44	0.72	0.47	0.30	0.13	0.08
	Mod	**1.17**	**0.72**	**0.91**	**0.88**	**1.01**	**0.78**	**0.57**	**0.68**	**0.69**
June	Obs	-	1.08	1.11	-	1.63	0.87	-	0.29	0.23
	Mod	**0.81**	**0.88**	**0.64**	**1.77**	**1.55**	**1.21**	**0.45**	**0.86**	**0.67**
July	Obs	-	0.42	0.64	0.55	0.80	0.53	-	0.42	0.63
	Mod	**0.63**	**0.52**	**0.36**	**1.64**	**1.56**	**0.96**	**0.26**	**1.28**	**1.16**

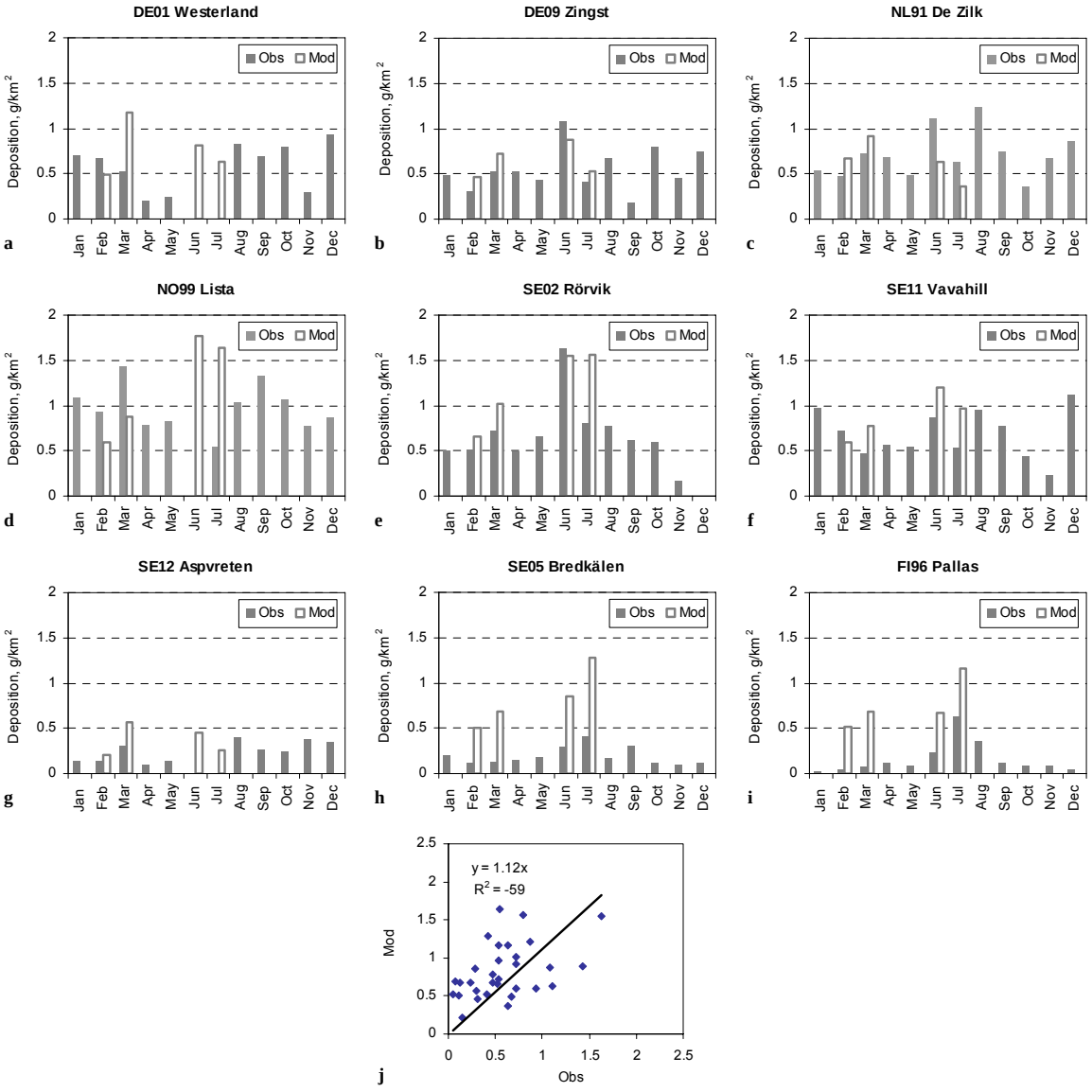


Fig. 7.1.4. *Comparison of mercury wet deposition values obtained on the basis of observations and calculated by ADOM for locations of EMEP stations*

One can see from Table 7.1.3 and Fig. 7.1.4, that the highest deviation between the observed and modelled results for wet deposition exceed a factor of 10. It means that all results obtained by ADOM lie within this factor. However, this value does not reflect degree of real reliability of the modelling data. A very high deviation value can be a result of combination of random parameters. Much more informative is conception of cumulative distribution of the deviation factors for the totality of the results. Such a cumulative curve is shown in Fig. 7.1.5 (where abscissa is logarithmic). The curve demonstrates that 72% of all results are within a factor of 2; 77% - within a factor of 3 and 93% - within a factor of 5. It means that probability to obtain an acceptable result (within a factor of 2) is high enough and probability of total failure is very low.

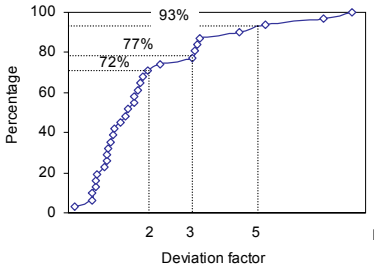


Fig. 7.1.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by ADOM (9 stations, 4 months)*

7.2. Community Multi-Scale Air Quality (CMAQ)

Calculations of concentrations and depositions at the site locations performed by CMAQ are restricted by two months: February (cold season) and August (warm season). Accordingly, the items of mercury atmospheric balances for the selected countries were calculated for these two months. Besides, the CMAQ domain does not cover the most northern sites: FI96 Pallas, NO42 Zeppelin and SE05 Bredkälen.

The observed and calculated values of GEM concentrations are presented in Table 7.2.1 and Fig. 7.2.1. The highest deviation of a modelled mean monthly value from a measured one is 1.2 (station Rörvik in February). Relative bias for all considered stations and months is only 2%. Unfortunately, the number of the results is statistically insufficient to judge about any correlation between the observational and modelled results for the individual stations.

Table 7.2.1. *Monthly mean observed (Obs) and modelled (Mod) concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station		
		NO99	SE02	IE31
February	Obs	1.7	1.33	1.70
	Mod	**1.53**	**1.61**	**1.53**
August	Obs	-	1.44	1.51
	Mod	**1.55**	**1.56**	**1.56**

nd – no data; ol - outlier

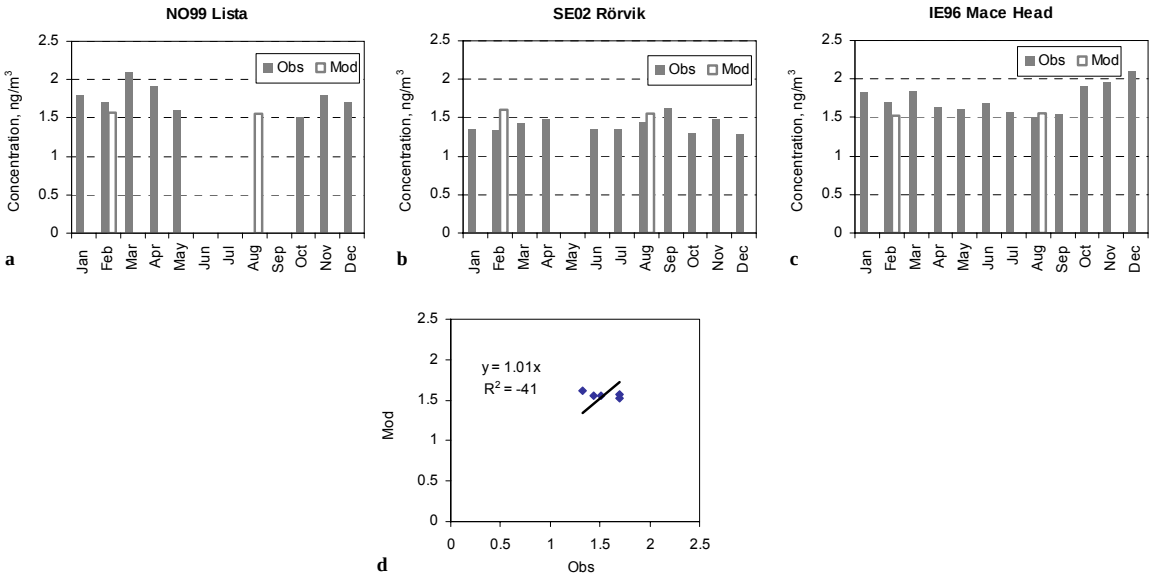


Fig. 7.2.1. *Comparison of measured and calculated by CMAQ concentrations of gaseous elemental mercury (GEM) at EMEP sites*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.2.2 and Fig. 7.2.2. Maximum deviation of the modelled value from the observed one reaches 4 times. However, 70% of all the results are within the factor of 2. The modelled mean weighted concentration (for the totality of the stations and of the months) is much higher than the measured mean weighted one (the relative bias is more than 70%).

Table 7.2.2. *Measured concentrations of Hg in precipitation at EMEP sites in 1999 (Obs) and modelled by CMAQ (Mod), ng/L*

Month	Obs/ Mod	Stations						
		DE01	DE09	NL91	NO99	SE02	SE11	SE12
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5
	Mod	**13.0**	**17.0**	**13.5**	**12.5**	**12.6**	**18.3**	**7.2**
August	Obs	10.6	8.5	13.1	14.1	6.4	6.9	10.3
	Mod	**19.2**	**17.5**	**21.9**	**21.0**	**14.9**	**25.1**	**24.5**

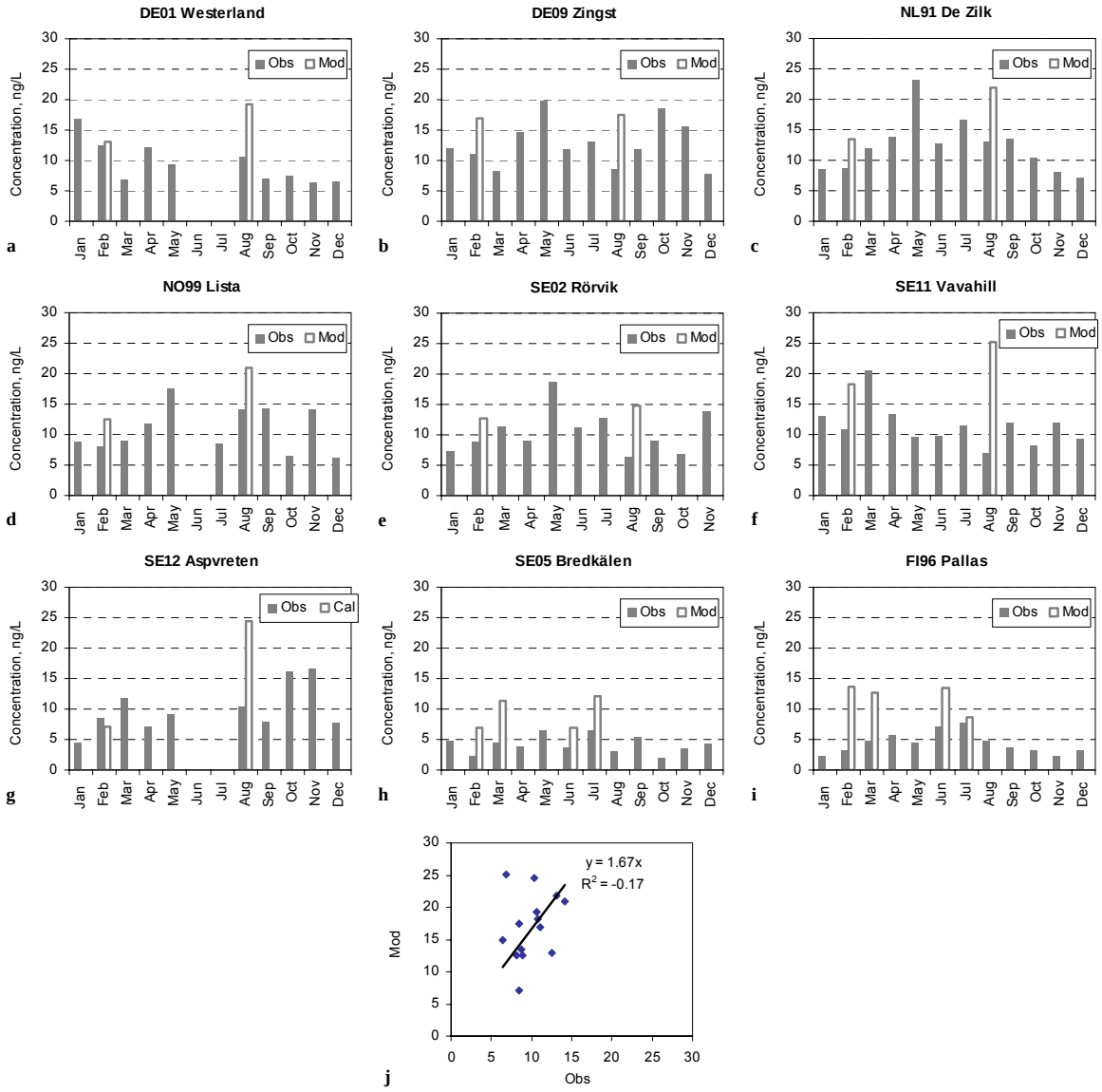


Fig. 7.2.2. *Comparison of measured and calculated by CMAQ concentrations of mercury in precipitation at EMEP sites*

CMAQ used meteorological data including precipitation fields obtained on the basis of mesoscale meteorological model MM5. Fig. 7.2.3 demonstrates correspondence of forecasted precipitation amounts by MM5 and observed ones at the stations. The forecasted and observed values are highly correlated (r = 0.76). However, the forecasted values are slightly underestimated (by the factor of 0.89).

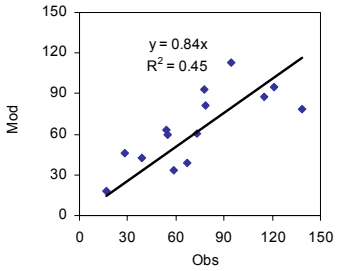


Fig. 7.2.3. *Monthly mean forecasted precipitation amounts used by CMAQ against observed precipitation amounts measured at the monitoring stations, mm*

Wet deposition fluxes calculated by CMAQ in comparison with the data obtained on the basis of the observations are presented in Table 7.2.3 and Fig. 7.2.4. Generally, the model overestimates the observed wet deposition values. It should be noted that such overestimation is much higher for

August (relative bias is 93%) than for February relative bias is 26%). For both months the correlation is very significant (CC = 0.78 and 0.76 correspondingly).

Table 7.2.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by CMAQ (Mod), g/km^2*

Month	Obs / Mod	Stations						
		DE01	DE09	NL91	NO99	SE02	SE11	SE12
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14
	Mod	**0.82**	**0.79**	**0.80**	**1.10**	**0.42**	**0.71**	**0.13**
August	Obs	0.82	0.67	1.23	1.03	0.78	0.96	0.40
	Mod	**1.79**	**1.42**	**2.47**	**1.26**	**1.41**	**1.97**	**1.03**

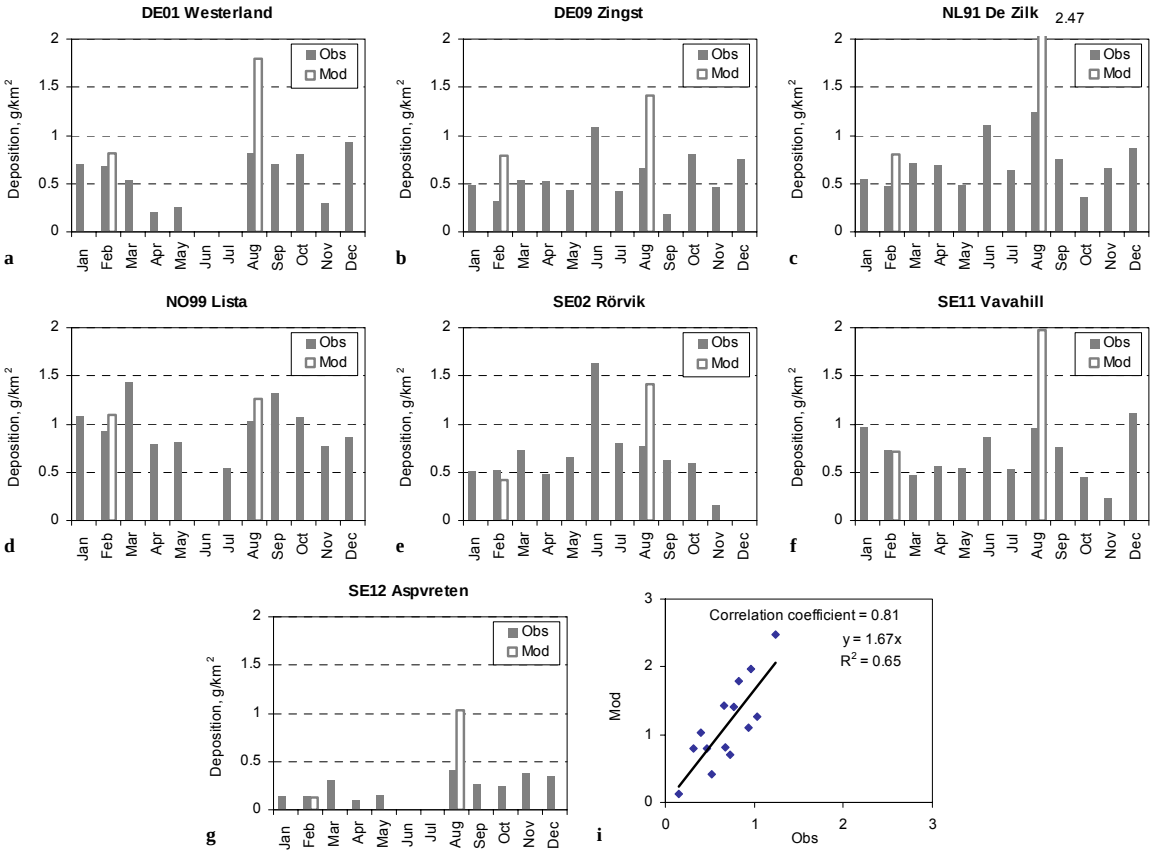


Fig. 7.2.4. *Comparison of mercury wet deposition values obtained on the basis of observations and calculated by CMAQ for locations of EMEP stations*

Number of the wet deposition results obtained by CMAQ is not statistically high (only 14 pairs). The highest deviation reaches a factor of 2.6. Cumulative distribution curve is shown in Fig. 7.2.5. One can see that 2/3 of all the results are within the factor of 2.

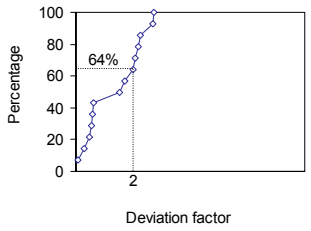


Fig. 7.2.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by CMAQ (7 stations, 2 months)*

7.3. Eulerian Model for Air Pollution (EMAP)

EMAP was used to simulate concentrations and depositions at the site locations during all months of 1999. Accordingly, the items of mercury atmospheric balances for the selected countries were calculated for all months and for 1999 as a whole. The measured and calculated concentrations of gaseous elemental mercury (GEM) are presented in Table 7.3.1 and Fig. 7.3.1. Both observations and modelled results do not reveal any obvious seasonal trends during 1999. The highest deviation of a modelled mean monthly value from a measured one reaches 2 times (station Rörvik in October). However, 86% of the results are within a factor of 1.5. In general, the model overestimates the measurements: relative bias for all considered sites and months is 18%. Especially high overestimation can be noted for station SE02 Rörvik (1.5 times on the annual basis). For 3 stations of 5 there is noticeable positive correlation between the observational and modelled results. Theoretically, the dots in Fig. 7.3.1 (f) should follow the straight line, which comes through the origin.

Table 7.3.1. *Monthly mean observed (Obs) and modelled (Mod) by EMAP concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station				
		NO99	SE02	FI96	NO42	IE31
January	Obs	1.8	1.35	1.5	2.13	1.83
	Mod	**2.00**	**2.05**	**1.65**	**1.73**	**1.77**
February	Obs	1.7	1.33	1.60	Ol	1.70
	Mod	**2.07**	**1.96**	**2.02**	**1.93**	**1.21**
March	Obs	2.1	1.43	1.85	1.83	1.85
	Mod	**2.22**	**2.57**	**2.07**	**1.71**	**1.84**
April	Obs	1.9	1.47	1.60	ol	1.64
	Mod	**2.00**	**1.93**	**1.87**	**2.25**	**1.88**
May	Obs	1.6	Nd	1.40	1.60	1.60
	Mod	**1.28**	**1.35**	**1.53**	**1.14**	**1.82**
June	Obs	Nd	1.35	1.30	nd	1.69
	Mod	**2.37**	**1.74**	**1.85**	**1.91**	**1.73**
July	Obs	Nd	1.35	1.23	nd	1.57
	Mod	**1.35**	**1.70**	**1.90**	**1.83**	**1.41**
August	Obs	Nd	1.44	1.33	nd	1.51
	Mod	**1.92**	**1.95**	**1.49**	**1.76**	**2.21**
September	Obs	Ol	1.62	1.10	1.75	1.54
	Mod	**2.21**	**1.94**	**1.16**	**2.28**	**2.43**
October	Obs	1.5	1.30	1.30	1.20	1.90
	Mod	**2.21**	**2.67**	**2.07**	**1.56**	**2.39**
November	Obs	1.8	1.48	1.38	nd	1.96
	Mod	**1.81**	**1.76**	**1.90**	**2.54**	**1.31**
December	Obs	1.7	1.28	1.50	1.65	2.10
	Mod	**2.34**	**2.31**	**1.57**	**1.88**	**1.72**
Year	Obs	**1.76***	**1.40***	**1.42**	**1.69***	**1.74**
	Mod	**1.98**	**1.99**	**1.76**	**1.88**	**1.82**
Correlation Coefficient		**0.24**	**-0.30**	**0.53**	**0.29**	**-0.24**

nd – no data; ol – outlier; * - corrected for 12 months

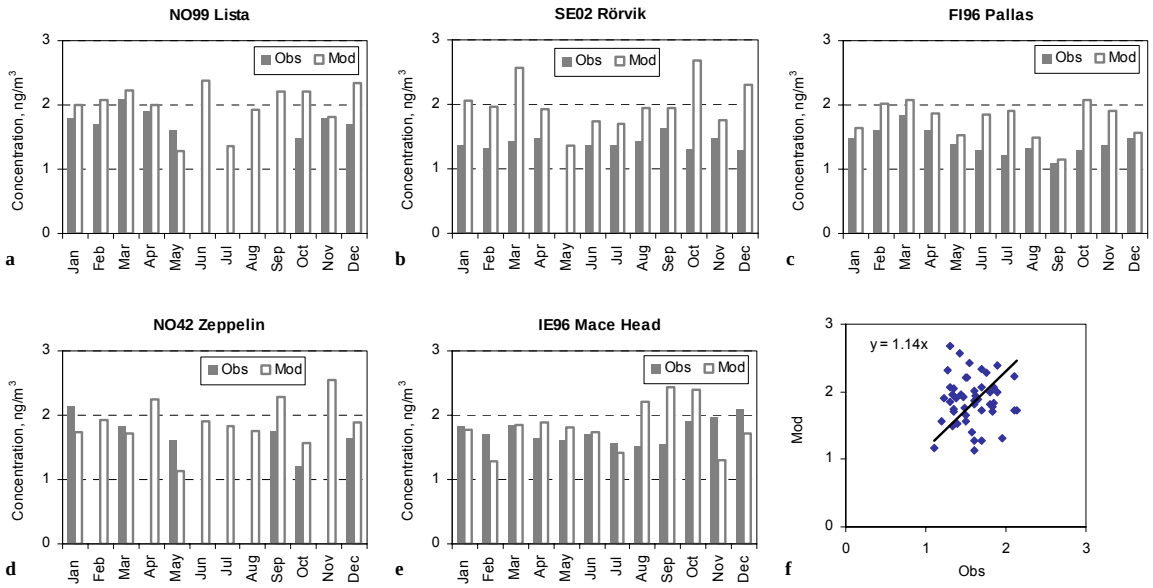


Fig. 7.3.1. *Comparison of measured and calculated by EMAP concentrations of gaseous elemental mercury (GEM) at EMEP sites*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.3.2 and Fig. 7.3.2. Variability of the data is high, maximum deviation of the modelled value from the observed one is 6.5 times. The modelled and the observed values are not practically correlated. In spite of high scattering, 80% of all the results are within the factor of 2. In general, the modelled mean weighted concentration (for the totality of the stations) closely agrees with the measured mean weighted one: modelled value is only 11% higher. The model tends to overestimate strongly (more than 2 times) the values for northern “background” stations. For these stations there is regularity: the observations demonstrate higher values during warm season, while the model predicts higher values for winter. This fact leads to obvious anti-correlation between the results for these two stations. One can see that the model overestimates strongly just low concentration values.

Table 7.3.2. *Measured concentrations of Hg in precipitation at EMEP stations in 1999 (Obs) and modelled by EMAP (Mod), ng/L*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	16.7	12.0	8.4	8.8	7.3	13.0	4.5	4.8	2.4
	Mod	**9.9**	**12.1**	**9.4**	**9.6**	**10.6**	**10.4**	**11.2**	**13.3**	**9.9**
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
	Mod	**11.4**	**11.9**	**10.3**	**11.1**	**10.7**	**11.2**	**11.3**	**13.1**	**17.4**
March	Obs	6.7	8.3	11.9	9.1	11.4	20.5	11.7	4.5	4.9
	Mod	**12.5**	**13.9**	**13.1**	**12.7**	**19.4**	**15.5**	**11.5**	**11.3**	**14.8**
April	Obs	12.2	14.7	13.9	11.8	9.0	13.2	7.2	3.8	5.8
	Mod	**10.0**	**9.4**	**9.6**	**18.2**	**10.6**	**10.4**	**15.3**	**11.7**	**9.8**
May	Obs	9.2	19.8	23.0	17.5	18.7	9.7	9.0	6.6	4.5
	Mod	**9.5**	**10.5**	**13.0**	**10.1**	**10.4**	**10.9**	**8.3**	**10.2**	**10.5**
June	Obs	Ol	11.7	12.6	Nd	11.1	9.9	Nd	3.6	7.1
	Mod	**8.2**	**8.1**	**8.7**	**11.4**	**7.4**	**8.2**	**6.4**	**8.7**	**7.5**
July	Obs	Ol	13.0	16.6	8.5	12.7	11.6	Ol	6.6	7.6
	Mod	**8.1**	**7.0**	**8.2**	**8.4**	**8.2**	**6.6**	**7.9**	**9.7**	**8.7**
August	Obs	10.6	8.5	13.1	14.1	6.4	6.9	10.3	3.0	4.9
	Mod	**9.3**	**8.2**	**10.2**	**8.7**	**10.5**	**9.5**	**7.1**	**8.9**	**7.8**
September	Obs	6.9	11.8	13.4	14.3	9.1	12.0	7.8	5.4	3.6
	Mod	**10.5**	**10.3**	**9.8**	**8.8**	**9.5**	**9.6**	**7.7**	**9.0**	**10.1**
October	Obs	7.5	18.4	10.3	6.5	6.8	8.2	16.1	2.0	3.2
	Mod	**8.8**	**9.3**	**9.4**	**9.9**	**13.6**	**11.7**	**8.9**	**12.9**	**8.8**
November	Obs	6.4	15.6	8.1	14.1	13.8	11.9	16.6	3.4	2.3
	Mod	**11.8**	**10.5**	**9.3**	**10.3**	**11.6**	**11.2**	**9.2**	**11.1**	**10.8**
December	Obs	6.6	7.6	7.1	6.1	Nd	9.3	7.6	4.3	3.3
	Mod	**11.1**	**11.2**	**10.5**	**10.9**	**11.5**	**12.0**	**12.1**	**12.9**	**9.0**
Corr. Coefficient		**-0.32**	**-0.24**	**0.34**	**-0.05**	**-0.06**	**0.54**	**-0.37**	**-0.36**	**-0.31**

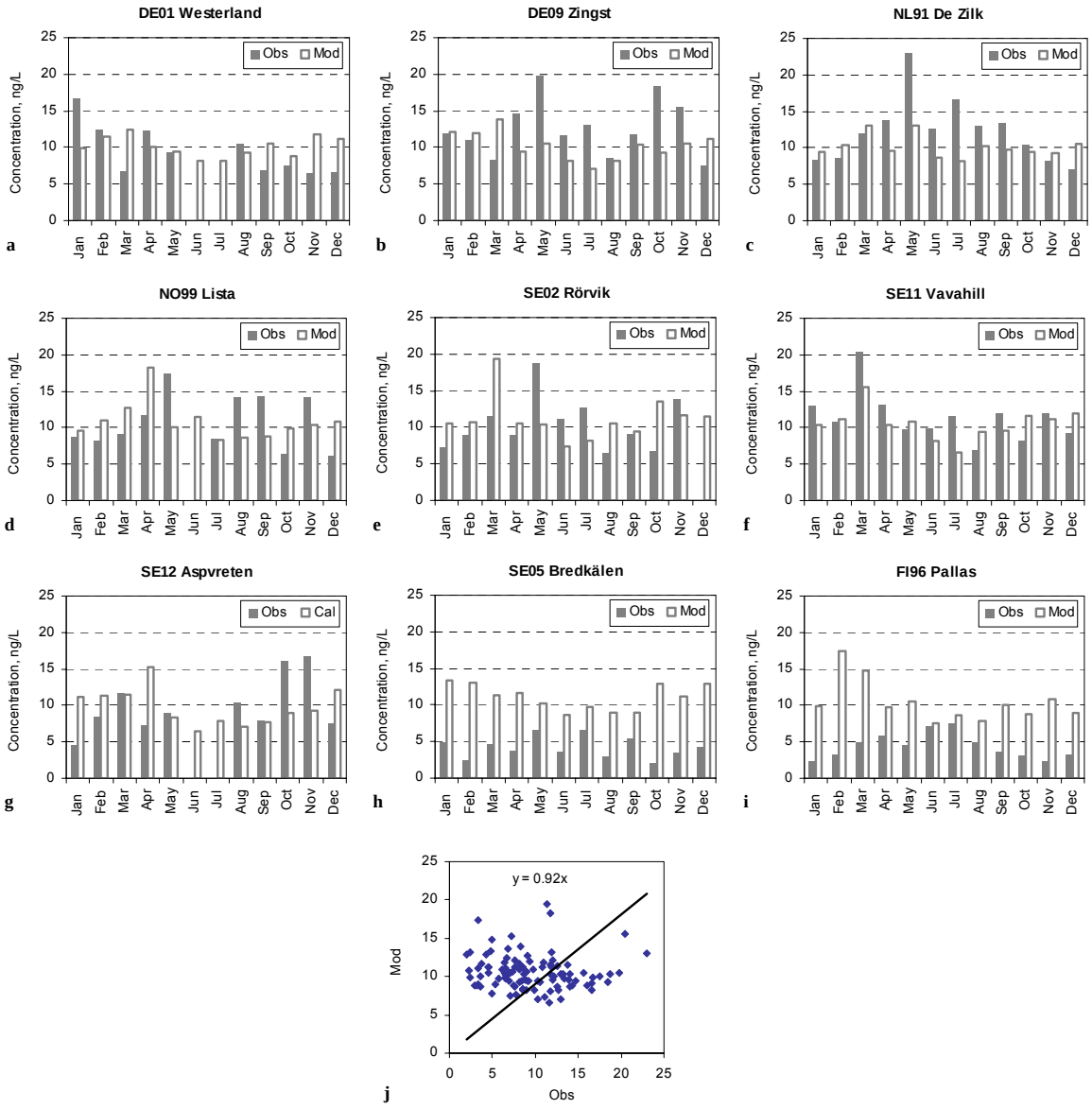


Fig. 7.3.2. *Comparison of measured and calculated by EMAP concentrations of mercury in precipitation at EMEP sites*

Fig. 7.3.3 demonstrates that the measured and forecasted values of monthly precipitation amounts are correlated (CC = 0.53), however, the scattering of the data is rather high. In some cases the difference between individual values exceeds 7 times. Naturally, such high scattering should result in corresponding scattering of wet deposition values. Nevertheless, mean values for the totality of the results (all the stations and all the months) are practically coincide: the difference is lower that 1%.

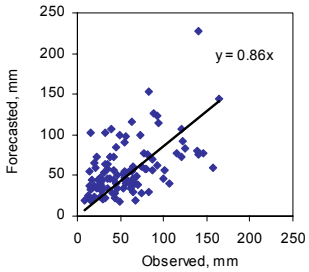


Fig. 7.3.3. *Monthly mean forecasted precipitation amounts used by EMAP against observed precipitation amounts measured at the monitoring stations, mm*

Table 7.3.3 and Fig. 7.3.4 presents the mercury wet deposition fluxes at the stations obtained on the basis of the observations and by the model. Correlation between the observed and modelled values for all the stations is positive and for some of them – very significant (up to CC=0.84). It should be noted that the results scatter significantly. The main reason of such scattering is corresponding scattering of observed and forecasted precipitation amounts. However, for the totality of the results (all months, all stations) there is very good agreement (relative bias is only 12%). Both observations and modelled results do not reveal any obvious seasonal cycles of mercury wet depositions at "polluted" and "regional" stations. One can find in Fig. 7.3.4 that there are elevated depositions during summer time at the "background" stations.

Table 7.3.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by EMAP (Mod), g/km^2*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	0.71	0.48	0.54	1.08	0.51	0.97	0.14	0.21	0.02
	Mod	**0.55**	**0.56**	**0.56**	**0.89**	**0.41**	**0.30**	**0.61**	**0.30**	**0.19**
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14	0.12	0.05
	Mod	**0.59**	**0.49**	**0.49**	**0.85**	**0.46**	**0.46**	**0.39**	**0.22**	**0.39**
March	Obs	0.53	0.53	0.72	1.44	0.72	0.47	0.30	0.13	0.08
	Mod	**0.73**	**0.73**	**0.73**	**0.76**	**0.73**	**0.54**	**0.46**	**0.49**	**0.29**
April	Obs	0.20	0.52	0.68	0.79	0.49	0.56	0.10	0.16	0.11
	Mod	**0.45**	**0.97**	**0.97**	**0.35**	**0.47**	**0.47**	**0.86**	**0.85**	**0.24**
May	Obs	0.25	0.44	0.48	0.82	0.66	0.55	0.15	0.18	0.09
	Mod	**0.45**	**0.76**	**0.76**	**0.35**	**0.36**	**0.43**	**0.18**	**0.24**	**0.68**
June	Obs	-	1.08	1.11	-	1.63	0.87	-	0.29	0.23
	Mod	**0.53**	**1.10**	**1.10**	**0.92**	**0.57**	**0.46**	**0.17**	**0.65**	**0.77**
July	Obs	-	0.42	0.64	0.55	0.80	0.53	-	0.42	0.63
	Mod	**0.43**	**0.59**	**0.59**	**0.49**	**0.46**	**0.33**	**0.37**	**1.12**	**1.33**
August	Obs	0.82	0.67	1.23	1.03	0.78	0.96	0.40	0.16	0.36
	Mod	**0.71**	**1.18**	**1.18**	**0.86**	**1.12**	**0.76**	**0.76**	**0.50**	**0.48**
September	Obs	0.70	0.18	0.75	1.33	0.62	0.77	0.27	0.31	0.12
	Mod	**0.60**	**0.96**	**0.96**	**1.09**	**0.47**	**0.28**	**0.45**	**0.48**	**0.22**
October	Obs	0.80	0.80	0.36	1.07	0.59	0.44	0.25	0.12	0.09
	Mod	**0.35**	**0.43**	**0.43**	**1.41**	**0.94**	**0.39**	**0.91**	**0.44**	**0.33**
November	Obs	0.29	0.45	0.67	0.77	0.17	0.23	0.39	0.10	0.09
	Mod	**0.47**	**0.28**	**0.28**	**0.94**	**0.29**	**0.42**	**0.41**	**0.24**	**0.50**
December	Obs	0.93	0.75	0.86	0.86	-	1.12	0.34	0.12	0.05
	Mod	**0.84**	**0.88**	**0.88**	**2.49**	**1.04**	**0.88**	**1.02**	**0.42**	**0.33**
Year	**Obs**	**7.1***	**6.6**	**8.5**	**11.6***	**8.2***	**8.2**	**3.0***	**2.3**	**1.9**
	Mod	**6.7**	**8.9**	**8.9**	**11.4**	**7.3**	**5.7**	**6.6**	**6.0**	**5.8**
Corr. Coefficient		**0.54**	**0.12**	**0.78**	**0.15**	**0.26**	**0.47**	**0.20**	**0.68**	**0.84**

* - corrected for 12 months

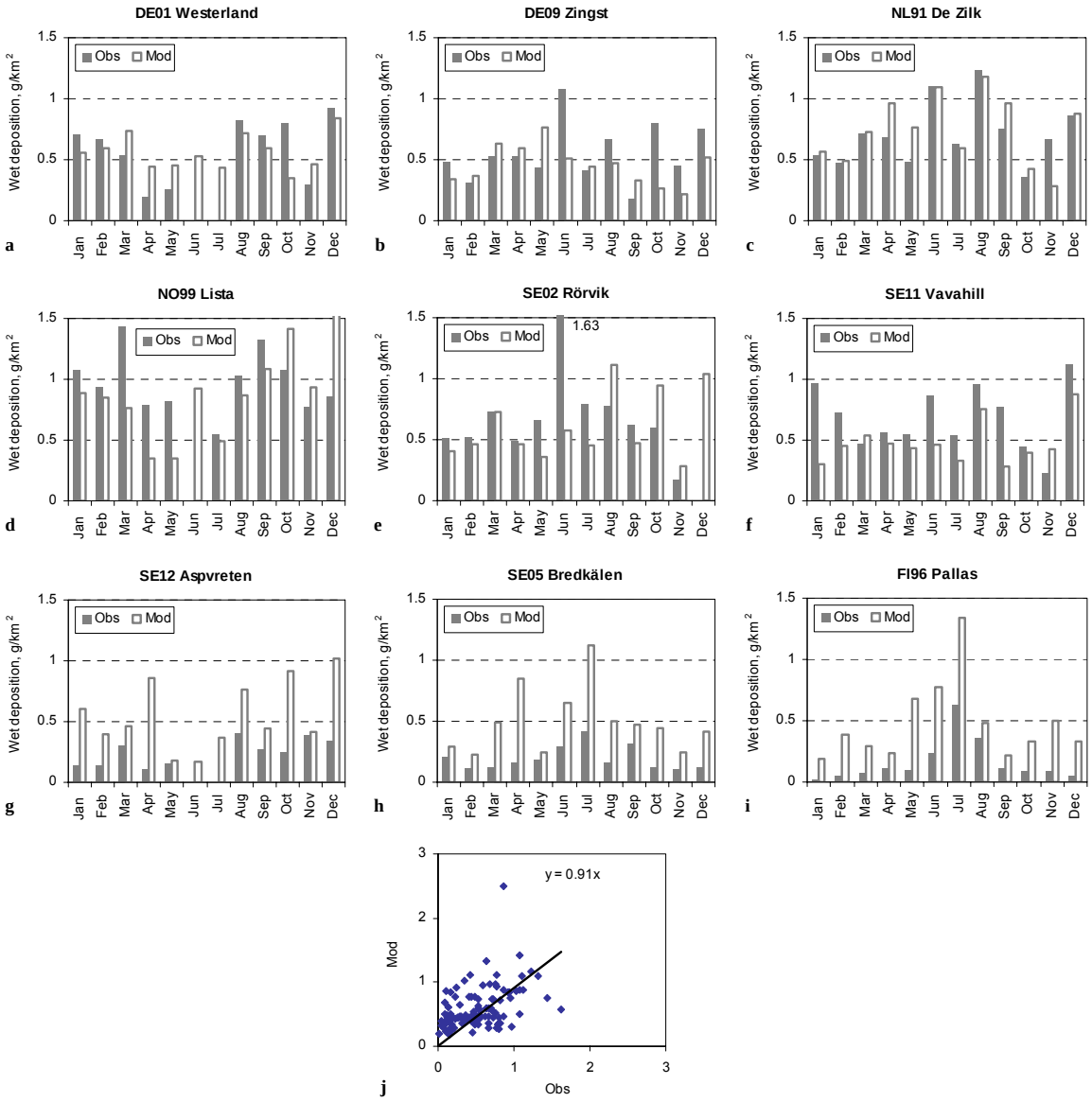


Fig. 7.3.4. *Comparison of mercury monthly mean wet deposition values obtained on the basis of observations and calculated by EMAP for locations of EMEP stations*

The highest deviation between the observed and modelled results for wet deposition reaches a factor of 9.9. Cumulative curve for all wet deposition results is shown in Fig. 7.3.5. The curve demonstrates that 66% of all results are within a factor of 2; 82% - within a factor of 3 and 94% - within a factor of 5. Hence, probability to obtain an acceptable result (within a factor of 2) is high enough and probability of total failure is very low.

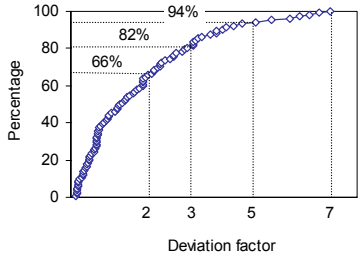


Fig. 7.3.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by EMAP (9 stations, 12 months)*

Month-to-month variations are smoothed when annual depositions are considered in Fig. 7.3.6. In this figure the monitoring stations are arranged in accordance with latitudes: from the most southern station NL91 (52°N) to the most northern one FI96 (68°N). The modelled and observed values are in close agreement for "polluted" and "regional" stations. However, the model overestimates significantly (2.5-3 times) mercury wet deposition at northern "background" stations. Nevertheless, correlation between the observed and modelled annual wet deposition values is high (r=0.72).

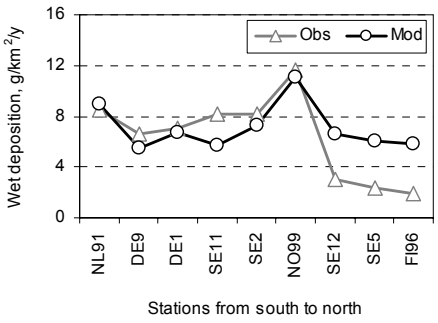


Fig 7.3.6. *Latitudinal trends of modelled by EMAP and observed annual wet depositions*

7.4. MSCE Heavy Metal model, Regional version (MSCE-HM)

MSCE-HM was used to simulate concentrations and depositions at all station locations during all months of 1999. The items of mercury atmospheric balances for the selected countries were also calculated for all months and for 1999 as a whole. The measured and calculated concentrations of gaseous elemental mercury (GEM) are presented in Table 7.4.1 and Fig. 7.4.1. Both observations and modelled results do not reveal any obvious seasonal trends during 1999 – the values for summer period are only slightly lower that for winter period. The highest deviation of a modelled mean monthly value from a measured one reaches 1.6 times (station Pallas in September). In general, the model somewhat overestimates the measurements: relative bias for all considered sites and months is 13%. The most obvious overestimation (36%) can be noted for station SE02 Rörvik. The scattering of the results is not high. The correlation coefficients for all the stations but SE02 Rörvik are significant (CC is between 0.51 and 0.98). Nevertheless, only 57% of the totality of the results lies within the factor of 1.2.

Table 7.4.1. *Monthly mean observed (Obs) and modelled (Mod) by MSCE-HM concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station				
		NO99	SE02	FI96	NO42	IE31
January	Obs	1.8	1.35	1.5	2.13	1.83
	Mod	**1.86**	**1.98**	**1.87**	**1.64**	**1.81**
February	Obs	1.7	1.33	1.60	ol	1.70
	Mod	**1.73**	**1.85**	**1.93**	**1.72**	**1.74**
March	Obs	2.1	1.43	1.85	1.83	1.85
	Mod	**1.94**	**2.04**	**2.06**	**1.93**	**1.83**
April	Obs	1.9	1.47	1.60	ol	1.64
	Mod	**1.80**	**1.86**	**1.81**	**1.68**	**1.70**
May	Obs	1.6	Nd	1.40	1.60	1.60
	Mod	**1.76**	**1.78**	**1.64**	**1.65**	**1.70**
June	Obs	Nd	1.35	1.30	nd	1.69
	Mod	**1.85**	**1.89**	**1.78**	**1.73**	**1.71**
July	Obs	Nd	1.35	1.23	nd	1.57
	Mod	**1.68**	**1.70**	**1.62**	**1.36**	**1.62**
August	Obs	nd	1.44	1.33	nd	1.51
	Mod	**1.72**	**1.85**	**1.56**	**1.47**	**1.65**
September	Obs	Ol	1.62	1.10	1.75	1.54
	Mod	**1.90**	**1.95**	**1.75**	**1.49**	**1.60**
October	Obs	1.5	1.30	1.30	1.20	1.90
	Mod	**1.84**	**1.91**	**1.79**	**1.39**	**1.83**
November	Obs	1.8	1.48	1.38	nd	1.96
	Mod	**1.91**	**2.03**	**1.84**	**1.56**	**1.90**
December	Obs	1.7	1.28	1.50	1.65	2.10
	Mod	**1.90**	**2.01**	**1.85**	**1.65**	**1.97**
Year	Obs	1.76*	1.40*	1.42	1.69*	1.74
	Mod	**1.83**	**1.90**	**1.79**	**1.61**	**1.76**
Correlation Coefficient		**0.51**	**0.18**	**0.73**	**0.55**	**0.98**

nd – no data; ol – outlier; * - accepted for 12 months

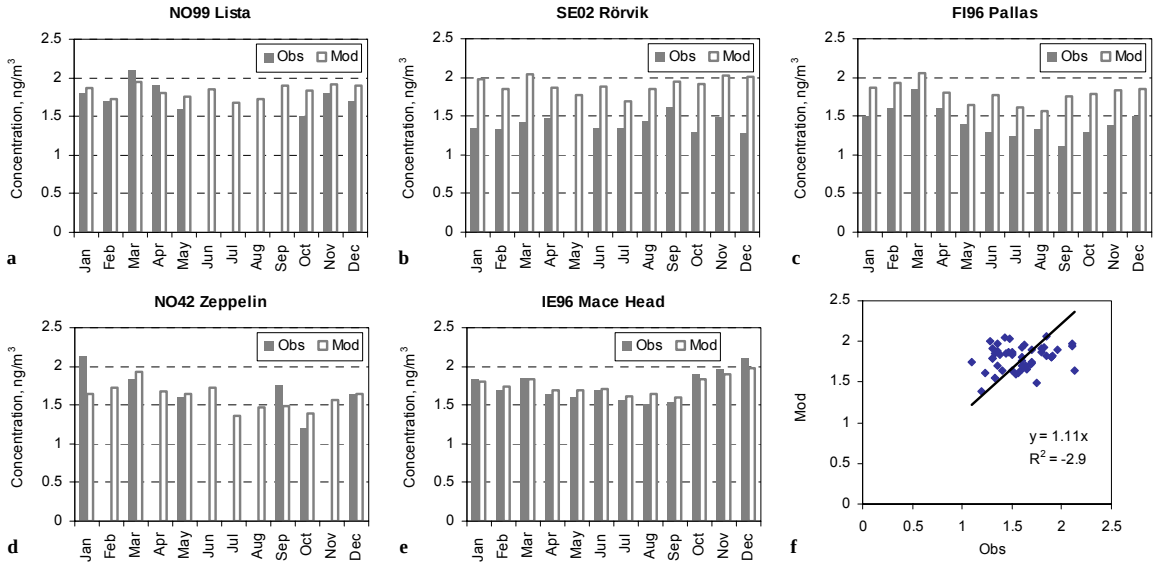


Fig. 7.4.1. *Comparison of measured and calculated by MSCE-HM concentrations of gaseous elemental mercury (GEM) at EMEP sites*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.4.2 and Fig. 7.4.2. Variability of the data is high, maximum deviation of the modelled value from the observed one reaches 7 times. In spite of high scattering, 72% of all the results are within the factor of 2. The modelled mean weighted concentration (for the totality of the stations) exceeds the measured mean weighted one: modelled value is 25% higher. The modelled and the observed values are slightly correlated for 7 of 9 stations. The model tends to overestimate obviously (about 2 times) the values for northern "background" stations. For these stations there is regularity: both the observations and the model demonstrate higher values during warm season.

Table 7.4.2. *Measured concentrations of Hg in precipitation at EMEP stations in 1999 (Obs) and modelled by MSCE-HM (Mod), ng/L*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	16.7	12.0	8.4	8.8	7.3	13.0	4.5	4.8	2.4
	Mod	**19.8**	**22.8**	**21.3**	**8.0**	**12.7**	**30.9**	**12.1**	**4.3**	**6.9**
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
	Mod	**12.8**	**19.6**	**23.5**	**6.9**	**12.3**	**25.8**	**11.4**	**7.5**	**7.1**
March	Obs	6.7	8.3	11.9	9.1	11.4	20.5	11.7	4.5	4.9
	Mod	**11.2**	**18.7**	**18.9**	**16.2**	**14.5**	**23.8**	**22.0**	**7.1**	**14.8**
April	Obs	12.2	14.7	13.9	11.8	9.0	13.2	7.2	3.8	5.8
	Mod	**17.8**	**10.3**	**18.7**	**6.0**	**13.8**	**13.6**	**6.5**	**5.4**	**8.0**
May	Obs	9.2	19.8	23.0	17.5	18.7	9.7	9.0	6.6	4.5
	Mod	**12.4**	**11.2**	**22.9**	**15.4**	**13.4**	**13.2**	**25.0**	**11.4**	**6.6**
June	Obs	ol	11.7	12.6	nd	11.1	9.9	nd	3.6	7.1
	Mod	**9.7**	**13.2**	**12.1**	**7.3**	**8.4**	**14.4**	**19.4**	**6.7**	**7.3**
July	Obs	ol	13.0	16.6	8.5	12.7	11.6	ol	6.6	7.6
	Mod	**12.2**	**7.8**	**21.1**	**9.3**	**9.3**	**9.7**	**7.3**	**4.8**	**6.2**
August	Obs	10.6	8.5	13.1	14.1	6.4	6.9	10.3	3.0	4.9
	Mod	**10.5**	**9.2**	**10.9**	**9.0**	**6.1**	**8.2**	**9.8**	**7.7**	**6.6**
September	Obs	6.9	11.8	13.4	14.3	9.1	12.0	7.8	5.4	3.6
	Mod	**11.4**	**11.7**	**18.3**	**7.1**	**7.7**	**26.4**	**8.0**	**11.9**	**12.8**
October	Obs	7.5	18.4	10.3	6.5	6.8	8.2	16.1	2.0	3.2
	Mod	**9.7**	**16.7**	**23.9**	**9.8**	**8.3**	**22.1**	**8.8**	**5.8**	**6.5**
November	Obs	6.4	15.6	8.1	14.1	13.8	11.9	16.6	3.4	2.3
	Mod	**36.1**	**38.3**	**47.4**	**11.4**	**27.7**	**80.8**	**24.2**	**6.7**	**8.0**
December	Obs	6.6	7.6	7.1	6.1	nd	9.3	7.6	4.3	3.3
	Mod	**9.5**	**13.2**	**16.3**	**4.9**	**7.5**	**16.3**	**9.1**	**4.9**	**6.7**
Corr. Coefficient		**0.05**	**0.13**	**-0.22**	**0.38**	**0.45**	**0.20**	**0.39**	**0.29**	**-0.05**

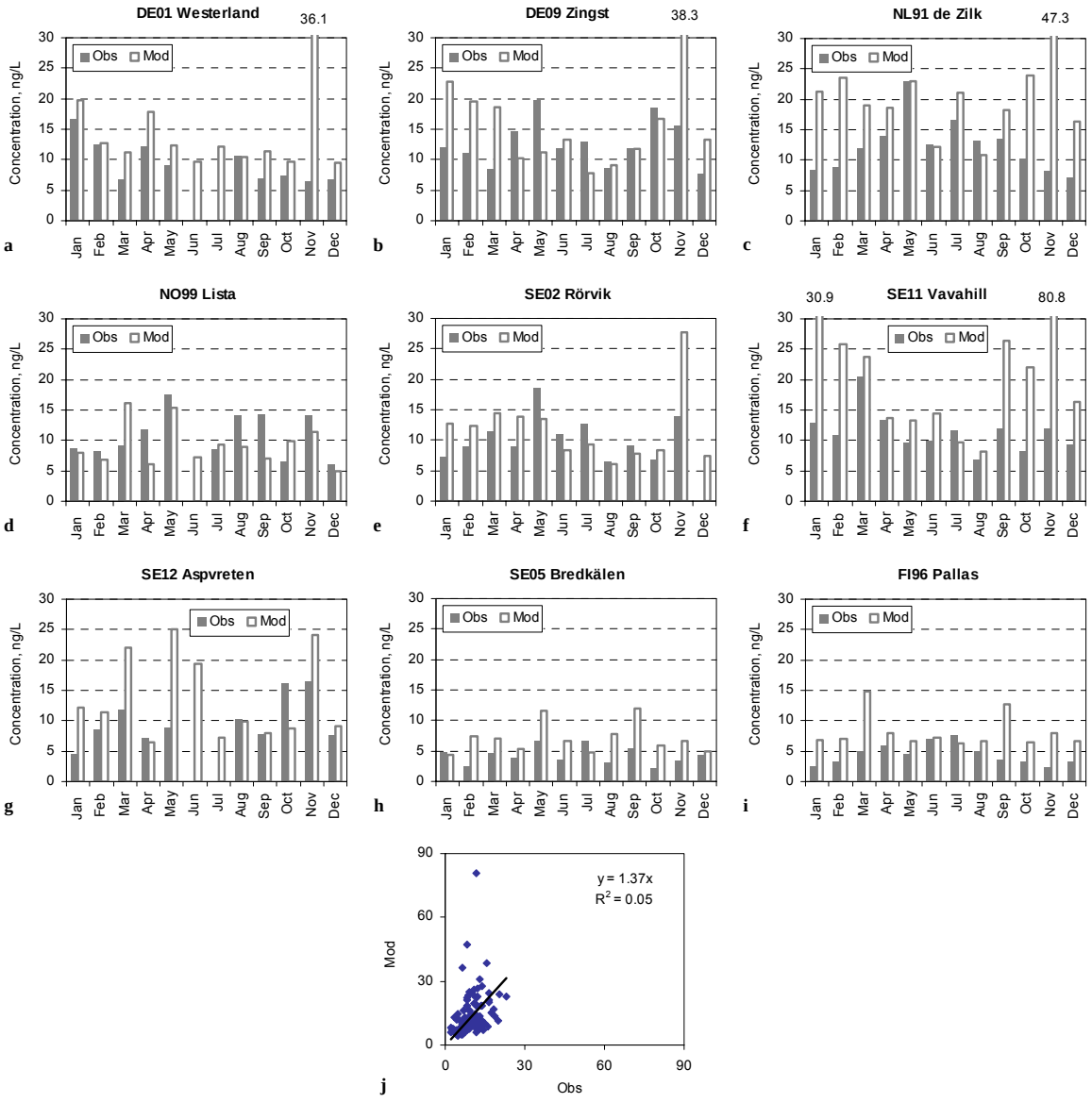


Fig. 7.4.2. *Comparison of measured and calculated by MSCE-HM concentrations of mercury in precipitation at EMEP sites*

Fig. 7.4.3 demonstrates that the measured and forecasted values of monthly precipitation amounts are correlated (CC = 0.52), however, the scattering of the data is rather high. In some cases the difference between individual values exceeds 6 times. About 27% of the results are outside the factor of 2. The forecasted values obtained by SDA meteorological system developed by Russian Hydrometeorological Center [*Rubinstein et al.*, 1997] are lower than the observed ones (in general, 22%). Naturally, such high scattering and general underestimation should result in corresponding scattering of wet deposition values.

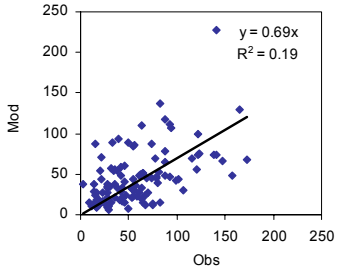


Fig. 7.4.3. *Monthly mean forecasted precipitation amounts used by MSCE-HM against observed precipitation amounts measured at the monitoring stations, mm*

Table 7.4.3 and Fig. 7.4.4 presents the mercury wet deposition fluxes at the stations obtained on the basis of the observations and by MSCE-HM. Correlation between the observed and modelled values is positive, and for 3 stations of 9 is very significant (up to CC = 0.84). However the results scatter significantly: maximum deviation for an individual pair is 4.6. Most probably, that the main reason of such scattering is corresponding scattering of observed and forecasted precipitation amounts. About 25% of the results lie outside the factor of 2. Nevertheless, for the totality of the results (all months, all stations) there is good agreement. Both observations and modelled results do not reveal any obvious seasonal cycles of mercury depositions at "polluted" and "regional" stations. One can find in Fig. 7.4.4 that there are elevated depositions during summer time at the "background" stations.

Table 7.4.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by MSCE-HM (Mod), g/km^2*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	0.71	0.48	0.54	1.08	0.51	0.97	0.14	0.21	0.02
	Mod	**0.57**	**0.41**	**1.08**	**0.60**	**0.34**	**0.42**	**0.42**	**0.07**	**0.10**
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14	0.12	0.05
	Mod	**0.41**	**0.40**	**0.81**	**0.48**	**0.28**	**0.64**	**0.20**	**0.06**	**0.10**
March	Obs	0.53	0.53	0.72	1.44	0.72	0.47	0.30	0.13	0.08
	Mod	**0.44**	**0.62**	**0.92**	**0.77**	**0.29**	**0.41**	**0.29**	**0.24**	**0.13**
April	Obs	0.20	0.52	0.68	0.79	0.49	0.56	0.10	0.16	0.11
	Mod	**0.49**	**0.57**	**1.66**	**0.77**	**0.32**	**0.44**	**0.24**	**0.32**	**0.14**
May	Obs	0.25	0.44	0.48	0.82	0.66	0.55	0.15	0.18	0.09
	Mod	**0.47**	**0.79**	**1.00**	**0.37**	**0.29**	**0.39**	**0.21**	**0.10**	**0.36**
June	Obs	-	1.08	1.11	-	1.63	0.87	-	0.29	0.23
	Mod	**0.46**	**0.62**	**1.42**	**0.49**	**0.55**	**0.69**	**0.24**	**0.35**	**0.65**
July	Obs	-	0.42	0.64	0.55	0.80	0.53	-	0.42	0.63
	Mod	**0.58**	**0.45**	**1.13**	**0.47**	**0.46**	**0.40**	**0.28**	**0.53**	**0.85**
August	Obs	0.82	0.67	1.23	1.03	0.78	0.96	0.40	0.16	0.36
	Mod	**0.73**	**0.41**	**1.16**	**0.79**	**0.61**	**0.61**	**0.91**	**0.34**	**0.30**
September	Obs	0.70	0.18	0.75	1.33	0.62	0.77	0.27	0.31	0.12
	Mod	**0.50**	**0.28**	**1.56**	**0.79**	**0.35**	**0.38**	**0.43**	**0.43**	**0.19**
October	Obs	0.80	0.80	0.36	1.07	0.59	0.44	0.25	0.12	0.09
	Mod	**0.30**	**0.34**	**0.89**	**1.27**	**0.54**	**0.46**	**0.76**	**0.16**	**0.21**
November	Obs	0.29	0.45	0.67	0.77	0.17	0.23	0.39	0.10	0.09
	Mod	**0.52**	**0.23**	**0.72**	**0.96**	**0.31**	**0.97**	**0.64**	**0.07**	**0.26**
December	Obs	0.93	0.75	0.86	0.86	-	1.12	0.34	0.12	0.05
	Mod	**0.70**	**0.55**	**1.21**	**1.10**	**0.59**	**0.91**	**0.55**	**0.13**	**0.19**
Year	**Obs**	**7.1***	**6.6**	**8.5**	**11.6***	**8.2***	**8.2**	**3.0***	**2.3**	**1.9**
	Mod	**6.2**	**5.7**	**13.6**	**8.9**	**4.9**	**6.7**	**5.2**	**2.8**	**3.5**
Corr. Coefficient		**0.30**	**0.31**	**0.44**	**0.23**	**0.57**	**0.12**	**0.74**	**0.79**	**0.84**

* - corrected for 12 months

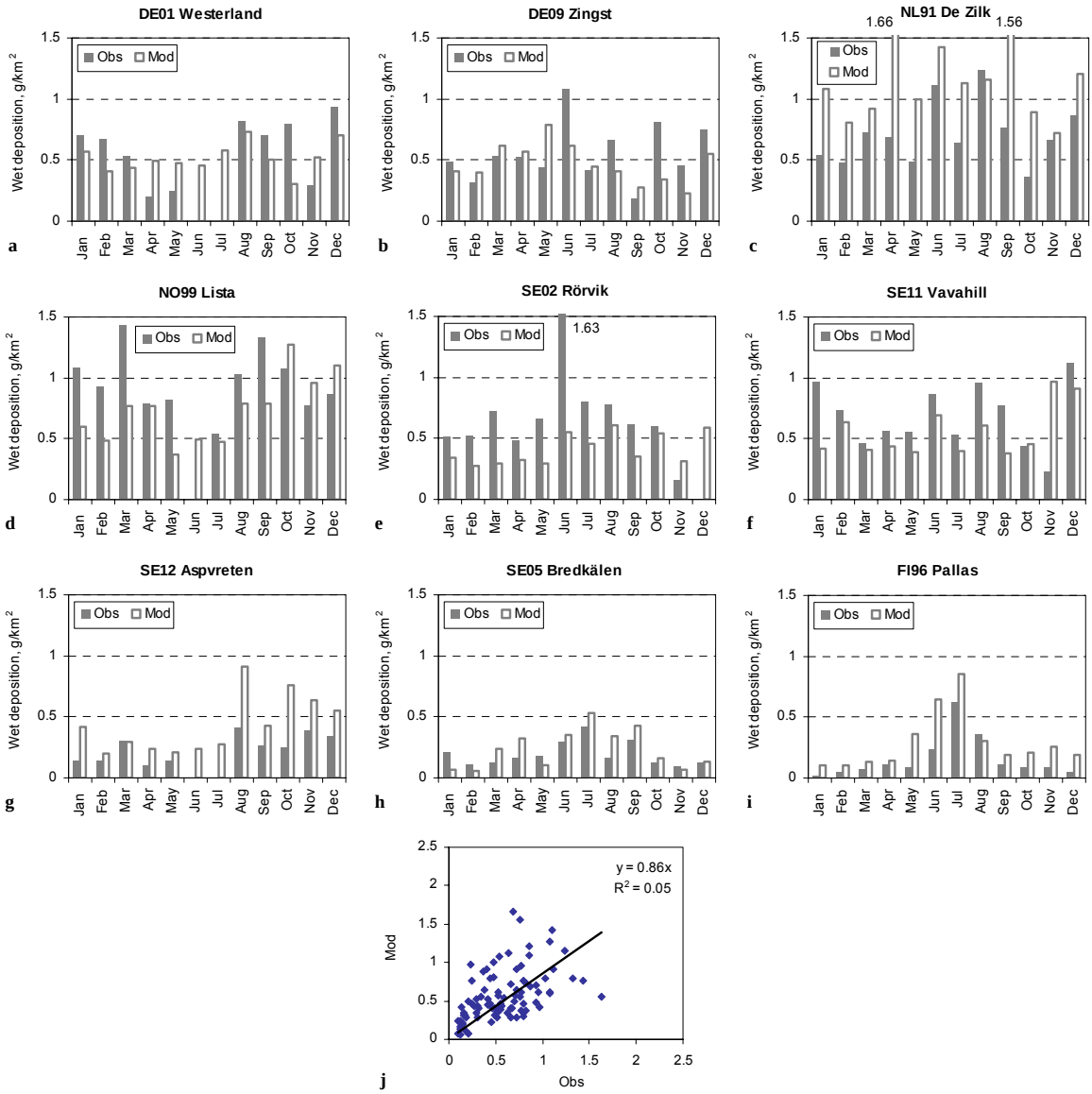


Fig. 7.4.4. *Comparison of mercury monthly mean wet deposition values obtained on the basis of observations and calculated by MSCE-HM for locations of EMEP stations*

The data presented in Table 7.4.3 and Fig. 7.4.4 shown, that the highest deviation between the observed and modelled results for wet deposition reaches a factor of 4.6. It means that all results obtained by MSCE-HM lie within this factor. Cumulative distribution of the deviation factors is shown in Fig. 7.4.5. The curve demonstrates that 71% of all results are within a factor of 2 and 91% - within a factor of 3. Hence, probability to obtain an acceptable result (within a factor of 2) is very high and probability of total failure is very low.

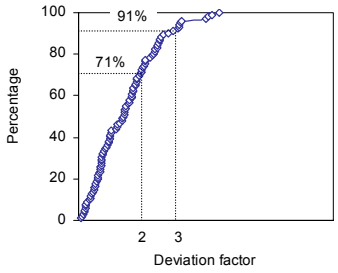


Fig. 7.4.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by MSCE-HM (9 stations, 12 months)*

Spatial trend of annual wet depositions is considered in Fig. 7.4.6. The monitoring stations are arranged in the figure in accordance with latitudes from south to north. The modelled and observed values are in close agreement for all stations – deviations do not exceed a factor of 2 in all cases. Correlation between the observed and modelled annual wet deposition values is very high (r = 0.67).

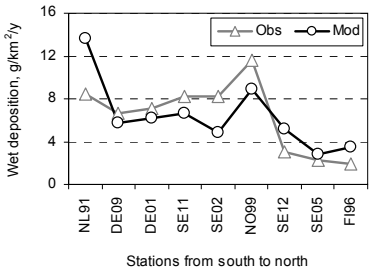


Fig. 7.4.6. *Latitudinal trends of modelled by MSCE-HM and observed annual wet*

7.5. MSCE Heavy Metal model, Global version (MSCE-HM-Hem)

MSCE-HM-Hem was used to simulate concentrations and depositions at all station locations during all months of 1999. The items of mercury atmospheric balances for the selected countries were also calculated for all months and for 1999 as a whole. The measured and calculated concentrations of gaseous elemental mercury (GEM) are presented in Table 7.5.1 and Fig. 7.5.1 (a-f). Both observations and modelled results do not reveal any obvious seasonal trends during 1999. The highest deviation of a modelled mean monthly value from a measured one is 1.4 times (station FI96 Pallas in September). The scattering of the results is not high, and 2/3 of the results are within the factor of 1.2. In general, the modelled results are in very good agreement with the observations: the overestimation is only 4%. The correlation between the observed and modelled results for 4 stations (but Mace Head) is rather high. For Mace Head the statistical analysis shows formally high anti-correlation. The reason of this is in the fact that the monthly average values here are not variable and any deviation for 2-3 months can change the correlation coefficient from highly positive to highly negative. One can note in the Fig. 7.5.1f that the model never predicts concentration values below 1.5 ng/m^3, while such low values (up to 1.1 ng/m^3) are observed at stations Rörvik and Pallas very often.

Table 7.5.1. *Monthly mean observed (Obs) and modelled (Mod) by MSCE-HM-Hem concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station				
		NO99	SE02	FI96	NO42	IE31
January	Obs	1.8	1.35	1.5	2.13	1.83
	Mod	**1.61**	**1.68**	**1.70**	**1.69**	**1.56**
February	Obs	1.7	1.33	1.60	ol	1.70
	Mod	**1.61**	**1.66**	**1.67**	**1.70**	**1.59**
March	Obs	2.1	1.43	1.85	1.83	1.85
	Mod	**1.69**	**1.78**	**1.74**	**1.70**	**1.59**
April	Obs	1.9	1.47	1.60	ol	1.64
	Mod	**1.72**	**1.74**	**1.66**	**1.70**	**1.62**
May	Obs	1.6	nd	1.40	1.60	1.60
	Mod	**1.74**	**1.73**	**1.61**	**1.64**	**1.71**
June	Obs	nd	1.35	1.30	nd	1.69
	Mod	**1.77**	**1.79**	**1.62**	**1.62**	**1.65**
July	Obs	nd	1.35	1.23	nd	1.57
	Mod	**1.68**	**1.67**	**1.58**	**1.58**	**1.62**
August	Obs	nd	1.44	1.33	nd	1.51
	Mod	**1.67**	**1.72**	**1.52**	**1.59**	**1.64**
September	Obs	ol	1.62	1.10	1.75	1.54
	Mod	**1.86**	**1.86**	**1.56**	**1.58**	**1.56**
October	Obs	1.5	1.30	1.30	1.20	1.90
	Mod	**1.63**	**1.66**	**1.57**	**1.62**	**1.62**
November	Obs	1.8	1.48	1.38	nd	1.96
	Mod	**1.61**	**1.68**	**1.59**	**1.62**	**1.55**
December	Obs	1.7	1.28	1.50	1.65	2.10
	Mod	**1.60**	**1.65**	**1.66**	**1.68**	**1.56**
Year	Obs	**1.76***	**1.40***	**1.42**	**1.69***	**1.74**
	Mod	**1.68**	**1.72**	**1.62**	**1.64**	**1.61**
Correlation Coefficient		**0.22**	**0.75**	**0.86**	**0.49**	**-0.53**

nd – no data; ol – outlier; * - corrected for 12 months

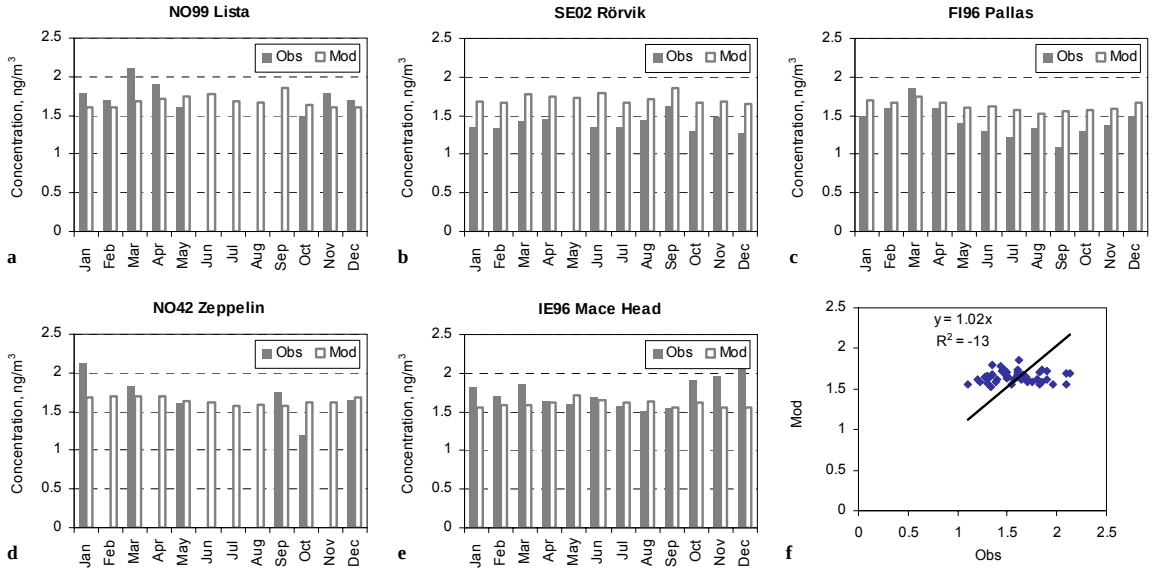


Fig. 7.5.1. *Comparison of measured and calculated by MSCE-HM-Hem concentrations of gaseous elemental mercury (GEM) at EMEP sites*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.5.2 and Fig. 7.5.2. Generally, variability of the data is rather high, maximum deviation of the modelled value from the observed one reaches 4.4 times. In spite of high scattering, 70% of all the results are within the factor of 2. The modelled mean weighted concentration (for the totality of the stations) slightly exceeds the measured mean weighted one: the modelled value is only 7% higher. The modelled and the observed values generally correlate (up to CC = 0.87). The model tends to overestimate (1.8 times) the values for northern "background" stations.

Table 7.5.2. *Measured concentrations of Hg in precipitation at EMEP stations in 1999 (Obs) and modelled by MSCE-HM-Hem (Mod), ng/L*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	16.7	12.0	8.4	8.8	7.3	13.0	4.5	4.8	2.4
	Mod	**16.6**	**27.0**	**13.2**	**7.9**	**18.0**	**21.6**	**12.1**	**7.7**	**7.8**
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
	Mod	**12.4**	**24.3**	**11.7**	**6.2**	**13.8**	**18.6**	**8.8**	**5.3**	**5.3**
March	Obs	6.7	8.3	11.9	9.1	11.4	20.5	11.7	4.5	4.9
	Mod	**15.2**	**19.3**	**14.1**	**10.7**	**22.6**	**20.8**	**19.7**	**11.4**	**8.4**
April	Obs	12.2	14.7	13.9	11.8	9.0	13.2	7.2	3.8	5.8
	Mod	**21.8**	**20.5**	**13.1**	**10.4**	**14.8**	**16.9**	**11.7**	**7.2**	**10.4**
May	Obs	9.2	19.8	23.0	17.5	18.7	9.7	9.0	6.6	4.5
	Mod	**20.2**	**20.4**	**25.0**	**15.5**	**20.8**	**20.0**	**19.4**	**8.0**	**9.7**
June	Obs	ol	11.7	12.6	nd	11.1	9.9	nd	3.6	7.1
	Mod	**12.3**	**13.8**	**11.1**	**8.4**	**11.7**	**12.9**	**12.4**	**6.8**	**7.2**
July	Obs	ol	13.0	16.6	8.5	12.7	11.6	ol	6.6	7.6
	Mod	**12.5**	**19.8**	**13.0**	**9.2**	**13.9**	**16.7**	**11.0**	**6.9**	**7.1**
August	Obs	10.6	8.5	13.1	14.1	6.4	6.9	10.3	3.0	4.9
	Mod	**12.3**	**13.3**	**9.5**	**10.7**	**15.0**	**13.3**	**11.9**	**7.0**	**5.8**
September	Obs	6.9	11.8	13.4	14.3	9.1	12.0	7.8	5.4	3.6
	Mod	**16.7**	**23.9**	**11.7**	**14.7**	**21.6**	**23.6**	**14.5**	**8.3**	**8.1**
October	Obs	7.5	18.4	10.3	6.5	6.8	8.2	16.1	2.0	3.2
	Mod	**17.5**	**23.8**	**11.7**	**9.7**	**16.2**	**18.2**	**15.5**	**6.2**	**10.0**
November	Obs	6.4	15.6	8.1	14.1	13.8	11.9	16.6	3.4	2.3
	Mod	**17.3**	**34.4**	**12.3**	**13.2**	**31.2**	**31.2**	**28.8**	**9.1**	**10.0**
December	Obs	6.6	7.6	7.1	6.1	nd	9.3	7.6	4.3	3.3
	Mod	**9.4**	**16.6**	**9.5**	**4.8**	**11.2**	**13.1**	**9.0**	**4.7**	**5.5**
Corr. Coefficient		**0.14**	**0.43**	**0.77**	**0.87**	**0.41**	**0.38**	**0.69**	**0.30**	**-0.09**

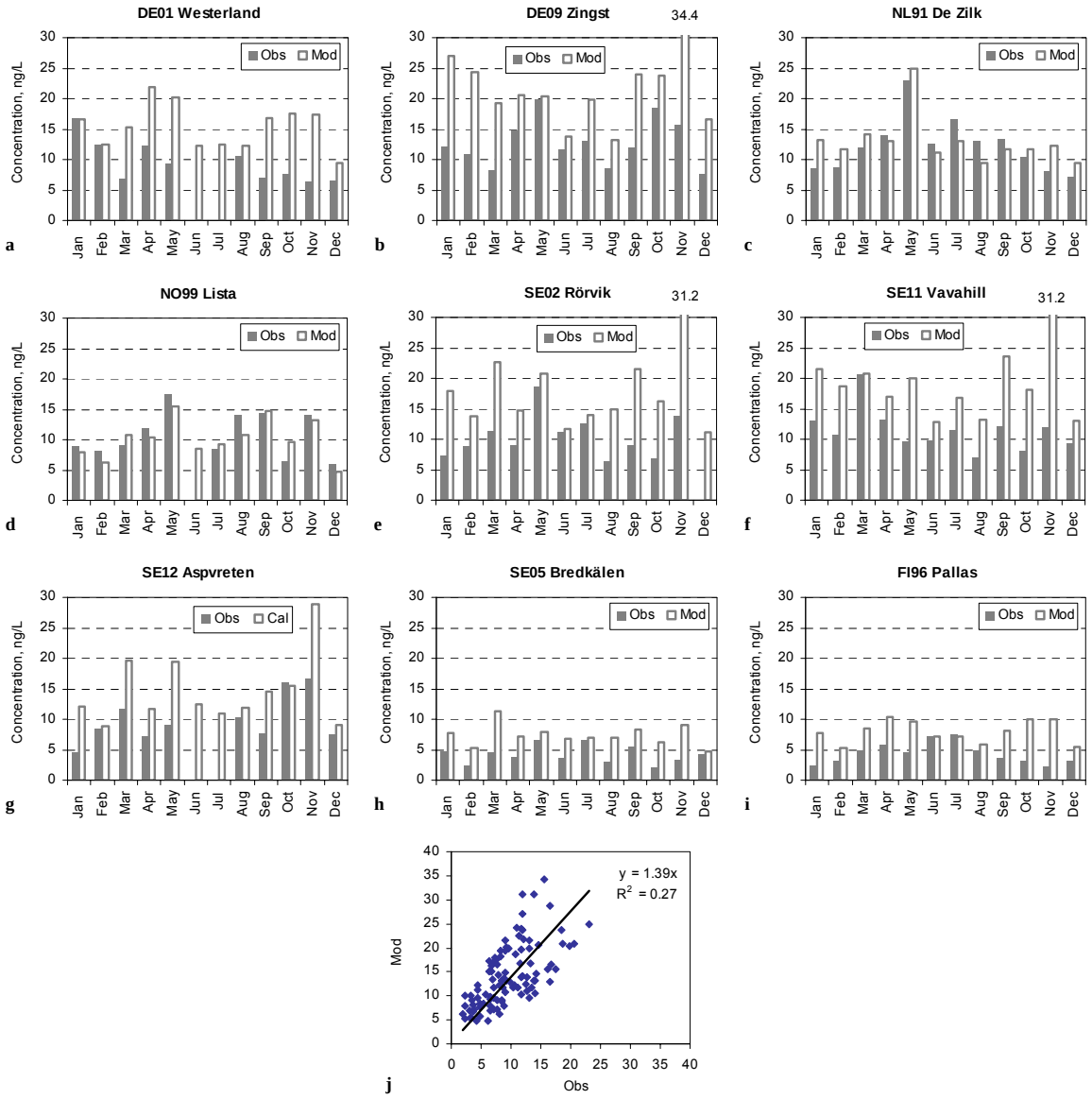


Fig. 7.5.2. *Comparison of measured and calculated by MSCE-HM-Hem concentrations of mercury in precipitation at EMEP sites*

Fig. 7.5.3 demonstrates that the measured and forecasted values of monthly precipitation amounts are obviously correlated (CC = 0.73), however, the scattering of the data is rather high. In one case the ratio of modelled to observed value reaches 17 times. Nevertheless, about 81% of the results are within the factor of 2. The forecasted values obtained by SDA meteorological system developed by Russian Hydrometeorological Center [*Rubinstein et al.,* 1997] are slightly higher than the observed ones (in general, 6%). Naturally, high scattering of precipitation amount data should result in corresponding scattering of wet deposition values.

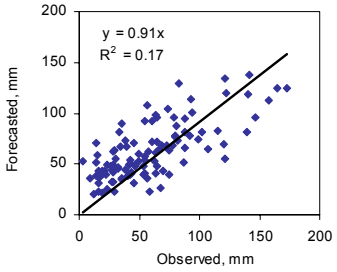


Fig. 7.5.3. *Monthly mean forecasted precipitation amounts used by MSCE-HM-Hem against observed precipitation amounts measured at the monitoring*

Table 7.5.3 and Fig. 7.5.4 presents mercury wet deposition fluxes at the stations obtained on the basis of the observations and by MSCE-HM-Hem. Correlation between the observed and modelled values is high practically for all the stations, however the results scatter significantly: maximum deviation for an individual pair is 13 times (FI96 in January). About 40% of the results lie outside the factor of 2. The model overestimates the observations (relative bias is 46%). Both observations and modelled results do not reveal any obvious seasonal cycles of mercury depositions at "polluted" and "regional" stations. One can find in Fig. 7.5.4 (h, i) that there are elevated depositions during summer time at the "background" stations.

Table 7.5.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by MSCE-HM-Hem (Mod), g/km^2*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	0.71	0.48	0.54	1.08	0.51	0.97	0.14	0.21	0.02
	Mod	**0.90**	**1.04**	**1.25**	**0.90**	**0.76**	**0.86**	**0.63**	**0.32**	**0.28**
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14	0.12	0.05
	Mod	**0.48**	**0.79**	**0.68**	**0.46**	**0.32**	**0.50**	**0.31**	**0.28**	**0.20**
March	Obs	0.53	0.53	0.72	1.44	0.72	0.47	0.30	0.13	0.08
	Mod	**1.17**	**1.17**	**1.28**	**1.17**	**0.92**	**0.99**	**0.84**	**0.48**	**0.37**
April	Obs	0.20	0.52	0.68	0.79	0.49	0.56	0.10	0.16	0.11
	Mod	**0.67**	**0.71**	**0.77**	**0.65**	**0.59**	**0.63**	**0.69**	**0.36**	**0.44**
May	Obs	0.25	0.44	0.48	0.82	0.66	0.55	0.15	0.18	0.09
	Mod	**0.89**	**0.81**	**1.06**	**0.71**	**0.74**	**0.71**	**0.42**	**0.26**	**0.37**
June	Obs	-	1.08	1.11	-	1.63	0.87	-	0.29	0.23
	Mod	**0.94**	**1.08**	**1.04**	**1.04**	**1.13**	**1.04**	**0.78**	**0.59**	**0.59**
July	Obs	-	0.42	0.64	0.55	0.80	0.53	-	0.42	0.63
	Mod	**0.61**	**0.92**	**0.58**	**0.62**	**0.72**	**0.80**	**0.56**	**0.67**	**0.92**
August	Obs	0.82	0.67	1.23	1.03	0.78	0.96	0.40	0.16	0.36
	Mod	**0.84**	**1.24**	**0.93**	**0.72**	**0.81**	**1.06**	**0.77**	**0.37**	**0.60**
September	Obs	0.70	0.18	0.75	1.33	0.62	0.77	0.27	0.31	0.12
	Mod	**1.33**	**0.91**	**1.26**	**1.61**	**1.46**	**1.12**	**0.97**	**0.52**	**0.37**
October	Obs	0.80	0.80	0.36	1.07	0.59	0.44	0.25	0.12	0.09
	Mod	**1.11**	**0.94**	**1.03**	**1.18**	**0.83**	**0.85**	**0.88**	**0.44**	**0.62**
November	Obs	0.29	0.45	0.67	0.77	0.17	0.23	0.39	0.10	0.09
	Mod	**0.49**	**0.75**	**0.86**	**0.97**	**0.62**	**0.64**	**0.70**	**0.55**	**0.73**
December	Obs	0.93	0.75	0.86	0.86	-	1.12	0.34	0.12	0.05
	Mod	**1.11**	**1.25**	**1.26**	**0.63**	**0.74**	**0.89**	**0.59**	**0.23**	**0.28**
Year	**Obs**	**7.1***	**6.6**	**8.5**	**11.6***	**8.2***	**8.2**	**3.0***	**2.3**	**1.9**
	Mod	**10.5**	**11.6**	**12.0**	**10.6**	**9.7**	**10.1**	**8.1**	**5.1**	**5.8**
Corr. Coefficient		**0.47**	**0.50**	**0.13**	**0.72**	**0.46**	**0.43**	**0.45**	**0.62**	**0.77**

* - corrected for 12 months

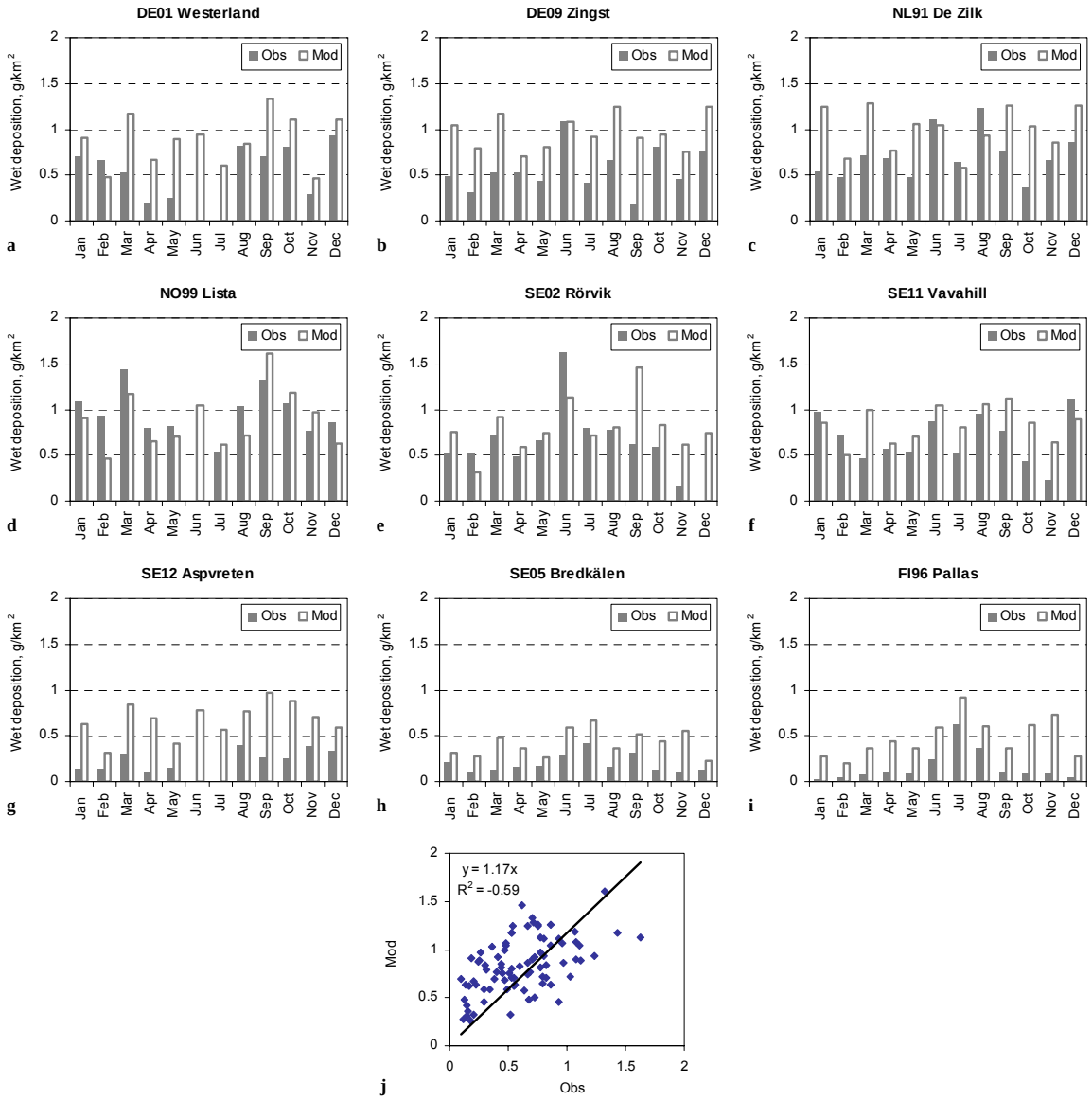


Fig. 7.5.4. *Comparison of mercury monthly mean wet deposition values obtained on the basis of observations and calculated by MSCE-HM-Hem for locations of EMEP stations*

It is derived from the data of Table 7.5.3. and Fig. 7.5.4., that the highest deviation between the observed and modelled results for wet deposition reaches a factor of 13. Cumulative distribution of the deviation factors is shown in Fig. 7.5.5. The curve demonstrates that 63% of all results are within a factor of 2; 78% - within a factor of 3 and 94% - within a factor of 5. Hence, probability to obtain an acceptable result (within a factor of 2) is high enough and probability of total failure is rather low.

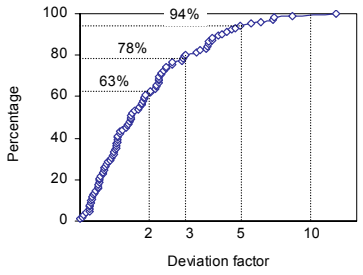


Fig. 7.5.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by MSCE-HM-Hem (9 stations, 12 months)*

Annual wet depositions at the monitoring stations are demonstrated in Fig. 7.5.6. The stations here are arranged in accordance with latitudes from south to north. The modelled and observed values are in close agreement for southern stations – deviations do not exceed a factor of 2. However, the model overestimates significantly the wet deposition values at northern stations. Nevertheless, correlation between the observed and modelled annual wet deposition values for all the stations is very high (r = 0.82).

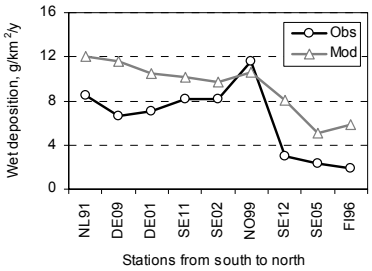


Fig. 7.5.6. *Latitudinal trends of modelled by MSCE-HM-Hem and observed annual wet*

7.6. Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT)

The HYSPLIT-Hg model is a Lagrangian puff model, in which the atmospheric fate and transport of emitted pollutants are simulated [*Cohen et al*., 2004]. In the simulations performed for this analysis, the model domain comprised the entire Northern Hemisphere, up to an elevation of 25 km in the atmosphere. The 1999 meteorological data used to drive the model were from the NCAR/NCEP Reanalysis, with a spatial resolution of 2.5x2.5 degrees and temporal resolution of 6 hours. The relative coarseness of the meteorological data used must be regarded as a significant potential limitation in the realism of the simulation. In some cases, discrepancies between modelled and measured values may be due largely to inaccuracies in the simulation due to the coarseness of the meteorological data.

Only two months during 1999 were simulated (February and August). However, a one-month "spin-up" was used for these simulations; i.e., the February 1999 episode was modelled starting January 1, 1999; the August 1999 episode was modelled starting July 1, 1999. In the HYSPLIT-Hg modelling system, a number of runs are made from "standard source locations". Estimates are then made for all sources based on interpolation from these base runs. For each location, nine individual runs were made – Hg0, HgII, and HgP, from elevations of 0, 100, and 200 meters.

Initially, a total of 183 standard source locations were used. A preliminary analysis of the February estimates suggested that some of the wet fluxes appeared to be underestimated. As a screening procedure, the February results for De Zilk were examined to check if there were any large contributions to the February model-estimated fluxes at NL91 De Zilk from grid cells that were not well represented by the chosen standard points. A number of issues of this nature were identified and 5 additional standard source locations were added to attempt to make the February simulation more realistic for De Zilk. A similar procedure was followed for the initial (183 std pt) February results for Westerland, Lista, and Rörvik, and a few additional standard point locations were added. A total of 13 pts were added, to make a total of 196 standard locations. Not all potential points were added, due to time limitations, but the addition of these points allows some estimate of the sensitivity of the results to such issues.

It was found that by adding the 13 additional standard source locations (of which 5 were specifically designed for De Zilk), the February model-estimated wet flux to De Zilk was increased by a factor of 2, and the resulting wet flux was significantly closer to the observed value. February estimates for the

other locations were not significantly changed. The results presented here are for the 196-point analysis. This sensitivity analysis suggests that the simulation might be improved by adding a number of additional standard points. Normally, a large number of standard points are included, but time constraints limited the extent of the calculations in this study. Experience with the model has shown that in most (but not all) cases, insufficient numbers of standard points leads to underestimates of concentrations and deposition, as the effects of large regional sources are sometimes ”missed”.

The measured and calculated concentrations of gaseous elemental mercury (GEM) are presented in Table 7.6.1 and Fig. 7.6.1 (a-f). Number of the data is too small (7 pairs) to make any deep statistical analysis. The highest deviation of a modelled mean monthly value from a measured one is a factor of 1.29 (stations NO99 Lista and IE31 Mace Head in February). In general, the modelled results are in very good agreement with the observations: the underestimation is only 4%. It should be mentioned again that the modelled values are the sum of the calculated concentrations and a constant background concentration (1.2 ng/m^3). Without the background the modelled values would be significantly underestimated. There are some possible reasons for such underestimation. First, global re-emissions were not included into consideration, as they were not included in the common datasets available for use in the project. Second, a “spin-up” of only one month was used. So, the simulation was not able reproduce the “full”, more realistic Hg0 concentration that one would get if a longer spin up was used. Third, the HYSPLIT model was configured to estimate the ambient concentrations at a height of 10 meters. To the extent that this was higher than the height that the GEM measurements were taken, this may have contributed somewhat to the GEM underestimation.

Table 7.6.1. *Monthly mean observed (Obs) and modelled (Mod) by HYSPLIT-Hg concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station				
		NO99	SE02	FI96	NO42	IE31
February	Obs	1.7	1.33	1.60	**ol**	1.70
	Mod (no background)	**0.12**	**0.26**	**0.19**	**0.10**	**0.12**
	Mod (with background)	**1.32**	**1.46**	**1.39**	**1.30**	**1.32**
August	Obs	**nd**	1.44	1.33	**nd**	1.51
	Mod (no background)	**0.23**	**0.35**	**0.26**	**0.43**	**0.39**
	Mod (with background)	**1.43**	**1.55**	**1.46**	**1.63**	**1.59**

nd – no data; **ol** - outlier

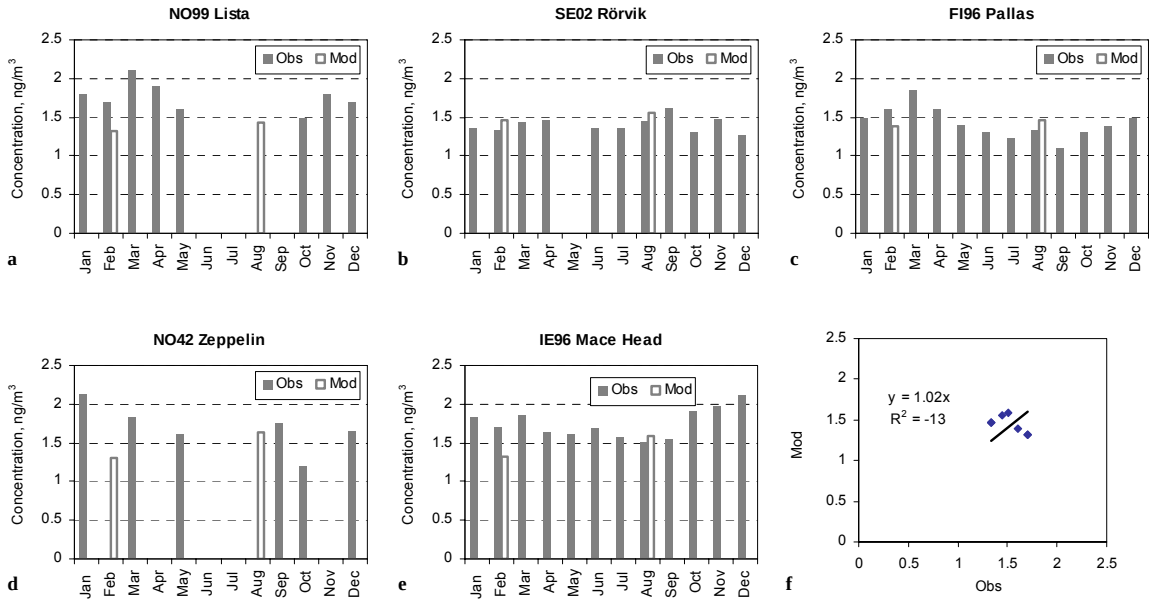


Fig. 7.6.1. *Comparison of measured and calculated by HYSPLIT-Hg concentrations of gaseous elemental mercury (GEM) at EMEP sites [model estimates were only made for Feb and Aug]*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.6.2 and Fig. 7.6.2. Generally, variability of the data is rather high (Fig. 7.6.2 j), maximum deviation of the modelled value from the observed one reaches 6.2 times. In spite of high scattering, 78% of all the results are within the factor of 2. The modelled mean weighted concentration (for the totality of the stations) is 21% lower that the observed one. There is practically no correlation between modelled and the observed values, however, it should be mentioned that the number of pairs is small. If three "outliers" are removed (out of the 18 data pairs), the correlation would improve significantly. On average, the model tends to underestimate the concentrations during wintertime (2.3 times). This underestimate is primarily driven by large underestimates for two stations (DE01 and NO99). These stations are in the same general region (Atlantic Ocean coast of north-central Europe), and this suggests that the underestimate at the two stations may have a common cause. Wet deposition is generally quite episodic, and small errors, - e.g., in meteorological data – can create large discrepancies if they occur at the particular times and places. One possibility is that the coarse-resolution (2.5^o x 2.5^o) weather data used as input to the HYSPLIT model could not adequately characterize the atmospheric flow fields upwind of the DE01/NO99 region for one or more of the precipitation events that occurred during the simulation period.

Table 7.6.2. *Measured concentrations of Hg in precipitation at EMEP stations in 1999 (Obs) and modelled by HYSPLIT-Hg (Mod), ng/L*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
	Mod	**2.4**	**8.6**	**6.3**	**1.3**	**4.1**	**8.1**	**4.3**	**3.9**	**6.2**
August	Obs	10.6	8.5	13.1	14.1	6.4	6.9	10.3	3.0	4.9
	Mod	**8.8**	**9.4**	**9.0**	**7.6**	**6.6**	**7.8**	**8.2**	**14.5**	**6.5**

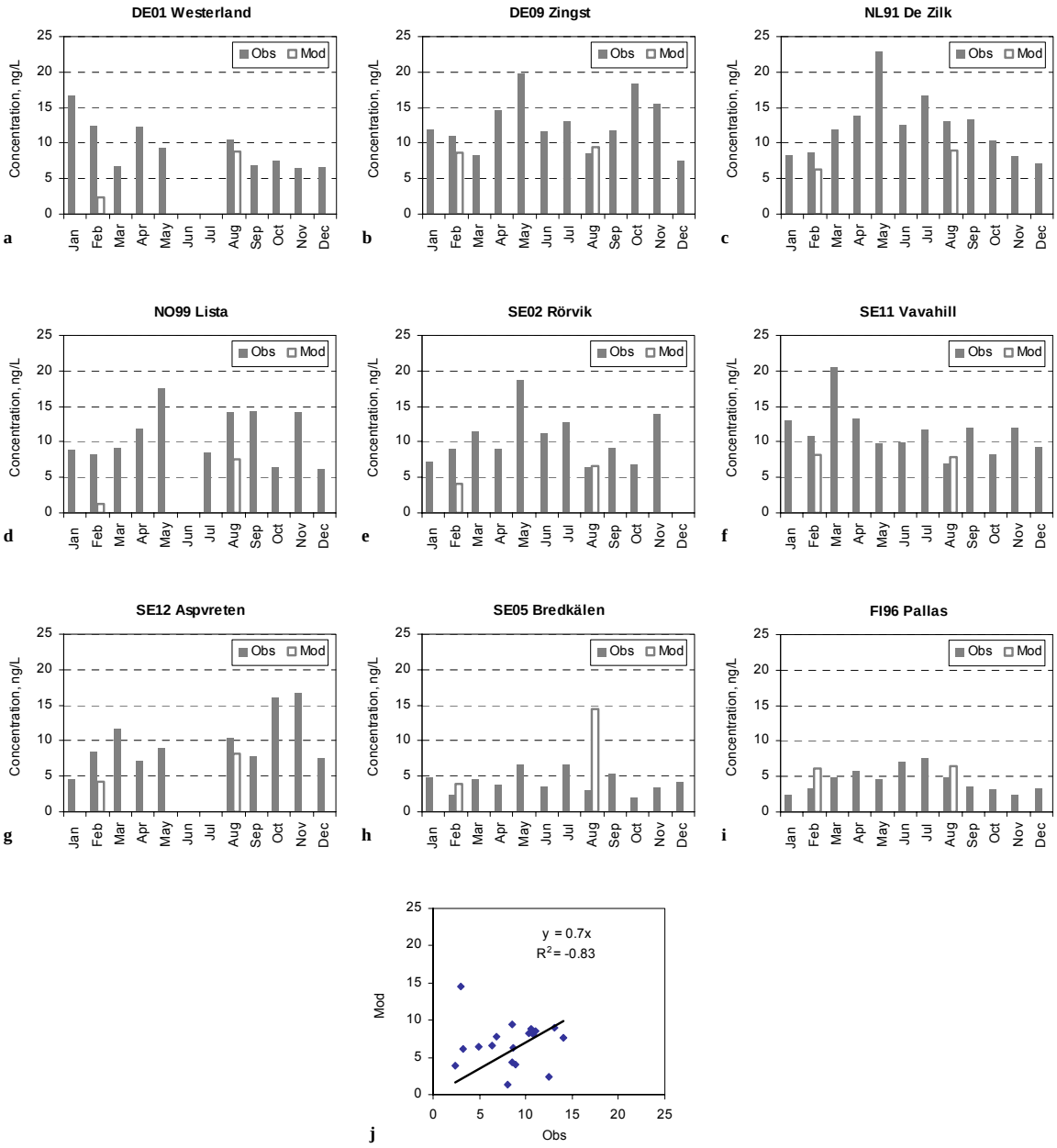


Fig. 7.6.2. *Comparison of measured and calculated by HYSPLIT-Hg concentrations of mercury in precipitation at EMEP sites [model estimates were only made for Feb and Aug]*

Zingst, Westerland and De Zilk are “wet only” stations. The model-estimated precipitation Hg concentrations at these three sites appear to be reasonably consistent with the observations except for the February estimate at Westerland. Pallas, Lista, Rörvik, Bredkälen, Vavihill, and Aspvreten are all “bulk-deposition” monitoring stations. Dry deposition could contribute here, and perhaps it is reasonable to “underestimate” the observed “wet” deposition at these stations. For example, the model estimated February wet flux for the Rörvik and Vavihill bulk monitoring sites was significantly underestimated. If the estimated dry deposition is included in the deposition estimates at these sites, the modelled deposition is much closer to the observed bulk deposition. A comparable occurrence

existed for the August deposition flux at Lista. This was not a constant occurrence at all the bulk sites, however, so it is not clear how much credence to give to this finding. For example, in some cases, if the dry deposition flux were to be included at all the bulk deposition sites, the model-estimated deposition would be even further above the existing overestimate based on wet flux alone. There are, of course, many sources of uncertainty in this model evaluation (emissions, meteorology, measurements) apart from the model itself. Therefore, with the limited number of measurements and model simulations, it is generally difficult to determine the precise reasons for differences between modelled and measured values.

Fig. 6.6.3 demonstrates that the measured and forecasted values of monthly precipitation amounts are obviously correlated (CC = 0.73), however, the forecasted values are in general somewhat (10%) underestimated. The highest deviation is 2.4 times (Aspvreten in February). Nevertheless, about 89% of the results are within a factor of 2. The uncertainty in precipitation amount will influence the accuracy of both the modelled Hg concentration and the total Hg wet deposition.

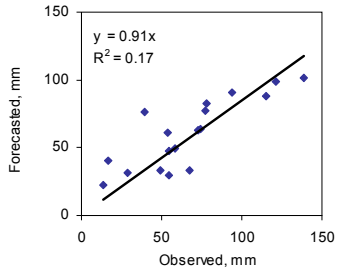


Fig. 7.6.3. *Monthly mean forecasted precipitation amounts used by HYSPLIT-Hg against observed precipitation amounts measured at the monitoring stations, mm*

Table 7.6.3 and Fig. 7.6.4 presents the mercury wet deposition fluxes at the stations obtained on the basis of the observations and as simulated by HYSPLIT. Correlation between the observed and modelled values is rather high (CC = 0.56). However, like other models, variability of the data is high: maximum deviation for an individual pair is about 8 times (NO99 in February). 66% of the results lie within a factor of 2. On average, the model tends to underestimates the observations (mean relative bias is 29%). One can find in the data that the bulk of the statistical underestimation is largely due to 3 out of the 18 individual data pairs – the February and August depositions at NO99 and the February deposition at DE01. The February values have a tendency to be underestimated (4 out of the 9 individual comparisons show an underestimate of a factor of about 2 or greater). In contrast the August values are much closer – all of the model estimates are within a factor of two of the measurements. This is an analogous pattern for the data regarding mercury concentrations in precipitation.

Table 7.6.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by HYSPLIT-Hg (Mod), g/km^2*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14	0.12	0.05
	Mod	**0.15**	**0.27**	**0.30**	**0.12**	**0.20**	**0.27**	**0.17**	**0.13**	**0.14**
August	Obs	0.82	0.67	1.23	1.03	0.78	0.96	0.40	0.16	0.36
	Mod	**0.67**	**0.77**	**0.82**	**0.48**	**0.65**	**0.79**	**0.62**	**0.43**	**0.41**

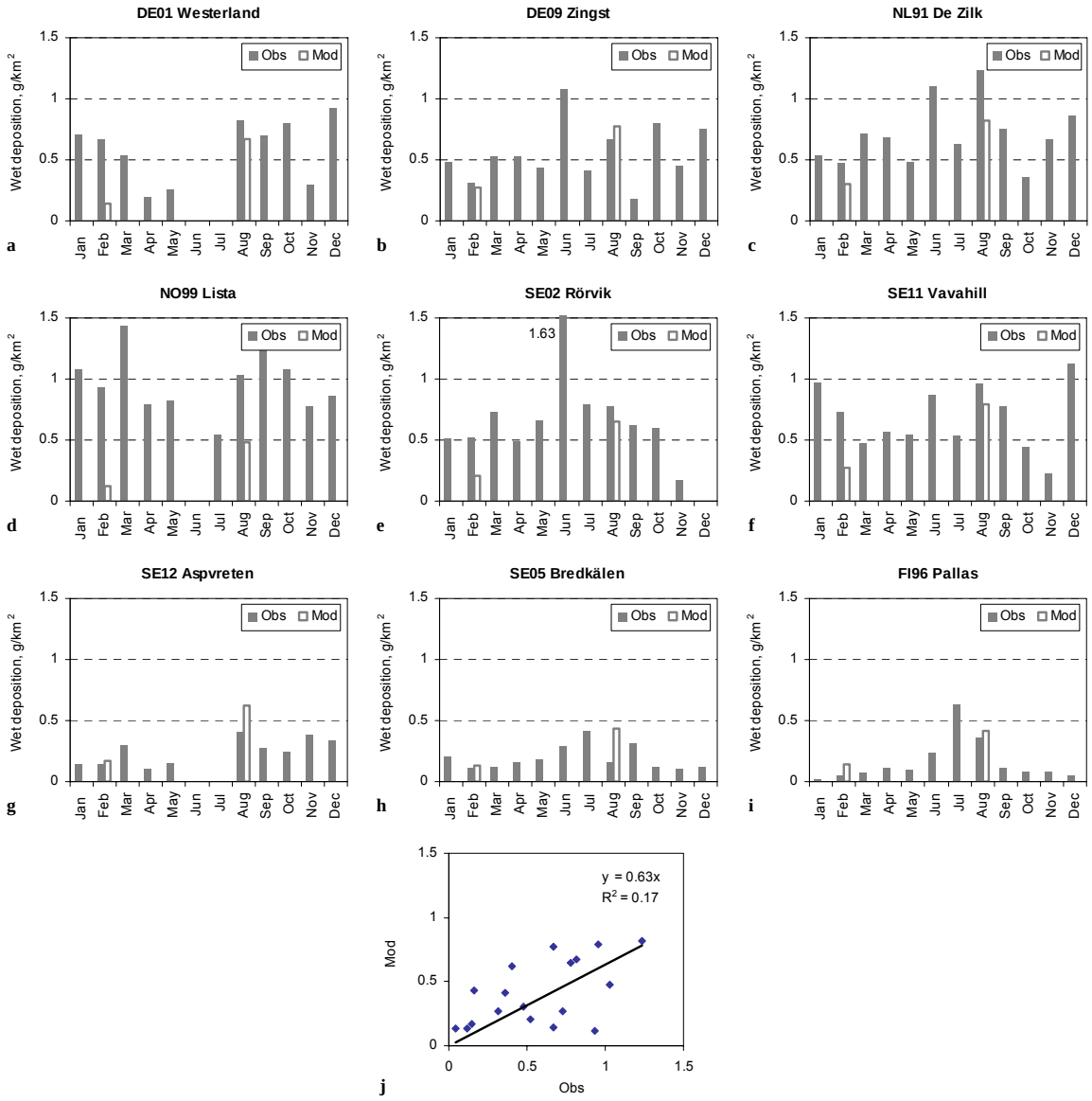


Fig. 7.6.4. *Comparison of mercury monthly mean wet deposition values obtained on the basis of observations and calculated by HYSPLIT-Hg for locations of EMEP stations [model estimates were only made for Feb and Aug]*

It is useful to examine the data in more detail. For example, consider the August wet deposition at SE05 and SE12, the sample representing the most significant August HYSPLIT-Hg overestimates. It can be noted that all of the models that simulated these stations for August overestimated the deposition significantly, comparable to the HYSPLIT overestimations. This suggests that the discrepancies may have been due to errors in emissions and/or meteorology. It is derived from the data of Table 7.6.3.

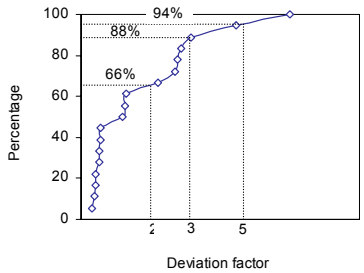


Fig. 7.6.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by HYSPLIT-Hg (9 stations, 2 months)*

and Fig. 7.6.4., that the highest deviation between the observed and modelled results for wet deposition reaches a factor of 8. Cumulative distribution of the deviation factors is shown in Fig. 7.6.5. The curve demonstrates that 66% of all results are within a factor of 2; 88% are within a factor of 3 and 94% are within a factor of 5. Hence, the probability of obtaining an acceptable result (within a factor of 2) is high enough and probability of total failure is rather low.

In summarising the HYSPLIT-Hg mercury concentration and wet deposition results, one can say that the model underestimates the observations by a factor of about 20-30%, on average. The order of magnitude of this discrepancy is certainly no greater than the uncertainty in the meteorology, emissions, and measurements used in the model evaluation. The overall tendency towards underestimation in this exercise may be due to the many of the same reasons that the GEM concentrations were underestimated, e.g., only a one-month spin-up was used and global re-emissions were not included. These factors were thought to be so significant for GEM that a "background" concentration was incorporated. No such factor was included for the wet deposition estimates, and the underestimation tendency may have partly resulted from this.

7.7. Danish Eulerian Hemispheric Model (DEHM)

DEHM simulated concentrations and depositions at all station locations during all months of 1999. The measured and calculated concentrations of gaseous elemental mercury (GEM) are presented in Table 7.7.1 and Fig. 7.7.1 (a-f). At all stations the model underestimates the observations. On the annual level such underestimation reaches 33% (Mace Head station). Very probably that the underestimation is connected with the accepted value of mercury background concentration at the domain borders. The highest deviation of a modelled mean monthly value from a measured one is 1.74 times (station IE31 Mace Head in December). The scattering of the results is high – only 55% of the results are within the factor of 1.2. The correlation between the observed and modelled results for 3 stations is rather high. At the same time for two stations there is no correlation.

Table 7.7.1. *Monthly mean observed (Obs) and modelled (Mod) by DEHM concentrations of GEM at the monitoring stations in 1999, ng/m^3*

Month	Obs/Mod	Station				
		NO99	SE02	FI96	NO42	IE31
January	Obs	1.8	1.35	1.5	2.13	1.83
	Mod	**1.50**	**1.54**	**1.48**	**1.47**	**1.45**
February	Obs	1.7	1.33	1.60	ol	1.70
	Mod	**1.46**	**1.48**	**1.48**	**1.44**	**1.42**
March	Obs	2.1	1.43	1.85	1.83	1.85
	Mod	**1.47**	**1.49**	**1.43**	**1.40**	**1.41**
April	Obs	1.9	1.47	1.60	ol	1.64
	Mod	**1.41**	**1.42**	**1.37**	**1.35**	**1.37**
May	Obs	1.6	nd	1.40	1.60	1.60
	Mod	**1.34**	**1.34**	**1.32**	**1.31**	**1.33**
June	Obs	nd	1.35	1.30	nd	1.69
	Mod	**1.33**	**1.34**	**1.30**	**1.28**	**1.29**
July	Obs	nd	1.35	1.23	nd	1.57
	Mod	**1.29**	**1.30**	**1.27**	**1.26**	**1.28**
August	Obs	nd	1.44	1.33	nd	1.51
	Mod	**1.27**	**1.27**	**1.24**	**1.23**	**1.25**
September	Obs	ol	1.62	1.10	1.75	1.54
	Mod	**1.29**	**1.30**	**1.23**	**1.22**	**1.24**
October	Obs	1.5	1.30	1.30	1.20	1.90
	Mod	**1.25**	**1.26**	**1.21**	**1.19**	**1.24**
November	Obs	1.8	1.48	1.38	nd	1.96
	Mod	**1.25**	**1.28**	**1.23**	**1.19**	**1.20**
December	Obs	1.7	1.28	1.50	1.65	2.10
	Mod	**1.25**	**1.27**	**1.24**	**1.24**	**1.21**
Year	Obs	**1.76***	**1.40***	**1.42**	**1.69***	**1.74**
	Mod	**1.34**	**1.36**	**1.32**	**1.30**	**1.31**
Correlation Coefficient		**0.56**	**-0.07**	**0.72**	**0.83**	**-0.15**

nd – no data; ol – outlier; * - corrected for 12 months

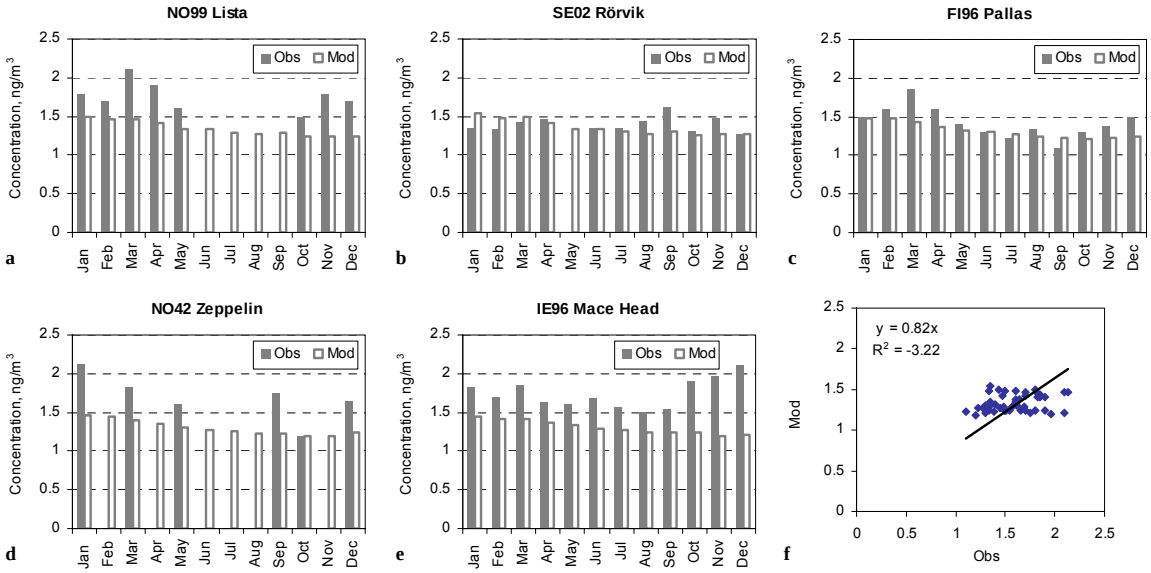


Fig. 7.7.1. *Comparison of measured and calculated by DEHM concentrations of gaseous elemental mercury (GEM) at EMEP sites*

Comparison of measured and calculated mercury concentrations in precipitation on monthly basis is presented in Table 7.7.2 and Fig. 7.7.2. One can see that the model strongly (more than 2 times) overestimates the observed values. Generally, variability of the data is rather high, maximum deviation of the modelled value from the observed one reaches 5.7 times (at FI96 in January). High scattering and overestimation by the model lead to the fact that only 37% of all the results are within the factor of 2. The modelled and the observed values for individual stations are generally slightly correlated (up to CC = 0.67).

Table 7.7.2. *Measured concentrations of Hg in precipitation at EMEP stations in 1999 (Obs) and modelled by DEHM (Mod), ng/L*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	16.7	12.0	8.4	8.8	7.3	13.0	4.5	4.8	2.4
	Mod	**19.4**	**31.9**	**25.7**	**16.2**	**22.4**	**25.8**	**23.6**	**11.2**	**13.6**
February	Obs	12.5	11.0	8.7	8.1	8.9	10.8	8.5	2.4	3.3
	Mod	**16.3**	**26.0**	**27.5**	**12.7**	**20.9**	**24.5**	**17.5**	**7.6**	**11.9**
March	Obs	6.7	8.3	11.9	9.1	11.4	20.5	11.7	4.5	4.9
	Mod	**23.6**	**33.2**	**28.4**	**16.1**	**28.4**	**32.8**	**29.4**	**19.3**	**21.8**
April	Obs	12.2	14.7	13.9	11.8	9.0	13.2	7.2	3.8	5.8
	Mod	**32.4**	**25.2**	**37.7**	**17.2**	**14.3**	**16.4**	**13.4**	**7.4**	**23.6**
May	Obs	9.2	19.8	23.0	17.5	18.7	9.7	9.0	6.6	4.5
	Mod	**36.2**	**27.9**	**35.2**	**18.0**	**19.4**	**22.8**	**22.8**	**6.4**	**21.4**
June	Obs	ol	11.7	12.6	nd	11.1	9.9	nd	3.6	7.1
	Mod	**23.8**	**31.1**	**34.8**	**23.9**	**20.8**	**24.0**	**18.0**	**11.2**	**12.4**
July	Obs	ol	13.0	16.6	8.5	12.7	11.6	ol	6.6	7.6
	Mod	**19.7**	**24.7**	**53.4**	**13.5**	**33.9**	**22.4**	**29.0**	**14.7**	**8.9**
August	Obs	10.6	8.5	13.1	14.1	6.4	6.9	10.3	3.0	4.9
	Mod	**24.9**	**37.6**	**25.9**	**21.9**	**23.9**	**22.5**	**22.7**	**11.7**	**8.9**
September	Obs	6.9	11.8	13.4	14.3	9.1	12.0	7.8	5.4	3.6
	Mod	**20.6**	**40.0**	**33.4**	**27.7**	**17.4**	**30.7**	**21.1**	**24.4**	**18.0**
October	Obs	7.5	18.4	10.3	6.5	6.8	8.2	16.1	2.0	3.2
	Mod	**19.3**	**32.3**	**27.7**	**18.3**	**30.5**	**36.5**	**20.7**	**10.7**	**9.4**
November	Obs	6.4	15.6	8.1	14.1	13.8	11.9	16.6	3.4	2.3
	Mod	**25.4**	**42.9**	**22.3**	**14.7**	**28.0**	**37.2**	**27.4**	**8.6**	**7.6**
December	Obs	6.6	7.6	7.1	6.1	nd	9.3	7.6	4.3	3.3
	Mod	**13.1**	**19.2**	**18.4**	**6.8**	**10.2**	**15.6**	**7.5**	**6.7**	**12.7**
Corr. Coefficient		**0.05**	**0.10**	**0.67**	**0.60**	**0.04**	**0.25**	**0.43**	**0.29**	**0.14**

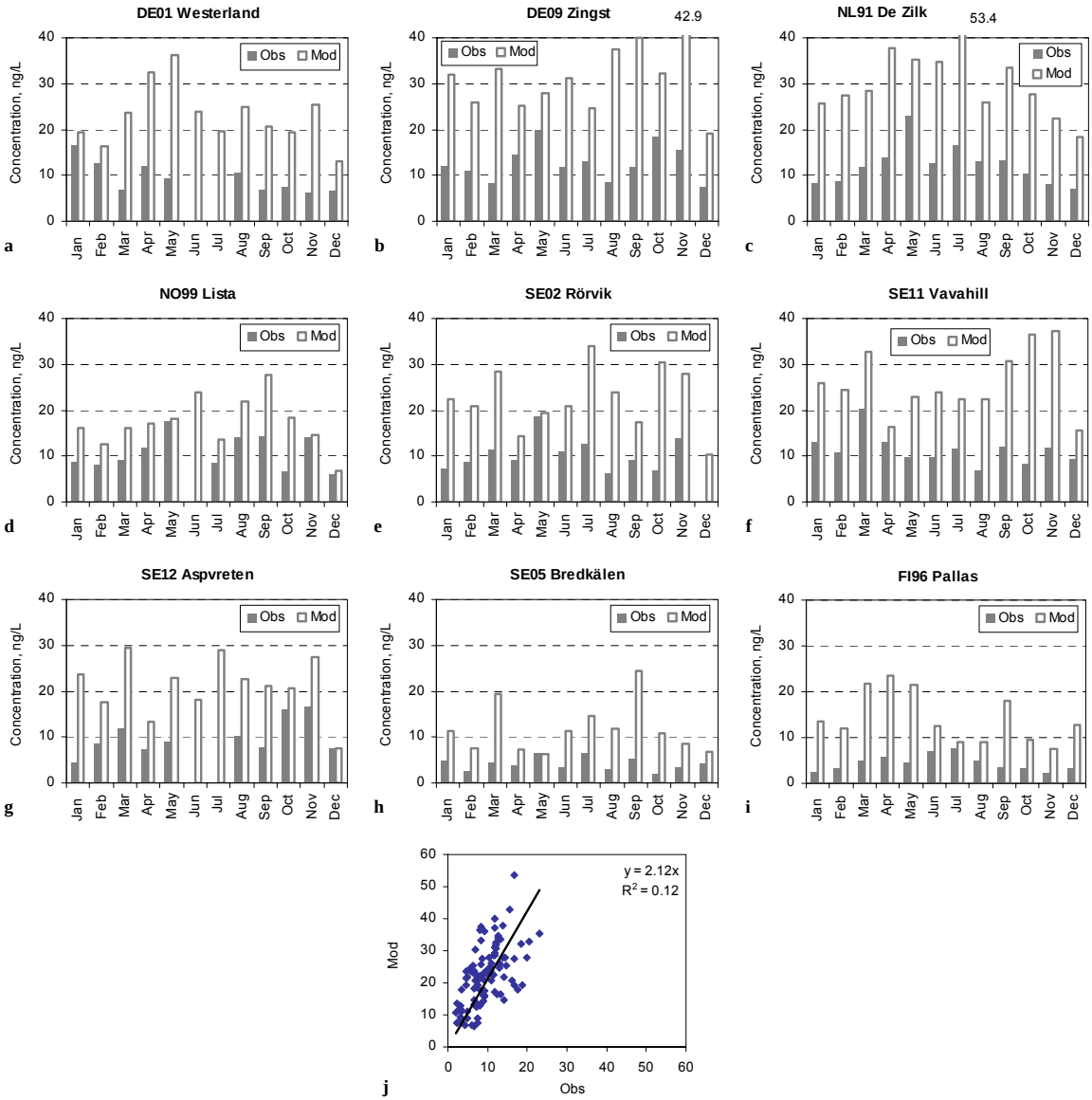


Fig. 7.7.2. *Comparison of measured and calculated by DEHM concentrations of mercury in precipitation at EMEP sites*

Fig. 7.7.3 demonstrates that the measured and forecasted values of monthly precipitation amounts are obviously correlated (CC = 0.73), however, the scattering of the data is rather high. Only 69% of the results are within the factor of 2. The forecasted values are noticeably lower (22% in general) than the observed ones. Approximation of the "modelling-observation" dependence by straight line is rather reliable.

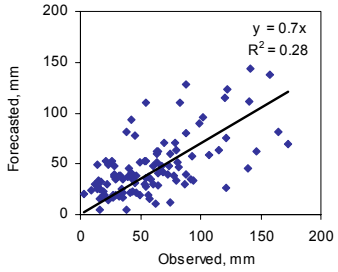


Fig. 7.7.3. *Monthly mean forecasted precipitation amounts used by DEHM against observed precipitation amounts measured at the monitoring stations, mm*

Table 7.7.3 and Fig. 7.7.4 presents the mercury wet deposition fluxes at the stations obtained on the basis of the observations and by DEHM. The model obviously overestimates the observations (relative bias is 74%). It should be noted that the concentrations in precipitation are overestimated much higher but this effect for the depositions is compensated by underestimation of precipitation amount. Correlation between the observed and modelled values is significant practically for all the stations (up to 0.79), however the scattering of the results is high. About half of all the results lie outside the factor of 2. One can pay attention to the fact that for northern "background" stations the correlation is very significant but the model overestimates the observations 2-3 times.

Table 7.7.3. *Measured Hg wet deposition at EMEP stations in 1999 (Obs) and modelled by DEHM (Mod), g/km^2/month (or g/km^2/y)*

Month	Obs/ Mod	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	Obs	0.71	0.48	0.54	1.08	0.51	0.97	0.14	0.21	0.02
	Mod	**1.80**	**1.44**	**1.58**	**1.99**	**1.58**	**1.55**	**0.84**	**0.39**	**0.33**
February	Obs	0.67	0.31	0.47	0.93	0.52	0.73	0.14	0.12	0.05
	Mod	**0.85**	**1.25**	**1.00**	**0.80**	**0.72**	**1.17**	**0.57**	**0.39**	**0.33**
March	Obs	0.53	0.53	0.72	1.44	0.72	0.47	0.30	0.13	0.08
	Mod	**1.49**	**1.70**	**1.35**	**2.20**	**1.70**	**1.63**	**1.55**	**0.71**	**0.54**
April	Obs	0.20	0.52	0.68	0.79	0.49	0.56	0.10	0.16	0.11
	Mod	**0.51**	**0.54**	**0.73**	**0.75**	**0.75**	**0.67**	**0.66**	**0.31**	**0.44**
May	Obs	0.25	0.44	0.48	0.82	0.66	0.55	0.15	0.18	0.09
	Mod	**0.70**	**0.84**	**1.83**	**0.69**	**0.32**	**0.45**	**0.12**	**0.11**	**0.40**
June	Obs	-	1.08	1.11	-	1.63	0.87	-	0.29	0.23
	Mod	**0.80**	**1.07**	**1.39**	**1.67**	**1.29**	**1.93**	**0.22**	**0.57**	**0.48**
July	Obs	-	0.42	0.64	0.55	0.80	0.53	-	0.42	0.63
	Mod	**0.43**	**0.94**	**0.25**	**0.55**	**0.38**	**0.49**	**0.59**	**0.91**	**0.97**
August	Obs	0.82	0.67	1.23	1.03	0.78	0.96	0.40	0.16	0.36
	Mod	**0.94**	**2.64**	**0.86**	**0.86**	**0.64**	**1.03**	**0.85**	**0.32**	**0.42**
September	Obs	0.70	0.18	0.75	1.33	0.62	0.77	0.27	0.31	0.12
	Mod	**1.96**	**1.32**	**1.23**	**1.59**	**0.73**	**0.90**	**0.53**	**0.37**	**0.32**
October	Obs	0.80	0.80	0.36	1.07	0.59	0.44	0.25	0.12	0.09
	Mod	**1.14**	**0.75**	**0.96**	**1.50**	**0.93**	**1.11**	**0.54**	**0.32**	**0.36**
November	Obs	0.29	0.45	0.67	0.77	0.17	0.23	0.39	0.10	0.09
	Mod	**0.78**	**0.80**	**1.04**	**1.62**	**0.83**	**0.83**	**0.38**	**0.29**	**0.62**
December	Obs	0.93	0.75	0.86	0.86	-	1.12	0.34	0.12	0.05
	Mod	**1.45**	**1.73**	**1.40**	**0.97**	**1.31**	**1.79**	**0.59**	**0.27**	**0.32**
Year	**Obs**	**7.1***	**6.6**	**8.5**	**11.6***	**8.2***	**8.2**	**3.0***	**2.3**	**1.9**
	Mod	**12.9**	**15.0**	**13.6**	**15.2**	**11.2**	**13.6**	**7.4**	**5.0**	**5.5**
Corr. Coefficient		**0.60**	**0.09**	**-0.05**	**0.75**	**0.19**	**0.54**	**0.20**	**0.61**	**0.79**

* - corrected for 12 months

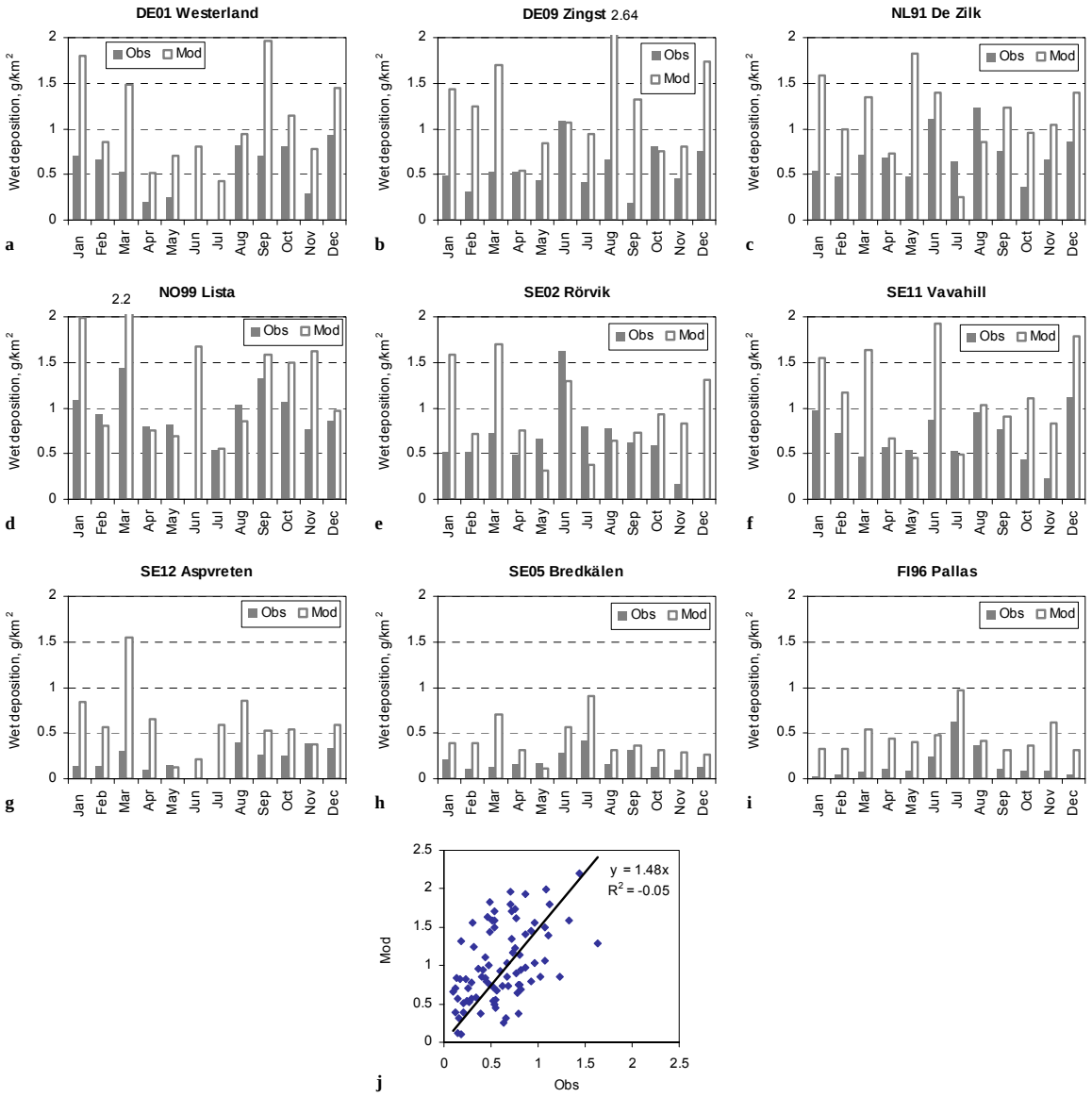


Fig. 7.7.4. *Comparison of mercury monthly mean wet deposition values obtained on the basis of observations and calculated by DEHM for locations of EMEP stations*

It is derived from the data of Table 7.7.3 and Fig. 7.7.4, that the highest deviation between the observed and modelled results for wet deposition reaches a factor of 15 times (FI96 in January). Cumulative distribution of the deviation factors is shown in Fig. 7.7.5. The curve demonstrates that 52% of all results are within a factor of 2; 77% - within a factor of 3 and 91% - within a factor of 5. Hence, probability to obtain an acceptable result (within a factor of 2) is high enough and probability of total failure is rather low.

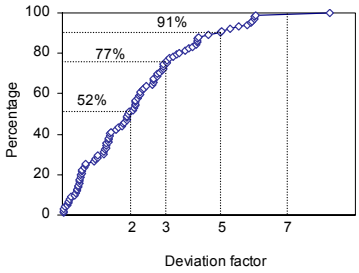


Fig. 7.7.5. *Cumulative distribution of deviation factors for the totality of wet deposition results obtained by DEHM (9 stations, 12 months)*

Fig. 7.7.6 shows comparison of annual wet deposition values at the stations. In this figure the monitoring stations are arranged in accordance with latitudes: from the most southern station NL91 (52°N) to the most northern one FI96 (68°N). The modelled and observed values reveal the same spatial decreasing trend of mercury wet deposition from south to north but the modelling results are about two times higher. Nevertheless, correlation between the observed and modelled annual wet deposition values for all the stations is very high (r = 0.90).

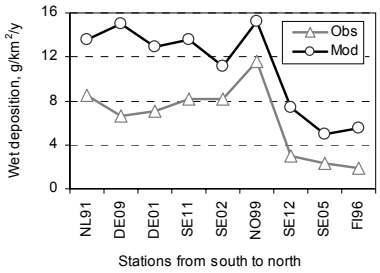


Fig. 7.7.6. *Latitudinal trends of modelled by DEHM and observed annual wet depositions*

8. MODEL-TO-MODEL COMPARISON

This intercomparison study joined most of known models of mercury atmospheric transport and depositions on regional and global levels. Besides, they cover all modelling approaches accepted within the scientific community. Comparison of results obtained by the participating models gives a possibility to get general concept on degree of reliability of modern mercury modelling.

There are three main types of parameters, which can characterize ability of a model to simulate mercury atmospheric transport and mercury atmospheric loads. The first type is atmospheric concentration, e.g., of gaseous elemental mercury. This type of parameter can show how this or that model can describe regional and global transport of mercury in the atmosphere, how they can take into account main sources and sinks of atmospheric mercury. The second kind of parameter is atmospheric mercury deposition. This allows an evaluation against the loading pathways through which mercury contamination is introduced to the biosphere. Deposition occurs by two mechanisms: deposition with precipitation (wet deposition) and uptake of mercury species by underlying surface (dry deposition). Dry deposition is a parameter, which is not measured on a routine basis and can be evaluated only by models. The third type of parameter is source-receptor relationships. In this comparison, all three types of parameters have been considered.

Not all the modelling groups had possibilities to simulate all months of 1999. Only February is a month, which was calculated by all the models. Six of the models simulated August. Four models of seven modelled mercury transport and deposition for all the months. The comparison below will be done separately for February, August and for the whole year.

8.1. Modelled GEM

Comparison of modelled values of GEM for five locations (monitoring stations) in February 1999 is shown in Table 8.1.1. Mean modelled value and observed value for each station are also presented. The modelled value, which is the closest one to the observed value, is highlighted in bold. Twice of five cases MSCE-HM is closer to the observations than the other models. The table demonstrates that mean deviation of modelled values is on the level of 10%. For three stations of four the mean modelled value agrees with the observed one better than 12%. For station SE02 all the models noticeably (about 30%) underestimated observations in February 1999.

Table 8.1.1. *Modelled values of GEM for five locations (monitoring stations) in February 1999, ng/m^3*

	NO99	SE02	FI96	NO42	IE31
ADOM	1.74	1.67	**1.60**	1.88	1.78
CMAQ	1.53	1.61	-	-	1.53
EMAP	2.07	1.96	2.02	1.93	1.28
MSCE-HM	**1.73**	1.85	1.93	1.72	**1.74**
MSCE-HM-Hem	1.61	1.66	1.67	1.70	1.59
HYSPLIT	1.32	**1.46**	1.39	1.30	1.32
DEHM	1.46	1.48	1.48	1.44	1.42
Mean modelled	**1.64**	**1.67**	**1.68**	**1.66**	**1.52**
Mean deviation of modelled values	**11%**	**8%**	**12%**	**12%**	**10%**
Observed	**1.70**	**1.33**	**1.60**	**-**	**1.70**
Deviation factor for the means	**1.04**	**1.25**	**1.05**	**-**	**1.12**

For August the comparison is shown in Table 8.1.2. The table demonstrates that mean deviation of modelled values is practically the same as for February (about 10%). For station SE02 all the models underestimated observations, as well as in February, however, the underestimation in this case is only 15%.

Table 8.1.2. *Modelled values of GEM for five locations (monitoring stations) in August 1999, ng/m^3*

	NO99	SE02	FI96	NO42	IE31
CMAQ	1.55	1.56			**1.56**
EMAP	1.92	1.95	1.49	1.76	2.21
MSCE-HM	1.72	1.85	1.56	1.47	1.65
MSCE-HM-Hem	1.67	1.72	1.52	1.59	1.64
HYSPLIT	1.43	**1.55**	1.46	1.63	1.59
DEHM	1.27	1.27	**1.24**	1.23	1.25
Mean modelled	**1.59**	**1.65**	**1.45**	**1.54**	**1.65**
Deviation of modelled values	**11%**	**12%**	**6%**	**10%**	**11%**
Observed	**-**	**1.44**	**1.33**	**-**	**1.51**
Deviation factor for the means		**1.15**	**1.09**		**1.09**

The comparison data for the whole year are shown in Table 8.1.3. The amount of data is not sufficient for reliable statistical treatment. Nevertheless, it is obvious (and not surprising) that on annual level the deviation of the modelled values is somewhat lower than for one month. All the modelling results are within about 10%. However, DEHM produces much lower results than the other models. For three stations of five the mean modelled values are close to the observed ones. For SE02 the models strongly (25%) overestimate the observations. In two cases of five MSCE-HM and DEHM demonstrate the closest agreement with the observations.

Table 8.1.3. *Modelled values of GEM for five locations (monitoring stations) mean for 1999, ng/m^3*

	NO99	SE02	FI96	NO42	IE31
EMAP	1.98	1.99	1.76	1.88	1.82
MSCE-HM	**1.83**	1.90	1.79	1.61	**1.76**
MSCE-HM-Hem	1.68	1.72	1.62	**1.64**	1.61
DEHM	1.34	**1.36**	**1.32**	1.30	1.31
Mean modelled	**1.71**	**1.74**	**1.62**	**1.61**	**1.63**
Deviation of modelled values	**11%**	**11%**	**9%**	**9%**	**10%**
Observed	**1.76**	**1.40**	**1.42**	**1.69**	**1.74**
Deviation factor for the means	**1.04**	**1.24**	**1.14**	**1.06**	**1.07**

The reason for the consistent overestimation of GEM at SE02 Rörvik by most of the models is not completely clear. The location of the station does not give any geophysical bases to assume that the concentration values could be lower than at Norwegian station NO99 or Finnish station FI96. In addition to the regular monitoring program carried out at SE02 in 1999, four 2-week campaigns were performed within the EU funded research project Mercury OVer Europe (MOE). These measurements included both automated and manual sampling methods and intercomparison with the regular monitoring. The automated measurements performed by an independent laboratory confirmed the unexpectedly low concentrations at SE02. The overall average for GEM during the 4 campaigns was 1.52 ng/m^3 i.e. slightly higher than the value for the full year but not as high as predicted by the models. One hypothesis to explain the unexpectedly low values obtained for SE02 in the MOE project was that Atlantic coastal sites (Mace Head Ireland, in the case of MOE, where higher concentrations

than at SE02 were found) were affected by re-emissions from the sea surface [*Munthe et al.,* 2004]. Whether this is true and also applicable for e.g. NO99, NO42 and IE31 remains to be determined by further research.

Table 8.1.4 gives an idea of probability distribution of the GEM results obtained by five of seven models (due to the shortness of their simulation periods and problems with the measurement data CMAQ and HYSPLIT yielded too few data pairs for reliable statistical treatment). The table shows that the probability to produce acceptable result (within F1.2) is more than 50% for four of five models. Probability to obtain a false result (>F.1.5) is very small for all the models.

Table 8.1.4. *Number of results and probability (%) to produce GEM results within a given factor of coverage*

Model / Number of results	F1.1	F1.2	F1.5
ADOM (14)	36	64	100
EMAP (49)	22	45	88
MSCE-HM (49)	41	57	96
MSCE-HM-Hem (49)	35	67	100
DEHM (49)	29	55	94

8.2. Modelled mercury wet deposition

The modelled data on mercury wet deposition for different locations (monitoring stations) in February 1999 are presented in Table 8.2.1. The table also includes mean modelled values and observed values for all the stations. The modelled value, which is the closest one to the observed value, is highlighted in bold. Deviations of the values modelled by different models from the mean modelled value are on the level of 40%. The depositions at Arctic stations (SE05 and FI96) are strongly overestimated in February by most models but MSCE-HM and HYSPLIT.

Table 8.2.1. *The modelled data on mercury wet deposition for different locations (monitoring stations) in February 1999, g/km^2*

Model	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
ADOM	0.49	0.46	0.68	0.60	0.66	0.59	0.21	0.51	0.52
CMAQ	0.82	0.79	0.80	1.10	0.42	**0.71**	**0.13**	-	-
EMAP	**0.59**	0.49	**0.49**	**0.85**	**0.46**	0.46	0.39	0.22	0.39
MSCE-HM	0.41	0.40	0.81	0.48	0.28	0.64	0.20	0.06	**0.10**
MSCE-HM-Hem	0.48	0.79	0.68	0.46	0.32	0.50	0.31	0.28	0.20
HYSPLIT	0.15	**0.27**	0.30	0.12	0.20	0.27	0.17	**0.13**	0.14
DEHM	0.85	1.25	1.00	0.80	0.72	1.17	0.57	0.39	0.33
Mean modelled	**0.54**	**0.64**	**0.68**	**0.68**	**0.44**	**0.62**	**0.28**	**0.27**	**0.28**
Mean deviation of modelled values	**34%**	**41%**	**24%**	**39%**	**35%**	**30%**	**43%**	**48%**	**48%**
Observed	**0.67**	**0.31**	**0.47**	**0.93**	**0.52**	**0.73**	**0.14**	**0.12**	**0.05**
Deviation factor for the means	**1.2**	**2.1**	**1.4**	**1.5**	**1.2**	**1.2**	**2.0**	**2.2**	**5.6**

The analogous modelled data on mercury wet deposition in August 1999 are presented in Table 8.2.2. In this case mean deviation of the values modelled by different models from the mean modelled value

is obviously lower than in February (on the level of 30%). In summer period overestimation of the depositions at the Arctic station FI96 is not so pronounced as in February.

Table 8.2.2. *The modelled data on mercury wet deposition for different locations (monitoring stations) in August 1999, g/km^2*

Model	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
CMAQ	1.79	1.42	2.47	1.26	1.41	1.97	1.03	-	-
EMAP	0.71	1.18	**1.18**	**0.86**	1.12	0.76	0.76	0.50	0.48
MSCE-HM	0.73	0.41	1.16	0.79	0.61	0.61	0.91	0.34	0.30
MSCE-HM-Hem	**0.84**	1.24	0.93	0.72	**0.81**	1.06	0.77	0.37	0.60
HYSPLIT	0.67	**0.77**	0.82	0.48	0.65	0.79	**0.62**	0.43	**0.41**
DEHM	0.94	2.64	0.86	**0.86**	0.64	**1.03**	0.85	**0.32**	0.42
Mean modelled	**0.95**	**1.28**	**1.24**	**0.83**	**0.87**	**1.04**	**0.82**	**0.39**	**0.44**
Deviation of modelled values	**30%**	**39%**	**33%**	**20%**	**30%**	**31%**	**13%**	**15%**	**18%**
Observed	**0.82**	**0.67**	**1.23**	**1.03**	**0.78**	**0.96**	**0.40**	**0.16**	**0.36**
Deviation factor for the means	**1.2**	**1.9**	**1.0**	**1.2**	**1.1**	**1.1**	**2.1**	**2.5**	**1.2**

The comparison of wet deposition data for the whole year is presented in Table 8.2.3. Number of the data is small to provide reliable statistical treatment. Comparison of Tables 8.2.1, 8.2.2 and 8.2.3 shows that on annual level the deviation of the modelled values is lower than for one month. The annual modelling results are on the level of 20%. For most stations the modelled values are close enough to the observed ones. However, for the northern stations all the models strongly overestimate the observations. Both for February, August and for the year as a whole EMAP produces the values that are closer to the observations.

Table 8.2.3. *Wet deposition comparison for year 1999, g/km^2*

Model	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
EMAP	**6.7**	8.9	**8.9**	**11.4**	**7.3**	5.7	6.6	6.0	5.8
MSCE-HM	6.2	**5.7**	13.6	8.9	4.9	**6.7**	**5.2**	**2.8**	**3.5**
MSCE-HM-Hem	10.5	11.6	12.0	10.6	9.7	10.1	8.1	5.1	5.8
DEHM	12.9	15.0	13.6	15.2	11.2	13.6	7.4	5.0	5.5
Mean modelled	9.1	10.3	12.0	11.5	8.3	9.0	6.8	4.7	5.2
Deviation of modelled values	**29%**	**29%**	**13%**	**16%**	**14%**	**31%**	**14%**	**20%**	**16%**
Observed	**7.1**	**6.6**	**8.5**	**11.6**	**8.2**	**8.2**	**3.0**	**2.3**	**1.9**
Deviation factor for the means	**1.3**	**1.6**	**1.4**	**1.0**	**1.0**	**1.1**	**2.3**	**2.0**	**2.7**

Table 8.2.4 presents factors of coverage, which gives an idea of probability distribution of the wet deposition results. Numbers of monthly mean results obtained by individual models are different, so reliability of the data on probability distribution is also different. The table shows that the probability distribution is very similar for all models. Probability to produce acceptable result (within F2) is about 2/3 for all of them. Probability to obtain a false result (>F5) is practically negligible for all the models.

Table 8.2.4. *Number of results and probability (%) to produce wet deposition results within a given factor of coverage*

Model	Number of results	Factor of coverage			
		F2	F3	F5	F10
ADOM	31	72	77	93	96
CMAQ	14	64	100	100	100
EMAP	102	66	82	93	100
MSCE-HM	102	71	91	100	100
MSCE-HM-Hem	102	63	78	94	99
HYSPLIT	18	66	88	94	100
DEHM	102	52	77	91	99

8.3. Modelled mercury dry and total deposition

As was mentioned above, mercury dry deposition fluxes cannot be directly measured but can be modelled. Hence, comparison of dry deposition values is possible only between the models. Table 8.3.1 gives an idea about dry deposition fluxes and their relative contributions to total deposition (sum of wet and dry deposition fluxes) calculated by ADOM. For the totality of the stations mean relative contribution of dry deposition to the total deposition amounts to 24%. However, for individual months at separate stations such contribution can reach 47%. During wintertime role of dry deposition is somewhat lower (22%) than during summer (27%).

Table 8.3.1. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by ADOM*

Month	Stations								
	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
February	0.11 (18)	0.09 (16)	0.10 (13)	0.36 (38)	0.26 (28)	0.05 (8)	0.03 (13)	0.33 (39)	0.36 (41)
March	0.18 (13)	0.11 (13)	0.16 (15)	0.38 (30)	0.23 (19)	0.10 (11)	0.03 (5)	0.35 (34)	0.39 (36)
June	0.27 (25)	0.24 (21)	0.26 (29)	0.62 (26)	0.56 (27)	0.14 (10)	0.03 (6)	0.35 (29)	0.39 (37)
July	0.30 (32)	0.27 (34)	0.32 (47)	0.98 (37)	0.75 (32)	0.13 (12)	0.04 (13)	0.77 (38)	0.41 (26)

Dry depositions and their contributions into total depositions for locations of the monitoring stations estimated by CMAQ are presented in Table 8.3.2. CMAQ predicts that dry deposition intensity is somewhat higher in summer. Relative contribution of the process to the total deposition varies as usual within 20-40%. Very high values of the contribution are connected with very low precipitation amount during a given month and as a result - with very low contribution of wet deposition.

Table 8.3.2. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by CMAQ*

Month	Station						
	DE01	DE09	NL91	NO99	SE02	SE11	SE12
February	0.24 (23)	0.37 (32)	0.56 (41)	0.25 (18)	0.27 (40)	0.60 (46)	0.28 (69)
August	0.29 (14)	0.41 (22)	0.68 (22)	0.32 (20)	0.28 (17)	0.71 (27)	0.48 (32)

Table 8.3.3 gives an idea about relative importance of dry deposition fluxes calculated by EMAP. For the totality of the stations mean relative contribution of dry deposition to the total deposition amounts for 21%. It should be noted that the model reveals a spatial trend of dry deposition intensity: the values for the "polluted" stations are about 2-3 times higher than for the "background" ones. It is impossible to find any noticeable seasonal trend of dry deposition intensities obtained by EMAP.

Table 8.3.3. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by EMAP*

Month	Station								
	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	0.15 (21)	0.20 (36)	0.21 (27)	0.17 (16)	0.24 (37)	0.18 (38)	0.16 (21)	0.10 (25)	0.09 (32)
February	0.11 (16)	0.25 (40)	0.13 (21)	0.17 (17)	0.17 (27)	0.18 (29)	0.11 (22)	0.09 (28)	0.14 (26)
March	0.17 (19)	0.22 (26)	0.24 (25)	0.22 (22)	0.40 (35)	0.27 (33)	0.14 (23)	0.13 (21)	0.17 (36)
April	0.15 (25)	0.15 (20)	0.25 (21)	0.14 (29)	0.16 (25)	0.15 (24)	0.13 (13)	0.11 (11)	0.12 (33)
May	0.13 (22)	0.14 (15)	0.23 (23)	0.05 (13)	0.08 (18)	0.11 (20)	0.03 (14)	0.04 (14)	0.07 (9)
June	0.12 (18)	0.10 (16)	0.17 (13)	0.21 (18)	0.13 (18)	0.11 (19)	0.04 (20)	0.05 (7)	0.11 (13)
July	0.09 (17)	0.12 (21)	0.12 (17)	0.05 (9)	0.12 (21)	0.11 (24)	0.07 (16)	0.08 (7)	0.12 (8)
August	0.16 (18)	0.18 (28)	0.29 (20)	0.14 (14)	0.18 (14)	0.16 (17)	0.09 (10)	0.02 (4)	0.07 (13)
September	0.31 (34)	0.25 (43)	0.34 (26)	0.24 (18)	0.20 (29)	0.19 (40)	0.05 (10)	0.06 (11)	0.03 (12)
October	0.17 (33)	0.22 (46)	0.32 (43)	0.21 (13)	0.41 (30)	0.27 (41)	0.14 (13)	0.09 (17)	0.15 (31)
November	0.14 (23)	0.15 (41)	0.16 (36)	0.12 (11)	0.16 (36)	0.13 (23)	0.09 (18)	0.09 (27)	0.13 (21)
December	0.16 (16)	0.24 (32)	0.21 (19)	0.26 (9)	0.29 (22)	0.28 (24)	0.16 (14)	0.08 (16)	0.07 (17)
Year	**1.85 (22)**	**2.22 (29)**	**2.68 (23)**	**1.98 (15)**	**2.56 (26)**	**2.13 (27)**	**1.22 (16)**	**0.95 (14)**	**1.27 (18)**

Table 8.3.4 presents the dry deposition data calculated by MSCE-HM. Within the year it is possible to determine a seasonal cycle of dry deposition values – they are higher in warm period. This fact is especially obvious for the northern stations. Such a cycle can be explained by role of snow cover because it is assumed in the model that snow at negative temperatures does not uptake elemental mercury. On the annual basis the dry deposition process accounts for 20-35% of the total mercury deposition in the region.

Table 8.3.4. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by MSCE-HM*

Month	Station								
	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	0.16 (22)	0.20 (33)	0.30 (22)	0.07 (10)	0.14 (29)	0.34 (45)	0.11 (21)	0.01 (13)	0.02 (17)
February	0.10 (20)	0.13 (25)	0.20 (20)	0.05 (9)	0.07 (20)	0.19 (23)	0.06 (23)	0.02 (25)	0.02 (17)
March	0.26 (37)	0.19 (23)	0.31 (25)	0.09 (10)	0.12 (29)	0.22 (35)	0.11 (28)	0.04 (14)	0.05 (28)
April	0.17 (26)	0.14 (20)	0.14 (8)	0.13 (65)	0.14 (30)	0.24 (35)	0.15 (38)	0.07 (18)	0.07 (33)
May	0.23 (33)	0.23 (23)	0.23 (19)	0.15 (29)	0.17 (37)	0.37 (49)	0.24 (53)	0.18 (64)	0.16 (31)
June	0.14 (23)	0.21 (25)	0.15 (10)	0.11 (18)	0.11 (17)	0.34 (33)	0.30 (56)	0.31 (47)	0.34 (34)
July	0.19 (25)	0.24 (35)	0.21 (16)	0.16 (25)	0.16 (26)	0.38 (49)	0.28 (50)	0.31 (37)	0.29 (25)
August	0.18 (20)	0.24 (37)	0.28 (19)	0.19 (19)	0.14 (19)	0.35 (36)	0.29 (24)	0.31 (48)	0.25 (45)
September	0.41 (45)	0.30 (52)	0.30 (16)	0.24 (23)	0.20 (36)	0.44 (54)	0.29 (40)	0.25 (37)	0.21 (53)
October	0.26 (46)	0.20 (37)	0.35 (28)	0.19 (13)	0.13 (19)	0.34 (43)	0.18 (19)	0.09 (36)	0.05 (19)
November	0.19 (27)	0.20 (47)	0.30 (29)	0.13 (12)	0.18 (37)	0.37 (28)	0.16 (20)	0.06 (46)	0.02 (7)
December	0.07 (9)	0.15 (21)	0.15 (11)	0.06 (5)	0.08 (12)	0.24 (21)	0.09 (14)	0.03 (19)	0.02 (10)
Year	**2.36 (28)**	**2.43 (30)**	**2.92 (18)**	**1.57 (16)**	**1.64 (25)**	**3.82 (36)**	**2.26 (30)**	**1.68 (38)**	**1.50 (30)**

Table 8.3.5 shows relative importance of dry deposition fluxes calculated by MSCE-HM-Hem. The model predicts a seasonal cycle of dry deposition values (maximum in summer) especially obvious for the northern stations. On the annual basis the dry deposition process accounts for 15-30% of the total mercury deposition in the region.

Table 8.3.5. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by MSCE-HM-Hem*

Month	Station								
	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	0.29 (24)	0.34 (25)	0.23 (16)	0.07 (7)	0.23 (23)	0.27 (24)	0.11 (15)	0.09 (21)	0.07 (21)
February	0.17 (26)	0.23 (22)	0.17 (20)	0.06 (11)	0.20 (38)	0.20 (28)	0.09 (23)	0.10 (26)	0.07 (24)
March	0.30 (20)	0.37 (24)	0.35 (21)	0.12 (9)	0.30 (25)	0.28 (22)	0.14 (14)	0.13 (21)	0.12 (25)
April	0.28 (29)	0.40 (36)	0.42 (35)	0.14 (18)	0.29 (32)	0.27 (30)	0.11 (13)	0.12 (25)	0.12 (21)
May	0.41 (32)	0.57 (41)	0.52 (33)	0.17 (19)	0.42 (36)	0.41 (36)	0.18 (29)	0.23 (47)	0.14 (28)
June	0.29 (23)	0.49 (31)	0.38 (27)	0.12 (11)	0.42 (27)	0.40 (27)	0.21 (22)	0.31 (34)	0.28 (33)
July	0.40 (40)	0.53 (36)	0.58 (50)	0.17 (21)	0.47 (40)	0.40 (33)	0.18 (25)	0.37 (36)	0.30 (25)
August	0.42 (33)	0.50 (29)	0.53 (37)	0.19 (21)	0.43 (35)	0.38 (26)	0.16 (17)	0.28 (43)	0.18 (23)
September	0.46 (26)	0.66 (42)	0.46 (27)	0.25 (13)	0.64 (30)	0.58 (34)	0.22 (19)	0.34 (40)	0.20 (36)
October	0.26 (19)	0.33 (26)	0.46 (31)	0.13 (10)	0.29 (26)	0.27 (24)	0.12 (12)	0.16 (26)	0.12 (16)
November	0.24 (34)	0.30 (29)	0.30 (26)	0.10 (9)	0.33 (35)	0.28 (30)	0.18 (21)	0.19 (26)	0.12 (14)
December	0.20 (15)	0.24 (16)	0.14 (10)	0.05 (7)	0.18 (20)	0.20 (18)	0.09 (13)	0.09 (28)	0.07 (20)
Year	**3.7 (26)**	**5.9 (30)**	**4.5 (24)**	**1.6 (27)**	**4.2 (13)**	**3.9 (30)**	**1.8. (32)**	**2.4 (28)**	**1.8 (18)**

Table 8.3.6 gives an idea about relative importance of dry deposition fluxes calculated by HYSPLIT-Hg. The model predicts that absolute values of dry deposition are generally higher in summer, while relative contribution of dry deposition is higher in winter. The dry deposition process accounts for 20-

50% of the total mercury deposition in the region. It should be mentioned that no background was added for the wet and dry deposition simulation results for the monitoring stations, as the deposition is largely due to RGM and TPM and the limitations described earlier (e.g. only a one month spin up) are not believed to be as significant for these forms of atmospheric mercury.

Table 8.3.6. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by HYSPLIT-Hg*

Month	Station								
	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
February	0.12 (45)	0.23 (47)	0.38 (56)	0.07 (38)	0.16 (44)	0.30 (52)	0.13 (42)	0.08 (39)	0.10 (41)
August	0.38 (36)	0.46 (37)	0.56 (41)	0.17 (27)	0.31 (33)	0.49 (39)	0.25 (28)	0.12 (22)	0.09 (18)

Table 8.3.7 shows relative importance of dry deposition calculated by DEHM. On the annual basis the dry deposition process accounts for 10-25% of the total mercury deposition in the region.

Table 8.3.7. *Mercury dry deposition (g/km^2) and relative dry deposition contribution into the total deposition (in brackets, %) calculated by DEHM*

Month	Station								
	DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
January	0.21 (10)	0.31 (18)	0.33 (17)	0.13 (6)	0.15 (9)	0.33 (18)	0.12 (13)	0.02 (5)	0.03 (8)
February	0.10 (11)	0.19 (13)	0.21 (17)	0.09 (10)	0.09 (11)	0.23 (16)	0.10 (15)	0.02 (5)	0.03 (8)
March	0.20 (12)	0.17 (9)	0.29 (18)	0.15 (6)	0.15 (8)	0.35 (18)	0.13 (8)	0.05 (7)	0.03 (5)
April	0.17 (25)	0.18 (25)	0.33 (31)	0.13 (15)	0.16 (18)	0.42 (39)	0.13 (16)	0.06 (16)	0.03 (6)
May	0.19 (21)	0.21 (20)	0.36 (16)	0.18 (21)	0.20 (38)	0.48 (52)	0.19 (61)	0.24 (69)	0.08 (17)
June	0.15 (16)	0.22 (17)	0.28 (17)	0.13 (7)	0.20 (13)	0.50 (21)	0.20 (48)	0.17 (23)	0.09 (16)
July	0.17 (28)	0.22 (19)	0.34 (58)	0.14 (20)	0.20 (34)	0.46 (48)	0.15 (20)	0.15 (14)	0.06 (6)
August	0.19 (17)	0.26 (9)	0.31 (26)	0.19 (18)	0.14 (18)	0.36 (26)	0.14 (14)	0.14 (30)	0.04 (9)
September	0.22 (10)	0.33 (20)	0.37 (23)	0.13 (8)	0.17 (19)	0.45 (33)	0.12 (18)	0.08 (18)	0.03 (9)
October	0.24 (17)	0.38 (34)	0.39 (29)	0.14 (9)	0.12 (11)	0.35 (24)	0.11 (17)	0.03 (9)	0.01 (3)
November	0.28 (26)	0.33 (29)	0.32 (24)	0.16 (9)	0.15 (15)	0.36 (30)	0.15 (28)	0.03 (9)	0.02 (3)
December	0.14 (9)	0.30 (15)	0.28 (17)	0.12 (11)	0.11 (8)	0.32 (15)	0.10 (14)	0.02 (7)	0.02 (6)
Year	**2.26 (15)**	**3.10 (17)**	**3.81 (22)**	**1.69 (10)**	**1.84 (14)**	**4.61 (25)**	**1.64 (18)**	**1.01 (17)**	**0.47 (8)**

Table 8.3.8 presents ranges of dry deposition contributions into total depositions obtained by all the models in February 1999, August 1999 and during the whole year. To some extent these ranges reflect conditions of dry deposition at different locations over the region of investigation. Another factor, which influences relative contribution of dry deposition, is intensity of wet deposition during the considered period. In spite of the disturbing factors these ranges can give an idea about relative importance of two deposition mechanisms.

One can see from the table that all the models predict that dry deposition contributes roughly 1/5 - 1/3 to total mercury deposition. Hence, in accordance with applied modelling schemes wet deposition scavenging is prevailing process of mercury removal from the atmosphere. It is impossible to reveal any seasonal difference: some models predict higher contributions during wintertime, some – conversely.

Table 8.3.8. *Ranges of dry deposition contributions into total depositions*

Model	Mean value and range of dry deposition contribution to the total deposition, %		
	February 1999	August 1999	1999 as a whole
ADOM	24 (8 – 41)	30* (12 – 47)	
CMAQ**	30 (18 – 69)	22 (14 – 32)	
EMAP	25 (16 – 40)	15 (4 - 28)	21 (14 – 29)
MSCE-HM	20 (9 – 25)	30 (19 – 48)	28 (16 – 38)
MSCE-HM-Hem	24 (11 – 38)	29 (17 – 43)	25 (13 – 32)
HYSPLIT	45 (38 – 56)	31 (18 – 41)	
DEHM	12 (5 - 17)	19 (9 - 30)	16 (8 - 25)

* - the data for July 1999;
** - the data for 7 stations (stations SE05 and FI96 were not calculated).

Table 8.3.9 shows the estimates of dry depositions and their mean modelled values obtained by the models for February and August 1999. Degree of scattering the modelling results on the depositions is presented by the maximum deviation (MD) from the average value. The results of individual models, which are the closest to the mean values are highlighted in bold. MSCE-HM-Hem more often produces the results, which are the closest to the means. One can see that the results of individual models vary rather widely. Especially great scattering of the model-to-model results are characteristic of the northern stations. Probably, this scattering is connected with different parameterisations of dry deposition process for snow cover. The table demonstrates clearly that all the models (but EMAP) predict higher dry deposition intensity during summer time. Most probably, it is connected with lower dry deposition velocity over snow surface and over non-foliage forest.

Table 8.3.9. *Predicted dry depositions during February and August at the monitoring stations (g/km^2/month) and factors of maximum deviations from the means*

Model Parameter	Month	Stations								
		DE01	DE09	NL91	NO99	SE02	SE11	SE12	SE05	FI96
ADOM *	Feb	0.11	0.09	0.10	0.36	0.26	0.05	0.03	0.33	0.36
	Jul	0.30	**0.27**	**0.32**	0.98	0.75	0.13	0.04	0.77	0.41
CMAQ	Feb	0.24	0.37	0.56	0.25	0.27	0.60	0.28	-	-
	Aug	**0.29**	0.41	0.68	**0.32**	0.28	0.71	0.48	-	-
EMAP	Feb	0.11	0.25	0.13	**0.17**	**0.17**	0.18	**0.11**	0.09	**0.14**
	Aug	0.16	0.18	0.29	0.14	0.18	0.16	0.09	0.02	0.07
MSCE-HM	Feb	0.10	0.13	0.20	0.05	0.07	0.19	0.06	0.02	0.02
	Aug	0.18	0.24	0.28	0.19	0.14	0.35	0.29	0.31	0.25
MSCE-HM-Hem	Feb	0.17	**0.23**	0.17	0.06	0.20	0.20	0.09	**0.10**	0.07
	Aug	0.42	0.50	0.53	0.19	0.43	**0.38**	0.16	**0.28**	**0.18**
HYSPLIT	Feb	**0.12**	**0.23**	0.38	0.07	0.16	0.30	0.13	0.08	0.10
	Aug	0.38	0.46	0.56	0.17	**0.31**	0.49	**0.25**	0.12	0.09
DEHM	Feb	0.10	**0.19**	**0.21**	0.09	0.09	**0.23**	0.10	0.02	0.03
	Aug	0.19	0.26	0.31	0.19	0.14	**0.36**	0.14	0.14	0.04
Mean	**Feb**	**0.13**	**0.21**	**0.25**	**0.15**	**0.17**	**0.25**	**0.11**	**0.11**	**0.12**
	Aug	**0.27**	**0.33**	**0.42**	**0.31**	**0.32**	**0.37**	**0.21**	**0.27**	**0.17**
Max. deviation	Feb	1.8	2.3	2.5	3.0	2.4	5.0	7.0	5.5	6.0
	Aug	1.7	1.8	1.6	3.2	2.3	2.8	5.3	13	4.3

* - The data for July were used.

All the models predict that monthly total depositions in February 1999 are on the level of 0.6 g/km^2 (Fig. 8.3.1a). Model-to-model variations are generally within a factor of two. In spite of high variability, the figure demonstrates very obvious spatial descending trend of mercury deposition from "polluted" south to "background" north (linear trend line in the figure). The data for August are given in Fig. 8.3.1b). All the models predict that the total Hg deposition values are noticeable higher (1.5-2 times) in August than in February. The south-to-north trend also takes place in August (see the trend line). The maximum values at southern stations are predicted by CMAQ, while for the northern ones ADOM produces the highest values. All the models (but CMAQ and MSCE-HM-Hem) predicted minimum values for this or that stations.

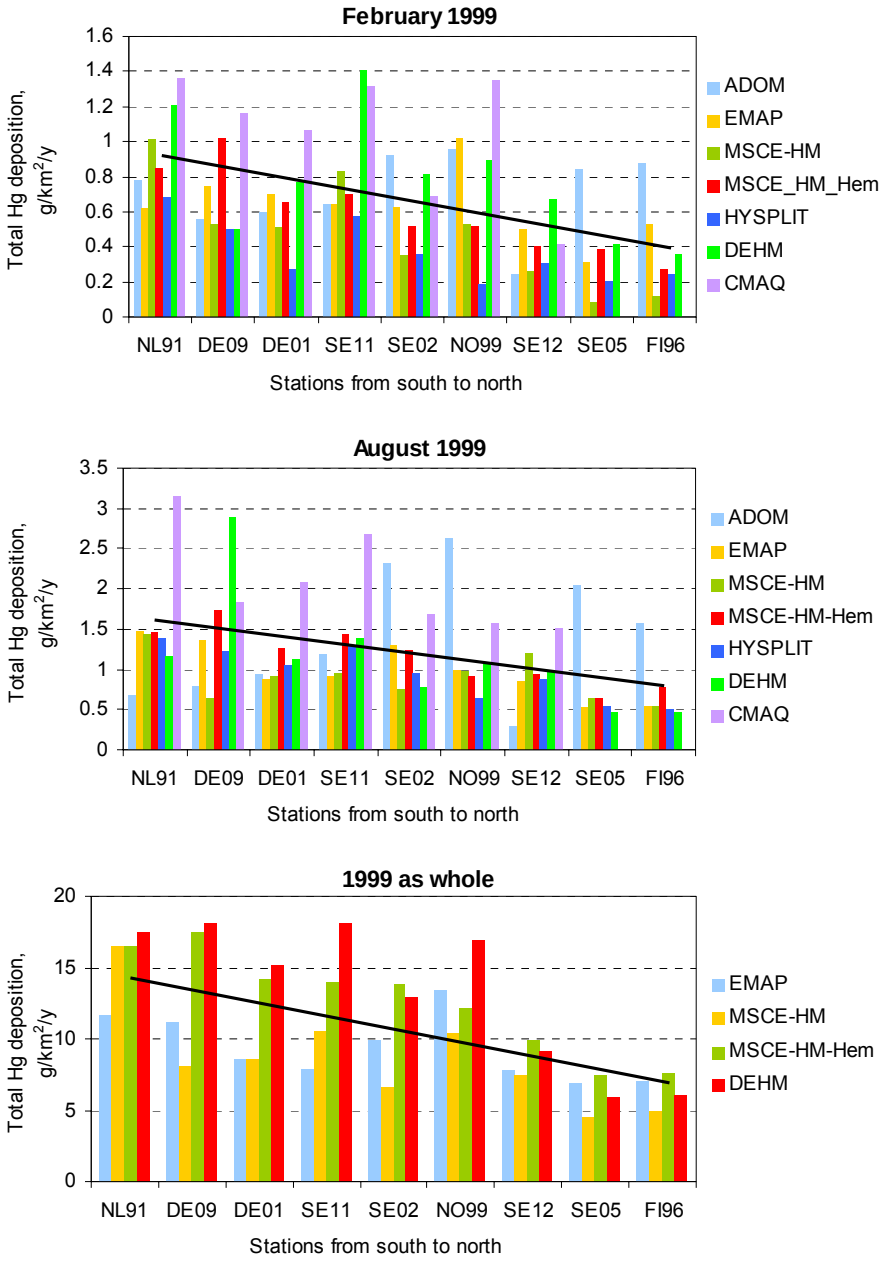


Fig. 8.3.1. *Predicted total mercury depositions at the monitoring stations during February (a), August (b) and 1999 as a whole (c). Note: ADOM data are taken for July instead of August.*

The data for the year as a whole can be compared only for four stations (Fig. 8.3.1c). As usual, the global version of the MSCE model and DEHM tend to predict higher total deposition values. However, the overall difference is not very high. The south-to-north descending trend of the total deposition is obvious (see the trend line). The models predict that the total deposition is two times higher in The Netherlands and Germany than in northern Sweden and Finland.

To assess acceptability of EMEP operational models for fulfilment of requirements of the HM Protocol to the Convention it is very important to evaluate, to what degree these operational models differ from the other scientific models. For this assessment the mean modelled and observed data for GEM concentrations in air, wet depositions and dry depositions were compared. The comparison was done for February because all the models produced the results for this month. Besides, only those stations were taken into account, for which all the models produced the results (CMAQ domain did not cover the Arctic stations).

Positions of the results produced by EMEP operational models among the others are presented in Table 8.3.10. One can see in the table that MSCE-HM-Hem never occupy any extreme positions (minimum or maximum). MSCE-HM in comparison with the other models tends to overestimate GEM concentrations in air and to underestimate dry depositions. The comparison also shows that the MSCE-HM-Hem results are very close to the mean observed values and the mean calculated values. In all cases the difference does not exceed 10%.

Table 8.3.10. *Positions of the results obtained by EMEP operational models for February 1999*

Model / Parameter	GEM, ng/m^3	Wet deposition, g/km^2/month	Dry deposition, g/km^2/month
Minimum value	1.37	0.21	0.11
MSCE-HM	**1.77**	**0.46**	**0.11**
MSCE-HM-Hem	**1.62**	**0.51**	**0.16**
Maximum value	1.77	0.91	0.37
Mean observed value	1.58	0.54	-
Mean modelled value	1.61	0.55	0.18

8.4. Mercury atmospheric balances for UK, Italy and Poland

This model comparison project included an analysis of the mercury atmospheric balances for three individual countries. The main items are deposition fluxes and outflow fluxes caused by a country's own anthropogenic sources. From the viewpoint of the Transboundary Convention it is important to know the extent of transboundary pollutant transport caused by mercury anthropogenic emissions in European region. Unfortunately, not all participating modelling groups could calculate all planned items for each month of 1999. This fact limits the comparison. Nevertheless, all the models simulated February 1999 and six out of seven of the models simulated August 1999. Below we will operate with the following parameters:

APS - depositions caused by **A**ll **P**ossible **S**ources including anthropogenic, natural and re-emission; in this case the regional models take into account global sources by prescribing boundary conditions, kg.

NAS - depositions caused by **N**ational **A**nthropogenic emission **S**ources of a given country, kg or per cent of ARS;

EAS - depositions caused by all **E**uropean **A**nthropogenic emission **S**ources without sources of a given country (European transboundary pollution), kg or per cent of ARS;

ROF – **R**elative **O**ut-**F**low determined as a fraction of national anthropogenic emission transported outside a given country, %.

Results of ADOM calculations of mercury depositions over the individual countries caused by different emission sources are presented in Table 8.4.1. The values show that the total depositions are the smallest in the UK and the highest in Poland. Own national anthropogenic sources contribute 30-50% to the total depositions over the UK, 15-25% over Italy. For Poland own anthropogenic sources give the main contribution into total mercury depositions of the country (50-65%). Obviously, the contributions depend mainly on two factors – intensity of own anthropogenic emission and geographical configuration of a given country. For the UK and Italy the contributions of external sources can exceed 50% while for Poland all external sources contribute less that half of the total deposition. ADOM predicts that all the countries during the considered months are net sources of atmospheric mercury. Own anthropogenic emissions several times exceed the total deposition values. Overwhelming parts of national anthropogenic mercury emissions in the countries are transported outside their territories. In all cases relative parameter ROF vary within 82 – 96%.

Table 8.4.1. *Mercury deposition over selected countries caused by national anthropogenic sources (NAS) and by all possible sources (APS), kg/month as well as relative outflow (ROF), % calculated by ADOM*

Month	United Kingdom			Italy			Poland		
	APS	NAS	ROF	APS	NAS	ROF	APS	NAS	ROF
February	95	43 (45%)	94	143	33 (23%)	96	403	258 (64%)	88
March	138	60 (43%)	92	233	48 (21%)	94	495	328 (66%)	85
June	170	53 (31%)	93	300	45 (15%)	94	718	393 (55%)	82
July	125	60 (48%)	92	283	43 (15%)	95	565	300 (53%)	86

Results of CMAQ calculations of mercury depositions over Poland caused by different emission sources during February 1999 are presented in Table 8.4.2. The values show that own anthropogenic sources contributed 61% to the total depositions over Poland. In accordance with CMAQ results 76% of Polish anthropogenic mercury emission is transported outside the national borders. The balance shows that Poland is net mercury anthropogenic source: total deposition is 2.5 times lower than the national anthropogenic emission.

Table 8.4.2. *Items of mercury atmospheric mercury balance for Poland in February 1999 calculated by CMAQ*

Item of the balance	Deposition or contribution
APS	835 kg/month
NAS	510 kg/month (61%)
ROF	76 %

Results of EMAP calculations of mercury depositions over the individual countries caused by different emission sources are presented in Table 8.4.3. The values show that on the annual basis own

anthropogenic sources contribute from 33% (Italy) to 72% (Poland) of the total depositions. It should be noted that for Poland own anthropogenic sources provide main contribution to the deposition. Such a difference between the countries is understandable because specific density of the anthropogenic emissions per area unit is much higher in Poland than in the UK or Italy. It is important to mention that about 3/4 of mercury emitted in Poland are deposited within the country. The model predicts that European anthropogenic sources provide minor contributions to the depositions over the countries in comparison with the contributions of national sources and globally distributed sources. It is especially rightly for the UK, which is located upwind to the main European mercury emission sources. On the contrary, for Poland the absolute values of transboundary depositions are an order of magnitude higher than for the UK. The model predicts an obvious seasonal cycle of the depositions for all the countries: maximum values are reached during wintertime.

The table shows also the roles of globally distributed sources pollution and outflow from given country. For the UK and Italy the contributions of globally distributed sources exceed 50%. In accordance with EMAP data the shares of outflow during individual months are practically the same as for the whole year. The main parts of national emissions are transported outside the countries. On the annual level all the considered countries are net atmospheric mercury sources: national anthropogenic emissions are higher that the total depositions 2.6 times for the UK, 2.2 times for Italy and 2 times for Poland.

Results of evaluation of mercury depositions over the individual countries calculated by MSCE-HM model are presented in Table 8.4.4. One can see that for all the countries the depositions caused by own anthropogenic sources dominate over European transboundary anthropogenic depositions. The transboundary anthropogenic contribution is minimal for the UK. It seems to be natural because the country is located in windward periphery of Europe, and probability of air mass transport from Central Europe to the UK is relatively low. On the contrary, for Poland transboundary depositions are several times higher although areas of the countries are similar. One can note that the model predicts an obvious seasonal cycle of transboundary pollution for the UK and Poland. In the UK the transboundary fluxes reach their highest values during spring and autumn, while in Poland maximum transboundary pollution fall on winter period. Such temporal distribution corresponds to general characteristics of air mass circulation.

The table shows the roles of own anthropogenic sources, transboundary pollution and outflow from given country. For Poland own anthropogenic sources give the main contribution into total mercury depositions of the country. For the UK and Italy the contribution of “out of EMEP” sources can exceed 50%. In accordance with MSCE-HM data the shares of outflow during individual months are practically the same as for the whole year. The main parts of national emissions are transported outside the countries. On the annual level all the considered countries are net atmospheric mercury sources: national anthropogenic emissions are higher that the total depositions 2.7 times for the UK, 2.2 times for Italy and 2 times for Poland.

Table 8.4.3. *Mercury deposition over selected countries caused by all possible sources (APS), by national anthropogenic sources (NAS) and by European anthropogenic sources (EAS) as well as relative outflow (ROF) calculated by EMAP*

Month	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
January	382	192 (50)	13 (3)	73	362	133 (37)	48 (13)	84	1348	836 (62)	209 (16)	61
February	286	151 (53)	11 (4)	79	424	121 (28)	65 (15)	85	1074	799 (74)	177 (16)	63
March	273	129 (47)	8 (3)	82	507	122 (24)	55 (11)	85	927	807 (87)	139 (15)	62
April	281	133 (47)	10 (3)	81	323	120 (37)	41 (13)	85	1167	779 (67)	105 (9)	63
May	200	101 (51)	12 (6)	86	351	128 (37)	38 (11)	84	949	742 (78)	83 (9)	65
June	209	115 (55)	8 (4)	84	299	99 (33)	29 (10)	88	1000	752 (75)	95 (10)	65
July	181	85 (47)	9 (5)	88	323	101 (31)	32 (10)	88	1064	759 (71)	72 (7)	64
August	263	121 (46)	19 (7)	83	369	153 (41)	44 (12)	81	1090	721 (66)	86 (8)	66
September	294	132 (45)	25 (9)	81	365	133 (36)	35 (10)	84	1068	713 (67)	78 (7)	67
October	304	136 (45)	32 (10)	81	304	108 (35)	41 (14)	87	849	742 (87)	99 (12)	65
November	214	112 (52)	12 (5)	84	478	129 (27)	52 (11)	84	1008	800 (79)	125 (12)	62
December	366	166 (45)	8 (2)	77	437	139 (32)	61 (14)	83	1253	778 (62)	178 (14)	64
Year	**3252**	**1575 (48)**	**167 (5)**	**82**	**4541**	**1485 (33)**	**543 (12)**	**85**	**12798**	**9229 (72)**	**1447 (11)**	**64**

Table 8.4.4. *Mercury deposition over selected countries caused by all possible sources (APS), by national anthropogenic sources (NAS) and by European anthropogenic sources (EAS) as well as relative outflow (ROF) calculated by MSCE-HM*

Month	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
January	332	175 (53%)	7 (2%)	75	336	128 (38%)	51 (15%)	84	1027	779 (76%)	197 (19%)	63
February	235	133 (57%)	3 (1%)	81	261	105 (40%)	54 (21%)	87	1068	832 (78%)	135 (13%)	61
March	238	133 (56%)	8 (3%)	81	344	130 (38%)	46 (13%)	84	1036	796 (77%)	142 (14%)	63
April	263	141 (54%)	12 (5%)	80	347	130 (37%)	47 (14%)	84	1114	785 (70%)	92 (8%)	63
May	232	115 (50%)	16 (7%)	84	312	118 (38%)	28 (9%)	86	1031	718 (70%)	79 (8%)	66
June	241	115 (48%)	5 (2%)	84	346	104 (30%)	38 (11%)	87	1222	771 (63%)	81 (7%)	64
July	147	65 (44%)	6 (4%)	91	387	117 (30%)	38 (10%)	86	1164	768 (66%)	77 (7%)	64
August	273	131 (48%)	21 (8%)	82	343	108 (31%)	39 (11%)	87	1033	734 (71%)	68 (7%)	66
September	301	143 (48%)	18 (6%)	80	449	141 (31%)	28 (6%)	83	965	687 (71%)	93 (10%)	68
October	278	118 (42%)	46 (17%)	83	375	133 (35%)	34 (9%)	84	953	723 (76%)	109 (11%)	66
November	239	109 (46%)	7 (3%)	85	520	167 (32%)	41 (8%)	80	963	749 (78%)	134 (14%)	65
December	371	168 (45%)	5 (1%)	76	389	148 (38%)	56 (14%)	82	1071	802 (75%)	191 (18%)	62
Year	**3149**	**1546 (49%)**	**154 (5%)**	**82**	**4411**	**1528 (35%)**	**500 (11%)**	**84**	**12646**	**9143 (72%)**	**1395 (11%)**	**64**

Results of calculations of mercury depositions over the individual countries obtained by MSCE-HM-Hem are presented in Table 8.4.5. The model predicts that for all the countries the depositions caused by own anthropogenic sources dominate over European transboundary anthropogenic depositions. The transboundary anthropogenic contribution is minimal for the UK because of windward location of the country in respect of the main European emission sources. In Italy the roles of own sources and transboundary European anthropogenic sources are practically equal. For Poland transboundary depositions are twice lower than caused by own sources.

For Poland own anthropogenic sources give about 50% into total mercury depositions of the country. On the contrary, for the UK and Italy the contribution of own sources is about only 25%. In accordance with MSCE-HM-Hem results the shares of outflow do not vary significantly from month to month. On the annual level all the considered countries are net atmospheric mercury sources: national anthropogenic emissions are higher that the total depositions 2.7 times for the UK, 3.1 times for Italy and 2.7 times for Poland. The model predicts an obvious seasonal cycle of transboundary pollution only for the UK: the transboundary fluxes reach their maximum values during spring and autumn.

Results of HYSPLIT calculations of mercury depositions over the individual countries caused by different emission sources are presented in Table 8.4.6. The model predicts that for the UK and Poland the depositions caused by own anthropogenic sources dominate over European transboundary anthropogenic depositions. For Italy European contribution is higher. The transboundary anthropogenic contribution is minimal for the UK in February because of windward location of the country in respect of the main European emission sources. For Poland transboundary depositions are 1.5 - 2 times lower than caused by own sources.

The table also shows the roles of own anthropogenic sources, transboundary pollution and outflow from given country. For Poland own anthropogenic sources give about 60% into total mercury depositions of the country. For the UK and Italy the contributions of own sources are lower – 37% and 30%, correspondingly. In accordance with HYSPLIT-Hg data the shares of outflow are higher than 90% for all countries. It is the highest for the UK and the lowest for Poland.

Table 8.4.5. *Mercury deposition over selected countries caused by all possible sources (APS), by national anthropogenic sources (NAS) and by European anthropogenic sources (EAS) as well as relative outflow (ROF) calculated by MSCE-HM-Hem*

Month	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
January	246	67 (27%)	6 (2%)	91	220	49 (22%)	41 (19%)	94	741	376 (51%)	216 (29%)	82
February	170	48 (28%)	5 (3%)	93	164	36 (22%)	48 (29%)	96	731	375 (51%)	190 (26%)	82
March	256	73 (29%)	15 (6%)	90	230	46 (20%)	49 (21%)	94	690	379 (55%)	182 (26%)	82
April	253	67 (26%)	24 (9%)	91	273	52 (19%)	51 (19%)	94	858	422 (49%)	198 (23%)	80
May	312	75 (24%)	38 (12%)	89	297	52 (18%)	41 (14%)	94	801	409 (51%)	185 (23%)	81
June	279	74 (27%)	16 (6%)	90	267	50 (19%)	48 (18%)	94	1065	473 (44%)	210 (20%)	78
July	238	66 (28%)	15 (6%)	91	274	50 (18%)	48 (18%)	94	853	403 (47%)	179 (21%)	81
August	339	80 (24%)	40 (12%)	89	289	51 (18%)	44 (15%)	94	778	410 (53%)	171 (22%)	81
September	343	81 (24%)	31 (9%)	89	307	50 (16%)	45 (15%)	94	769	386 (50%)	180 (23%)	82
October	277	68 (25%)	41 (15%)	90	281	52 (19%)	41 (15%)	94	756	380 (50%)	189 (25%)	82
November	208	48 (23%)	14 (7%)	93	335	54 (16%)	63 (19%)	93	793	405 (51%)	221 (28%)	81
December	211	56 (27%)	11 (5%)	92	239	44 (18%)	51 (21%)	95	751	370 (49%)	230 (31%)	83
Year	**3130**	**803 (26%)**	**257 (8%)**	91	**3174**	**586 (18%)**	**570 (18%)**	94	**9589**	**4788 (50%)**	**2351 (25%)**	81

Table 8.4.6. *Mercury deposition over selected countries caused by all possible sources (APS), by national anthropogenic sources (NAS) and by European anthropogenic sources (EAS) as well as relative outflow (ROF) calculated by HYSPLIT*

Month	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
February	76	28 (37%)	10 (13%)	96	153	46 (30%)	76 (50%)	94	300	170 (57%)	91 (30%)	92
August	173	48 (28%)	40 (23%)	93	297	74 (25%)	108 (36%)	91	548	250 (46%)	172 (31%)	88

Results of evaluation of mercury depositions over the individual countries calculated by DEHM are presented in Table 8.4.7. One can see that for all the countries the depositions caused by own anthropogenic sources dominate over European transboundary anthropogenic depositions. The transboundary anthropogenic contribution is minimal for the UK probably because of the fact that the country is located in windward periphery of Europe, and probability of air mass transport from Central Europe to the UK is relatively low. On the contrary, for Poland transboundary depositions are much higher although areas of the countries are similar. One can note that the model predicts an obvious seasonal cycle of transboundary pollution for the UK and Poland. In the UK the transboundary fluxes reach their highest values during spring and autumn, while in Poland maximum transboundary pollution fall on winter period. Such temporal distribution corresponds to general characteristics of air mass circulation.

For Poland own anthropogenic sources give the main contribution into total mercury depositions of the country. For the UK and Italy the contribution of “non-European” anthropogenic sources can exceed 50%. In accordance with DEHM data the shares of outflow during individual months are practically the same as for the whole year. However, contributions of own and European anthropogenic sources into the depositions can differ noticeably from the pattern for the whole year. The main parts on the national emissions are transported outside the countries. On the annual level all the considered countries are net atmospheric mercury sources: national anthropogenic emissions are higher that the total depositions 2.0 times for the UK, 1.5 times for Italy and 2 times for Poland.

The data obtained by the participating models show rather wide range of assessment of mercury atmospheric balances for the selected countries. Tables 8.4.8 and 8.4.9 presents the comparison of different items for February and August, while Table 8.4.9 – for the whole year. The modelling results, which are the closest to the average, are highlighted in the tables in bold.

Three countries considered here being similar in their areas are very different in their locations relatively main European sources of mercury transboundary pollution. Own mercury anthropogenic emissions are practically similar in the UK and Italy but 3 times higher in Poland. Hence, the participating models should catch such peculiarities of mercury atmospheric balances for the countries. Degree of scattering the modelling results on the depositions is presented by the maximum deviation (MD) from the average value. In most case for February EMAP produces the highest values of deposition, while the lowest values are produced by HYSPLIT.

For the UK the predicted items of mercury atmospheric balances on the monthly basis can vary within a factor higher than 3. For Italy and Poland the maximum deviation factors are usually lower than two. On the annual basis the variations are not so pronounced, however, it can be explained by lower number of the compared data. Generally, the predicted total deposition values are higher in summer period and their model-to-model variations are lower.

The depositions caused by own national sources in all cases are the lowest in Italy (about 30%) and the highest in Poland (60-65%). Depositions caused by European transboundary pollution are the lowest for the UK and the highest for Poland. This fact is quite understandable because the UK is in the windward periphery of Europe while Poland is in the central part. The scattering of these results is smaller than the factor of 2.

Table 8.4.7. *Mercury deposition over selected countries caused by all possible sources (APS), by national anthropogenic sources (NAS) and by European anthropogenic sources (EAS) as well as relative outflow (ROF) calculated by DEHM*

Month	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
January	370	124 (34%)	25 (7%)	83	440	137 (31%)	89 (20%)	83	979	576 (59%)	273 (28%)	73
February	240	69 (29%)	15 (6%)	90	334	113 (34%)	75 (22%)	86	1188	707 (60%)	278 (23%)	67
March	303	105 (35%)	30 (10%)	85	515	149 (29%)	105 (20%)	82	893	573 (64%)	197 (22%)	73
April	361	123 (34%)	59 (16%)	83	616	173 (28%)	107 (17%)	79	1126	709 (63%)	194 (17%)	67
May	428	128 (30%)	65 (15%)	82	767	203 (26%)	117 (15%)	75	1238	768 (62%)	212 (17%)	64
June	417	149 (36%)	44 (11%)	79	680	168 (25%)	123 (18%)	79	1713	1040 (61%)	238 (14%)	51
July	325	110 (34%)	21 (6%)	85	640	162 (25%)	134 (21%)	80	1335	820 (61%)	200 (15%)	62
August	441	140 (32%)	85 (19%)	80	610	152 (25%)	136 (22%)	81	836	529 (63%)	145 (17%)	75
September	429	141 (33%)	88 (21%)	80	568	141 (25%)	130 (23%)	83	769	528 (69%)	128 (17%)	75
October	299	95 (32%)	83 (28%)	87	430	148 (34%)	76 (18%)	82	882	588 (67%)	174 (20%)	72
November	286	93 (33%)	26 (9%)	87	577	177 (31%)	134 (23%)	78	1033	691 (67%)	226 (22%)	68
December	333	120 (36%)	26 (8%)	83	400	150 (38%)	83 (21%)	82	1124	694 (62%)	311 (28%)	67
Year	**4232**	1397 (33%)	567 (13%)	84	**6577**	1873 (28%)	1309 (20%)	81	**13116**	8223 (63%)	2576 (20%)	68

Table 8.4.8. *Comparison of items of mercury atmospheric balances obtained by the participating models for February 1999 (for the averages in brackets – factors of deviations from the average values)*

Model	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
ADOM	95	43 (45%)		94	143	33 (23%)		96	403	258 (64%)		88
CMAQ									**835**	**510** (61%)		**76**
EMAP	286	151 (53%)	11 (4%)	79	424	121 (28%)	**65** (15%)	85	1074	799 (74%)	**177** (16%)	63
MSCE-HM	235	133 (57)	3 (1%)	81	**261**	**105** (40%)	54 (21%)	87	1068	832 (78%)	135 (13%)	61
MSCE-HM-Hem	**170**	48 (28%)	5 (3%)	93	164	36 (22%)	48 (29%)	96	731	375 (51%)	190 (26%)	82
HYSPLIT	76	28 (37%)	**10** (13%)	96	153	46 (30%)	76 (50%)	**94**	300	170 (57%)	91 (30%)	92
DEHM	240	**69** (29%)	15 (6%)	**90**	334	113 (34%)	75 (22%)	86	1188	707 (60%)	278 (23%)	67
Average	**184**	**79 (43%)**	**9 (5%)**	**89**	**247**	**76 (31%)**	**64 (26%)**	**91**	**800**	**522 (65%)**	**174 (22%)**	**76**
Maximum deviation	F2.4	F2.8	F3.0		F1.7	F2.3	F1.3		F2.7	F3.1	F1.9	

Table 8.4.9. *Comparison of items of mercury atmospheric balances obtained by the participating models for August 1999 (for the averages in brackets – factors of deviations from the average values)*

Model	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
ADOM *	125	60 (48%)	-	92	283	43 (15%)	-	95	565	300 (53%)	-	86
EMAP	263	121 (46%)	19 (7%)	83	**369**	153 (41%)	**44** (12%)	81	1090	721 (66%)	86 (8%)	66
MSCE-HM	**273**	131 (48%)	21 (8%)	82	343	**108** (31%)	39 (11%)	**87**	1033	734 (71%)	68 (7%)	66
MSCE-HM-Hem	339	**80** (24%)	**40** (12%)	**89**	289	51 (18%)	**44** (15%)	94	778	410 (53%)	171 (22%)	81
HYSPLIT	173	48 (28%)	**40** (23%)	93	297	74 (25%)	108 (36%)	91	548	250 (46%)	172 (31%)	88
DEHM	441	140 (32%)	85 (19%)	80	610	152 (25%)	136 (22%)	81	**836**	**529** (63%)	**145** (17%)	**75**
Average	**269**	**97 (36%)**	**41 (15%)**	**87**	**365**	**97 (27%)**	**74 (20%)**	**88**	**808**	**491 (61%)**	**128 (16%)**	**77**
Maximum deviation	F2.2	F2.0	F2.2		F1.7	F2.3	F1.9		F1.5	F1.6	F1.9	

* - The data for July were used.

Table 8.4.10. *Comparison of items of mercury atmospheric balances obtained by the participating models for year 1999 as a whole (in brackets – factors of deviations from the average values)*

Model	United Kingdom				Italy				Poland			
	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF	APS	NAS	EAS	ROF
EMAP	**3252**	1575 (48%)	167 (5%)	82	**4541**	**1485** (33%)	543 (12%)	**85**	12798	9229 (72%)	1447 (11%)	64
MSCE-HM	3149	1546 (49%)	154 (5%)	82	4411	1528 (35%)	500 (11%)	84	**12646**	9143 (72%)	1395 (11%)	64
MSCE-HM-Hem	3130	803 (26%)	**257** (8%)	91	3174	586 (18%)	**570** (18%)	94	9589	4788 (50%)	**2351** (25%)	81
DEHM	4232	**1397** (33%)	567 (13%)	**84**	6577	1873 (28%)	1309 (20%)	81	13116	**8223** (63%)	2576 (20%)	**68**
Average	**3441**	**1330 (39%)**	**286 (8%)**	**85**	**4676**	**1368 (29%)**	**731 (16%)**	**86**	**12037**	**7846 (65%)**	**1942 (16%)**	**69**
Maximum deviation	F1.2	F1.7	F2.0		F1.5	F2.3	F1.8		F1.3	F1.6	F1.4	

Total deposition values are included mercury from all possible sources. Hence, this parameter depends not only on European anthropogenic pollution but also on natural emissions, on calculated or accepted concentrations of different mercury forms at the borders of the EMEP domain. Contributions of natural sources and remote anthropogenic sources are practically equal for the countries (7.9 $g/km^2/y$ for the UK, 8.6 for Italy and 7.3 for Poland). On the annual basis the scattering between the modelling results is on the level of a factor of 1.5.

All the models predict that the main part of national anthropogenic emissions is transported in the atmosphere outside the national territory. Percentages of mercury transported outside the country vary in rather narrow range. All the countries are net sources of mercury for the global atmosphere: 85-90% of British and Italian anthropogenic mercury and 70-75% of Poland's emissions leave these countries.

The comparison of the tables shows that the scattering the results is lower for the annual data than for the data obtained for a single month. It is not surprising, because of smoothing the highly variable meteorological factors during twelve months' period. Hence, it is possible to conclude very generally that the participating models can predict monthly values of items of mercury atmospheric budgets for individual countries with accuracy on the level of a factor of 2.5. As to yearly atmospheric budgets, such accuracy can be expressed by a factor of 2 or better. However, one should remember, that the number of models calculated monthly data and yearly data is different. Comparing the modelling results for February and August, it appears that the models giving results most often closest to the average include MSCE-HM, MSCE-HM-Hem, EMAP and DEHM. Among the four models that produced annual deposition results, the model that the most often gave values closest to the average was EMAP. Generalizing the tables it is possible to mention that EMEP operational models among the other models do not occupy as usual any extreme positions.

9. SUMMARY AND CONCLUSIONS

A five-year project on the intercomparison of mathematical models designed to simulate mercury long-rang atmospheric transport and deposition has been completed. The main task of the project was to assess the capability of the models to reproduce measurements of mercury concentrations in air and atmospheric precipitation and to get a clear idea of range of uncertainties of results obtained by modern models. The third stage of the project presented in this report was aimed at comparisons of modelling results with long-term (month – year) observations. The most important constituent of the third stage was an attempt to compare capabilities of the participating models to simulate integrated items of mercury atmospheric balances for individual countries. Just such calculations are of the particular interest from ecological viewpoint and for the implementation of Heavy Metal Protocol to the Convention on Long-range Transboundary Air Pollution.

It is known that there are not more than a dozen of mercury models designed for evaluation of mercury atmospheric transport on regional (continental) and global levels. Most of them partly of fully participated in the project. They are different in many aspects: in methods of atmospheric transport simulation (Eulerian or Lagrangian types), in approaches to simulate deposition mechanisms, in conception of mercury chemistry in the atmosphere. The main interest of the study is focused on European region (EMEP domain) but some models consider mercury fate on hemispheric level. Accordingly, spatial resolution of the models is different – from 36x36 km to 2.5x2.5 geographical degrees. Vertical coverage of the models varies from 5 to 30 km. At the same time all the models deal with three main physico-chemical forms of mercury in the atmosphere. The intercomparison program envisages to use common input parameters (mainly, the emissions) to the greatest extent possible for all the models.

To compare the models with observations, results of routine monitoring of mercury in the atmosphere and atmospheric precipitation obtained at EMEP monitoring stations in 1999 are used. The temporal resolution of the observational data is one month. The measurement data were critically considered and some “outlier” values have been removed. Some mercury emission fields common for all the models have been prepared and used in the study. The fields include mercury anthropogenic emissions of individual mercury forms at different vertical levels in Europe with 50x50 km resolution, mercury anthropogenic emissions on a global level, global natural emissions and secondary anthropogenic emission (re-emission) in Europe. Different models used input meteorological information obtained and prepared individually. Some of the models additionally simulate chemical reactants involved into mercury chemistry (ozone, sulphur dioxide), while some of the models use predetermined concentration values of such reactants.

The results of the third stage of the project lead to the following conclusions.

1. Calculations of elemental mercury concentrations in air show that the concentration levels over Europe only partly are determined by European anthropogenic sources. For many European countries the main contribution to the levels is given by “hemispheric background”.

2. For most of the monitoring stations the models can predict the observations with accuracy within a factor of 1.2. Model-to-model variations of the results are within 12%. Probability to obtain a false result (>F.1.5) is very small for all the models.

3. Comparison of calculated and measured data on deposition with precipitation for one month shows that for the “polluted” stations (Germany, The Netherlands, Southern Scandinavia) the modelled values are in agreement with the observations within a factor of 2. However, for the northern “background” stations the models typically overestimate the observations (up to 10 times). Partly such overestimation can be explained by very pronounced discrepancies between measured and

forecasted amounts of precipitation. Typically, meteorological models used strongly (1.5-2 times) overestimate precipitation amounts in the Arctic region. It leads to corresponding overestimation of mercury wet depositions. Model-to-model variations of predicted monthly wet deposition values are within ±50%. On the annual basis the agreement is much better. For the "polluted" stations the ratios are within a factor of 1.5. However, for the "background" stations the factor exceeds 2. Model-to-model variations for annual values are small: from 13% to 40%. Probability to produce a result within a factor 2 for all the models is about 2/3 and probability to obtain a false result (>F5) is negligible.

4. Significant uncertainty to modelled wet deposition values is introduced by difference between measured amounts of precipitation and forecasted by meteorological models. Special attention should be paid to usage of correct meteorological data on precipitation field in investigated regions.

5. Comparison of the modelling results for mercury dry deposition shows that most of the models predict contribution of this process into total deposition on the level of 30%. Range of variations is high – maximum deviation factors for February 1999 vary from 1.8 to 7.0. The highest variability is typical for the northern part of the European region. The models predict that dry deposition intensity is higher in summer than in winter. In any case, dry deposition contributes significantly to the total deposition, however, degree of reliability of the modelling results is still not known.

6. The program of the model comparison has proposed to calculate some items of mercury atmospheric balances for three individual countries – the UK, Italy and Poland. The models predict that for the UK and Italy, national anthropogenic sources account for about 1/3 of the total mercury deposition over the country. In the case of Poland all the models predict that more than half of deposited mercury is of Polish origin. Depositions caused by European transboundary pollution are the lowest for the UK and the highest for Poland. Contributions of natural sources, re-emission and remote anthropogenic sources are practically equal for the countries (7.9 g/km^2/y for the UK, 8.6 for Italy and 7.3 for Poland). It is possible to conclude very generally that the participating models can predict monthly values of items of mercury atmospheric budgets for individual countries with accuracy on the level of a factor of 2.5. As to yearly atmospheric budgets, such accuracy can be expressed by a factor of 2 or better.

7. All the models predict that the main part of national anthropogenic emissions is transported in the atmosphere outside the national territory. Percentages of mercury transported outside the country vary in rather narrow range. All the countries are net sources of mercury for the global atmosphere: about 85-90% of British and Italian anthropogenic mercury and 70-75% of Polish one quit the corresponding countries.

8. The EMEP Meteorological Synthesising Center "East" presented in the study two models – a regional (for the EMEP domain) and hemispheric one. The comparison reveals that for the main calculated parameters both models predict quite similar results. The agreement between them is as a rule better than a factor of 1.5. Both models do not occupy any extreme positions among the other participating models. It is possible to conclude that both EMEP models are acceptable to comply with demands of Heavy Metal Protocol to the Convention. At the same time The Parties of the Convention should keep in mind that accuracy of modern mercury modelling cannot exceed a factor of two.

9. The study reveals some most important gaps in our knowledge. Uncertainty of emission estimations for individual mercury physico-chemical forms can exceed 50%. Reliability of the data on natural emission and re-emission is not sufficient for modelling assessment of mercury fate in the atmosphere, especially on hemispheric level. There are urgent needs to obtain on a routine basis reliable monitoring data for all three mercury forms to validate the models. Physico-chemical properties of gaseous compounds of oxidised mercury are poorly known. Correspondingly, parameterisation of scavenging processes for such compounds needs significant improvement.

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