Chapter-29

EMEP REGIONAL/HEMISPHERIC MERCURY MODELLING: ACHIVEMENTS AND PROBLEMS

Alexey Ryaboshapko, Sergey Dutchak, Alexey Gusev, Ilia Ilyin and Oleg Travnikov

EMEP Meteorological Synthesizing Center "East", Arhitektor Vlasov Str., 51, Moscow 117393, Russia

INTRODUCTION

Investigation and control of transboundary air pollution in Europe has a relatively deep history. Considerable practical and scientific progress in this direction has been achieved in the framework of the Convention on Long-Range Transboundary Air Pollution. A number of international binding instruments (Protocols to the Convention) on reduction of air pollution were developed and entered into force during the last 25 years. Scientific support for the evaluation of long-range air pollution, as well as development and implementation of the Protocols is provided by the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP).

EMEP was established in 1977 under the United Nations Economic Commission for Europe. After the Convention entered into force in 1983 EMEP became an operational programme of the Convention. Routine activity of EMEP is based on joint efforts of the participating countries and 4 international centres of EMEP. One of these centres – the Meteorological Synthesizing Centre - East (MSC-E) is responsible for the development and application of atmospheric transport deposition models for the assessment of air pollution by heavy metals and persistent organic pollutants.

Heavy metals in line with acid compounds are included in the priority list of substances considered under the Convention. A Protocol on Heavy Metals was signed by 36 Parties to the Convention in 1998. The Protocol is aimed at control of heavy metal emissions into the atmosphere to reduce their transboundary transport and to prevent adverse effects on human health and the environment. In accordance with the Protocol EMEP is responsible for use of appropriate models and measurements for providing to European countries calculations of transboundary fluxes and depositions of lead, cadmium and mercury.

This paper is focused on the assessment of the long-range transboundary transport of mercury. Mercury is widely recognized as a global pollutant. To evaluate mercury pollution of Europe mercury emissions all over the globe and intercontinental transport should be taken into account. To meet these requirements MSC-E is developing mathematical models for the evaluation of mercury atmospheric transport on regional (Europe) and hemispherical (Northern Hemisphere) scales.

EMEP REGIONAL MODEL

The EMEP regional model considers basic processes governing the transport and deposition of mercury - advection, diffusion, dry and wet removal and chemical transformations. This is an Eulerian three-dimensional atmospheric transport model. The model operates within the so

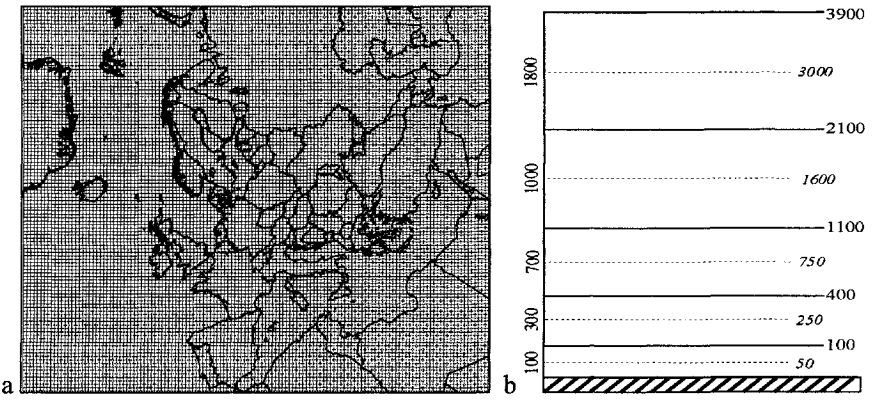

Figure 1. Horizontal (a) and vertical (b) structure of EMEP regional model.

called EMEP domain (Posh et al., 1997). This region includes the European continent, the northern part of Africa, part of the Middle East, the North Atlantic and part of the Arctic (see Figure 1). The EMEP grid consists of 135x111 grid cells with spatial resolution 50 km at 60°N. latitude.

As seen from Figure 1, the model domain consists of five non-uniform layers along the vertical. The top of the model is at a height of about 4 km. Therefore, the model domain covers the entire atmospheric boundary layer and part of the middle troposphere. Depths of the layers are 100, 300, 700, 1000 and 1800 m (from bottom to top). The advection scheme is conservative, stable and positively defined (Pekar, 1996). The model description of the vertical turbulent diffusion is based on the first order closure approach.

The model deals with three physical-chemical mercury forms: gaseous elemental mercury (GEM), reactive (oxidised) gaseous mercury (RGM) and total particulate mercury (TPM). They possess very different characteristics, which determine very different lifetimes of each form in the atmosphere. Detailed description of the parameterisation of all modelled processes can be found in MSC-E technical reports (Ilyin et al., 2001; 2002; www.msceast.org).

Scavenging of all mercury forms encompasses wet removal and uptake by the underlying surface. Wet removal of TPM and RGM is described using a washout ratio approach. It is accepted that particles containing mercury behave like sulfate particles and the equilibrium washout ratio is equal to 5×10^5 (Petersen et al., 1998; Iversen et al., 1989). Washout of RGM by the liquid phase is prescribed by the equilibrium washout ratio characteristic of nitric acid: 1.4×10^6 (Petersen et al., 1995; Jonsen and Berge, 1995). The reason for this is the similar solubilities of these two species in water. Besides, RGM and GEM can be dissolved in the aqueous phase of clouds.

Dry deposition of TPM is described in the framework of an electric resistance analogy. Mercury containing particles are in the submicron size range (Milford and Davidson, 1985; Keeler et al., 1995), hence the effect of the gravity can be ignored. Dry deposition efficiency is differentiated with regard to the land-cover category of the underlying surface and depends basically on properties of the underlying surface and atmospheric stability (Ruijgrok et al., 1997; Wesely and Hicks, 2000).

Dry uptake of GEM depends on a number of parameters. On the basis of literature data (Lindberg et al., 1992; Xu et al., 1999; Petersen et al., 2001; Seigneur et al., 2001) it is assumed that at negative temperatures uptake does not occur. Uptake also does not occur by water and vegetation-free surfaces.

For the surface covered with forest the velocity is accepted to be equal to 0.03 cm/s at 20°C and higher. For other types of vegetation the maximum value is 0.01 cm/s. The uptake velocity decreases linearly to zero as temperature decrease to 0°C.

When describing the dry uptake of gaseous oxidised mercury a similarity of dry uptake velocity to that of nitric acid is assumed (Petersen et al., 1998). This assumption comes from their similar solubilities in water. Keeping in mind the obvious lack of knowledge on this, the dry uptake velocity for RGM assumed in the model is 0.5 cm/s for all seasons and types of underlying surfaces.

Parameterisation of chemical processes includes both aqueous-phase and gaseous-phase reactions and equilibria. It is based on the chemical scheme suggested by Petersen et al. 1998. However, the scheme has been simplified - only key reactions are used in the model. They are gas-phase oxidation of GEM by ozone, dissolution of GEM and RGM in cloud droplets, aqueous phase oxidation of GEM by ozone with further sorption of the reaction products on insoluble particles within droplets, and mercury reduction to the elemental state through decomposition of mercury-sulfite complexes. All products of gaseous-phase oxidation are treated as aerosol particles. It is accepted (Brosset and Lord, 1991; Iverfeldt, 1991; Lamborg et al., 1995) that half of TPM being captured by cloud or rainwater droplets can be dissolved. After drop evaporation an aerosol particle is formed containing in its composition all earlier dissolved and insoluble mercury compounds.

An important distinction of the scheme from analogous ones typically used in atmospheric mercury models (Petersen et al, 1998; Bullock and Brehme, 2002; Shia et al., 1999) is in usage of temperature dependencies of reaction rates and equilibrium constants. For Henry's law constants the following equations are used (Sander, 1997; Ilyin et al., 2002):

For GEM: $$K_{Hg^0} = 0.00984 \cdot T \cdot \exp\left(2800 \cdot \left(\frac{1}{T} - 0.003356\right)\right),$$

for RGM: $$K_{HgCl_3} = 1.054 \cdot 10^5 \cdot T \cdot \exp\left(5590 \cdot \left(\frac{1}{T} - 0.003356\right)\right),$$

for O_3: $$K_{O_3} = 0.000951 \cdot T \cdot \exp\left(2325 \cdot \left(\frac{1}{T} - 0.003356\right)\right).$$

The dependence of the rate of gas-phase GEM oxidation by ozone on temperature is described by the following equation delivered from the data published by Hall [1995]:

$$k = 2.1 \cdot 10^{-18} \cdot \exp\left(-\frac{1246}{T}\right) \quad cm^3/molec/s$$

Since temperature in the troposphere can vary within the range of 100 degrees, the accepted dependencies can significantly change rates of mercury chemical transformations and its removal from the atmosphere.

EMEP HEMISPHERIC MODEL

The EMEP hemispheric model has been developed in order to evaluate the atmospheric transport of mercury over the Northern Hemisphere. This is a three-dimensional chemical transport model of Eulerian type. The detailed description of the model can be found in MSC-E technical reports (Travnikov and Ryaboshapko, 2002; www.msceast.org).

The model computation domain covers the whole Northern Hemisphere with a spatial resolution of 2.5° both in zonal and meridional directions. The surface grid structure of the model domain is shown in Figure 2. To avoid a singularity at the pole point, peculiar to the spherical co-ordinates, the grid has a special circular mesh of radius 1.25° including the North Pole. In the vertical direction the model domain consists of eight irregular levels of terrain-following sigma-pressure co-ordinates defined as a ratio of local atmospheric pressure to the ground surface pressure (Jacobson, 1999). The vertical grid structure of the model is presented in Figure 2.

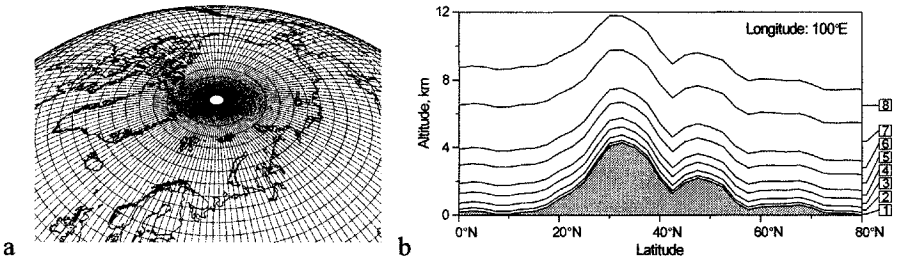

Figure 2. Horizontal (a) and vertical (b) structure of EMEP hemispheric model.

Advection is treated using the Bott flux-form advection scheme (Bott, 1992). This scheme is mass conservative, positive-definite, monotone, and is characterised by comparatively low artificial diffusion. In order to reduce the time-splitting error in strong deformational flows the scheme has been

modified according to (Easter, 1993). The vertical movements are solved using the original Bott scheme generalised for a grid with variable step. Non-linear diffusion has been approximated by the second-order implicit numerical scheme in order to avoid restrictions of the time step caused by possible sharp gradients of species mixing ratio.

The modelling domain has two borders - upper layer and the Equator. At the upper boundary a uniform distribution of GEM concentration of 0.185 pptv (corresponding to about 1.5 ng/m^3 at 1 atm and 20°C) is prescribed. Within the equatorial zone some gradient of gaseous mercury was observed between the Northern and Southern Hemispheres (Slemr, 1996). In the model the gradient of GEM is set to 0.05 ng/m^3/degree at the equatorial boundary. Since the atmospheric residence times of the other mercury species are considerably shorter their input through the boundaries is neglected.

The parameterisation of mercury scavenging processes in the hemispheric model does not differ from that used in the regional model. The hemispheric model takes into account the same chemical transformations of mercury as the regional model described above. However, GEM oxidation by chlorine is introduced into the chemical scheme because in the oceanic atmosphere this reaction can give a noticeable effect.

EMISSIONS AND OTHER INPUT PARAMETERS

The Convention envisages that all participating countries should evaluate their national emissions using the same inventory methodology. Currently national data on total mercury emissions (for at least one year for the period of 1990-2000) were submitted by 34 countries. For the other countries, which have not reported national emission data, expert estimates are applied (Berdowski et al., 1997; Pacyna and Pacyna, 2002). Mercury emission data for 2000 used for the assessment of pollution level in Europe are demonstrated in Table 1 (Ilyn and Travnikov, 2003). During the last decade mercury emissions into the atmosphere in most European countries were reduced. Thus, the total European mercury emission decreased from 463 t/yr in 1990 (Berdowski et al., 1997) to 201 t/yr in 2000 (Table 1). Accuracy of the emission data is quite uncertain. It is believed that the expert estimates can be within ±30% (Pacyna and Pacyna, 2002).

Table 1. Mercury anthropogenic emissions* (AE) to the atmosphere in the EMEP region in 2000, t/y.

Country	AE	Country	AE	Country	AE
Albania	0.5	Georgia	0.5	Portugal	4.85
Armenia	0.001	Germany	29	Moldova	0.18
Austria	1.15	Greece	13	Romania	6.55
Azerbaijan	0.6	Hungary	4.21	Russia	10
Belarus	0.36	Iceland	0.05	Serbia & Mont.	3.3
Belgium	2.88	Ireland	1.95	Slovakia	4.37
Bosnia & Herz.	0.2	Italy	13.2	Slovenia	0.58
Bulgaria	4.19	Kazakhstan	0.1	Spain	23.4
Croatia	0.41	Latvia	0.21	Sweden	0.81
Cyprus	0.30	Lithuania	0.25	Switzerland	2.63
Czech Rep.	3.84	Luxembourg	0.27	Macedonia	0.05
Denmark	1.96	Monaco	0.08	Turkey	4.30
Estonia	0.55	Netherlands	0.58	Ukraine	9.03
Finland	0.6	Norway	1.00	UK	8.79
France	15	Poland	25.6	Total	201

* Values obtained by extrapolation of official data and on the base of expert estimates are in italic.

For modelling purposes the emissions need to be spatially distributed over the domain. Some European countries assess the distribution of their national emissions in accordance with the EMEP grid (50x50 km resolution). For the others the total national emissions were distributed in accordance with (Berdowski et al., 1997).

During recent years the emission density has changed very significantly. In 1990 the emission density in "hot spots" reached 3200 g/km^2/yr. A very detailed emission inventory of the three mercury forms for each European country was implemented recently by Pacyna et al. (2003). The emissions were spatially distributed in accordance with the EMEP grid taking into account the locations of main point sources. In the vertical direction three emission layers were distinguished: <50, 50-150, and >150 m. It is possible to see (Figure 3b) that from 1990 to 2000 emissions declined in most European countries.

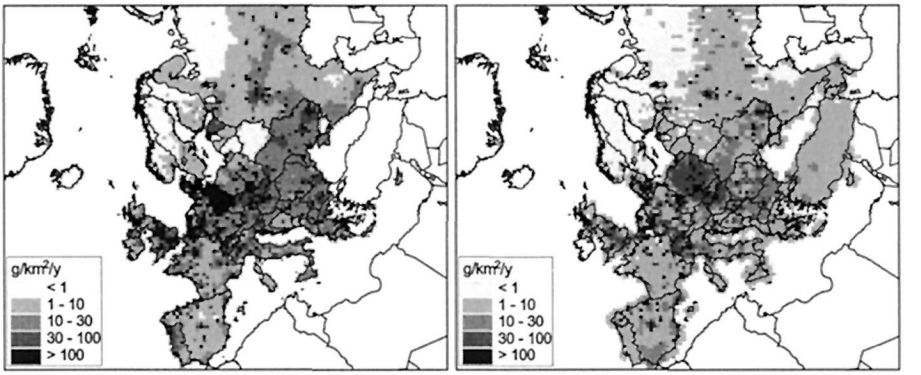

Figure 3. Spatial distribution of mercury anthropogenic emissions in Europe: (a) for 1990 (Berdowski et al., 1997) and (b) for 2000 (b) Pacyna et al. (2003).

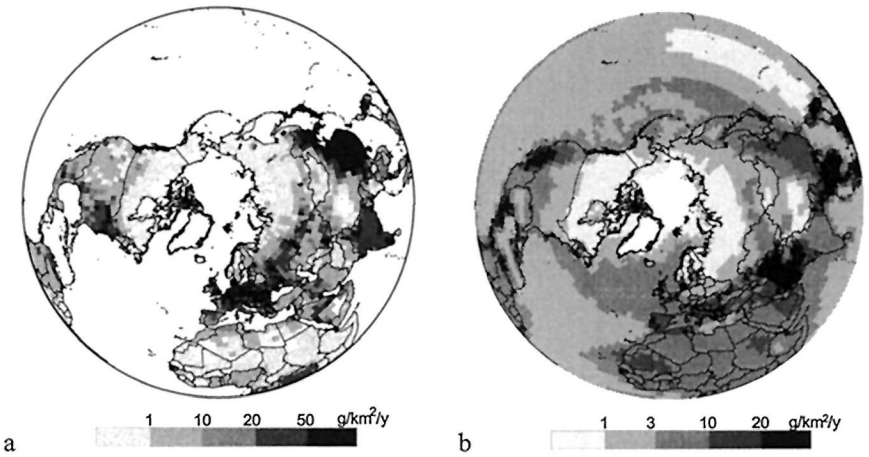

Figure 4. Hemispheric distribution of anthropogenic (a) and natural (b) mercury emissions.

To simulate mercury atmospheric transport on the hemispheric level a global emission inventory prepared by Pacyna and Pacyna (2001) is used. The inventory includes the data for three mercury forms. The spatial resolution of the emission field is 1x1 degree. In accordance with these estimates the mercury anthropogenic emission in the Northern Hemisphere totalled 1900 tons in 1995. Its spatial distribution is shown in Figure 4a.

It is well-known that mercury enters the atmosphere from different natural emission sources. For modelling purposes this emission should be

assessed and spatially distributed. Lamborg et al. (2002) suggested that global natural emissions reach about 1800 t/y (1000 over land and 800 over the ocean). In order to obtain a spatial distribution of natural emission fluxes the total emission value was scattered throughout the globe depending on the mercury content in soils and the surface temperature. It was assumed that mercury evasion from the ocean was proportional to the primary organic carbon production (Travnikov and Ryaboshapko, 2002).

The obtained distribution of natural mercury emissions in the Northern Hemisphere is shown in Figure 4b. The highest emission values are typical of the so-called geochemical mercuriferous belts with increased content of mercury in soils. The emission flux from seawater is lowest in the Middle Pacific and highest in internal seas and coastal waters at low latitudes. The total natural emission of mercury in the Northern Hemisphere constitutes about 1220 t/y.

In addition to the emission data the models require some other input information like meteorological data, characteristics of underlying surface, concentrations of different reactants involved into mercury atmospheric chemistry and so on. Meteorological data used in the calculations on the regional level are based on the Re-analysis project data (Kalnay et al., 1996). These data are prepared by National Centers for Environmental Predictions together with National Center of the Atmospheric Research (NCEP/NCAR). Meteorological data necessary for the hemispheric model are based on NCEP/NCAR Re-analysis data and processed by the low atmosphere diagnostics system (SDA) developed in co-operation with Hydro-meteorological Centre of Russia. The system provides 6-hour weather prediction data along with estimates of the atmospheric boundary layer parameters and covers the Northern Hemisphere.

To take into account information on land cover of the Earth surface the regional model uses data on fifteen types of underlying surface based on (Posh et al., 2001). In hemispheric model 25-category land cover data set from NCAR Mesoscale Modelling System (MM5) is used (Guo and Chen, 1994). Since the model formulation does not require highly detailed specification, the original 25-category data were reduced to five general categories (urban, forests, grassland, bare land, and glaciers) and redistributed over the model grid.

To describe chemical transformations one has to know spatial and temporal distribution of the reactants concentration (such as ozone and sulfur dioxide) in the atmosphere. The regional model uses the calculated fields of main reactants provided by EMEP Meteorological Synthesizing Centre - West. Global monthly mean data on ozone and SO_2 concentrations

in the atmosphere were kindly presented by Dr. Malcolm Ko (Wang et al., 1998; Chin et al., 1996). Besides, for the aqueous-phase chemistry cloud water was characterised by pH value equal to 4.5 and chloride ion concentration in cloud water equal to $7 \cdot 10^{-5}$ M (Acker et al., 1998). Following Seigneur et al. (2001) air concentration of molecular chlorine in the lowest model layer over the ocean is assumed to be 100 ppt at night-time, 10 ppt during the day and zero concentration over land.

POLLUTION LEVELS IN EUROPE

In accordance with the EMEP work programme MSC-E carries out a modelling assessment of mercury transboundary pollution within Europe. The main objective of the work is to evaluate mercury concentration levels in air and in precipitation. Besides, the modelling approach gives a possibility to calculate dry and wet deposition of mercury over Europe and transboundary transport between countries.

The atmospheric mercury budget for the EMEP region can be described by the following items: emissions, depositions, inflow and outflow fluxes. The main items of the budget of mercury emitted in Europe for the year 2000 are shown in Table 2. As seen from the table GEM enters mainly into the global mercury cycle. Since other mercury forms - TPM and RGM have lifetimes from hours to days, most of these species are deposited within the region. In general Europe is a net source of mercury for the global atmosphere (its emission exceed deposition). It should be kept in mind that a huge mass of atmospheric mercury (in comparison with annual European emission) enters and leaves the EMEP reservoir via lateral boundaries. This is conditioned mostly by the long-living form of elemental mercury. However, TPM can also be generated in the atmosphere due to chemical transformations. This can explain the fact that deposition of TPM plus its transport outside the EMEP region is higher than TPM emission.

Levels of mercury concentration in air are rather smooth due to its long lifetime in the atmosphere and due to the significant contribution of globally distributed mercury (Figure 5a). The highest concentration values exceed the global background only by a factor of 2-3. Maximum values of GEM concentrations in 2000 were obtained in Greece (6.3 ng/m^3), Slovakia (4.2 ng/m^3), Poland (4.1 ng/m^3) and the eastern part of Germany (4.0 ng/m^3). Regions with relatively low air concentrations are in the north of Scandinavia, where computed concentrations lay within 1.7 - 1.9 ng/m^3.

Table 2. Atmospheric budget of mercury emitted in Europe for EMEP region in 2000, t/y.

Budget item	Mercury forms			
	GEM	RGM	TPM	ΣHg
Total emission	369	59	31	459
Natural and re-emission	258	0	0	258
Direct anthropogenic	111	59	31	201
Total depositions *	4	57	29	90
Output from EMEP domain	360	2	7	369

* taking into account chemical transformations within the atmosphere: 5 tonnes of GEM are oxidized and deposited as TPM.

In contrast to concentrations in air mercury depositions reveal high gradients from "hot spots" in Central Europe to the periphery of the continent (Figure 5b). The deposition intensity in different parts of Europe can differ by more than an order of magnitude. Total depositions of mercury are mostly formed by depositions of oxidised mercury forms - TPM and RGM. Since a considerable fraction of these forms has basically anthropogenic origin, the deposition maxima are usually strongly associated with the anthropogenic sources. High deposition levels are characteristic of Central and Southern Europe. The highest values of average mercury deposition per country – about 40 g/km^2/y are in Slovakia, Poland and Belgium. This is caused both by high national emissions and transboundary transport from neighbouring countries. It should be kept in mind that for individual grid cells the deposition values could exceed country average ones by an order of magnitude (the maximum value was 250 g/km^2/y).

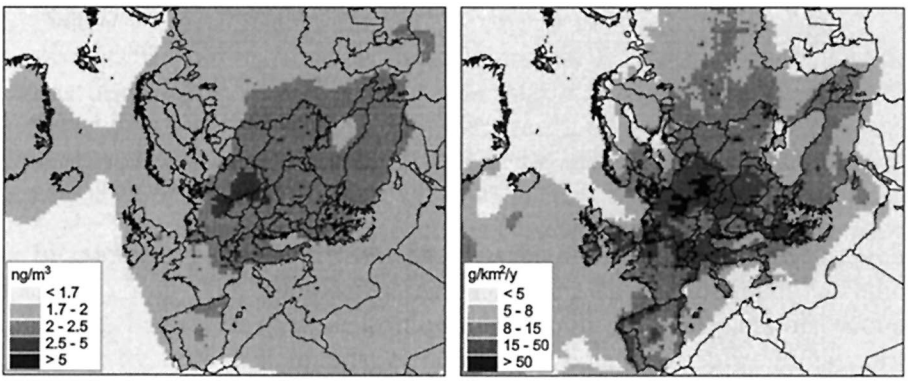

Figure 5. Annual mean concentrations of GEM (a) and total deposition (b) within EMEP in 2000.

In most part of the European region the levels of mercury deposition fluxes are between 10-20 g/km^2/y. Relatively low depositions are seen in the European North. In the central part of Scandinavia and in Northern Russia deposition fluxes as a rule do not exceed 10 g/km^2/y.

TRANSBOUNDARY POLLUTION

Assessment of transboundary depositions for each European country due to long-range atmospheric transport is the main task of EMEP model calculations. The results of the calculations are presented as a matrix of country-to-country depositions (Ilyin and Travnikov, 2003). In Table 3 a simplified version of the mercury deposition matrix is presented. Here for each EMEP country the two major sources countries of transboundary pollution are shown for each receiving country. In addition to this, contributions from totality of natural emission, secondary anthropogenic re-emission and remote (non-European) anthropogenic sources (NSR sources) are given. It is important to stress that this fraction does not indicate the pure natural contribution to depositions, but in fact it is a combination of natural inputs, inputs from remote anthropogenic sources and inputs due to previous anthropogenic pollution (re-emission).

Analysis of the table demonstrates that transboundary pollution can be very important for most European countries. For example, two neighbouring countries - France and Germany - contribute 40% of total deposition to Belgium. In some countries the main contribution is given by national sources. The highest absolute input of transboundary transport to mercury pollution (above 1 t/y) is characteristic of countries with large territories such as Russia, Poland, France, etc. It is typical for all countries that a considerable share of mercury deposition is caused by NSR sources, located all over the globe.

POLLUTION BUDGETS FOR INDIVIDUAL COUNTRIES

EMEP should provide each member-country with a detailed analysis of transboundary pollution.

Table 3. Mercury depositions on countries-receptors and contributions of different sources into the depositions (a fragment of total country-to-country matrix).

Country - receptor	Total deposition, (tonnes)	Contribution to the deposition from different sources, %				
		Main countries – sources*		Other EMEP countries*	Own sources*	NSR sources
Austria	1.35	Italy 8	Germany 4	17	15	56
Belgium	1.05	France 38	Germany 4	8	30	20
Bulgaria	1.88	Romania 9	Greece 7	8	36	40
Czech Rep.	1.97	Germany 18	Poland 14	11	31	26
Denmark	0.65	Germany 16	Poland 3	10	40	31
Finland	2.41	Poland 4	Germany 3	8	3	82
France	8.43	Spain 7	Switzerl. 2	5	43	43
Germany	10.48	France 4	Switzerl. 2	10	61	23
Greece	3.04	Bulgaria 3	Romania 1	3	69	24
Hungary	1.87	Slovakia 14	Romania 3	12	42	29
Italy	4.82	France 3	Spain 2	2	52	41
Netherlands	0.69	France 20	Belgium 13	21	16	30
Norway	2.60	Germany 3	Poland 3	5	6	83
Poland	11.99	Germany 10	Czech R. 4	7	61	18
Romania	3.69	Hungary 4	Poland 3	13	41	39
Russia	26.92	Ukraine 3	Poland 3	5	13	76
Slovakia	1.61	Hungary 11	Poland 6	10	49	24
Slovenia	0.43	Italy 12	Austria 2	10	24	52
Spain	6.65	Portugal 3	France 1	1	55	40
Sweden	2.88	Germany 7	Poland 7	12	1	73
Switzerland	1.09	France 10	Italy 7	3	47	33
Ukraine	7.92	Poland 7	Romania 3	12	32	46
UK	3.43	France 2	Ireland 2	3	56	37

* Only anthropogenic emissions.

Examples of two countries - Austria and Poland are considered below. The first one is a typical country-receiver of mercury pollution while the second one is a country-source (see Table 3). Information on pollution of any other European countries is available on the Internet: www.msceast.org/countries/. This information is intended to help national experts in developing abatement strategies concerning mercury emissions. Indeed, even very significant reduction of national emission can give no effect in a given country if the pollution levels are determined mainly by transboundary pollution.

The pie diagrams in Figure 6 present mercury depositions to Austria and Poland caused by national and external sources in 2000. In the case of Austria the main contribution to the deposition is made by neighbouring countries and NSR. Own sources give only 15% of the total value. The opposite situation is seen in Poland. Here national emission sources dominate. Nevertheless, the contribution of NSR is significant.

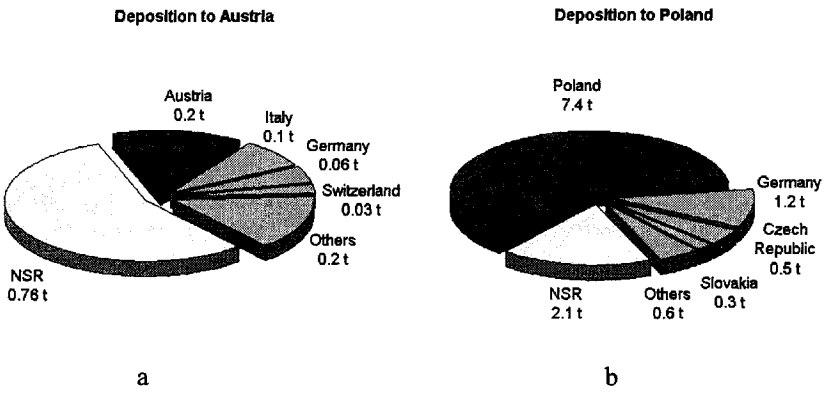

Figure 6. Mercury depositions over Austria (a) and Poland (b) from different emission sources in 2000.

The contribution of transboundary transport is non-uniformly distributed over a country. To develop national abatement strategies it is important to know the spatial distribution of transboundary pollution within a given country. Figure 7 illustrates the patterns of transboundary contributions from anthropogenic sources over the territories of the considered countries. Their regions neighbouring the countries with powerful emission sources are mostly impacted by external anthropogenic sources. In some regions of Austria the external contribution can reach 50%. In Poland noticeable contribution of transboundary mercury pollution (up to 85%) can be found in western parts of the country.

LONG-TERM POLLUTION TRENDS

According to the modelling results, emission reductions have resulted in the decrease of heavy metal depositions over the major part of the European territory. On the whole, in the period from 1990 to 2000, mercury deposition in Europe decreased 1.5 times. During the period of 1990–2000 anthropogenic mercury emissions in Europe reduced more than 2 times – from 420 to 201 t/y.

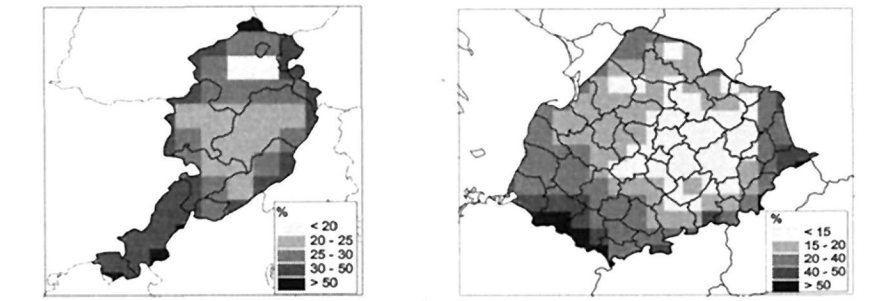

Figure 7. Contribution of external anthropogenic sources to mercury depositions to Austria (a) and Poland (b) in 2000, % (calculated in 50*50 km grid).

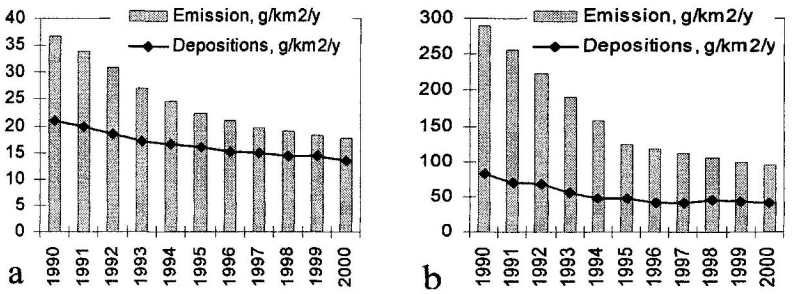

Figure 8. Trends of mercury emissions and calculated depositions during the period of 1990-2000: in Europe as a whole (a) and in Belgium (b).

The smaller decrease of deposition in comparison with anthropogenic emissions is conditioned by the contribution of natural sources and re-emission, as well as by growing global emissions. This difference in the trends for Europe as a whole is demonstrated by Figure 8a.

Some countries reduced their emissions significantly. However, the effects of the reduction on mercury deposition can be different. In Belgium, for example, the national emission dropped 3.5 times (Figure 8b). However, this did not lead to the same decrease in deposition, which reduced less than twice. Such lack of correspondence can be explained by the fact that mercury deposition in Belgium is determined primarily by European transboundary pollution.

ROLE OF MERCURY SPECIATION

From the viewpoint of developing abatement strategies, both on national and pan-European levels, it is very important to know which individual mercury forms are mostly responsible for elevated levels of deposition. Figure 9a demonstrates the mercury deposition field caused by emissions of only elemental mercury. The field is even and the deposition values are not high. This means that the reduction of emissions of this mercury form could not lead to a considerable decline of the deposition in Europe. If only oxidised mercury emissions are considered (Figure 9b) one can see that just these forms are primarily responsible for the elevated mercury depositions. Hence, emission reduction of oxidised mercury forms is more important to decrease atmospheric loads in the most polluted areas of Europe. At the same time reduction of GEM emissions is more important in a global context.

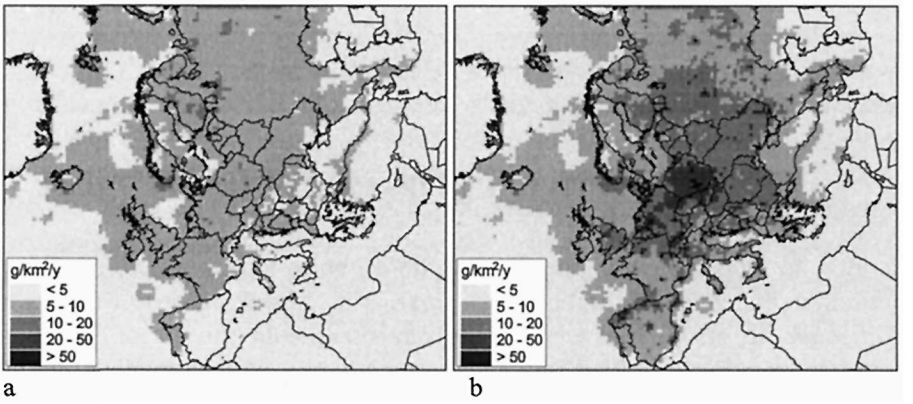

Figure 9. Mercury depositions caused by elemental mercury emission only (a) and oxidized mercury emission only (b).

HEMISPHERIC TRANSPORT AND DEPOSITION

As shown above the role of globally distributed anthropogenic sources can be significant for mercury deposition levels in different regions of the Northern Hemisphere. The results obtained by the hemispheric model demonstrate that the contribution of intercontinental transport to mercury deposition over Europe is about 40% of the total value. About half of mercury deposition to such a remote region as the Arctic is due to long-

range atmospheric transport from anthropogenic emission sources. Asian emissions nowadays play the most important role on the global level.

Figure 10a displays the calculated distribution pattern of mercury concentrations in the surface air for the Northern Hemisphere. On the global level it possible to distinguish some "hot spots". The highest concentrations are typical of Europe and South-eastern Asia. As a result of mixing processes, levelling of mercury concentrations in the troposphere takes place, and the global mercury background is established. Even in the remote parts of the Atlantic and Pacific oceans, as well as in the Arctic, mercury concentration in the surface air does not fall below 1.4 ng/m^3. In accordance with the calculations the background mercury concentrations in air masses coming to Europe from the Atlantic are about 1.6 ng/m^3 for GEM, 10 pg/m^3 for TPM and 0.3 pg/m^3 for RGM. The background values calculated by the hemispheric model are used as boundary conditions for regional calculations.

The distribution of mercury depositions in the Northern Hemisphere is shown in Figure 10b. The highest deposition values are characteristic of regions of high anthropogenic emissions (Eastern Asia, Europe, North America). Depositions in different parts of the hemisphere depend significantly on intercontinental atmospheric transport. For example, the deposition levels in Europe from global non-European sources amount to about 10 g/km^2/yr. This value is comparable with those from European sources.

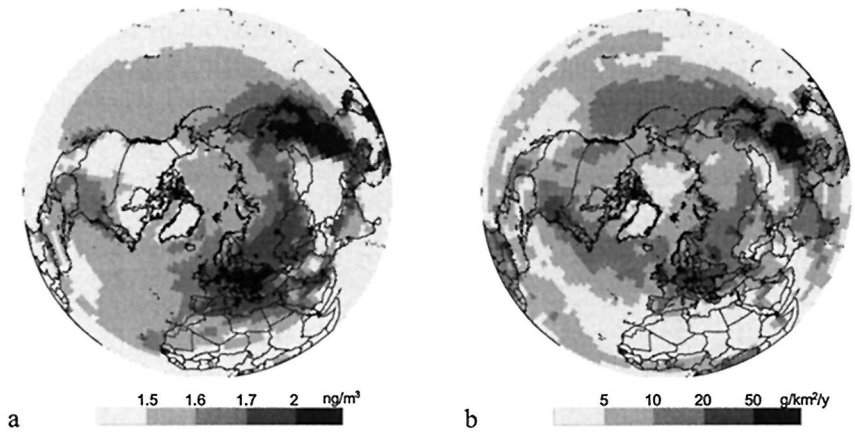

Figure 10. Hemispheric distribution of mean annual concentration of total gaseous mercury, ng/m^3 (a) and total depositions, g/km^2/yr (b).

Relative contributions of own and external sources to mercury deposition in Europe are presented in Table 4. The main contribution is given by European anthropogenic sources. However, the contribution of external sources is comparable and makes up about 40%. The most significant non-European input is made by Asian sources (15%) and mercury evasion from the ocean surface (12%). American sources contribute about 5%. It should be noted that the anthropogenic component of mercury deposition to Europe considerably exceeds the natural one and amounts to 75% of the total.

Table 4. Contributions of different regions to the total annual mercury deposition over the European region, %.

Source	Europe	Asia	Americas	North Africa	World Ocean	SH*
Anthropogenic	59	11	3	1	-	4
Natural	2	4	2	1.5	12	

* SH – Southern Hemisphere

MODEL VALIDATION

To confirm the quality and reliability of the modelling results the modelled data were compared with monitoring data obtained mainly by the EMEP monitoring network. Unfortunately, only few EMEP monitoring stations measure mercury on a routine basis, and practically all of them are located in North-western Europe. Locations of EMEP monitoring stations are shown in Figure 11a. The co-ordinates and description of the stations can be found in EMEP technical reports (Ilyin et al., 2002; www.nilu.no/projects/ccc). The comparison was carried out for annual mean mercury concentrations in air and in precipitation. To verify the hemispheric model long-term measurements performed in Europe and in North America (NADP/MDN, 2002) are used. In addition, data from short-term measurements performed during episodic measurements over the Atlantic and in Eastern Asia are considered. The locations of monitoring stations and sites of episodic measurements are shown in Figure 11b.

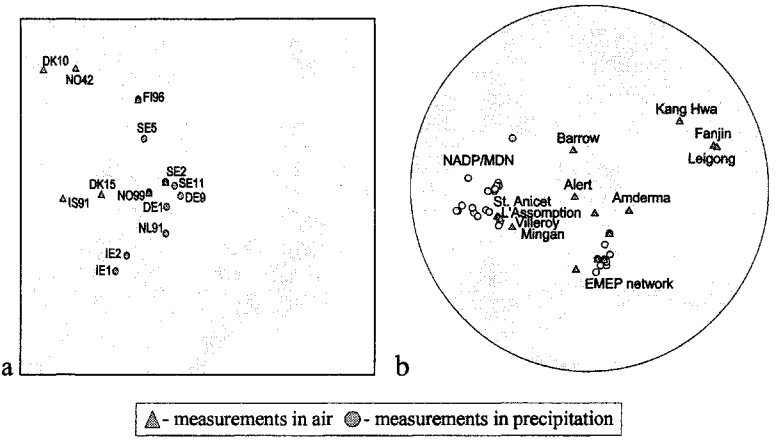

Figure 11. Locations of monitoring stations and sites of episodic measurements.

As shown in Figure 12a, the measured and calculated GEM concentrations vary within narrow limits. The agreement between average measured and calculated GEM concentrations is within 10-15%, however, the difference for individual stations can exceed 25%. Generally, the model somewhat overestimates measured values. For almost all stations measuring mercury content in precipitation the measured and calculated deposition fluxes agree within a factor of 2 (Figure 12b). On the whole mercury wet deposition fluxes measured at stations in 2001 were slightly overestimated by the model. An appreciable overestimation is noted for German station DE9. The reason for this can be connected with uncertainties in spatial distribution of the anthropogenic emissions of different mercury forms.

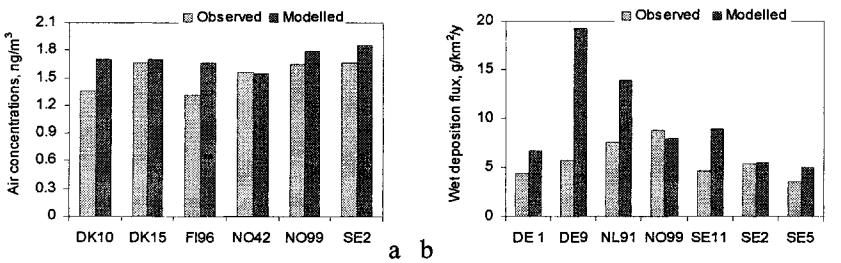

Figure 12. The comparison of measured and calculated values: (a) for GEM concentrations; (b) for wet deposition.

A 2-week measuring campaign focused on TPM and RGM (Schmolke et al., 1999; Munthe et al., 2003; Wängberg et al., 2003) gave a unique opportunity to verify the model for these mercury forms which are not measured on a routine basis. The measurements were performed simultaneously at two German, two Swedish and one Irish monitoring stations. The results of the comparison for TPM and RGM measured at the most polluted German station are presented in Figure 13. In the case of RGM the model generally overpredicts the measurements by a factor of about 2. For TPM the agreement is much better – the difference makes up less than 25%, and the correlation factor is high (0.72). It should be kept in mind that the measurements of TPM and especially RGM are very uncertain. Hence, the mentioned disagreement can be partly explained by this fact.

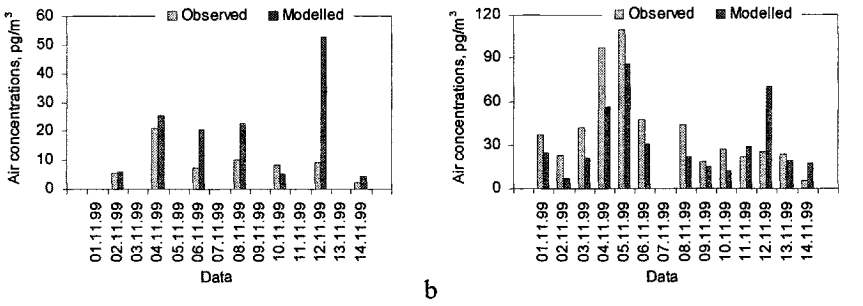

Figure 13. Modelled RGM (a) and TPM (b) concentrations by EMEP regional model against observations at German monitoring station Neuglobsow.

For the hemispheric model the results of the comparison of calculated and measured GEM concentrations are given in Figure 14a. As seen from these data, the model predicts air concentrations of mercury in background regions (~1.5 ng/m^3) rather accurately. Some underestimation of measured values takes place in the regions with an increased concentration of mercury (South-eastern Asia). In general, the difference between measured and predicted values does not exceed 30%.

The difference between predicted and measured values of the annual wet depositions of mercury is displayed in Figure 14b. The accuracy of model prediction in this case is somewhat lower, because deposition fluxes highly depend on the precipitation amount - the model input parameter with a considerable degree of uncertainty. However, in general, the ratio between measured and predicted values is close to unity, and the maximum difference between them does not exceed a factor of two.

A very useful approach to model validation is the comparison of the EMEP operational model with other scientific models used by national

experts. Nine different models have been included in this intercomparison study: ADOM (Germany), CMAQ, HYSPLIT, AER (USA), GRAHM (Canada), EMAP (Bulgaria), MCM (Sweden), DEHM (Denmark) and EMEP operational model. Their descriptions can be found in (Ryaboshapko et al., 2002; Ryaboshapko et al., 2003).

At the first stage of the study only schemes of chemical transformations were compared. The results demonstrated that all the models predicted increases in mercury concentration in cloud water during the first hours of modelling experiment. The range of the predicted maximum concentrations was from 80 to 150 ng/L.

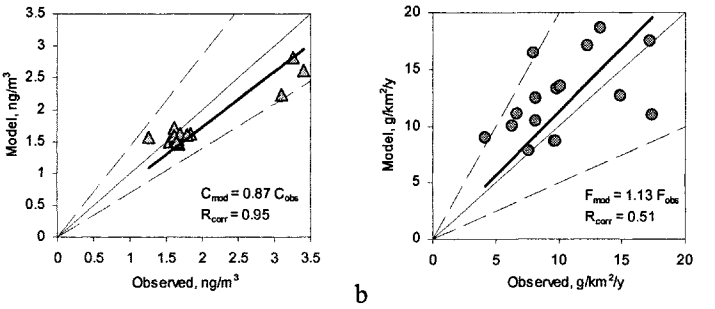

Figure 14. Comparison of measured and calculated by the hemispheric model: (a) GEM concentrations; (b) wet depositions. Dashed lines: (a) – ±30%, (b) – factor 2.

For the second stage of the comparison the data from the short-term measuring campaign mentioned above was used. Seven models calculated EGM, TPM and RGM on sample-by-sample basis for four monitoring stations in Germany, Sweden and Ireland. The scattering of the calculated values for TPM and especially for RGM in the individual samples was very high. However, the mean values for all stations and for all samples were in rather good agreement (Ryaboshapko et al., 2003). Figure 15 presents comparison results for "models vs. observations" and "models vs. models". In the case of elemental mercury all the models are in good agreement both with the observations and between each other. The data for RGM and TPM are characterised by much larger scattering. In the case of RGM the difference between the lowest and the highest modelling values reaches an order of magnitude. For all parameters of the comparison the EMEP operational model demonstrated acceptable agreement both with the observations and with the results of the other scientific models. This gives some confidence that the model can be used for the purposes of the

Convention and can provide the participating countries with information of acceptable reliability.

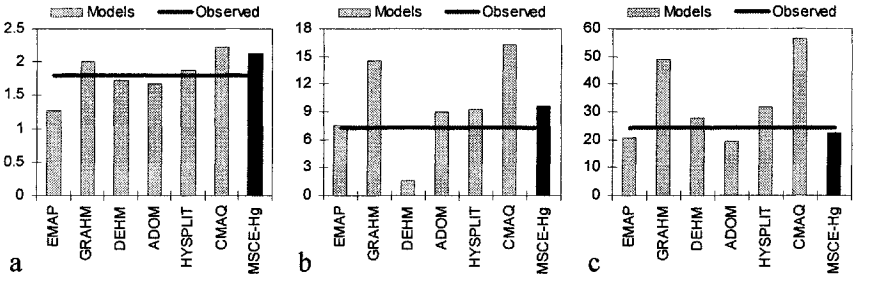

Figure 15. Comparison of different models between each other and with measurements for GEM (a), TPM (b) and RGM (c).

FURTHER DIRECTIONS OF MODEL DEVELOPMENT

It is possible to foresee two very important directions of further development of EMEP mercury models. The first one is connected with the necessity to assess mercury accumulation in environmental compartments and its secondary emission or re-emission to the atmosphere. The second direction is consideration in the modelling scheme of a newly discovered phenomenon - Arctic mercury depletion.

During a period of active usage of mercury in human activity more than one million tonnes was extracted from the lithosphere, and at least half of that came to the atmosphere (Travnikov and Ryaboshapko, 2002). A great amount of mercury was emitted in the process of coal combustion. Fitzgerald and Mason (1996) believe that 95% of previously emitted mercury has being accumulated in soil over the globe. The enhanced content of mercury in soils should inevitably lead to its re-emission to the atmosphere.

Consideration of mercury re-emission processes is very important for operational modelling of mercury transport in the atmosphere. One of the possible ways to describe this process is application of a dynamic multi-component model describing the mercury cycle in the environment during the entire period of pronounced anthropogenic impact (about 500 years Hylander & Meili, 2003). Due to very long period of supposed calculations and the contemporary level of knowledge on mercury behaviour in different environmental compartments the model cannot be very detailed. In this

content a box-modelling approach seems to be the most acceptable for estimation of mercury accumulation in the environment and re-emission. On the other hand, it should have enough spatial resolution to provide input information for operational atmospheric transport models at regional and global levels. The concept of the box-modelling approach was evolved in works by Jonasson and Boyle (1971), Ribeyre et al. (1991), Hudson et al. (1995), Jackson (1997), and Lamborg et al. (2002).

The first attempt to assess mercury re-emission from European soils was done using the EMEP regional mercury model in conjunction with a simple box model (Ryaboshapko and Ilyin, 2000). Accumulated depositions during the last century were calculated by the regional model. The box model considered European soils as a single reservoir with two output fluxes – re-emission and hydrological leaching. Mercury lifetime in the box according to re-emission was assumed to be 400 yr, and according to the leaching – 950 yr. Under accepted assumptions the model predicted that by the end of 20^{th} century the total re-emission in Europe could make up 50 t/yr. The value seems to be not very high but one should keep in mind that it is the total value for the whole European territory. In some heavily polluted areas (Eastern Germany, for example) the re-emission can nowadays exceed the current direct anthropogenic emission.

To provide more accurate re-emission assessment a modeller should possess quantitative information on the mercury cycle in soils, because the retention time of mercury in soils is the most crucial parameter. Besides, mercury deposition and accumulation should be calculated on the global scale. Finally, the fate of mercury in the environment compartments should be considered at least during last 500 years (Hylander & Meili, 2003).

Modelling assessment of the role of mercury depletion events (MDE) in the Arctic is a very challenging problem. Measurements show that MDE can provide very significant fluxes of mercury from the atmosphere into vulnerable Arctic ecosystems. To what extent this phenomenon is connected with anthropogenic influence on the mercury cycle is still an open question. Unfortunately, the mechanism of MDE is not fully understood.

Two attempts to model MDE were made recently (Christensen, 2001; Ilyin and Travnikov, 2003). EMEP calculations show that additional deposition of mercury due to MDE can be significant – about 50 t/yr or 20% of the total deposition. Figure 16 demonstrates the pattern of annual mercury deposition in the Arctic and the effect caused by MDE. One can see that the effect is the most pronounced along the shoreline of the Arctic Ocean. Here MDE can contribute more then 50% of the total deposition, and just here the life in the Arctic is the most active.

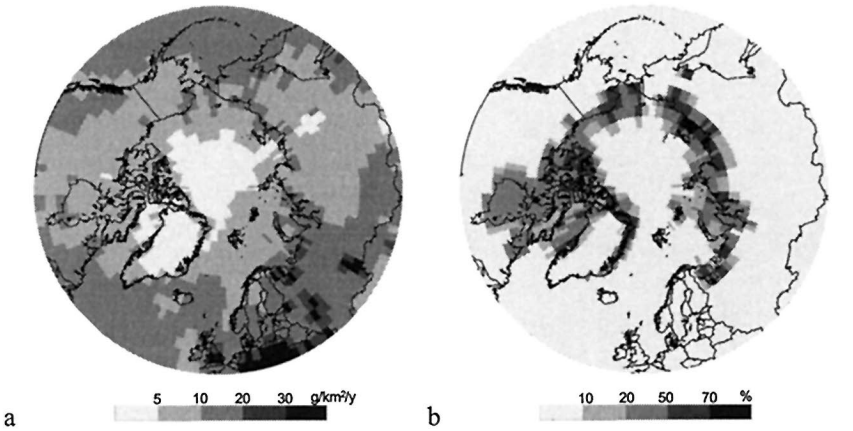

Figure 16. Spatial distribution of annual mercury depositions in the Arctic (a) and the contribution of MDE to the total deposition (b).

The main problem for a modeller to parameterise MDE is in the fact that the real "trigger mechanism" of the phenomenon is unknown. Christensen 2001 used a certain sun zenith angle as the trigger to start MDE. In the EMEP hemispheric model (Ilyin and Travnikov, 2003) prescribed temperature changes were applied. In both cases the approaches are phenomenological. They give a possibility to tie modelled MDE with known geophysical parameters. However, they do not allow one to predict a moment of the real onset of MDE. Probably, the beginning of the depletion is connected with explosive emission into the atmosphere of some bromine species from open water when leads appear very quickly during the spring drift of Arctic ice cover.

REFERENCES

Acker, K., Möller, D., Wieprecht, W., Kalass, D., and Auel, R. Investigations of ground-based clouds at the Mt. Brocken. *Fresenius J. Anal. Chem.*, 361, 59-64, 1998.

Berdowski, J.J.M., Baas, J., Bloos, J.P.J., Visschedijk, A.J.H., Zandveld, P.Y.J. The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, 1997; UBA-FB report 104 02 672/03, Apeldoorn, 239, 1997.

Bott, A. Monotone flix limitation in the area-preserving flux-form advection algorithm. *Month. Weath. Rev.*, 120, 2592-2602, 1992.

Brosset, C., Lord E. Mercury in precipitation and ambient air - A new scenario. *Water, Air Soil Pollut.*, 56, 493-506, 1991.

Bullock Jr. O.R., Brehme K.A. Atmospheric mercury simulation using the CMAQ model: formulation, description and analysis of wet deposition results. *Atmos. Environ.*, 36, 2135-2146, 2002.

Chin, M., Jacob D.J., Gardner, G.M., Foreman-Fowler M.S., Spiro, P.A., Savoie, D.L. A global three-dimensional model of tropospheric sulfate. *J. Geophys. Res.*, 101 (D13), 18,667-18690, 1996.

Christensen, J. Modelling of Mercury with the Danish Eulerian Hemispheric Model. The International Workshop on Trends and Effects of Heavy Metals in the Arctic, 18-22 June 2001, McLean, Virginia, USA, 2001

Easter, R.C. Two modified versions of Bott's positive-definite numerical advection scheme. *Month. Weath. Rev.*, 121, 297-304, 1993.

Fitzgerald, W.F., Mason, R.P. "The global mercury cycle: oceanic and anthropogenic aspects." In Global and Regional Mercury Cycles: Sources, Flaxes and Mass Balances." W.Baeyens, R.Ebinghaus and O.Vasiliev, eds. Dordrecht: NATO ASI Series, 2. Environment, Vol. 21., Kluwer Academic Publ., 1996; 85-108.

Guo, Y.-R., Chen, S. Terrain and land use for the fifth-generation Penn State/NCAR mesoscale modeling system (MM5): Pragram TERRAIN. NCAR Technical Note NCAR/TN-397+IA, National Center for Atmospheric Research, Boulder, Colorado, USA, 1994.

Hall, B. The gas phase oxidation of mercury by ozone. *Water, Air Soil Pollut.*, 80, 301-315, 1995.

Hudson, R.J.M., Gherini, S.A., Fitzgerald, W.F., Porcella, D.B. Anthropogenic influences on the global mercury cycle: a model-based analysis. *Water, Air Soil Pollut.*, 80, 265-272, 1995.

Hylander, L.D., Meili, M. 500 years of mercury production: global annual inventory by region until 2000 and associated emissions. *Sci. Tot. Env.*, 304, 13-27, 2002.

Ilyin, I., Ryaboshapko, A., Afinigenova, O., Berg, T., Hjellbrekke, A.-G. Evaluation of Transboundary Transport of Heavy Metals in 1999. Trend Analysis. EMEP MSC-E/CCC Technical Report 3/2001, 129 p., 2001.

Ilyin, I., Ryaboshapko, A., Afinigenova, O., Berg, T., Hjellbrekke, A.-G., Lee, D. Lead, Cadmium and Mecury Transboundary Pollution in 2000. EMEP MSC-E/CCC Technical Report 5/2002, 131 p., 2002.

Ilyn, I., Travnikov, O. Heavy Metals: Transboundary Pollution of the Environment. EMEP MSC-E Technical Report 5/2003, 96 p., 2003.

Iverfeldt, Å. Occurence and turnover of atmospheric mercury over the Nordic countries. *Water, Air Soil Pollut.*, 56, 252-265, 1991.

Iversen, T., Saltbones, J., Sandnes, H., Eliassen, A., Hov, O. Airborn tronsboundary transport of sulphur ans nitrogen over Europe - model descriptions and calculations. EMEP MSC-W Report 2/89, 1989; 92 p.

Jackson, T.A. Long-range atmospheric transport of mercury to ecosystems, and the importance of anthropogenic emissions - a critical review and evaluation of the published evidence. *Environ. Rev.*, 5, 99-120, 1997.

Jacobson, M.Z. Fundamentals of Atmospheric Modeling. Cambridge University Press, 656 p., 1999.

Jonasson I. R., Boyle R. W. Geochemistry of mercury. Spatial Symposium on Mercury in Man's Environment, Environment Canada, 15-16 February 1971, Ottawa, Canada, 1971.

Jonsen, J.E., Berge, E. Some preliminary results on transport and deposition of nitrogen components by use of Multilayer Eulerian Model. EMEP Meteorological Synthesizing Centre - West, Report 4/95, 25 p., 1995.

Kalnay, E., Kanamitsu, M., Kistle, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha S., White, G., Woollen, J., Zhu, Y., Leetmaa, A., Reynolds, R., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K.C., Ropelewski, C., Wang, J., Jenne, R., Dennis, J. The NCEP/NCAR 40-Year Reanalysis Project. *Bull. Am. Met. Soc.*, 77, 437-471, 1996.

Keeler, G., Glinsorn, G., Pirrone N. Particulate mercury in the atmosphere: its significance, transport, transformation and sources. *Water, Air and Soil Pollut.*, 80, 159-168, 1995.

Lamborg, C.H., Fitzgerald, W.F., Vandal, G.M., Rolfhus, K.R. Atmospheric mercury in northern Wisconsin: sources and species. *Water, Air and Soil Pollut.*, 80, 189-198, 1995.

Lamborg, C.H., Fitzgerald, W.F., O'Donnell, J., Torgersen, T. A non-steady-state compartmental model of global-scale mercury biogeochemistry with interhemispheric atmospheric gradients. *Geochim. Cosmochim. Acta*, 66, 1105-1118, 2002.

Lindberg, S.E., Meyers, T.P., Taylor, G.E., Turner, R.R., Schroeder, W.H. Atmospheric-surface exhange of mercury in a forest: results of modeling and gradient approaches. *J. Geophys. Res.*, 97, 2519-2528, 1992.

Milford, J.B., Davidson, C.I. The sizes of particulate trace elements in the atmosphere – a review. *J. Air Pollut. Control Assoc.*, 35, 1249-1260, 1985.

Munthe, J., Wängberg, I., Iverfeldt, Å., Lindqvist, O., Strömberg, D., Sommar, J., Gårdfeldt, K., Petersen, G., Ebinghaus, R., Prestbo, E., Larjava, K., Siemens, V. Distribution of atmospheric mercury species in Northern Europe: Final Results from the MOE Project. *Atmos. Environ.*, 304, S9-S20, 2003.

NADP/MDN. National Atmospheric Deposition Program (NRSP-3) / Mercury Deposition Network 2002. (Online Version: http://nadp.sws.uiuc.edu/mdn/)

Pacyna, E., Pacyna, J. Global emission of mercury from anthropogenic sources in 1995. *Water, Air and Soil Pollut.*, 137, 149-165, 2002.

Pacyna, J., Pacyna, E., Steenhuisen, F., Wilson, S. Global Mercury emissions. International Workshop "Long Range Transport", 16-17 September 2003, Ann Arbor, Michigan, USA. 2003.

Pekar, M. Regional models LPMOD and ASIMD. Algorithms, parameterization and results of application to Pb and Cd in Europe scale for 1990. 1996; MSC-E/EMEP, Technical Report 9/96.

Petersen, G., Munthe, J., Pleijel, K., Bloxam, R., Kumar, A.V. A comprehensive Eulerian modeling framework for airborne mercury species: Development and testing of the tropospheric chemistry module (TCM). *Atmos. Environ.*, 32, 829-843, 1998.

Petersen, G., Bloxam, R., Wong, S., Munthe, J., Krüger, O., Schmolke, S., Kumar, A.V.. A comprehensive Eulerian modelling framework for airborne mercury species: model development and applications in Europe. *Atmos. Environ.*, 35, 3063-3074, 2001.

Posh, M., Hettelingh, J.-P., de Smet, P.A.M., Downing, R.J. Calculations and mapping of critical thresholds in Europe. Status report 1997. RIVM, National Institute of Public Health and the Environment. P.O. Box 1, 3720 BA Bilthoven, The Netherlands, 1997.

Ribeyre, F., Boudou, A., Maury-Brachet, R. Multicompartment ecotoxicological models to study mercury bioaccumulation and transfer in freshwater system. *Water, Air and Soil Pollut.*, 56, 641-652, 1991.

Ruijgrok, W., Davidson, C.I., Nicholson, K.W. Dry deposition of particles. Implications and recommendations for mapping of deposition over Europe. *Tellus*, 47B, 587 – 601, 1995.

Ryaboshapko, A., Ilyin I. Mercury re-emission to the atmosphere in Europe. In: Proceedings of EUROTRAC Symposium 2000 Transport and Chemical Transformation in the Troposphere. P.M.Midgley, M.J.Reuther, M.Williams eds. Springer-Verlag Berlin Heidelberg (CD-annex), 2001.

Ryaboshapko, A., Bullock, R., Ebinghaus, R., Ilyin, I., Lohman, K., Munthe, J., Petersen, G., Seigneur, C., Wängberg, I. Comparison of mercury chemistry models. *Atmos. Environ.*, 36, 3881- 898, 2002.

Ryaboshapko, A., Artz, R., Bullock, R, Christensen, J., Cohen, M., Dastoor, A., Davignon, D., Draxler, R., Ebinghaus, R., Ilyin, I., Munthe, J., Petersen, G., Syrakov, D. Intercomparison study of numerical models for long-range atmospheric transport of mercury. Stage II. Comparison of modeling results with observations obtained during short-term measuring campaigns. MSC-E Technical Report 1/2003, 2003.

Sander, R. Henry's law constants available on the Web. EUROTRAC Newsletter 1997; No.18, 24-25

Schmolke, S.R., Schroeder, W.H., Munthe, J., Kock, H.H., Schneeberger, D., Ebinghaus, R. Simultaneous Measurements of Total Gaseous Mercury at Four Sites on a 800 km Transect: Spatial Distribution and Short Time Variability of Total Gaseous Mercury over Central Europe. *Atmos. Environ.*, 33, 1725-1734, 1999.

Seigneur, C., Karamchandani, P., Lohman, K., Vijayaraghavan, K., Shia, R.-L. Multiscale modeling of the atmospheric fate and transport of mercury. *J. Geophys. Res.*, 106 (D21), 27795-27809, 2001.

Shia, R.-L., Seigneur, Ch., Pai, P., Ko, M., Sze, N.D. Global simulation of atmospheric mercury concentrations and deposition fluxes. *J. Geophys. Res.*, 104 (D19), 23747-23760, 1999.

Slemr, F. Trends in atmospheric mercury concentrations over the Atlantic ocean and at the Wank summit, and the resulting constraints on the budget of atmospheric mercury. In Global and Regional Mercury Cycles: Sources, Flaxes and Mass Balances. W.Baeyens, R.Ebinghaus and O.Vasiliev eds. NATO ASI Series, 2. Environment, Vol. 21. Kluwer Academic Publ., Dordrecht, 33-84, 1996.

Travnikov, O., Ryaboshapko, A. Modelling of Mercury Hemispheric Transport and Depositions. EMEP MSC-E Technical Report 6/2002, 2002, 67 p.

Wang, Y., Logan, J.A., Jacob, D.J. Global simulation of tropospheric O3-NOx-hydrocarbon chemistry, 2, Model evaluation and global ozone budget. *J. Geophys. Res.*, 103 (D9), 10727-10755, 1998.

Wesley, M.L., Hicks ,B.B. A review of the current status of knowledge on dry deposition. *Atmos. Environ.*, 34, 2261-2282, 2000.

Wängberg, I, Munthe, J., Ebinghaus, R., Gårdfeldt, K., Sommar, J. Distribution of TPM in Northern Europe. *Sci. Tot. Env.*, 304, 53-59, 2003.

Xu, X., Yang X., Miller, D.R., Helble, J.J., Carley, R.J. Formulation of bi-directional atmosphere-surface exchanges of elemental mercury. *Atmos. Environ.*, 33, 4345-4355, 1999.