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Heavy metals:
transboundary pollution of the environment

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EXECUTIVE SUMMARY

In accordance with the EMEP Workplan for 2006 [EB.AIR/GE.1/2005/10/Rev.1] Meteorological Synthesizing Centre – East (MSC-E) and Chemical Co-ordinating Centre (CCC) continued their research activities in the field of heavy metal (HMs) atmospheric pollution assessment. The major goal of this work is to assess regional-scale levels of pollution in Europe of lead, cadmium and mercury by means of monitoring and atmospheric modelling. In October 2005 in Moscow EMEP Task Force on Measurements and Modelling (TFMM) held a workshop devoted to the review of MSC-E models. MSC-E paid special attention to further model development and analysis of its input data, following the recommendations of the workshop. Furthermore, MSC-E supported the Task Force on Heavy Metals (TFHM) in their preparatory activity for the review of 1998 Protocol on Heavy Metals, and collaborated with the Working Group on Effects in the field of development of the critical loads approach. Besides, MSC-E took part in preparation of the report, coordinated by the Task Force on Health, which is devoted to evaluation of health effects caused by long-range transport of heavy metals.

At present the EMEP monitoring network contains 63 stations measuring lead and cadmium, of which 26 stations measure concentrations of these metals both in air and precipitation. There are 18 stations where at least one mercury form is measured. The monitoring stations, however, are non-uniformly distributed over European territory: they are mainly located in Central and Northern Europe. Annual analytical intercomparison of national laboratories treating measurements of heavy metals indicate an essential improvement of data quality during the period of 1995-2004, but still there is need for improvement for some countries.

Measurements of heavy metal pollution levels in 2004 showed that the lowest concentrations of lead, cadmium and mercury were observed in Northern Scandinavia. In general, concentration levels increase towards the southeast of Europe.

In 2006, national data on emissions of priority metals (lead, cadmium and mercury) for 2004 were reported by 28 European countries, i.e. by about 60% Parties to the CLRTAP located in the EMEP region. Among them 16 countries re-calculated previously reported data. Because of these recalculations the emissions of some metals in some countries changed by more than 100%. Completeness of reported national total emissions is not sufficed in many countries. It leads to underestimation of national emission totals. According to officially reported data supplemented with unofficial expert estimates emissions in Europe between 1990 and 2004 decreased about 6 times for lead and almost twice for cadmium and mercury. However, in individual countries the magnitude of emission changes is highly variable.

MSCE-HM model has been reviewed at the EMEP/TFMM Workshop held in Moscow in October 2005. The Workshop concluded that the MSCE-HM model is suitable for the evaluation of the long-range transboundary transport and depositions of heavy metals in Europe. However, significant difficulties still remain with the official emissions for Pb and Cd. Besides, a number of recommendations, aimed at further improvement of heavy metal atmospheric modelling was drawn. The results of the workshop were adopted at the 7th Meeting of the Task Force on Measurements and Modelling (Helsinki, May 2006).

Following the recommendations of TFMM, a tentative parameterisation of wind re-suspension of particle-bound heavy metals (Pb, Cd, As, Ni and Cr) from natural surfaces (soil and seawater) was developed. Preliminary calculations demonstrate that wind re-suspension of some heavy metals can significantly contribute to airborne pollution in Europe. Another activity aimed at improvement of the model formulation was connected with evaluation of input meteorological data. The procedure of the

evaluation was initiated and preliminary results demonstrate that the meteorological fields used for the modelling process reasonably well agree with reference ECMWF data. MSC-E extended the model parameterisation for calculations of the atmospheric transport and depositions of the second priority metals (As, Cr, and Ni). Pilot calculations of concentrations and depositions of these metals demonstrated a reasonable agreement with available measurements.

Evaluation of lead, cadmium and mercury depositions, concentrations and source-receptor relationships for 2004 was carried out on the basis of officially reported emission data. For countries wihs national emissions not available, the expert estimates of [*Denier van der Gon et al*., 2005] were applied. According to the modelling results the spatial distribution of environmental pollution levels of heavy metals in 2004 was highly non-uniform. The deposition flux in different parts of Europe can differ by more than an order of magnitude. High deposition levels are characteristic of Central and Southern Europe, the lowest levels – of Northern Europe.

The influence of the transboundary transport on heavy metal depositions in Europe is significant. More than a half of anthropogenic depositions of heavy metals is defined by the transboundary transport in 33, 30 and 23 European countries for lead, cadmium and mercury, respectively. Fraction of national emissions of lead and cadmium contributing to the transboundary transport in Europe ranges from 60% to 80% for different countries. In case of mercury, this fraction is commonly higher than 80%.

Modelled concentrations of lead and cadmium in air and in precipitation are well correlated with the observed data. The modelled levels of lead underestimate measurements by around 30%, of cadmium – by a factor of 2, which can be caused by underestimated anthropogenic or natural and historical emissions. Mercury concentrations in air were reproduced by the model with high accuracy. The difference between modelled and measured values does not exceed 10%. Modelled concentrations of mercury in precipitation satisfactorily agree (within 25%) with measurements at most of the stations.

The modelling results for lead and cadmium were also extensively evaluated using different emission scenarios. Their analysis have shown that the addition of natural and historical emissions of lead resulted in significant improvement of modelling results against measurements. In general, modelling results based on the unofficial expert emission estimates developed in the EU ESPREME project significantly better agree with measurements than those based on the official emissions data.

Mercury hemispheric transport and depositions were evaluated at the hemispheric scale. Comparison of pollution levels estimated for 1996 and 2000 showed that the most significant decrease of mercury depositions took place in Europe and North America because of reduction of anthropogenic emissions. Along with the estimates of the intercontinental transport, the hemispheric model was applied for implementation of the one-way nesting to the regional modelling of mercury pollution. For this purpose monthly mean concentrations of three mercury forms (elemental, reactive gaseous and particulate) were calculated with the hemispheric model at the EMEP domain boundaries and were assimilated by the regional model.

In the framework of cooperation with the Working Group on Effects MSC-E performed calculations of atmospheric depositions of heavy metals to ecosystems for the development of the effects-based approach. The depositions were calculated for three emission scenarios: emissions for 2000 based on officially reported data with TNO expert estimates, and two TNO emission projections for 2020.

MSC-E contributed to the work of the Task Force on Heavy Metals by preparation of relevant information on modelled and measured pollution levels of lead, cadmium and mercury, as well as of overview of heavy metal emissions within UN ECE region. These contributions were included in TFHM background document prepared as a scientific basis for the review of the Protocol on Heavy Metals. MSC-E also supported the organization of the second meeting of the EMEP/TFHTAP (Task Force on Hemispheric Transport of Air Pollution) (Moscow, Russia, June 2006) and presented information on its activities in regional and hemispheric mercury modelling.

The EMEP Centres were also involved in cooperation with other subsidiary bodies to the Convention as well as international organizations and national programmes (European Commission, HELCOM, OSPAR, UNEP). The main results were discussed at a number of scientific conferences, workshops and expert meetings.

CONTENTS

INTRODUCTION

Pollution of the environment by heavy metals (HMs) is the subject of concern of a number of national and international bodies. In 1998 a number of Parties to the Convention on Long-Range Transboundary Air Pollution (CLRTAP, hereinafter the Convention) signed the Protocol on Heavy Metals (Protocol). The aim of the Protocol is to control atmospheric emissions of toxic metals (lead, cadmium and mercury). In accordance with the Protocol the Co-operative Programme for Monitoring and Evaluation of Long-Range Transmission of Air Pollutants in Europe (EMEP) provides the assessment of pollution levels of heavy metals in the European region. Measurements of heavy metal concentrations in the air and precipitation are carried out at the EMEP monitoring network under the methodological guidance of the Chemical Coordinating Centre (CCC). Along with that Meteorological Synthesizing Centre – East (MSC-E) performs the model assessment of depositions and air concentrations of heavy metals throughout the European region as well as the transboundary fluxes between the European countries.

The goal of this report is to summarize activities of EMEP countries and technical centres (first of all, CCC and MSC-E) concerning emissions, monitoring and modelling of heavy metals within the EMEP region.

Primary information about levels of heavy metals and their temporal and spatial trends in the environment is obtained via monitoring. For the needs of the Convention the monitoring of HMs is performed in the EMEP network. Since 1990 around 90 stations have been involved in the measurement activity. The stations measure concentrations in air, and in precipitation and estimate wet deposition fluxes. Measurement samples are analysed by about 20 laboratories located in various European countries. The laboratories take part in annual intercalibration studies, supervised by CCC. The aim of these studies is evaluation of quality of measurement information.

Modelling is used to assess HMs pollution levels along with measurements. Measurements are performed at limited number of stations. Besides, it is hardly possible to deduce information about source-receptor relationships from measured data. Modelling, in its turn, allows providing information about pollution levels over entire EMEP region with identification of individual source countries. Obviously, the modelled information has to be reliable. In particular, the model should be capable to reproduce measured values and their trends. Modelling of HMs transport and deposition for the purposes of the Convention is carried out by MSC-E.

In order to evaluate the reliability of the MSC-E regional-scale model for heavy metals (Pb, Cd, Hg) the procedure of the model review has been launched. The process of the review included uncertainty analysis, model intercomparison studies and comparison of modelled and measured concentrations and depositions. In October 2005 in Moscow EMEP Task Force on Measurements and Modelling (THMM) held a Workshop on the review of the EMEP models on HMs and POPs. The main conclusion of the workshop was that “MSC-E model is suitable for the evaluation of long range transboundary transport and depositions of HMs in Europe” [*ECE/EB.AIR/GE.1/2006/1*]. In addition to this, the workshop formulated recommendations for further improvement of MSC-E models. Following these recommendations, MSC-E started a number of long-term activities aimed at improving of its models. In particular, MSC-E has developed a parameterisation of meteorologically-driven emissions from land and sea surfaces, has started evaluation of meteorological data and included other metals (arsenic, nickel and chromium) to modelling schemes in addition to Pb, Cd and Hg.

Main directions of EMEP activity concerning heavy metals for 2006 are outlined in the EMEP Workplan for 2006 [*EB.AIR/GE.1/2005/10/Rev.1*]. According to the workplan, MSC-E carried out evaluation of pollution levels of lead, cadmium, and mercury in the EMEP region and mercury over the

northern hemisphere. Source-receptor relationships for the atmospheric transport of lead, cadmium and mercury between countries of Europe were established. Besides, pilot calculations of depositions and concentrations of arsenic, chromium and nickel were performed. Modelled results were compared with available measurements from EMEP network. Modelling tests with the use of different emission scenarios were undertaken. The results of collaboration with the Task Force on Measurements and Modelling (TFMM), the Working Group on Effects (WGE), the Task Force on Heavy Metals (TFHM) and the Task Force on Hemispheric Transport of Air Pollution (TFHTAP) are highlighted in the report.

The progress in the field of heavy metals, achieved by CCC and MSC-E is summarized in this status report. The report consists of introduction, five sections, conclusions, and annexes. Brief content of the sections of the report is given below.

Section 1 is focused on the monitoring activities in the field of heavy metals. The measurement network is described, and the measured pollution levels (concentrations in the air and precipitation) are presented. Special attention is devoted to the quality of measurement data. Reliable data were suggested for further use in the analysis of pollution levels and model validation. Results of the recent intercomparison of the analytical methods are outlined.

Section 2 describes emission data officially reported to UN ECE by countries within the EMEP region. The section deals with official reporting of emission data in 2006, trends of European emissions between 1990 and 2004, and emissions changes in individual countries for this period. In addition to this, the results of emission re-calculations of the official data submitted in previous year, done by some countries, are presented.

Section 3 is focused on the progress in model development and input data analysis achieved by MSC-E as a response to the recommendations of the TFMM workshop held in Moscow in 2005. In particular, the section describes emissions from land and marine surfaces driven by meteorological processes, approaches to validate meteorological data used by models for pollution levels simulations, and preliminary results of modelling of arsenic, nickel and chromium concentrations and depositions.

Section 4 is devoted to modelling results for 2004. The concentrations and depositions of lead, cadmium and mercury in Europe and their transboundary transport to European countries are described. The modelled concentrations and depositions are compared with measurement data collected at EMEP monitoring network and brief statistical analysis of the results is performed. Besides, model experiments aimed at evaluation of model performance for different emission scenarios are presented. Results of mercury modelling at the hemispheric scale are discussed.

Section 5 deals with the results of cooperation of MSC-E with subsidiary bodies to the Convention, national experts and international programmes and projects. In particular, the section deals with the contribution of MSC-E to the background document for the review of Protocol on Heavy Metals, prepared under the Task Force on Heavy Metals. Furthermore, the section describes the contribution of MSC-E to the report on health effects of long-range transport, coordinated by the Task Force on Health and contribution to the Task Force on Hemispheric Transport of Air Pollution. Besides, results of cooperation of MSC-E and WGE are presented in this section. Finally, cooperation with Helsinki Commission, the Task Force on Measurements and Modelling, and the European Commission is overviewed.

The main outcomes of CCC and MSC-E work regarding heavy metals are summarised in *Conclusions*. The supplementary information, such as tables with national emissions in countries etc. is given in *Annexes*.

1. MONITORING OF HEAVY METALS IN EMEP

1.1. Measurement network

Heavy metals were included in EMEP's monitoring program in 1999. However, earlier data has been available and collected, and the EMEP database thus also includes older data, even back to 1987 for a few sites. A number of countries have been reporting heavy metals within the EMEP area in connection with different national and international programmers such as HELCOM, AMAP and OSPARCOM.

The locations of the measurement sites, which have delivered data on heavy metals for 2004, are found in Fig. 1.1. Detailed information about the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [*EMEP/CCC report 7/2006*]. In the figures, the sites are divided in those measuring both concentrations in air and in precipitation, and those measuring only one of them. In 2004 it was 26 sites measuring heavy metals in both compartments, and altogether it was 63 measurement sites. It was 18 sites measuring at least one form of mercury. Notice that Nuuk in Greenland is outside the map in Fig. 1.1.

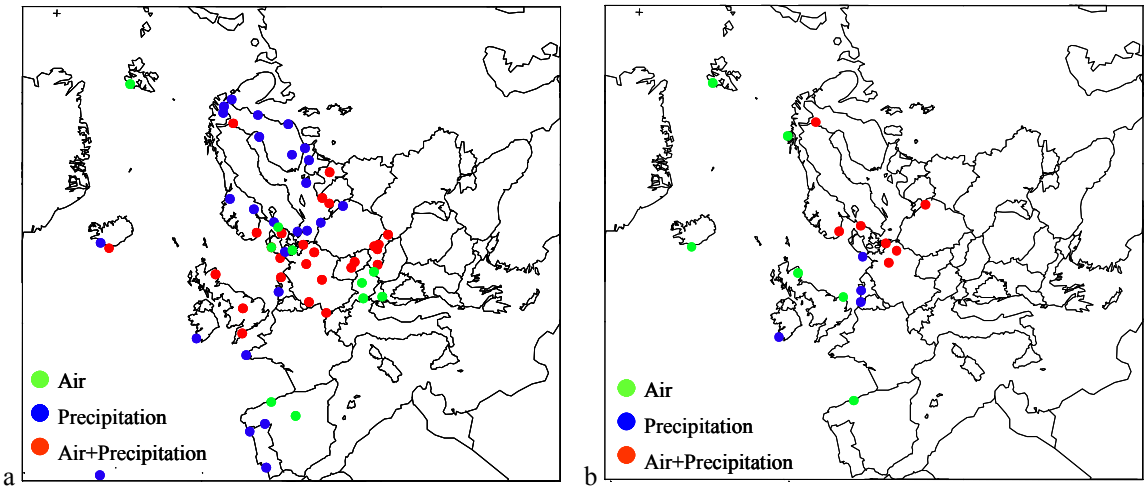


Fig. 1.1. *Measurement network of lead, cadmium (a) and mercury (b) in 2004*

From Fig. 1.1 one can see that the spatial resolution in eastern and southern Europe is unsatisfactory. In addition, it is too few sites measuring both in air and precipitation. The adopted EMEP monitoring strategy for 2004-2009 (EB.AIR/GE.1/2004/5) will expectantly improve this situation.

1.2. Monitoring of Pb, Cd and Hg in 2004

Annual averages of Pb, Cd and Hg concentrations in precipitation and in air in 2004 are presented in Fig. 1.2-1.7. The lowest concentrations for all elements in air as well as precipitation are found in northern Scandinavia. An increasing gradient can in general be seen southeast, but the concentration levels are not evenly distributed, there are some "hotspots" for some elements and the distribution is not always similar in air and precipitation.

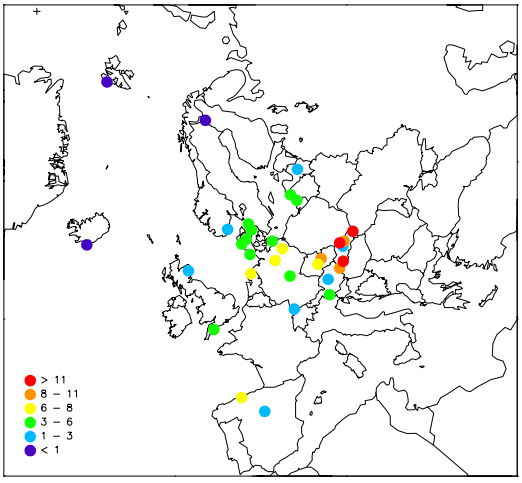


Fig. 1.2. *Pb in aerosol, ng/m^3*

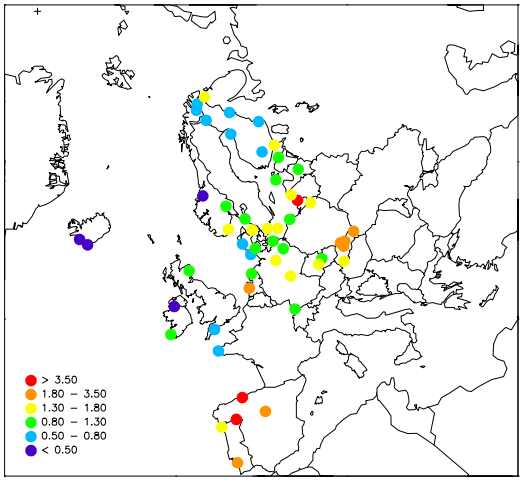


Fig. 1.3 *Pb in precipitation, μg/L*

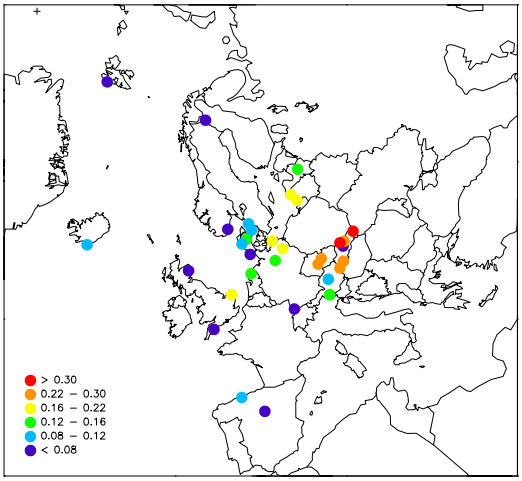


Fig. 1.4*. Cd in aerosol, ng/m^3*

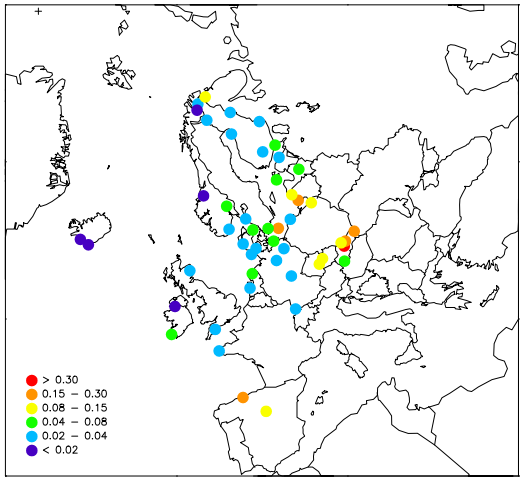


Fig. 1.5. *Cd in precipitation, μg/L*

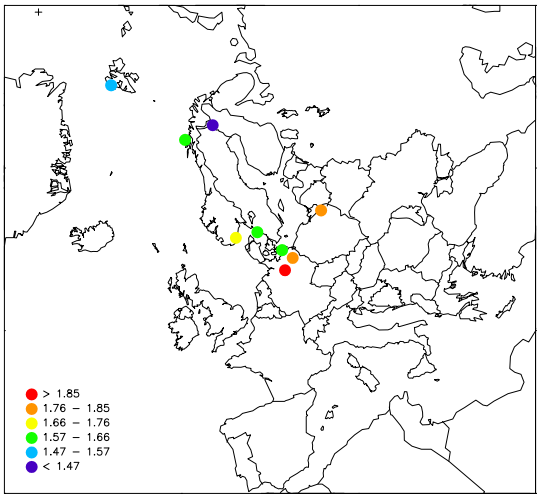


Fig. 1.6*. Hg in air, ng/m^3*

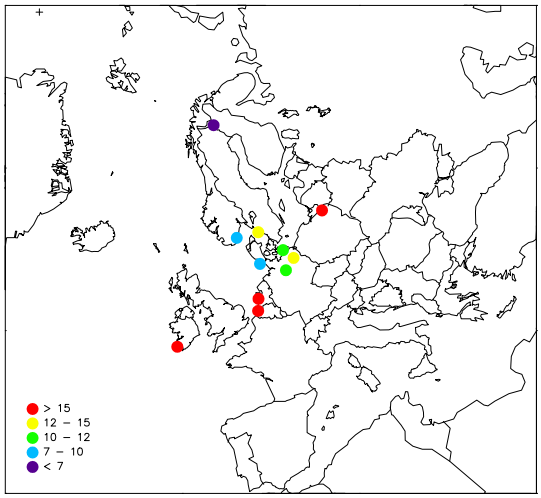


Fig. 1.7. *Hg in precipitation, ng/L*

The highest Cd concentrations in precipitation are seen in Slovakia; in air the highest concentrations are observed in Slovakia as well as in Austria and The Czech Republic. Elevated levels of Cd are also seen in Spain, Latvia and Lithuania. The highest levels in precipitation are actually seen in Portugal, but they have a very high detection limit for Cd (0.85 ng/L), so these data are not shown in Fig. 1.4. The lowest concentrations are seen in North of Norway and Finland. For Pb the highest levels in air are also here in Slovakia and Austria. In precipitation it is also very high levels in Latvia, Spain, Portugal and the Netherlands. In Portugal and Spain the high levels can partly be explained by relatively high detection limit. There are only a few stations measuring mercury in Europe, and most of them are related to the OSPAR program CAMP. The concentrations of mercury at the different sites are decreasing from north to south, but these differences are quite small. The precipitation data from Ireland, Poland and Belgium has too high detection limits and are not really useful, they should be looked upon as a concentration maximum. After these countries, the highest level is observed in the Netherlands.

1.3. Data quality

Most of the Portuguese and Irish heavy metal measurements have in general very high detection limits and these data are regarded as very uncertain. Also other countries like Spain, Latvia, Estonia and Austria experience relatively high concentration levels but a large fraction of the data are undetectable which increases the uncertainty also of some of these measurements. In Norway, Denmark and Sweden the concentration levels are relatively low, and a high percentage of these data in both air and precipitation are also below the detection limits. The data quality objectives (DQO) in EMEP states that the accuracy in the laboratory should be better than 15% and 25% for high and low concentrations of heavy metals, respectively. One important measure to check the data quality is laboratory ring test. There is a marked improvement in the laboratory performance for both lead and cadmium since the beginning of the laboratory comparison in 1995. The intercomparison completed last year [*Aas and Breivik*, 2004] is representative for the 2004 data. In Table 1.1, there is a summary of the results from this laboratory intercomparison. Sweden and Iceland were not participating because these measurements were analyzed in Norway. Denmark and France have problems with many of their heavy metal measurements, while Poland has large errors in the priority compounds Cd and Pb. The large errors can be due to reporting errors, i.e. wrong unit, and it is not necessarily an analytical problem. But since correct reporting is an important part of the complete QA/QC these errors are shown. In general, the measurements of high concentration samples are of fewer problems, but these samples are not very representative for many EMEP sites. E.g. Estonia can't measure the low concentrations of many of the element due to too high detection limit. In addition, there are some countries like Ireland, Portugal, and Spain that report measurements data without participation in the laboratory intercomparison. Data from these countries are of unknown quality; and it is therefore strongly recommended that they take part in the annual laboratory intercomparison.

Table 1.1. *Average per cent error (absolute) in low and high concentration samples, results from the laboratory intercomparison. DQO is EMEPs data quality objectives*

	Cr		Ni		Cu		Zn		As		Cd		Pb	
	low	high	low	high	low	high	low	high	low	high	low	high	low	high
AT	1	3	1	2	8	5	7	0	3	1	5	8	6	4
BE	20	1	10	10	5	1	78	8	5	7	3	1	2	4
CS	3	5	6	4	2	5	6	2	4	4	3	4	6	2
DK	0	3	15	8					31	33	860	829	46	3
FI	3	4	2	1	2	0	3	1	1	1	5	1	2	4
FR	14	2	400	12	164	8	259	3	48	46	471	35	9	12
DE	0	0		6	3	2	1	2		5	0	0	5	0
NL	21	4	15	3	11	1	0	2	3	1	12	2	4	4
NO	3	5	4	5	12	10	6	2	2	3	8	2	2	5
PL	0	0	0	2	0	2	3	2			22	61	37	78
GB	13	5	15	2	10	1	20	0	8	6	18	10	0	1
SK	19	12	26	10	25	8			18	12	13	9	9	6
LT	13	18	0	8	12	12	21	24	13	11	25	17	3	4
SI	7	5	0	3	3	5	30	12	0	2	7	4	7	4
EE		10		8	21	5	69	8		27		11	22	23

1/2 - 1 DQO 1 - 2 DQO > 2 DQO

Selection of measurement sites for model verification

Measurement data are highly important for the evaluation of modelling results. However, for this purpose the most reliable measurement data should be selected. Comparison with low quality measurements (e.g., if measured values are below detection limit) or with data sampled in non-representative locations does not characterize model performance. Hence, these data should not be used in the model validation. That is why some of measurement data were not involved into comparison of modelling results with measurements. The criteria of selection of measurement data for the comparison have already been formulated in MSC-E report [*Ilyin and Travnikov*, 2005]. Table 1.2 summarizes the data excluded from the comparison for 2004 and gives reasons for this. The decision to exclude the data was taken in agreement with the experts from CCC.

Table 1.2. *Measurement data for 2004 from EMEP stations not included into model verification*

Station	Pollutant	Media	Comments
NO47	all metals	precipitation	not EMEP site
PT1, 3, 4, 10	all metals	precipitation	Values mostly below detection limit
PL5	Hg	air	Too large and unrealistic variability
PL5	Hg	precipitation	Too high detection limit
ES8, 9	Pb, Cd	precipitation	Too high detection limit
IE1	Pb, Cd, Hg	precipitation	Most of values are below detection limit
BE4	Hg	precipitation	Too high detection limits
LT15	Pb	precipitation	Uncertain data quality, very high monthly mean for several of the months (e.g, April, May, August, September, October)

2. EMISSIONS OF HEAVY METALS IN EUROPE

This section provides information on trends of anthropogenic emissions of lead, cadmium and mercury in the EMEP region in the period of 1990-2004, as well as information on emission trends and emission reductions in individual countries. Emission changes caused by backward recalculations of emission data are also presented. Besides, temporal variations of arsenic, chromium and nickel emissions in the period of 1990-2004 are shown.

Official submissions of emission data

In 2006, national total and sector data on lead, cadmium and mercury emissions for 2004 reporting year were submitted to the UNECE Secretariat by 28 European countries. These countries are Austria, Belarus, Belgium, Bulgaria, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Ireland, Latvia, Lithuania, Monaco, the Netherlands, Norway, Poland, Portugal, Republic of Moldova, the Russian Federation, Slovakia, Slovenia, Sweden, Switzerland, Ukraine and the United Kingdom. Among them 16 countries recalculated previously reported emission data on Pb, Cd and Hg for all or for selected years from the period of 1990-2003.

Emission trends in the EMEP region (1990-2004)

Changes of Pb, Cd and Hg anthropogenic emissions in the EMEP region in the period of 1990-2004 are shown in Fig. 2.1. Emission trends were assessed on the base of the reported emission data. In the absence of official data emission expert estimates [*Berdowski et al.,* 1997; *Denier van der Gon et al.,* 2005] were used. Between 1990 and 2004 total anthropogenic emissions in the EMEP region decreased for all the three metals: for lead by about 84%, for cadmium by about 47% and for mercury by about 44%.

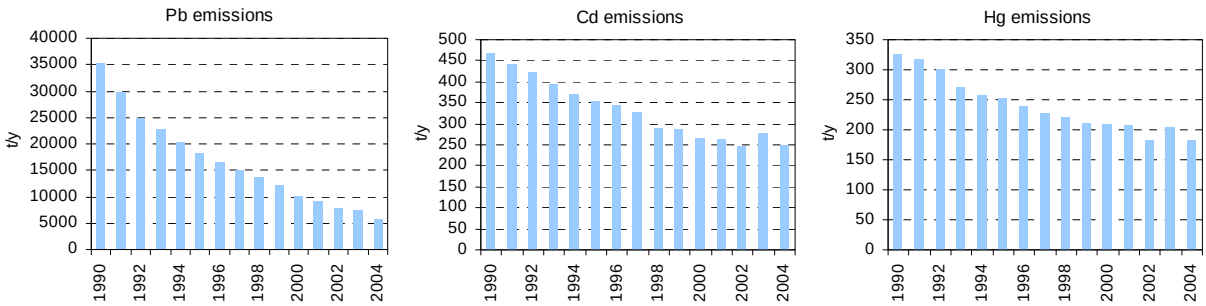


Fig. 2.1. *Total anthropogenic emissions of lead, cadmium and mercury in the EMEP region in the period 1990-2004 according to the official data combined with expert estimates*

Emission reductions from 1990 to 2004 in individual countries

The extent of emission reductions varies significantly from country to country. The relative emission reduction was estimated as $100*(E_{1990}-E_{2004})/E_{1990}$ (%), where E_{1990} and E_{2004} are emissions in 1990 and 2004, respectively. Figure 2.2 shows the relative reductions of lead, cadmium and mercury emissions between 1990 and 2004 in the 27 countries of the EMEP region, which reported their emissions both for 1990 and 2004. Emissions of lead decreased in all the countries varying from about 5% (Latvia) to 99% (Republic of Moldova). The lowest reduction of cadmium is in Portugal

(1%), the highest – in Republic of Moldova (95%). In Germany and Cyprus emissions of cadmium have increased about 2 and 5 times respectively. The reductions of mercury emissions vary from about 16% (Slovenia) to 90% (Republic of Moldova). In Portugal and Belarus mercury emissions have increased by about 1% and 32% respectively. In Cyprus, emissions of mercury have increased almost 4 times, and in Lithuania – almost 23 times.

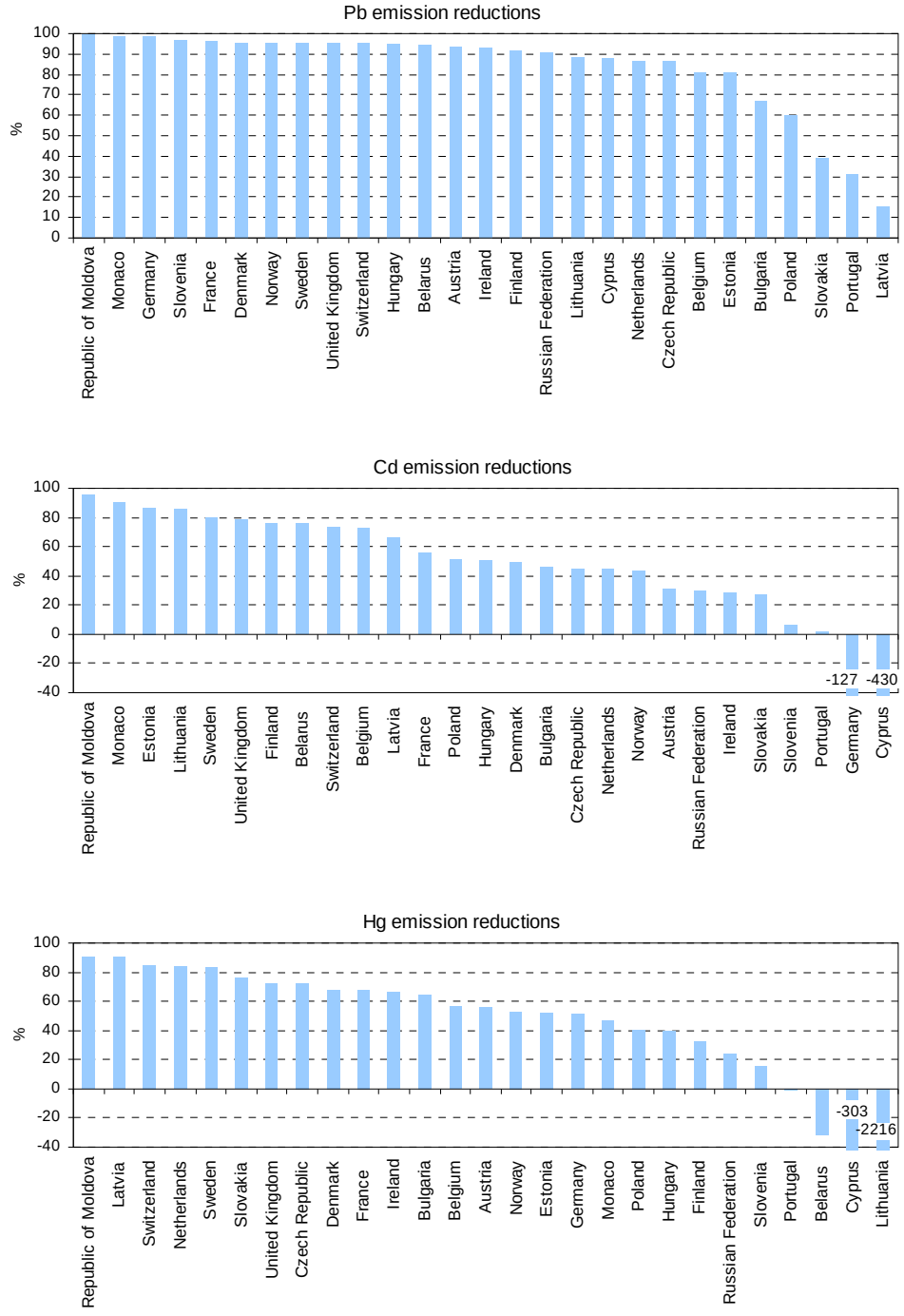


Fig. 2.2. *Reductions of lead, cadmium and mercury emissions in the 27 countries of the EMEP region for the period of 1990-2004 based on data reported for both 1990 and 2004, %*

Emission trends in individual countries

Temporal variations of lead, cadmium and mercury emissions for the period 1990-2004 in all countries that reported emission data are shown in Annex B. In most countries downtrends of emissions take place.

The analysis of emission data completeness shows that in some countries not all emission sources are estimated. This leads to underestimation of national total emissions.

Trends Pb, Cd and Hg emissions in Germany, built in accordance with the officially reported data, are considered here as example. In 2006, Germany reported national data on lead, cadmium and mercury emissions for each year of the period 1990-2004. In 2005 Germany reported emission data only for lead (1990-1997). The changes of lead, cadmium and mercury emissions in the country for the period 1990-2004 are shown in Fig. 2.3. In accordance with the reported data emissions of Pb and Hg have decreased between 1990 and 2004 by about 99% and 51% respectively. Cadmium emissions have grown almost twice in 2004 relatively 1990.

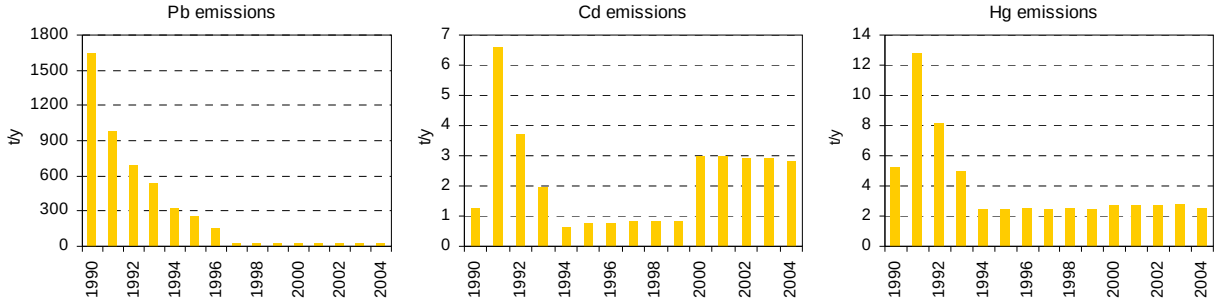


Fig. 2.3. *Emissions of lead, cadmium and mercury in Germany*

Analysis of total cadmium emission components shows that national totals include emissions of 5 from 80 sectors incoming in national totals. These sectors are "Public Electricity and Heat Production", "Petroleum refining", "Manufacture of Solid Fuels and Other Energy Industries", "Other, Manufacturing Industries and Construction" and "Residential plants" (Fig. 2.4). For all years (1990-2004) emissions from 22 sectors are indicated in reporting tables as "NE" (not estimated), from 46 sectors as "NA" (not applicable) and from 7 sectors as "NO" (not occurring). Thus, Cd emissions from 22 sectors are not estimated and hence are not included in national totals. Moreover, emissions from sector "Petroleum refining" are not estimated for 1990-1999. Emissions from this sector are included in national totals since 2000. In this situation the increase of cadmium emissions from 1990 to 2004 is formal because of inconsistency in time series of the sector data. Incompleteness of national totals and inconsistency in time series of some sector data are also characteristic for lead and mercury.

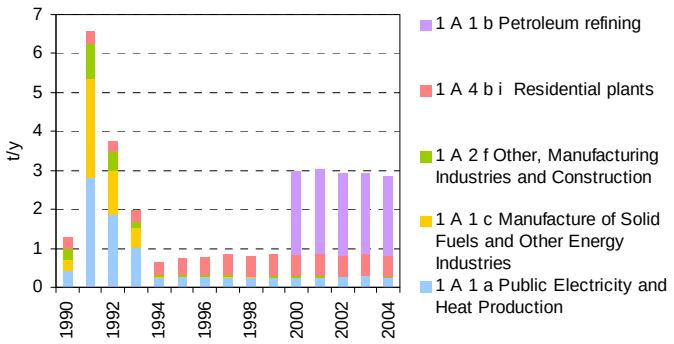


Fig. 2.4. *Cadmium emissions by source sectors in Germany*

Recalculations of national emission totals

In 2006, recalculations of previously submitted data on Pb, Cd and Hg emissions for the period 1990-2003 were reported by 16 countries (Austria, Belgium, Denmark, Estonia, France, Germany, Hungary, Ireland, Latvia, Norway, Portugal, Republic of Moldova, Slovenia, Sweden, Switzerland and United Kingdom). Percentage changes in the total national emissions after recalculations in these countries for the period 1990-2003 are given in Annex C.

The changes of lead, cadmium and mercury emissions in each country are expressed as $100*(E_{current} - E_{previous})/E_{previous}$ (%), where $E_{current}$ and $E_{previous}$ are current and previous emissions in the specific year, respectively. Maximum changes are illustrated in Figs 2.5 – 2.7. Negative values indicate a decrease in emissions after the recalculations, while positive values illustrate an increase in emissions.

Lead. For Austria, Denmark, France, Norway, Republic of Moldova and the United Kingdom the recalculations do not exceed ±10%. In Germany and Latvia the recalculations are more than 100%. Emissions increased about 14 times in Germany (1997) and about 29 times in Latvia (2003). The maximum emission reduction after recalculation is 75% (Switzerland, 2003) (Fig. 2.5).

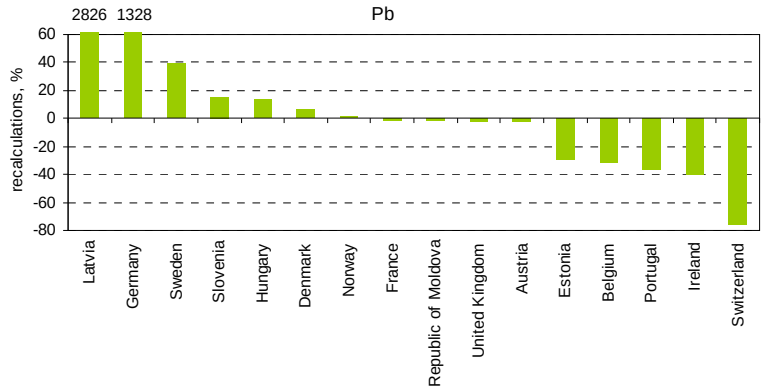


Fig. 2.5. *Maximum relative changes in the official lead emissions after the recalculations, %*

Cadmium. For Austria, Denmark, France, Slovenia, Sweden and the United Kingdom the recalculations are below ±10%. In Estonia, Portugal and Ireland recalculations exceed 100%. In Estonia (1994) and Portugal (1996) emission increased about 3 times, in Ireland (2002) – about 2 times. The maximum emission reduction after recalculation is 45% (Belgium, 2003) (Fig. 2.6).

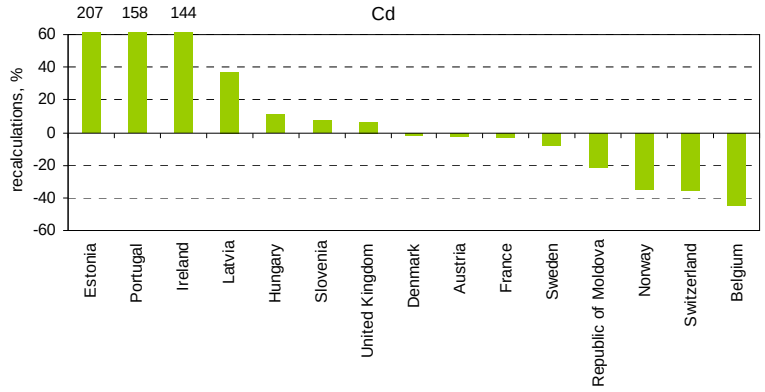


Fig. 2.6. *Maximum relative changes in the official cadmium emissions after the recalculations, %*

Mercury. For nine countries (Austria, Denmark, France, Hungary, Norway, Portugal, Slovenia, Sweden and United Kingdom) the recalculations are less ±10%. The highest increase of emissions after recalculations is about 16% (Belgium, 2003) and the highest decrease is about 76% (Ireland, 2003) (Fig. 2.7).

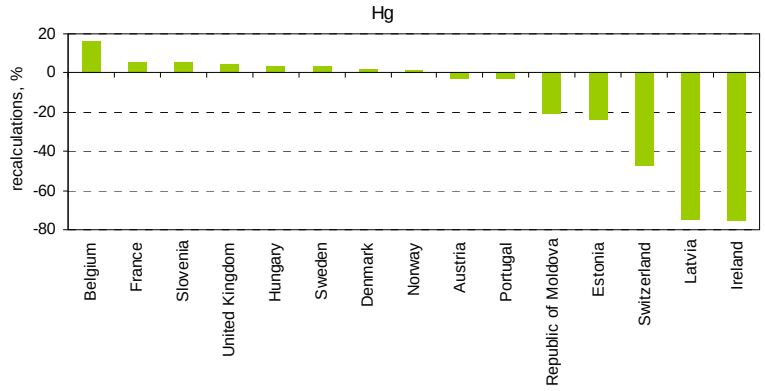


Fig. 2.7. *Maximum relative changes in the official mercury emissions after the recalculations, %*

Other heavy metals

Emission data on other heavy metals, such as arsenic (As), chromium (Cr), copper (Cu), nickel (Ni), selenium (Se) and zinc (Zn) are reported by countries to UN ECE as additional information. At this stage attention is focused on three (As, Cr, Ni) of these metals.

In 2006 and in previous years data on emissions of As, Cr and Ni at least for one year in the period of 1990-2004 were reported by 30 European countries, that is about 65% of Parties to the CLRTAP located in the EMEP region.

The changes of As, Cr and Ni emissions in the EMEP region for the period of 1990-2004 were estimated on the basis of available official data and expert estimates [*Berdowski et al*., 1997; *Denier van der Gon et al.,* 2005] (Fig. 2.8). Emissions of all considered metals have decreased between 1990 and 2004: for arsenic by about 53%, for chromium by about 37% and for nickel by about 57%.

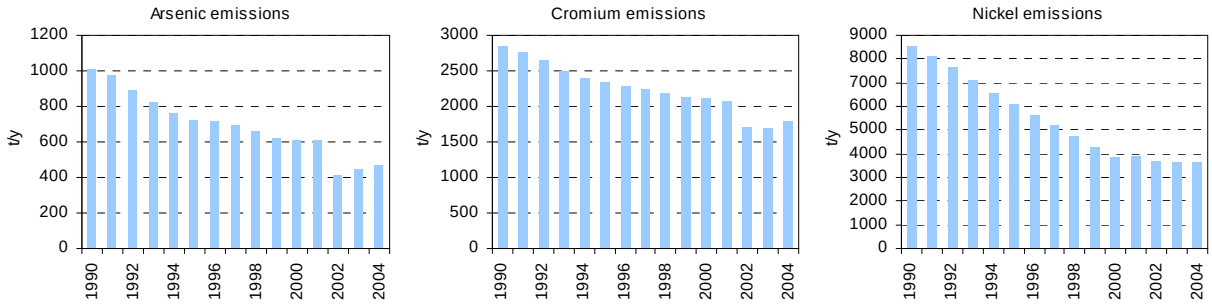
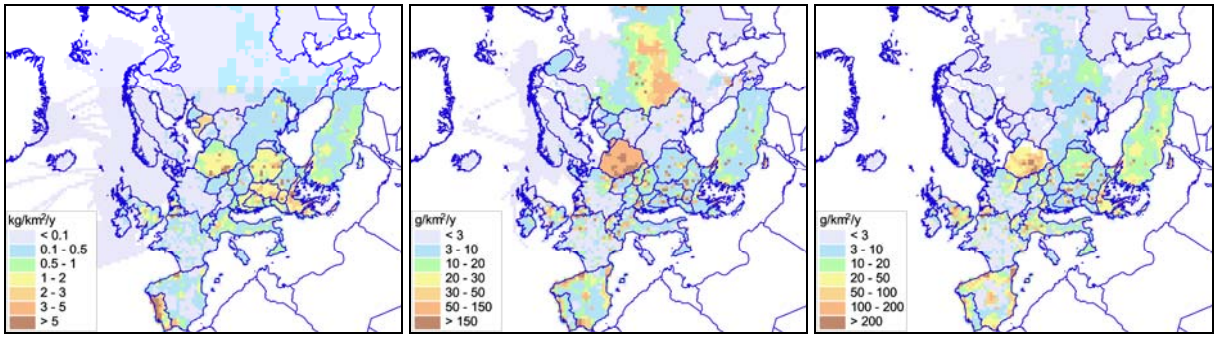


Fig. 2.8. *Total anthropogenic emissions of arsenic, chromium and nickel in the EMEP region in the period 1990-2004 according to the official data combined with expert estimates*

Input emission data for modelling

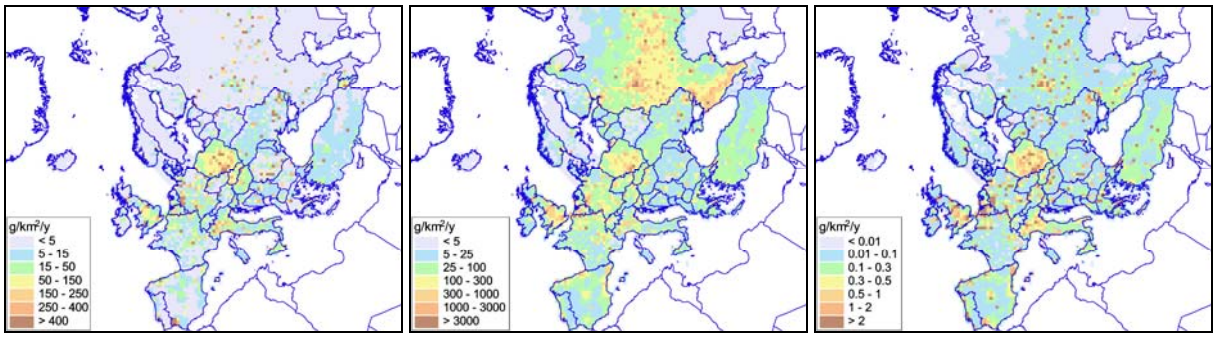
In order to provide European countries with data on depositions, concentrations and source-receptor relationships, MSC-E needs emission data from all countries in Europe. The emission data used by modelling for 2004 are based on officially reported information to the UN ECE Secretariat. For countries, which official data are not available, emission totals for 2004 were estimated by interpolation between 2000 and 2010 of expert estimates and projections made by the Dutch TNO institution [*Denier van der Gon et al.,* 2005]. Total emissions of lead, cadmium and mercury for 2004, derived through this combination of official data and expert estimates were around 5600, 250 and 180 t/y, respectively. Spatial distributions of these emissions are demonstrated in Fig. 2.9. Temporal and vertical distribution of the emission data and speciation of mercury emissions, employed in MSCE-HM model, are described in [*Travnikov and Ilyin*, 2005].



a b c

Fig.2.9. *Spatial distribution of anthropogenic emissions of lead (a), cadmium (b) and mercury (c) for 2004*

According to Workplan of EMEP for 2006 (Annex A), MSC-E is responsible for undertaking pilot modelling of transport and depositions of arsenic, nickel and chromium. The modelling was performed for 2000. Spatial distributions of emissions of these metals for 2000 used for modelling are demonstrated in Fig. 2.10. The emission fields were made on the basis of official gridded data and expert estimates of spatial resolution for 2000 [*Denier van der Gon et al.,* 2005].



a b c

Fig. 2.10 *Spatial distribution of anthropogenic emissions of arsenic (a), chromium (b) and nickel (c) for 2000*

3. MODEL DEVELOPMENT

The EMEP/TFMM Workshop on the review of MSC-E HM and POP models (Moscow, October 2005) recommended a number of activities aiming to improve MSCE-HM model formulation and its input data [*TFMM Workshop minutes*, 2005]. In particular, it was recommended to develop emission algorithms and models for representations of meteorological processes driven emissions, such as resuspension of particle-bound heavy metals. Besides, the workshop requested to carry out validation of meteorological data used by MSC-E for modelling at European scale. Finally, one of TFMM recommendations was "extension of the MSC-E HM model to the consideration of other elements and heavy metals, including: Ni, Cu, Cr, As, Zn and Se". This section is focused on description of wind-blown emission (re-suspension) of heavy metals from natural surfaces, preliminary results of meteorological data evaluation and results of pilot calculations of the second priority metals (Ni, Cu, Cr, As, Zn and Se).

3.1. Wind re-suspension of particle-bound heavy metals

Analysis of long-term trends of measured depositions and estimated emissions as well as the atmospheric balance for the Europe as a whole revealed significant inconsistencies between measured levels of lead and cadmium and their officially reported European emissions [*Ilyin and Travnikov*, 2005]. These inconsistencies could be explained by either underestimation of the anthropogenic emissions data, or significant unaccounted influence of natural emissions and re-emissions of historic depositions, or by both reasons. Therefore, a tentative parameterisation for the re-suspension of particle-bound heavy metals (Pb, Cd, As, Cr, Ni) from soil and seawater was developed and included into the MSCE-HM model. This parameterisation is to be improved and refined further in future.

The process of wind erosion and suspension of dust aerosol from the ground was incorporated to the model as combination of two major processes: saltation and sandblasting. The first process (saltation) presents horizontal movement of large soil aggregates driven by wind stress. Indeed, in natural soils small particles (below 20 μm) never occur in free state, but are embedded in larger soil aggregates by cohesion forces (up to a few centimeters). These aggregates are too heavy to be directly suspended by wind in usual conditions. Instead, they are moved by wind stress close to the surface jumping from one place to another. When the saltating aggregates impact the ground they can eject much smaller particles (few micrometers), which can be easily suspended by wind and transported far away from the source region. This process is called the sandblasting.

Parameterization of mentioned above processes are based on approaches applied in contemporary mineral dust production models [e.g. *Gomes et al*., 2003; *Zender et al*., 2003; *Gong et al*., 2003]. The dust suspension was estimated for the following types of non-vegetated land cover:

- deserts and bare soils;
- agricultural soils (during the cultivation period);
- urban areas.

For estimation of heavy metal emission with dust suspension from soils detailed measurement data on heavy metals concentration in topsoil from the Geochemical Atlas of Europe developed under the auspices of the Forum of European Geological Surveys (FOREGS) [*www.gtk.fi/publ/foregsatlas/*] were used. The data cover most parts of Europe (excluding Eastern European countries) with more than

2000 measurement sites. The kriging interpolation was applied to obtain spatial distribution of heavy metal concentration in soil. For Eastern Europe as well as for the rest of the model domain (Africa, Asia) we used default concentration values based on the literature data.

In order to estimate heavy metal re-suspension from seawater with sea-salt aerosol the empirical Gong-Monahan parameterization was applied [*Gong*, 2003] along with the emission factors derived from the literature. More detailed description of heavy metal re-suspension from soil and seawater is available in [*Gusev et al.*, 2006].

Estimates of resuspension of particle-bound heavy metals from soil and seawater were performed for Europe and adjacent territories in 2000. Spatial distributions of the annual resuspension flux of Pb, Cd, As, Cr, and Ni are presented in Fig. 3.1. In general, the resuspension fluxes from soil are significantly higher those from seawater for all the metals. High re-suspension fluxes were obtained from desert areas of Africa and Central Asia because of significant dust production in these regions. Elevated fluxes are also characteristics of some countries of Western, Central, and Southeast Europe, which are conditioned by combination of relatively high concentration in soil and significant dust suspension from urban and agricultural areas.

Heavy metals bound to re-suspended particles may have purely natural origin or come from previous long-term (historical) depositions of metals from anthropogenic sources. It is not possible to distinguish between these two components. Therefore, the release of metals to the atmosphere due to re-suspension will further be called natural and historical emission.

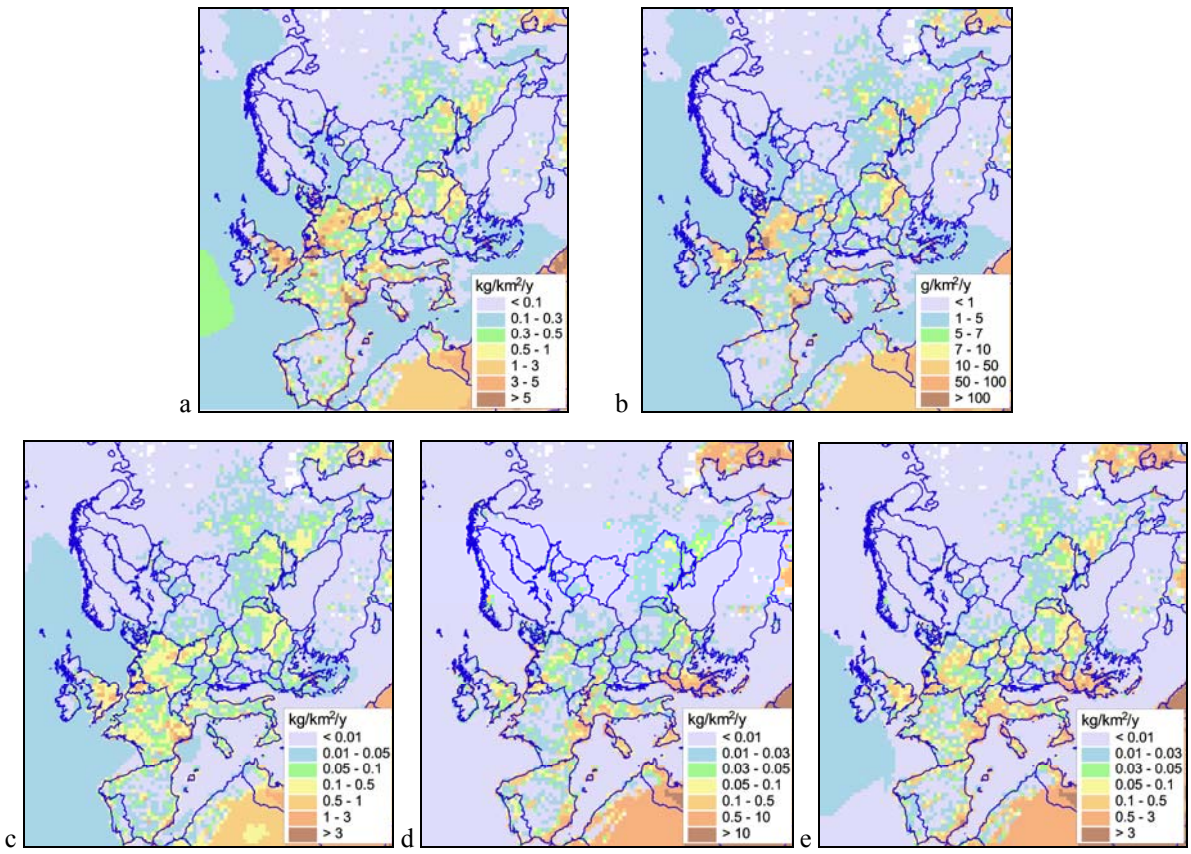
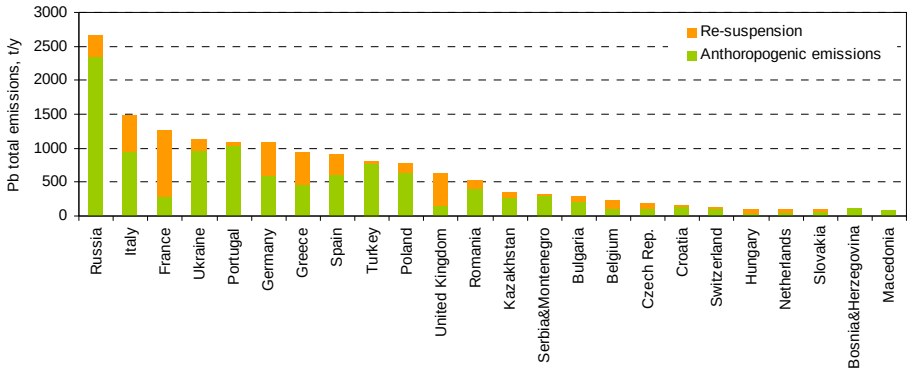
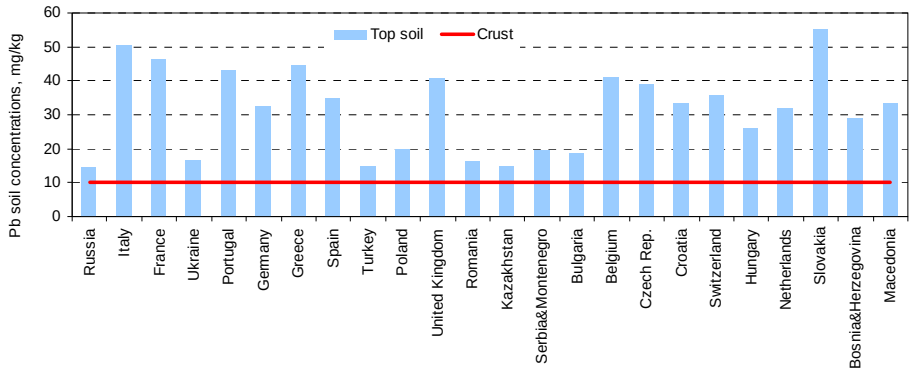


Fig. 3.1. *Spatial distribution of annual resuspension flux of heavy metals in Europe in 2000: (a) – Pb; (b) – Cd; (c) – As; (d) – Cr; (e) – Ni*

Aggregated values of lead resuspension from soil in different European countries are presented in Fig. 3.2a along with total anthropogenic emissions based on official data. As seen the estimated contribution of lead resuspension is comparable or even higher than anthropogenic emissions in such countries as Italy, France, Germany, Greece, Spain, the United Kingdom etc., where observed concentration of this metal in soil considerably exceeds its average natural content in the Earth's crust (Fig. 3.2b) [see *Gusev et al*., 2006 and references therein]. The most probable reason for this is long-term accumulation of historical depositions.



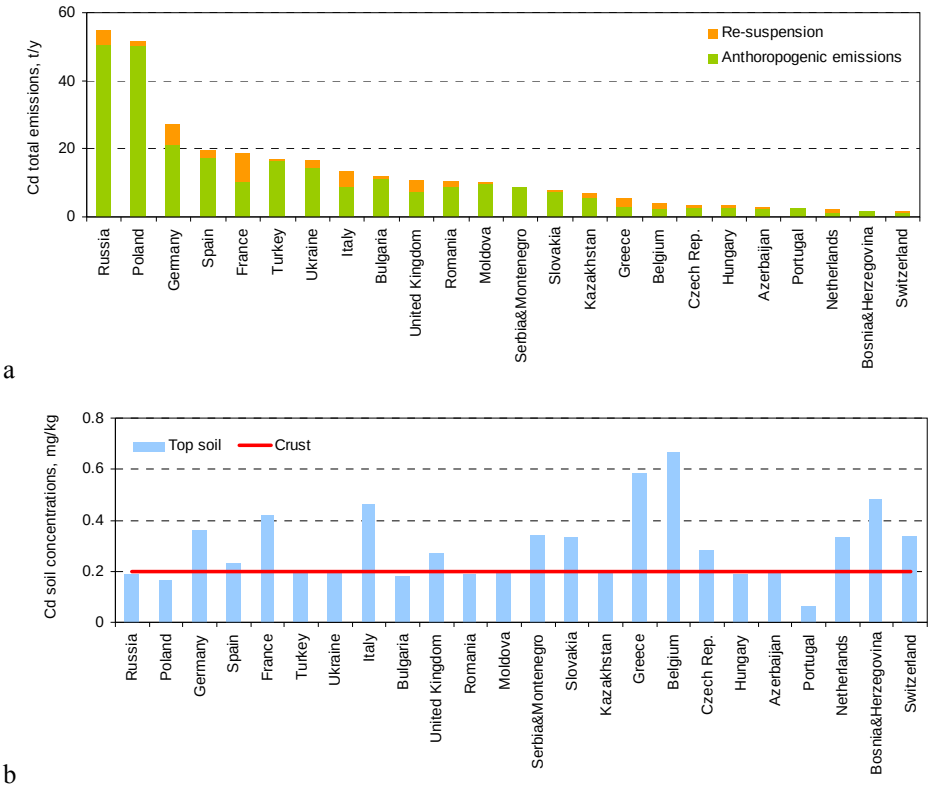
a



b

Fig. 3.2. *Lead total anthropogenic emissions and resuspension from soil (a) and average topsoil concentration (b) in some European countries*

Contrary to lead, cadmium resuspension from soil insignificantly contribute to total emission of this metal in most European countries (Fig. 3.3a). The reason for this is in relatively low cadmium concentrations measured in European soils. Only in a few countries of Europe (France, Italy, Greece, Belgium etc.) mean topsoil concentration noticeably exceeds cadmium natural content in the crust, and natural and historical emissions are comparable with anthropogenic ones (Fig. 3.3.b).



a

b

Fig. 3.3. *Cadmium total anthropogenic emissions and resuspension from soil (a) and average topsoil concentration (b) in some European countries*

3.2. Evaluation of meteorological input data

Meteorological Synthesizing Centre East uses MM5 as a meteorological pre-processor to prepare meteorological data for heavy metal and POP regional transport models. MM5 system is described in detail in [*Grell et al*., 1995; *http://www.mmm.ucar.edu/mm5/overview.html*]. The configuration of the pre-processing system used in MSC-E was overviewed in MSC-E Technical Report [*Travnikov and Ilyin,* 2005].

Following the recommendations of the EMEP/TFMM Workshop on MSC-E models review the procedure of evaluation of meteorological fields produced by MM5 for the modelling purposes was initiated. This section briefly describes the first results of this activity. In particular, the influence of different parameterisations of physical processes on the simulated meteorological parameters was tested. Additionally, first results of evaluation of meteorological parameters produced by MM5 via comparison with ECMWF re-analysis data are presented.

MM5 allows a number of parameterizations of atmospheric physical processes. Calculations of meteorological fields for January and July 2000 were performed with the use of various physical options. The processed meteorological fields were compared with the data of ECMWF Re-analysis project (ERA-40) with free access through the Internet [*www.ecmwf.int*]. The details of the experiment are available in MSC-E technical report 4/2006. The main preliminary conclusions of the experiment are as follows:

1. The best agreement between the results of computations and re-analysis data was obtained for meteorological parameters that are involved in the data assimilation procedure (temperature, air humidity, wind speed). Coefficients of correlation in time and space are normally within the range

of 0.7 -0.9. The total amount of monthly precipitation is fairly well reproduced by the model, however the temporal coefficients of correlation are lower.

2. All meteorological parameters except precipitation amount are sensitive to the change in boundary layer and soil parameterization. Parameterization of clouds and microphysical processes influences air humidity and precipitation amount first of all.
3. The use of “polar physics” option improves the MM5 performance for polar regions. However, the effect of this option seems to be small.
4. There are several combinations of parameterizations of physical processes in MM5 that provide similar high quality of results in a reasonable computing time. One of these combinations is currently used by MSC-E for operational modelling of transboundary transport and depositions of heavy metals and POPs.

Additional task was aimed at more detailed analysis of meteorological data used by MSC-E for calculations of atmospheric transport of pollutants. This activity is not completed yet, and the section is focused on first available results. Horizontal wind velocity components, precipitation amounts and air temperature, were compared against similar parameters available in ECMWF reanalysis database. These parameters were selected because of their importance for pollution transport modelling and because of their availability in ECMWF reanalysis. The comparison was performed for 2000. The details of the analysis are present in [*Gusev et al.,* 2006].

Comparison of near-surface air temperatures, produced by MM5 and derived from ECMWF demonstrated good agreement between them. It is confirmed by high spatial and temporal correlation coefficients. Besides, the differences in annual-mean air temperatures are not high, with the exception of few gridpoints located in mountainous regions (Fig. 3.4). Precipitation amounts modelled by MM5 were compared with precipitation derived from two different sources: ECMWF re-analysis and results of Global Precipitation Climatology Project (GPCP, http://cics.umd.edu/~yin/GPCP/main.html). Spatial correlations of monthly sums of precipitation are higher for ECMWF, than for GPCP data (Fig. 3.5). The agreement between annual sums of precipitation from MM5 and ECMWF on one hand, and between MM5 and GPCP on another hand significantly differs across Europe. Temporal correlations of wind components and wind magnitudes, derived from MM5 and ECMWF, are relatively high (>0.65) over most of Europe. In mountainous regions the correlation is lower, presumably because of different orography data.

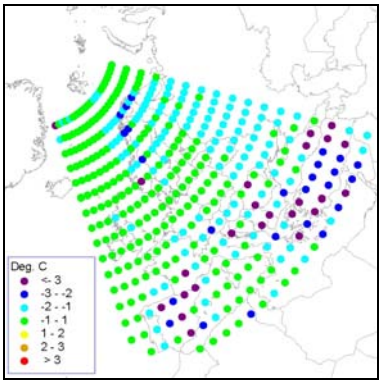


Fig. 3.4 *Difference between annual-mean near-surface air temperatures produced by MM5 and ECMWF (T_{MM5} - T_{ECMWF})*

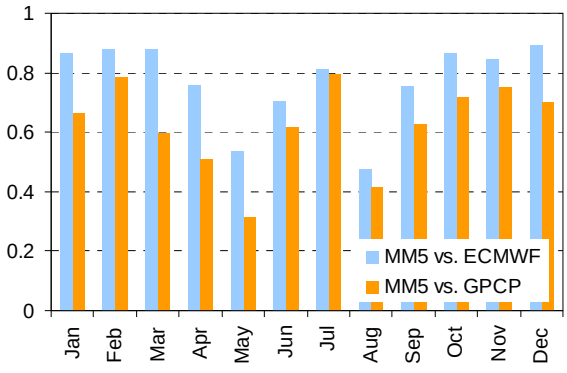


Fig. 3.5. *Spatial correlation coefficients of monthly-mean precipitation sums processed by MM5 with those from ECMWF and GPCP*

Further steps of evaluation of meteorological data should include analysis of spatial distributions and temporal variability for wider range of meteorological fields, which are important for modelling of atmospheric transport. Furthermore, analysis of frequency distributions of meteorological parameters in MM5 and ECMWF/GPCP data could give information about data quality. Finally, the transition to the use of ECMWF data as input information for meteorological preprocessor has already been initiated.

3.3. Pilot calculations of the second priority metals

MSCE-HM model parameterisation was extended to evaluate atmospheric transport and depositions of the second priority metals (As, Cr, Ni). Dry deposition velocities for these metals were calculated on the base of parameterization, developed for particle-bound metals [*Travnikov and Ilyin*, 2005], using mass median diameters available from the literature [*Milford and Davidson*, 1985]. Parameters of in-cloud and below-cloud scavenging were selected by analogy with those for lead and cadmium. Similar to lead and cadmium, any possible chemical transformations of these metals were not considered, because these transformations can hardly change properties of particles with regard to removal from the atmosphere.

Calculations of atmospheric transport and depositions of these metals were performed for 2000 on the base of officially reported data supplemented with TNO emission expert estimates [*Denier van der Gon et al,* 2005]. Besides, for arsenic, chromium and nickel European emission expert estimates prepared within EU project ESPREME [*http://espreme.ier.uni-stuttgart.de/data.html*] were used as an alternative emission scenario. In both cases the same parameterisation of wind re-suspention of these heavy metals was used. The origin of heavy metals bound to re-suspended particles can be purely natural, or they can be resulted from long-term accumulation of depositions from anthropogenic sources. Further these wind-blown releases of heavy metals to the atmosphere will be called natural and historical emissions. Total annual emissions from European region involved in modelling are summarized in Table 3.1. The results showing model performance for each of these metals are demonstrated below.

Table 3.1. *Total emissions from Europe in 2000, t/y*

	As	Ni	Cr
Official/TNO	440	3840	1780
ESPREME	760	4800	2700
Natural and historical*	1040	990	1450

*Includes emissions from seas surrounding Europe: the North, Baltic, Mediterranean, Black and Caspian Seas

Arsenic

Depositions of arsenic computed with official/TNO emissions over most of Europe ranged within 30–250 g/km^2/y (Fig. 3.6a). Similar to map of air concentrations, the lowest depositions occur over the Scandinavian Peninsula and north of Russia (below 30 g/km^2/y). The highest depositions were computed for areas with high emission sources, such as Poland, north Italy, southern Spain, Greece and others. Spatial distribution of modelled depositions based on the ESPREME emissions resemble those based on official/TNO data. However, their magnitudes are significantly higher (60-400 g/km^2/y), because of higher anthropogenic emissions (Fig. 3.6b).

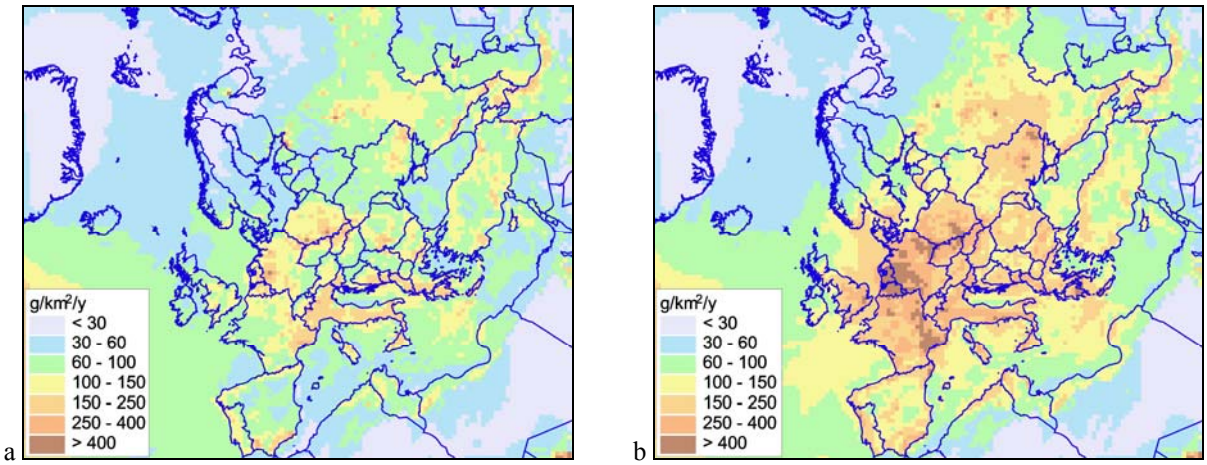


Fig. 3.6. *Modelled total depositions of arsenic in 2000 based on official/TNO (a) and the ESPREME emissions (b)*

Modelled and observed at EMEP stations concentrations of arsenic in air and precipitation were compared. Air concentrations of arsenic based on official/TNO emissions are lower than observed values by a factor about 2.5 (Fig. 3.7). Concentrations in precipitation were underestimated by similar factor. If ESPREME emissions were used, the agreement between modelled and measured quantities is better. Air concentrations derived from ESPREME emissions are higher than the observed ones by 20 – 30%, whereas concentrations in precipitation are around 30% lower. Correlation coefficients for air concentrations are significantly higher compared to those for concentrations in precipitation. These pilot results demonstrate that special attention should be paid to wet scavenging parameters of arsenic and quality of its measured concentrations in precipitation.

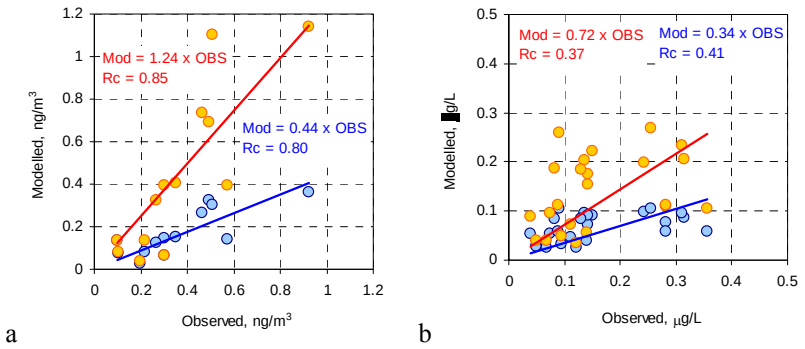


Fig. 3.7. *Comparison of modelled and measured arsenic concentrations: in air (a) and concentrations in precipitation (b) (official/ TNO – blue; ESPREME – red). Natural and historical emissions are included*

Chromium

Depositions of chromium in 2000 based on official/TNO emissions mostly ranges from 100 to 1000 g/km^2/y (Fig. 3.8a). In regions with significant anthropogenic emissions (Greece, central Russia) the depositions were higher than 1500 g/km^2/y. Significant depositions to the Mediterranean Sea resulted from the transport of re-suspended dust from northern Africa. Relatively high depositions to the west of Balkan Peninsula were caused by combination of high natural and historical emissions and large precipitation amounts. The use of the ESPREME emission estimates led to higher depositions of chromium compared to those based on official/TNO emissions (Fig. 3.8b), although spatial patterns of these fields are similar. The exception for this is Russia, where depositions based on the official/TNO emissions are higher than those derived from the ESPREME estimates. The reason for that is 2.5 times higher TNO emission estimates used for Russia in the former case.

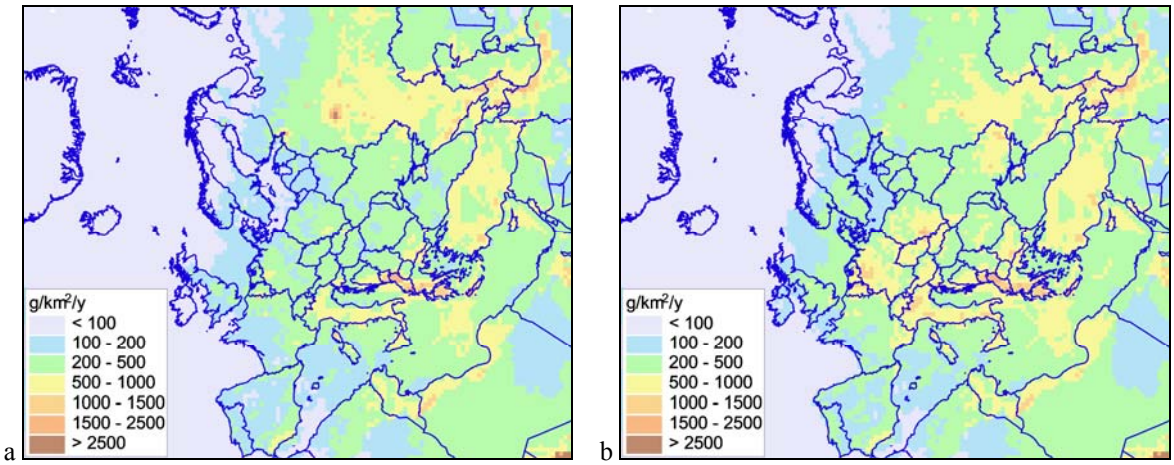


Fig. 3.8. *Modelled total depositions of chromium in 2000 based on official/TNO (a) and the ESPREME emissions (b)*

Modelled concentrations of chromium in air and in precipitation were compared with the observed quantities. Concentrations in air, derived from the official/TNO emissions underestimate observations by a factor of 2.5 (Fig. 3.9a). For concentrations in precipitation two-fold underestimation took place (Fig. 3.9b). On the other hand, modelled concentrations in air and precipitation based on the ESPREME emissions favourably matched the observed values. Correlation coefficients were also significant: 0.83 for concentrations in air and 0.63 for concentrations in precipitations.

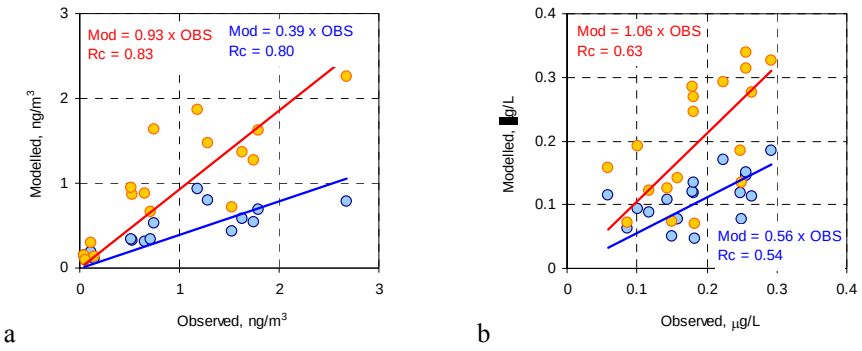


Fig. 3.9. *Comparison of modelled and measured chromium concentrations: in air (a) and concentrations in precipitation (b) (official and TNO – blue; ESPREME – red). Natural and historical emissions are included*

Nickel

Depositions of nickel over Europe for 2000, modelled on the base of official and TNO emissions, vary from 0.1 to 0.8 kg/km^2/y (Fig. 3.10a). Only over northern parts of EMEP region the depositions were below 0.1 kg/km^2/y. In regions associated with significant emission sources (north-west of Germany, Poland, central part of European Russia, north of Italy, Greece), the depositions exceeded 0.8 kg/km^2/y. Fields of nickel depositions derived from the ESPREME emissions are similar to those obtained through official and TNO emissions (Fig. 3.10b). Nevertheless, over most part of Europe the ESPREME-based levels are higher than those based on official/TNO emissions because of higher anthropogenic emissions. The exception is Russia, whose official emissions are higher than ESPREME estimates.

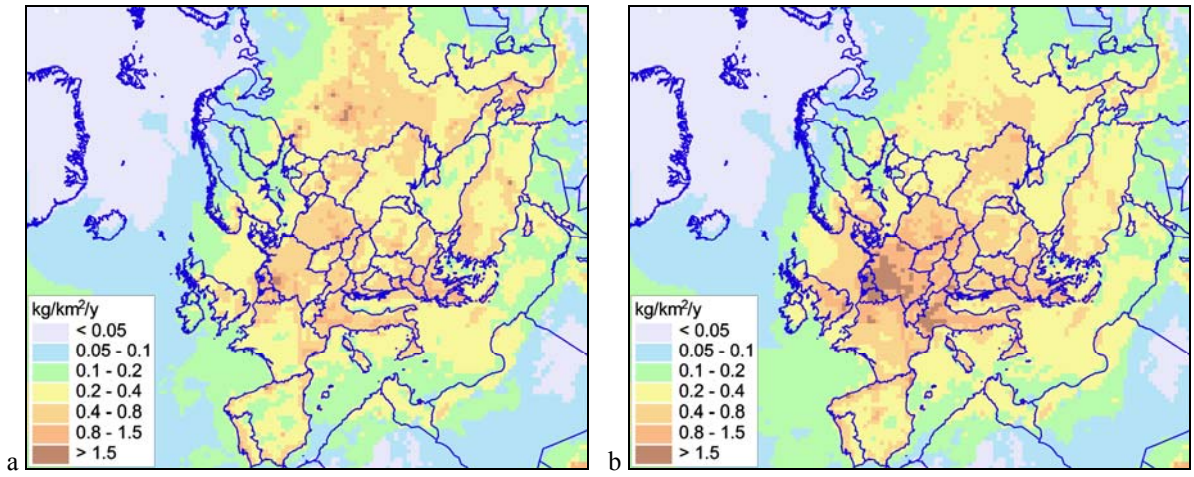


Fig. 3.10. *Modelled total depositions of nickel in 2000 based on official/TNO (a) and the ESPREME emissions (b)*

Comparison of observed nickel concentrations in air and in precipitation with those modelled on the base of official and TNO emissions reveals that modelled values are about 30% lower than the observed ones. On the contrary, the use of ESPREME emissions leads to some (~25%) overestimation of measured concentrations in air and in precipitation (Fig. 3.11). Correlation coefficients for concentrations in precipitation are relatively low, compared to those for concentrations in air. More detailed analysis of the comparison results is available in MSC-E Technical Report [*Gusev et al*., 2006].

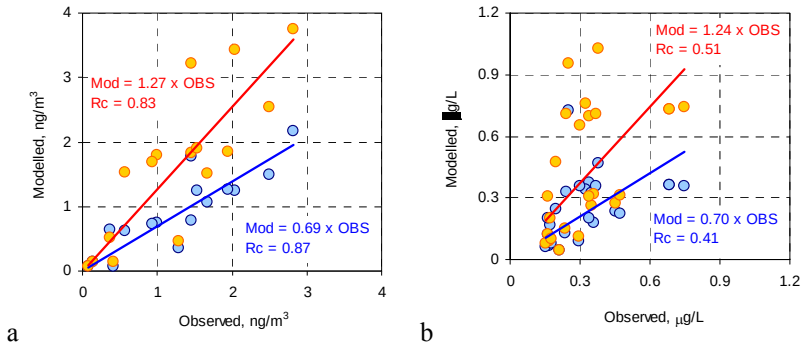


Fig. 3.11. *Comparison of modelled and measured nickel concentrations: in air (a) and concentrations in precipitation (b) (official and TNO – blue; ESPREME – red). Natural and historical emissions are included*

Zinc, Copper, Selenium

This year MSC-E has started preparatory activity for modelling of other metals: zinc, copper and selenium. Pilot parameterisations of their natural and historical emissions as well as other model parameters were developed. Measurement data for verification of modelling results were derived from the CCC database. Preliminary attempts to model atmospheric concentrations and depositions of zinc and copper resulted in significant (up to an order of magnitude) underestimation of the observed concentrations in air and in precipitation. Other attempts to model transport and depositions of zinc [e.g., *Nijenhuis et al.*, 2001; *Alcamo et al.*, 1992; *Sofiev et al.*, 2001, *Bartnicki et al*., 1998] or copper [*Nijenhuis et al.*, 2001] resulted in similar underestimation of observed values. It was suggested in these studies that low emissions of zinc were the main reason of the underestimation. In case of selenium, even preliminary verification of the obtained results is hampered by extremely limited measurement data. The work on modelling of zinc, copper and selenium is to be continued.

4. MODEL ASSESSMENT OF HEAVY METAL POLLUTION

This Section is devoted to the model assessment of the long-range transport of heavy metals in the atmosphere of Europe and the northern hemisphere. The first section contains the analysis of lead, cadmium and mercury pollution levels in Europe in 2004, information on transboundary fluxes between European countries, and heavy metal atmospheric loads to the regional seas. Besides, the modelling results for 2004 are compared with available measurements. Detailed information on source-receptor relationships for European countries is presented in Annex D. In the second section performance of the MSCE-HM model is extensively tested using different emission scenarios. The third section includes evaluation of mercury atmospheric transport and depositions in the northern hemisphere.

4.1. Heavy metal pollution levels in Europe in 2004

Lead

According to official emissions data total anthropogenic emissions of lead in European countries in 2004 were 5580 tonnes. It is lower by 35% than official emission estimates for 2003. Besides, a new pilot parameterisation of natural emission and re-suspension from soil and seawater was implemented this year for 2004 calculations. For Europe and the marginal seas (North, Black, Baltic, Mediterranean and Caspian) these natural and historical emissions made up as much as 6860 tonnes. It is almost twice higher previous rough estimates of natural emissions for 2003. Total deposition of lead to European countries from anthropogenic sources in 2004 is estimated at 3580 tonnes. It is lower by 38% than those estimated for 2003 based on official emissions data [*Bartnicki et al*., 2005]. However, significantly higher natural and historical emissions estimated for 2004 results in about 40% increase of lead total deposition from all (anthropogenic, natural, global) sources (10960 tonnes) in comparison with 2003 (7820 tonnes).

Along with the model estimates of lead pollution levels in 2003 based on official emissions data [*Bartnicki et al*., 2005] calculations of lead depositions were evaluated using the adjusted emission scenarios [*Ilyin et al*., 2005]. These scenarios were inversely derived from available measurements of lead concentration in air and precipitation in Europe [*Ilyin and Travnikov*, 2005] and more likely correspond to realistic pollution levels in Europe. According to these estimates total lead emission in Europe in 2003 is 2.5 times higher than that base on official emissions data. However, these estimates can include the natural component since it is not possible to separate it from the anthropogenic one. Estimated total depositions of lead to European countries in 2003 based on the adjusted scenarios (15280 tonnes) are 2 times higher than those based on official data and 1.4 times higher than current estimates for 2004.

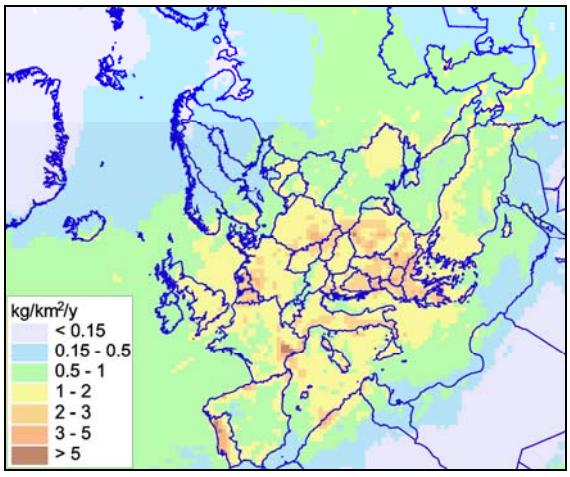


Fig. 4.1. *Spatial distribution of lead depositions in Europe in 2004*

Spatial distribution of lead depositions over the EMEP region to significant extent corresponds to the anthropogenic and natural emissions patterns (Fig. 4.1). In general, annual deposition fluxes in Europe ranges from 0.3 to 3 kg/km^2/y. In the most pollution loaded areas of such countries as

Belgium, Germany, Portugal, Poland, Greece etc. deposition fluxes often exceed 2 kg/km^2/y. In the northern part of Europe (Iceland, north of Scandinavia Peninsula) deposition fluxes of lead are typically less than 0.5 kg/km^2/y.

The largest total deposition flux took place in Belgium, followed by the FYR of Macedonia, Greece and Portugal (Fig. 4.2). Portugal is characterized by the highest depositions from anthropogenic emissions, which are mostly defined by national sources. As seen from the figure, the contribution of natural and historical emissions to total depositions is comparable to the anthropogenic one for the majority of countries. This contribution consists of two fractions, which cannot be separated: purely natural emission, and re-suspension of long-term (historic) depositions from anthropogenic sources. It is also important to stress that present estimates of natural and historical emissions are highly uncertain. The contribution of external sources (natural and anthropogenic) located outside EMEP region varies from less than 6% (Belgium, Slovakia) to about 45% (Armenia, Iceland). However, for the majority of countries their contribution is less than 20%. The contribution of these global sources was estimated via prescribed observation-based air concentrations of lead at the EMEP region boundaries. Although the contribution of global sources is not dominant, it is substantial in many countries. Hence, hemispheric modelling approach is preferable to derive more scientifically rigorous contributions of global sources.

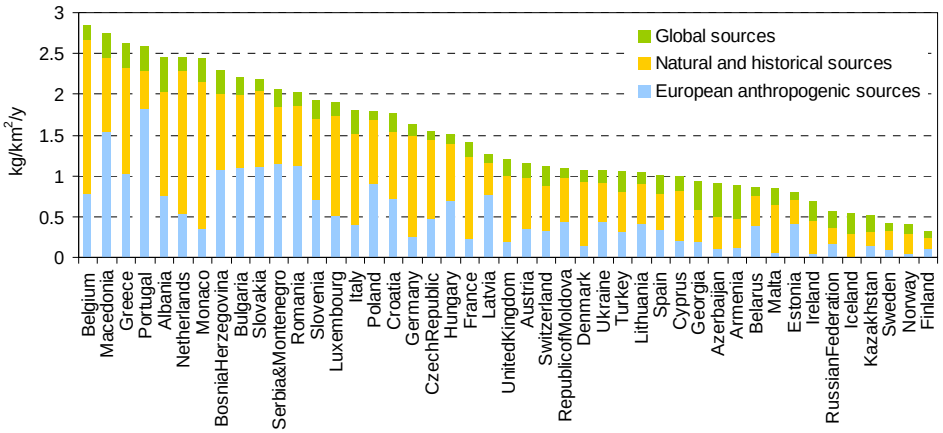


Fig. 4.2. *Country-averaged deposition fluxes of lead from European anthropogenic, natural/historical and global sources in 2004*

Relative contribution of the transboundary transport from European sources to anthropogenic depositions in such countries as Monaco, Republic of Moldova, Lithuania, Denmark exceeds 90% (Fig. 4.3). In 33 countries this contribution exceeds 50% and in 14 countries - 80%. Contribution of the transboundary transport to depositions is relatively low for countries which are characterized by high emission, and hence, high depositions from national sources (e.g., Poland, Greece, Romania), or countries remote from main anthropogenic sources (e.g., Spain, United Kingdom), or where both factors seems to be significant (e.g. Portugal).

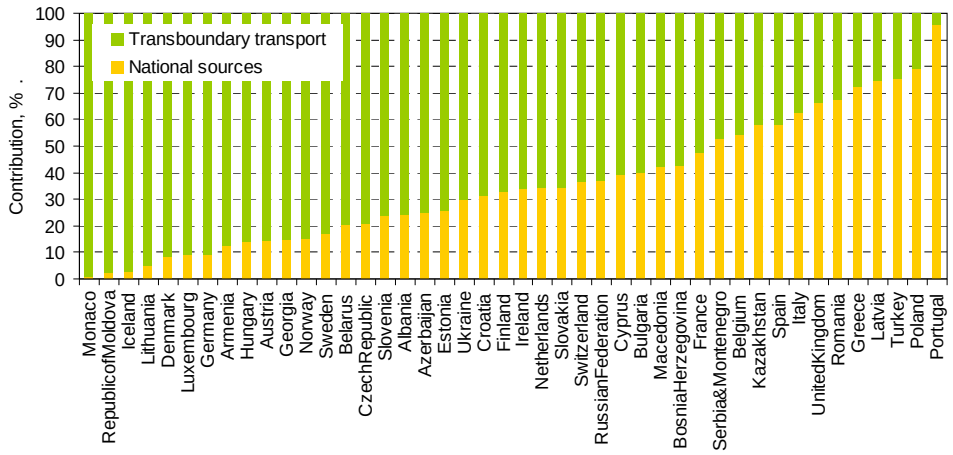


Fig. 4.3 *Relative contribution of the transboundary transport and national sources to anthropogenic lead depositions in European countries in 2004*

Contribution of countries to transboundary pollution was evaluated as mass of the pollutant, emitted by national sources and transported outside country's territory. This contribution depends on magnitude of national emissions, area and configuration of a country territory, predominant meteorological conditions etc. In 2004 this contribution varies from about 500 tonnes (Portugal) to some few kg (Monaco) (Fig. 4.4). Other countries contributing significantly to the transboundary transport are Turkey, Greece, Poland, Romania. Fraction of national emission, involved in transboundary transport, are mostly between 60% and 80%. For Russia its fraction is around 30%, for Cyprus, Luxembourg and Monaco this fraction exceeds 90%.

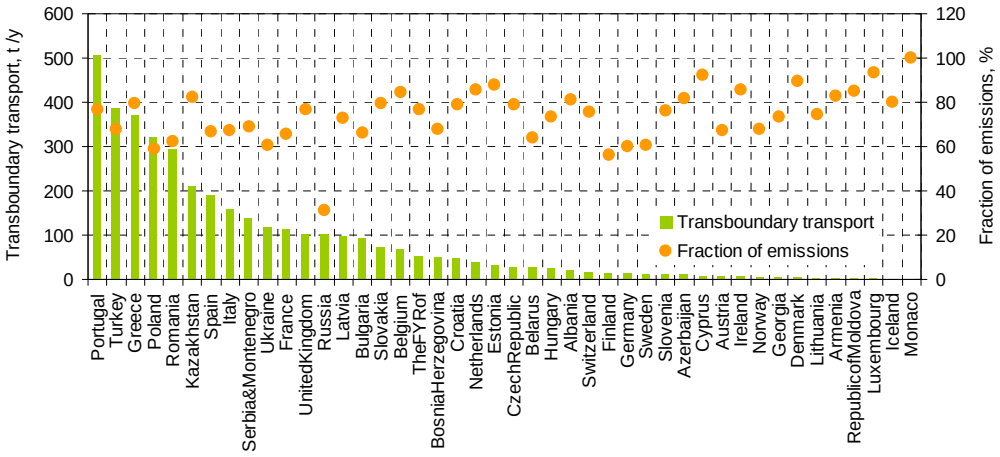


Fig. 4.4 *Absolute contribution of European countries to lead transboundary transport in Europe in 2004 and relative fraction of national emissions in this contribution*

Modelled mean annual concentrations of lead in air and in precipitation were compared with available observations. Modelled concentrations in air and in precipitation underestimate measurement data by approximately 30% (Fig. 4.5). Correlations coefficients are satisfactory high: 0.79 and 0.69, for air and precipitation, respectively.

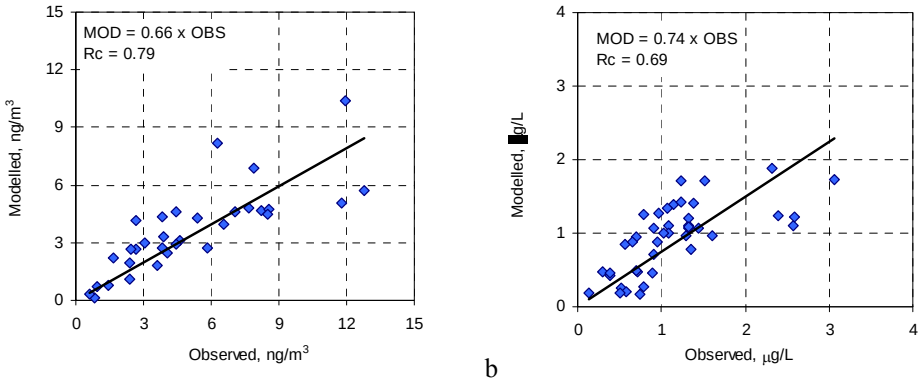


Fig. 4.5. *Comparison of modelled concentrations of lead in air (a) and in precipitation (b) with measurements from the EMEP monitoring network. Rc is the correlation coefficient*

Cadmium

According to official emissions data total anthropogenic emissions of cadmium in European countries in 2004 were 250 tonnes. It is approximately equal to official emission estimates for 2003. On the other hand, a new pilot parameterisation of natural emission and re-suspension from soil and seawater was implemented this year for 2004 calculations. For Europe and the marginal seas (North, Black, Baltic, Mediterranean and Caspian) these natural and historical emissions made up as much as 65 tonnes. It is even somewhat lower than previous rough estimates of natural emissions for 2003. Total deposition of cadmium to European countries from anthropogenic sources in 2004 is estimated at 181 tonnes. It is lower only by 4% than those estimated for 2003 based on official emissions data [*Bartnicki et al*., 2005]. Besides, lower natural and historical emissions estimated for 2004 results in about 6% decrease of cadmium total deposition from all (anthropogenic, natural, global) sources (293 tonnes) in comparison with 2003 (313 tonnes).

Along with the model estimates of cadmium pollution levels in 2003 based on official emissions data [*Bartnicki et al*., 2005] calculations of cadmium depositions were evaluated using the adjusted emission scenarios [*Ilyin et al*., 2005]. These scenarios were inversely derived from available measurements of cadmium concentration in air and precipitation in Europe [*Ilyin and Travnikov*, 2005] and more likely correspond to realistic pollution levels in Europe. According to these estimates total cadmium emission in Europe in 2003 is 2.3 times higher than that based on official emissions data. However, these estimates can include the natural component since it is not possible to separate it from the anthropogenic one. Estimated total depositions of cadmium to European countries in 2003 based on the adjusted scenarios (438 tonnes) are 1.4 times higher than those based on official data and 1.5 times higher than current estimates for 2004.

Spatial distribution of cadmium depositions over the EMEP region is shown in Fig.4.6.

Country-averaged depositions from anthropogenic, natural/historical and global sources are demonstrated in Fig. 4.7. As seen,

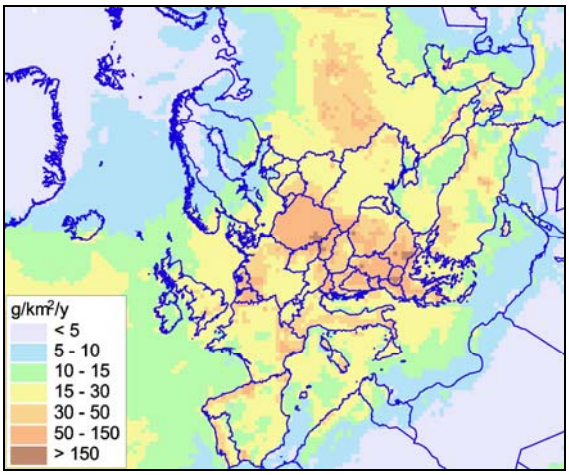


Fig. 4.6. *Spatial distribution of cadmium depositions in Europe in 2004*

among 45 countries as much in 35 countries the depositions from anthropogenic sources dominate over those from natural and historical ones. The highest anthropogenic depositions were obtained for the FYR of Macedonia, then Slovakia, Bulgaria and Poland. High depositions in these countries are mainly caused by significant national emissions. The contribution of global sources to European countries ranges from less than 4% in Poland to almost 70% in Iceland. However, in most of countries this contribution does not exceed 25% of total deposition.

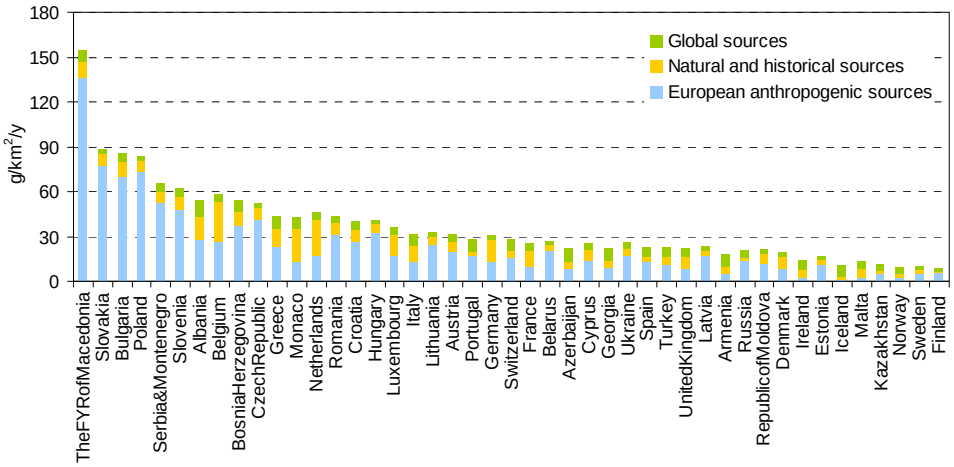


Fig. 4.7. *Country-averaged deposition fluxes of cadmium from European anthropogenic, natural/historical and global sources in 2004*

The contribution of the transboundary transport to cadmium anthropogenic depositions in European countries in 2004 varied from 13 to 97% (Fig. 4.8). In 30 countries it exceeded 50%, in 16 countries it is higher than 80%. The countries most affected by the transboundary transport of cadmium are Monaco, Albania, Republic of Moldova, Lithuania, Luxembourg and Sweden. The lowest contributions are estimated for remote countries such as Spain and countries with high national emissions – e.g. Poland.

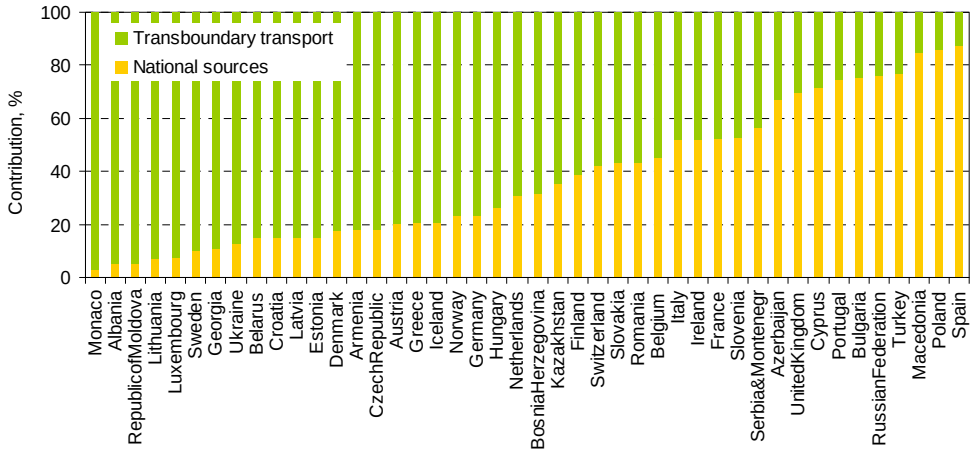


Fig. 4.8. *Relative contribution of the transboundary transport and national sources to anthropogenic cadmium depositions in European countries in 2004*

Contribution of each country to the atmospheric transboundary transport of cadmium was also estimated (Fig. 4.9). The largest absolute contribution to the transboundary transport of cadmium in 2004 is made by Poland (about 26 t/y), followed by Russia (16 t/y), Spain and Turkey (11 t/y). The

main reason for these high amount of atmospheric cadmium is high national emissions, compared to other countries of Europe. Similar to lead, the fraction of national emission involved into transboundary transport mostly ranges from 60% to 80%. The relative fraction in Russia does not exceed 30%, whereas in Monaco and Luxembourg it is higher than 90%.

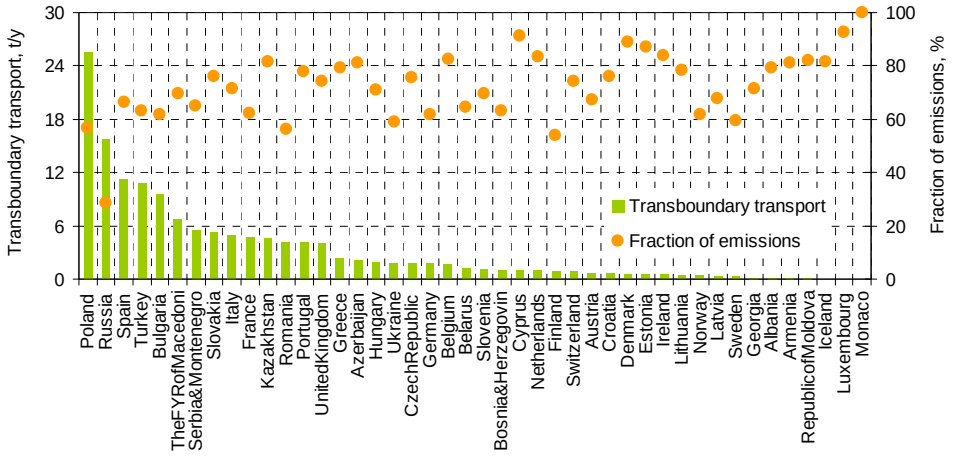


Fig. 4.9. *Absolute contribution of European countries to cadmium transboundary transport in Europe in 2004 and relative fraction of national emissions in this contribution*

The model evaluation against observations showed that monitored air concentrations of cadmium were underestimated by the model about twice, and concentrations in precipitation – 3 times (Fig. 4.10). Most probably, this underestimation is connected with underpredicted emission estimates (either anthropogenic or natural and historical or both). High uncertainty and possible incompleteness of cadmium anthropogenic emissions based on official data was repeatedly highlighted during last time [e.g. see *Minutes of the 7th TFMM Meeting, Helsinki*, 2006]. Besides, comparison of the official emissions data with different unofficial expert estimates demonstrates significant discrepancies of evaluated cadmium emissions. On the other hand, contribution of natural emissions and re-suspension of cadmium could be also significantly underestimated because of high uncertainty in the current understanding of the re-suspension process [*Gusev et al*., 2006]. In spite of considerable underestimation of observations, modelling results demonstrate high correlation with measured values: 0.76 for air concentrations and 0.71 for concentrations in precipitation. It means that the model satisfactory reproduced the spatial pattern of cadmium levels. More thorough analysis of the model evaluation is presented in Section 4.2 based on different emissions scenarios.

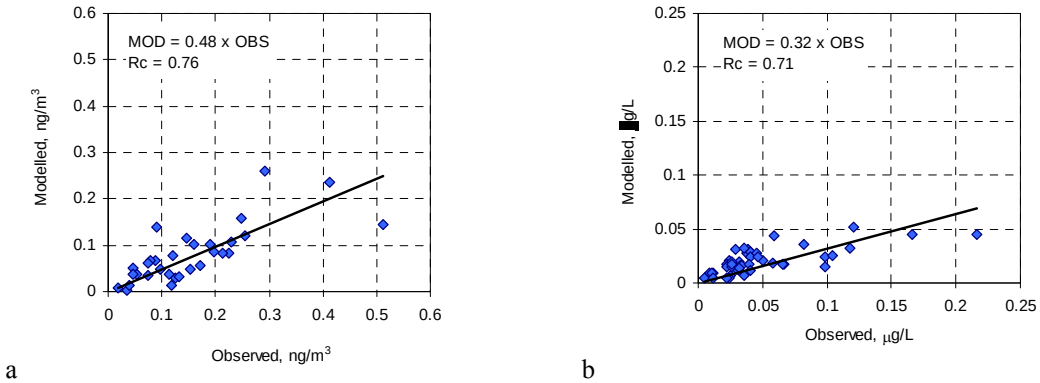


Fig. 4.10. *Comparison of modelled concentrations of cadmium in air (a) and in precipitation (b) with measurements from the EMEP monitoring network. Rc is the correlation coefficient*

Mercury

Total anthropogenic emissions of lead in European countries in 2004 according to official emissions data were 182 tonnes. It is by 7% lower than official emission estimates for 2003. Contribution of natural sources and re-emission of mercury from territories of European countries and the marginal seas (North, Black, Baltic, Mediterranean and Caspian) was about 115 tonnes. Total deposition of lead to European countries from European anthropogenic sources in 2004 is estimated at 57 tonnes. It is lower by 10% than those estimated for 2003 [*Ilyin and Travnikov*, 2005]. However, somewhat higher contribution of natural and global sources estimated for 2004 results in practically the same mercury total deposition from all (anthropogenic, natural, global) sources in comparison with 2003 (148 tonnes).

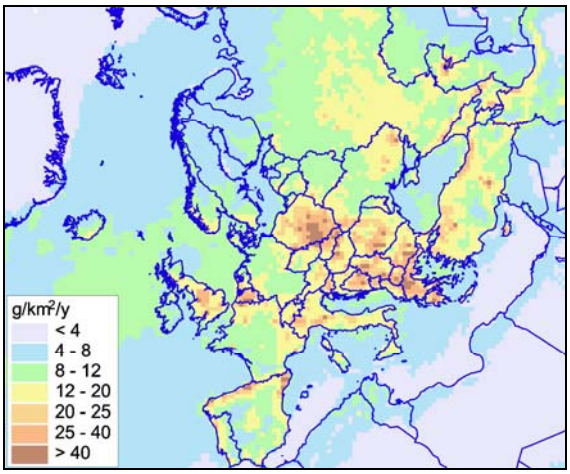


Fig. 4.11. *Spatial distribution of mercury depositions in Europe in 2004*

Generally, mercury depositions in Europe in 2004 were higher than 4 g/km^2/y but rarely exceeded 40 g/km^2/y (Fig. 4.11). The highest deposition fluxes took place in Poland, Belgium, the United Kingdom, and Balkan countries, where depositions are as large as 25 g/km^2/y and higher. Over central parts of the Scandinavia Peninsula and the Arctic the depositions did not exceed 8 g/km^2/y.

Country-averaged deposition fluxes and contribution of different types of emission sources to mercury deposition to European countries in 2004 is presented in Fig. 4.12. The highest average fluxes are estimated for the FYR of Macedonia, Greece, Slovakia, Belgium and Poland. In 18 European countries anthropogenic emissions made up the major contribution to mercury depositions, whereas in the other 27 countries contribution of global emissions prevails over the anthropogenic ones. The contribution of natural emissions and re-emissions from the EMEP region is comparatively small. It is expected that mercury is emitted from natural sources and re-emitted in the elemental gaseous form. This mercury form is characterized by very long residence time in the atmosphere. As a result the most part of these emissions is transported behind the model domain not being deposited.

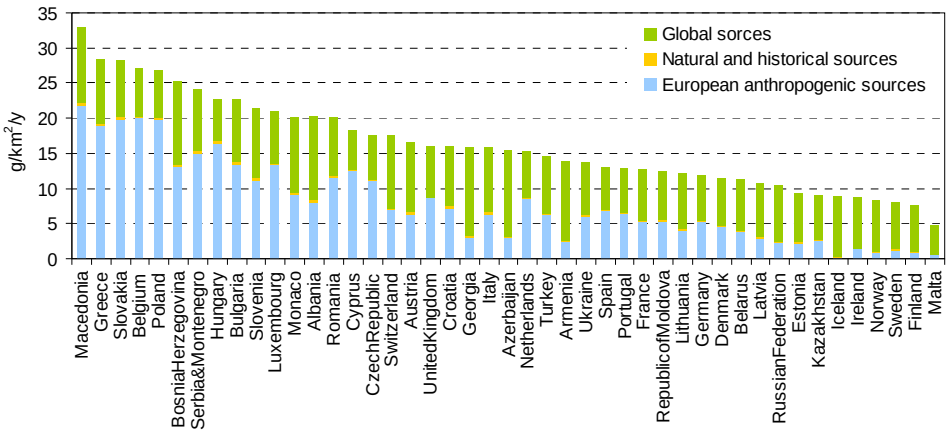


Fig. 4.12. *Country-averaged deposition fluxes of mercury from European anthropogenic, natural/historical and global sources in 2004*

The role of the transboundary transport in mercury pollution from anthropogenic sources in Europe varies greatly from country to country (Fig. 4.13). The highest contribution of the transboundary transport to anthropogenic mercury deposition took place in Iceland and Latvia (more than 95%). In 23 European countries this contribution exceeds 50% and in 8 countries – 80%. Similar to lead and cadmium, the highest input from external anthropogenic sources is typical for countries with small national emissions. As a rule, low contribution of external European sources is typical for countries with high national emissions (e.g. Poland) and countries located far from main anthropogenic sources (Spain, United Kingdom).

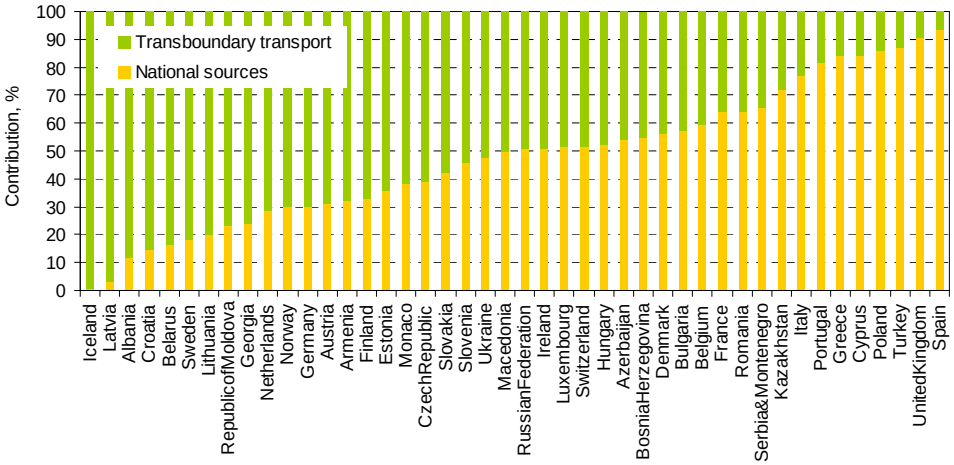


Fig. 4.13. *Relative contribution of the transboundary transport and national sources to anthropogenic mercury depositions in European countries in 2004*

Contribution of different countries to mercury transboundary transport in Europe in 2004 is illustrated in Fig. 4.14. As seen from the figure Spain was the largest contributor of mercury to long-range transport. As much as 18 tonnes of mercury emitted by Spanish sources were transported outside its territory. Other major contributors are Turkey (16 tonnes), Poland (15 tonnes) and Greece (11 tonnes). Significant transboundary transport from these countries is mainly caused by their large national emissions. The fraction of mercury emissions contributed to the transboundary transport was typically higher than that of lead and cadmium and commonly exceeds 80%.

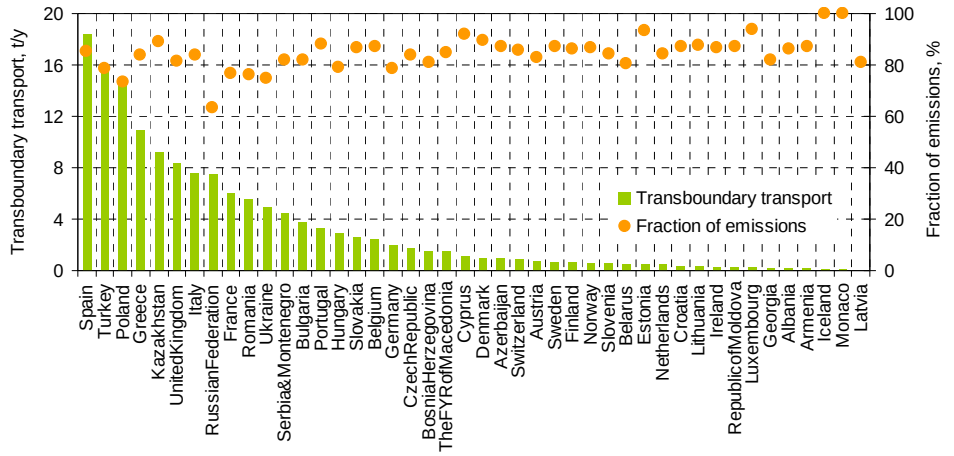


Fig. 4.14. *Absolute contribution of European countries to mercury transboundary transport in Europe in 2004 and relative fraction of national emissions in this contribution*

Observed concentrations of gaseous elemental mercury (GEM) in air were relatively well captured by the model. The difference between measured and modelled values is within ±10% (Fig. 4.15). Due to long atmospheric lifetime mercury is well mixed over the globe and does not exhibit significant variability in space and time on regional scale. Because of this feature its spatial and temporal variability is comparable with the model intrinsic uncertainty (~20%) [*Travnikov and Ilyin,* 2005].

Concentrations of mercury in precipitation, measured at the EMEP monitoring stations were somewhat underestimated by the model (Fig. 4.16). For German and Norwegian stations DE1, DE9, DE2, DE7 and NO1 the underestimation did not exceed 25%. At Swedish station SE14 and Dutch NL91 the underprediction is about 40 – 45%, which is comparable with uncertainty of the modelled concentrations in precipitation.

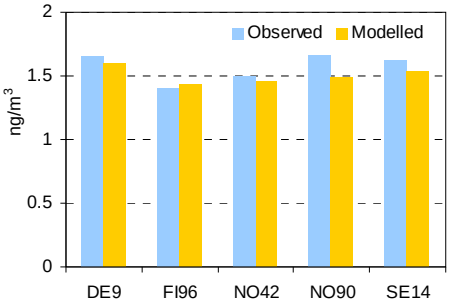


Fig. 4.15. *Comparison of modelled GEM concentrations in air with measurements from the EMEP monitoring network*

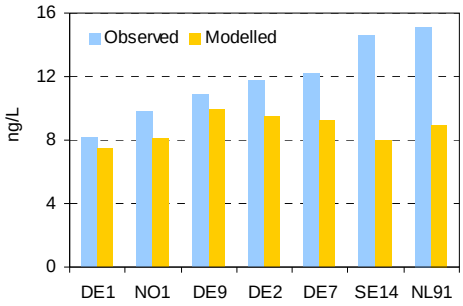


Fig. 4.16. *Comparison of modelled concentrations of mercury in precipitation with measurements from the EMEP monitoring network*

Previous results of MSC-E modelling of mercury [e.g., *Ilyin and Travnikov*, 2005] tended to somewhat overestimate mercury concentrations in precipitation. The following reasons can lead to these changes in the modelling results for mercury. First, in previous calculations of MSC-E emission from Germany was selected on the base of expert estimates [*Berdowski et al*., 1997, 1998]. Germany was characterized by the highest mercury emissions in EMEP region, and it contributed about 13% of total anthropogenic emission in Europe [*Ilyin and Travnikov*, 2005]. Hence, the contribution of Germany to mercury levels over own territory and in neighbouring countries was considerable. In calculations for 2004 officially reported data for Germany were used, and its contribution to total anthropogenic emission in Europe was only about 1.5%. This led to significant reduction of German influence on modelled mercury levels in and near Germany, and in particular, at monitoring stations.

Another reason, which can result in the deviation between modelled and measured concentrations, is connected with the temporal variations of the observed concentrations. Mercury concentrations in precipitation measured at stations SE14, NL91 and DE2 demonstrated unexpected increase by a factor 1.5–2 between 2003 and 2004 (Fig. 4.17) which was not captured by the model. The reasons of this sharp increase are not clear at the moment and should be further investigated.

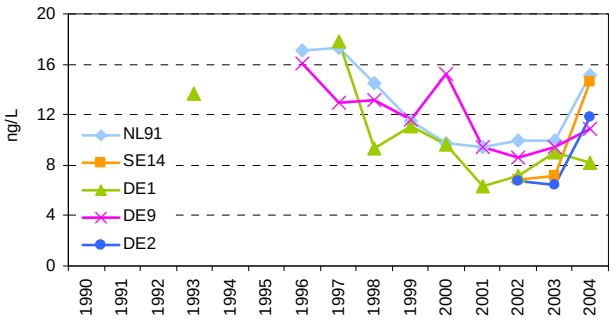


Fig. 4.17. *Long-term variability of mercury concentrations in precipitation at some monitoring stations.*

4.2. MSCE-HM model evaluation based on different emission scenarios

Preparatory work of MSC-E to the review of MSCE-HM model has shown that the modelled concentrations and depositions of lead and cadmium based on officially reported emissions lead to significant (2–3 times) underestimation of the observed values [*Ilyin and Travnikov*, 2005]. *O.Travnikov and I.Ilyin* [2005] have also demonstrated that emission data are the major source of uncertainties of model estimates. On the other hand, the issue of emission data quality has been raised and discussed at meetings under the Convention. In particular, the Executive Body to the Convention "expressed concern at the insufficient reporting of emission data on HMs and urged Parties to work to improve this" [*ECE/EB.AIR/87*]. The meeting of Task Force on Measurements and Modelling (Helsinki, Finland, 2006) also recognized that the significant difficulties with official emission data remain and further work to improve national emission estimates is needed.

MSC-E carried out model experiments aimed at evaluation of model performance under different emission scenarios of lead and cadmium. Two anthropogenic emission datasets were involved into the model experiments. One set is based on officially reported emission data for 2000. For countries, which have not provided their emission data for 2000, TNO emission expert estimates were applied [*Denier van der Gon et al*., 2005]. An alternative emission data set for 2000, developed in the framework of EU project ESPREME [*http://espreme.ier.uni-stuttgart.de/data.html*] was used for deposition modelling of lead and cadmium.

In addition to anthropogenic emissions, natural and historic emissions were used in the model experiments. This natural and historical emission represents re-suspension of particle-bound heavy metals from the Earth's surface to the atmosphere, caused by wind erosion. The approach developed in MSC-E to quantify these emissions is briefly described in section 3.1 of this report, and more thoroughly in MSC-E technical report [*Gusev et al*., 2006].

Three model experiments were performed for lead and cadmium. In the first one official/TNO emissions were used, in the second natural and historical emissions were added and in the third one the official/TNO emissions were replaced by emissions of the ESPREME project. The emission totals for Europe, used in these experiments, are given in Table 4.1. Totals for re-suspension of natural and historical emissions, presented in the table, include emissions from territory of Europe and from the surrounding seas. As seen, for Europe as a whole natural and historic emission of lead is comparable with the anthropogenic ones, while for cadmium it is much less. It is also clear, that ESPREME emissions of lead are slightly higher, and for cadmium - much higher than those based on official and TNO data. Differences in spatial distributions of these emission datasets are described in [*Gusev et al*., 2006].

Table 4.1. *Total emissions from Europe in 2000, t/y, used in the experiments*

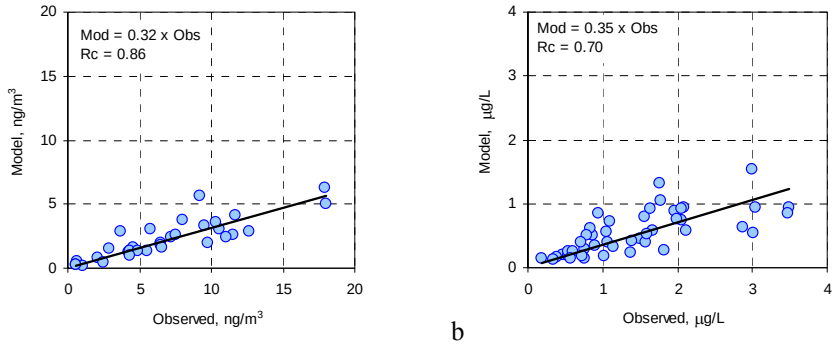
	Pb	Cd
Official/TNO	11180	280
ESPREME	13160	580
Natural and historical*	6800	65

* Includes emissions from the seas surrounding Europe: the North, Baltic, Mediterranean, Black and Caspian Seas.

Main results of this activity are summarized in this section. Detailed description of the experiments is available in the MSC-E technical report [*Gusev et al*., 2006].

Lead

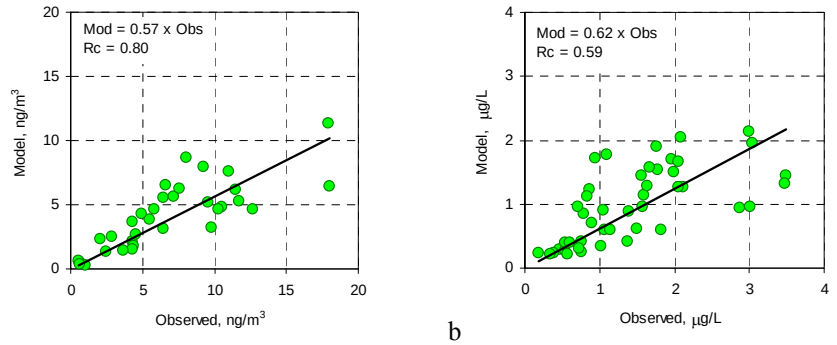
Concentrations of lead in air and precipitation, calculated on the base of only anthropogenic official/TNO emissions underestimate approximately by a factor of 3 (Fig. 4.18). Therefore, the use of only anthropogenic emissions provided by Parties to the Convention is not enough to explain the existing levels. Hence, two other tests were carried out.



a b

Fig. 4.18. *Comparison of modelled and measured concentrations of lead in air (a) and in precipitation (b) based on official/TNO emissions*

The addition of natural and historical emission leads to much better agreement with observations (Fig. 4.19). Measured concentrations both in air and in precipitation are underestimated by about 40%. If official/TNO emission is replaced by ESPREME emission, the agreement between measured and modelled quantities improves, although some underestimation (~20 – 25%) of observations still remains (Fig. 4.20).



a b

Fig. 4.19. *Comparison of modelled and measured concentrations of lead in air (a) and in precipitation (b) based on official/TNO emissions supplemented with natural and historic emissions*

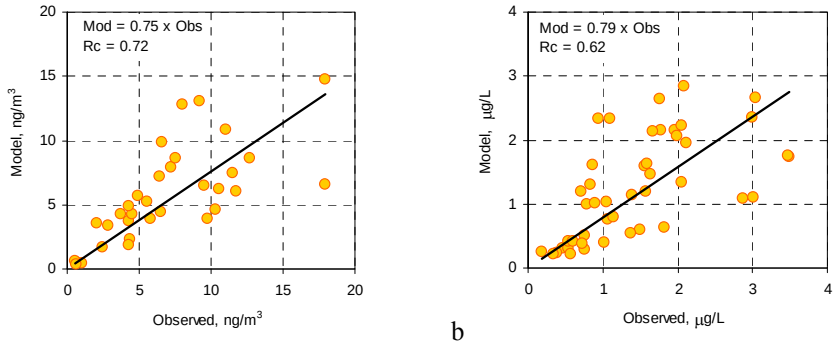


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Fig. 4.20. *Comparison of modelled and measured concentrations of lead in air (a) and in precipitation (b) based on ESPREME emissions supplemented with natural and historic emissions*

Cadmium

Similar set of simulations was carried out for cadmium. Annual mean concentrations of cadmium in air and in precipitation are also underestimated by a factor of 3 if only anthropogenic official/TNO emissions were used (Fig. 4.21). Similar to lead, in other model test run natural and historical emissions were included, and in another one official/TNO emission data were replaced by ESPREME emission estimates.

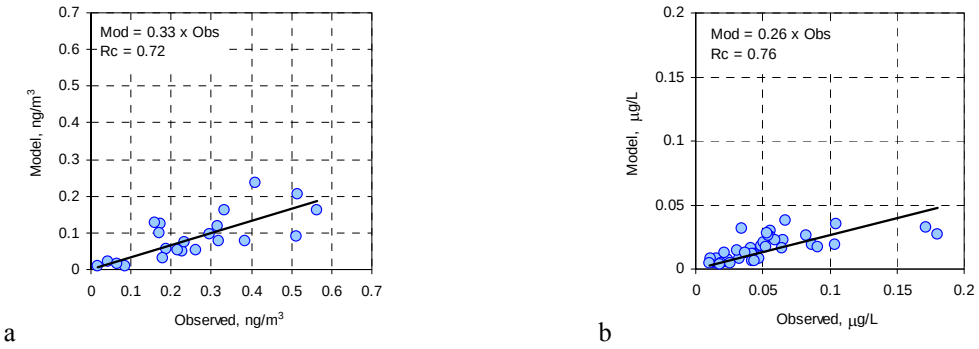

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Fig. 4.21. *Comparison of modelled and measured concentrations of cadmium in air (a) and in precipitation (b) based on official/TNO emissions*

The use of official/TNO emissions together with natural and historical emissions only slightly improves the model performance (Fig. 4.22). Although correlation coefficients increased, regression coefficients improved insignificantly: from 0.33 to 0.39 for air concentrations, and from 0.26 to 0.32 for concentrations in precipitation. This small increase of regression coefficients is explained by small contribution of cadmium natural and historical emissions (see Table 4.1), used in these experiments. ESPREME emissions of cadmium are significantly larger than official/TNO ones. Therefore, the use of these data resulted in much smaller underestimation of measured concentrations in air and precipitation (Fig. 4.23). However, the scatter of the results is high and correlation coefficients are lower compared to those obtained for official/TNO emissions. More detailed analysis of model results based on different cadmium emissions should be further continued.

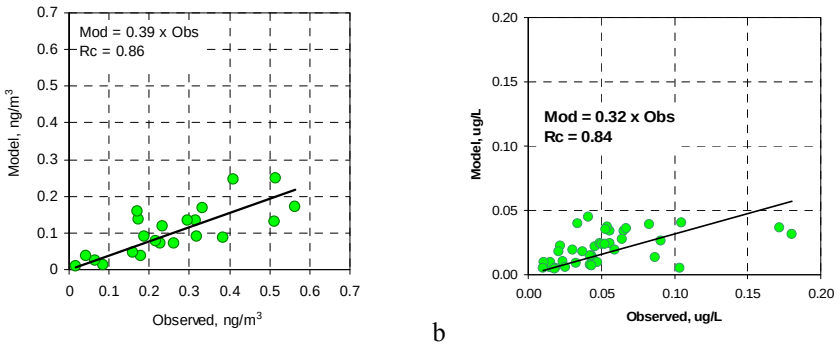

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Fig. 4.22 *Comparison of modelled and measured concentrations of cadmium in air (a) and in precipitation (b) based on official/TNO emissions supplemented with natural and historical emissions*

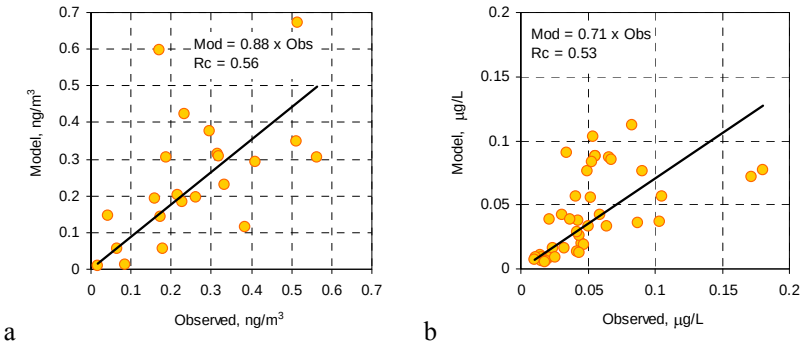

Fig. 4.23. *Comparison of modelled and measured concentrations of cadmium in air (a) and in precipitation (b) based on ESPREME emissions supplemented with natural and historical emissions*

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4.3. Hemispheric modelling of mercury pollution

Mercury is well recognized by the environmental community as a global pollutant capable of dispersing all over the globe. Because of its very long residence time in the atmosphere it can be easily transported by air masses between the continents and can significantly affect regional depositions both in industrially developed and in remote regions. Therefore mercury pollution levels in Europe cannot be assessed without consideration of the intercontinental transport.

Modelling of mercury atmospheric transport and depositions at the hemispheric/global scale greatly depends on availability of worldwide estimates of anthropogenic and natural emissions. Recently developed the new global mercury anthropogenic emissions dataset for 2000 [*Pacyna et al.,* 2006; *Wilson et al.,* 2006] allows updating the mercury hemispheric pollution estimates as well as evaluating changes of mercury depositions in the northern hemisphere. Fig. 4.24a shows spatial distribution of mercury anthropogenic emissions in the northern hemisphere in 2000 according to the estimates by *Wilson et al.* [2006]. Elevated emission fluxes are characteristics of Europe, Eastern part of North America, Southeastern and Southern Asia. As seen from Fig. 4.24b the most significant reductions of mercury emissions for the period 1995-2000 took place in Europe and North America. Emissions in Southeastern Asia decreased slightly and even increased in Southern Asia (e.g. in India).

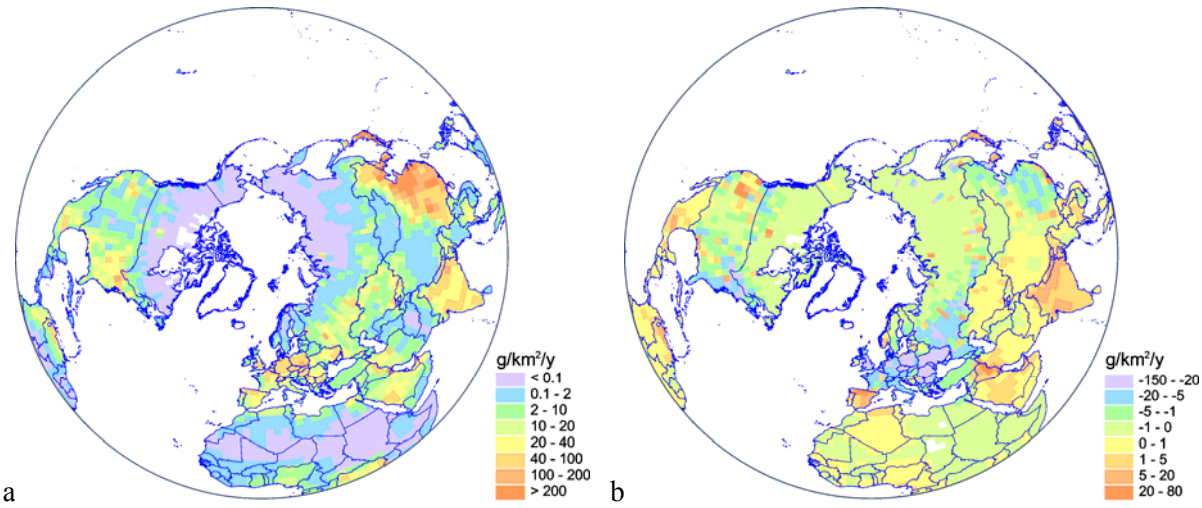


Fig. 4.24. *Spatial distribution of Hg anthropogenic emissions in the northern hemisphere in 2000 (a) and changes in Hg emissions for the period 1995-2000 (b). Positive values of changes correspond to increase*

The available anthropogenic mercury emissions datasets [*http://www.amap.no/Resources/HgEmissions/*] along with estimates of global natural emissions [*Travnikov and Ryaboshapko*, 2002] were utilized for evaluation of mercury hemispheric pollution in 2000 and changes in mercury depositions between 1996 and 2000. One-year model spin-up was performed prior to the calculations in order to fill in the model domain with mercury. Calculated pattern of total mercury deposition in the northern hemisphere in 2000 is shown in Fig. 4.25a. The spatial distribution of depositions well corresponds to the emission field (Fig. 4.24a): Elevated depositions (more than 15 g/km^2/y) are in Europe, North America, Eastern and Southern Asia. It can be explained by the fact the mercury depositions are defined to a greater extent by short-lived mercury forms (oxidized gaseous and particulate mercury), which, in their turn, make up to 50% of direct anthropogenic emissions. On the other hand, significant depositions (up to 10 g/km^2/y) are also predicted over the oceans as a result of oxidation of long-lived elemental mercury in the atmosphere by ozone, hydroxyl radical and reactive halogens.

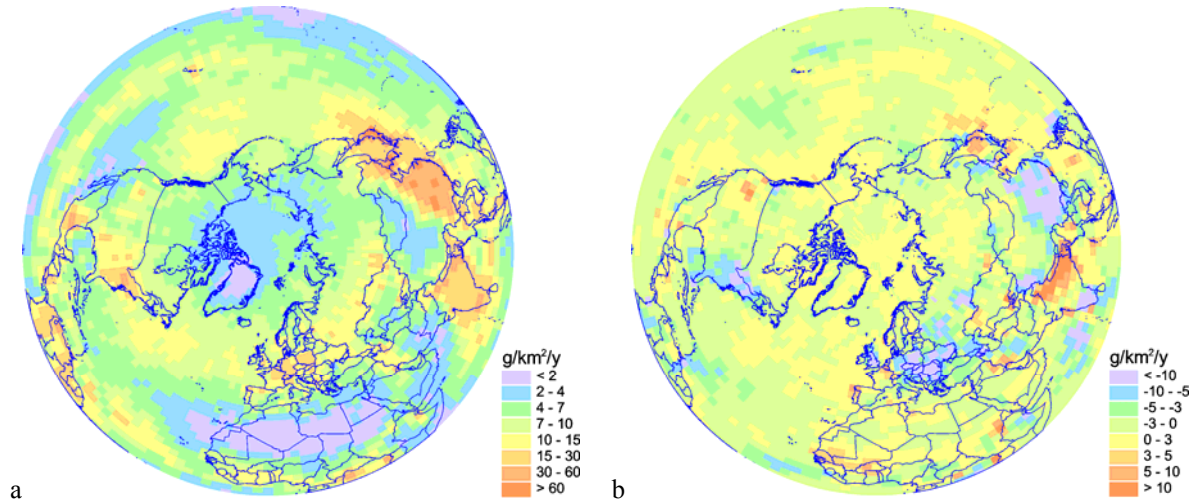


Fig. 4.25. *Spatial distribution of Hg total deposition in the northern hemisphere in 2000 (a) and changes in Hg depositions between 1996 and 2000 (b). Positive values of changes correspond to increase*

Estimated changes in mercury depositions between 1996 and 2000 in the northern hemisphere are illustrated in Fig. 4.25b. It should be noticed that in some cases the changes could be affected by the inter-annual meteorological variability. In general, mercury depositions did not change significantly for this period. Over the most part of the northern hemisphere the changes do not exceed ±3 g/km^2/y. The most significant decrease of mercury depositions (by 10 g/km^2/y) took place in Europe and North America because of reduction of anthropogenic emissions. Significantly lower depositions in 2000 in comparison with 1996 are also predicted in Southeastern Asia, but in this case the decrease can be explained by considerably smaller precipitation amount in 2000 in this

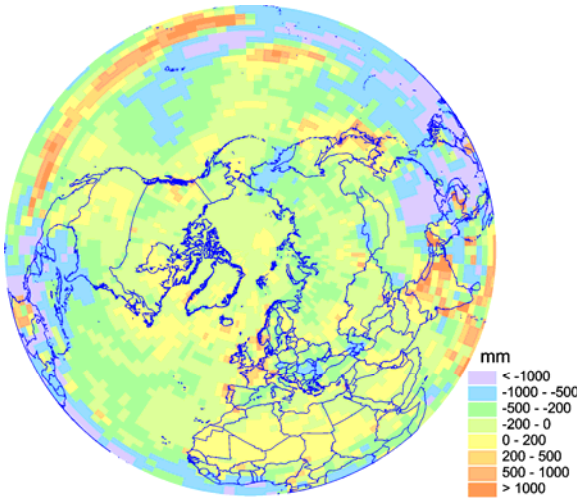


Fig. 4.26. *Difference in precipitation amount between 1996 and 2000. Positive values correspond to higher precipitation in 2000*

region (see Fig. 4.26). Substantial increase of mercury depositions in India resulted from both increase of emissions (Fig. 4.24b) and also larger precipitation amount (Fig. 4.26).

Pathways and intensity of mercury intercontinental transport to Europe can vary from season to season depending on typical synoptic conditions. Calculated seasonal variation of mercury inflows to Europe and outflows from Europe in 2000 through the Western, Eastern, Northern and Southern boundaries is shown in Fig. 4.27a. In the figure inflows correspond to positive values, whereas outflows – to negative ones. The European domain boundaries used in the assessment schematically depicted in Fig. 4.27b. As seen from the Fig. 4.27a the most significant incoming and outgoing fluxes are through the western and eastern boundaries, respectively. The input of mercury through the western boundary is almost twice higher during winter months than in summer. The elevated mercury input in winter is accompanied by significant outflow through the northern boundary to the Arctic. The net flux through the southern boundary varies around zero during the whole year.

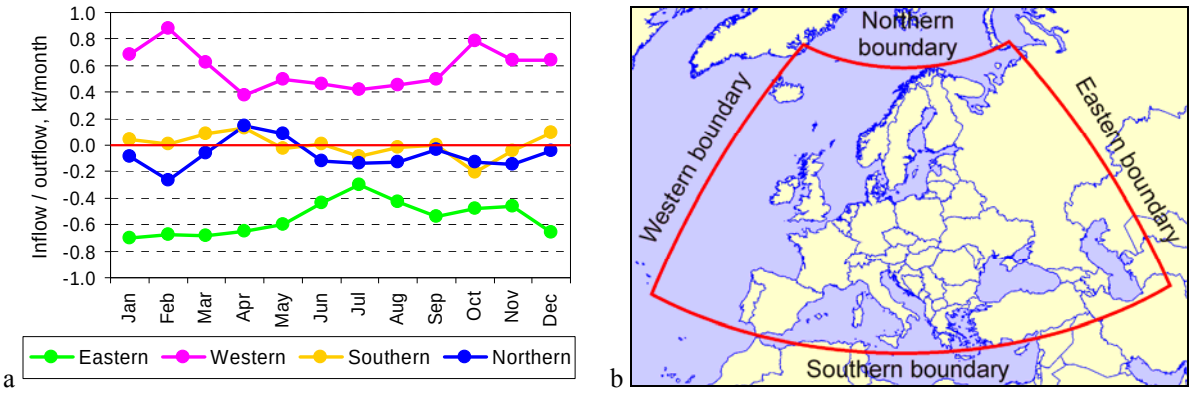
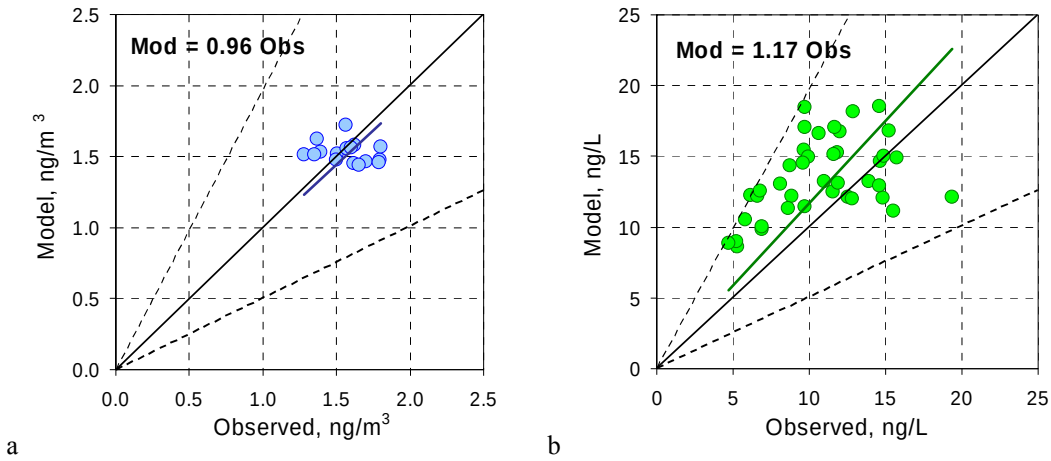


Fig. 4.27. *Seasonal variation of mercury inflow to and outflow from European domain through Western, East, Northern and Southern boundaries (a) and a sketch of the domain boundaries used in the assessment (b)*

In order to evaluate the modelling results calculated mercury concentrations in air and precipitation were compared with available measurements. For this purpose we used long-term observations from the EMEP monitoring network in Europe as well as the NADP/MDN and CAMNet networks in North America. Besides, measurements at the Arctic stations Barrow, Alert and Amderma were involved in the comparison. Location of monitoring sites used in the model evaluation is shown in Fig. 4.28.

Results of the comparison are presented in Fig. 4.29. As seen modelled concentrations of gaseous elemental mercury (GEM) well agree with observed ones (Fig. 4.29a). Discrepancy between calculated and measured values does not exceed 20% because of low spatial and temporal variability of GEM concentrations in the ambient air.

Fig. 4.28. *Location of monitoring sites used in the model evaluation: Triangles – gaseous elemental mercury (GEM) concentration; circles – concentration in precipitation; squares – both GEM and concentration in precipitation*



a b

Fig. 4.29. *Calculated vs. measured values of mean annual concentration of gaseous elemental mercury (a) and mercury concentration in precipitation (b). Dashed lines depict two-fold difference interval*

The model somewhat overestimates observed mercury concentration in precipitation (Fig. 4.29b). Besides, scattering of modelled and observed concentration in precipitation is more significant than in the case of GEM concentration. Nevertheless, difference between the calculated and measured values does not exceed a factor of two.

Along with the estimates of the intercontinental transport, the hemispheric model was applied for implementation of the one-way nesting to the regional modelling of mercury pollution. For this purpose monthly mean concentrations of three mercury forms (elemental, reactive gaseous and particulate) were calculated with the hemispheric model at the EMEP domain boundaries and were assimilated the regional model.

5. CO-OPERATION

5.1. Task Force on Measurements and Modelling

According to the EMEP Workplan for 2006 [*EB.AIR/GE.1/2005/Rev.1/*], MSCE-HM model has been reviewed at the EMEP/TFMM Workshop, held in Moscow in October 2005. The results of the workshop were adopted at the 7th Meeting of the Task Force on Measurements and Modelling (Helsinki, May 2006). The Task Force concluded that "…the MSC-E HM model is suitable for the evaluation of the long range transboundary transport and deposition of HMs in Europe". However, a number of recommendations, aimed at further improvement of heavy metal atmospheric modelling were drawn [*ECE/EB.AIR/GE.1/2006/4*]:

- to extend MSCE-HM model to consideration of other heavy metals (Ni, Cu, Cr, As, Zn and Se)
- to develop emission algorithms and models for representation of emissions driven by meteorological processes
- to extent MSC-E mercury model to the global scale
- to investigate potential influence of climate change on the fate and behaviour of mercury
- to provide some validation of meteorological fields generated
- to consider increasing of number of layers along vertical and time resolution of meteorological data employed
- to consider moving to meteorological data of European Centre for Medium Range Weather Forecasts, and to increase spatial resolution of input meteorological data from 2.5°x2.5° to 1°x1°.

Essential part of MSC-E work in 2006 was focused on solving the tasks posed by the TFMM workshop. The preliminary results of this work were also presented and approved at the 7th TFMM meeting in Helsinki. Besides, TFMM noted that further work is needed on improving national emission inventories.

5.2. Working Group on Effects

Working Group on Effects (WGE) has requested MSC-E to take part in a collaborative activity aimed at evaluation of exceedances of critical loads of heavy metals. The considered metals include those listed in the Protocol on heavy metals (lead, cadmium, mercury) as well as other metals – arsenic, chromium, nickel, zinc, copper and selenium. Four emission scenarios were considered. The first one is based on the emission data for 2000 officially reported by Parties to the Convention to UNECE. Other scenarios are based on various emission projections or 2020, which are described in detail in [*Denier van der Gon et al*., 2005].

In addition to anthropogenic emissions, natural and historic emissions of particulate metals, described in Section 3, were used. MSC-E calculated ecosystem-dependent depositions of heavy metals using meteorological data for 2000 for all four emission scenarios. MSC-E also carried out calculations of ecosystem-dependent depositions of lead, cadmium, mercury, arsenic, nickel and chromium on the base of alternative emission data set. This set was prepared within ESPREME project. The information about the project is available through the website [*http://espreme.ier.uni-stuttgart.de/data.html*].

The detailed analysis of the obtained results of this activity is planned to be presented in collaborative report led by Coordinating Center for Effects (CCE) of WGE. This section deals with some examples of the results provided for CCE.

To evaluate exceedances, CCE uses ecosystem-dependent depositions of heavy metals. MSC-E calculates depositions to 18 land-cover categories. Map of spatial distribution of land-cover categories was prepared by CCE and its description is available in [*Travnikov and Ilyin,* 2005]. Figure 5.1 shows examples of spatial distribution of cadmium to crops and coniferous forests in 2000. As seen, over major part of Europe depositions of cadmium to crops vary from 3 to 50 g/km^2/y (Fig. 5.1a). Over the Scandinavian Peninsular and Spain depositions are below 10 g/km^2/y. In Poland, east of Germany, east of Ukraine, Belgium and some areas of the Balkan Peninsular the depositions exceed 50, and in some cases – 150 g/km^2/y. Depositions to coniferous forests are essentially higher compared to those over crops (Fig. 5.1b). They mainly ranged from 10 to 150 g/km^2/y. The regions with elevated depositions are the same as those for crops. Similar maps were available for other metals.

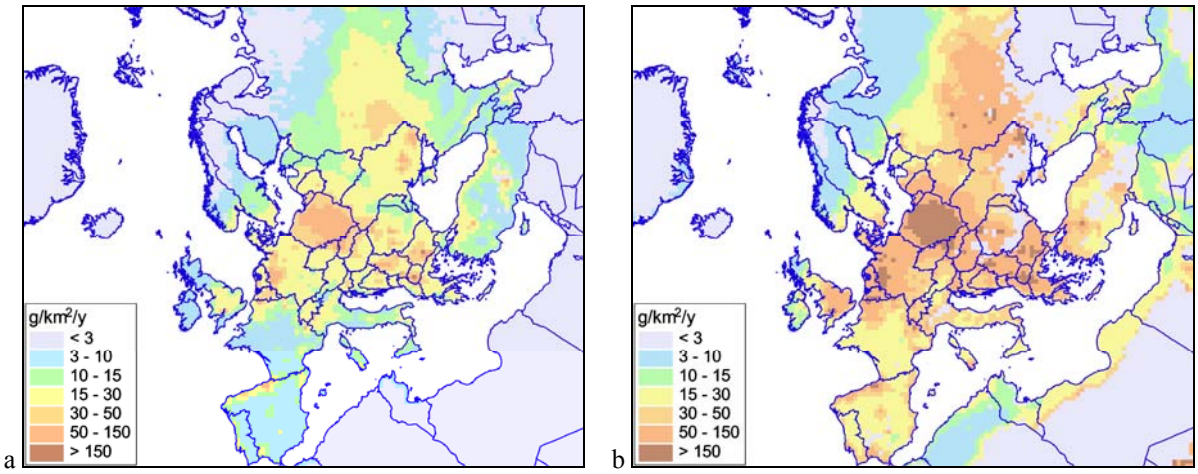


Fig.5.1. *Cadmium depositions to crops (a) and to coniferous forests (b) for 2000*

Statistical regularities of depositions to various land-cover categories were also analysed using cumulative distribution functions. These functions demonstrate per cent of i-th land-cover area S(di) where depositions are below a certain value di. For example, modelled depositions below 30 g/km^2/y occur over about 72% of coniferous forests ($1.53 \cdot 10^6$ km^2) and over 86% of croplands ($3.35 \cdot 10^6$ km^2) (Fig. 5.2). Therefore, over 28% of coniferous forests and 14% of croplands the depositions exceed 30 g/km^2/y. Similar plots for other metals, and for depositions based on the ESPREME estimates, were also prepared and transferred to CCE.

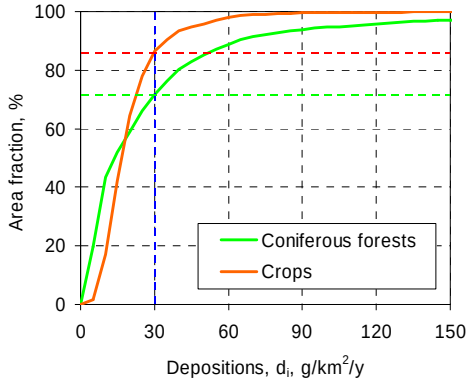
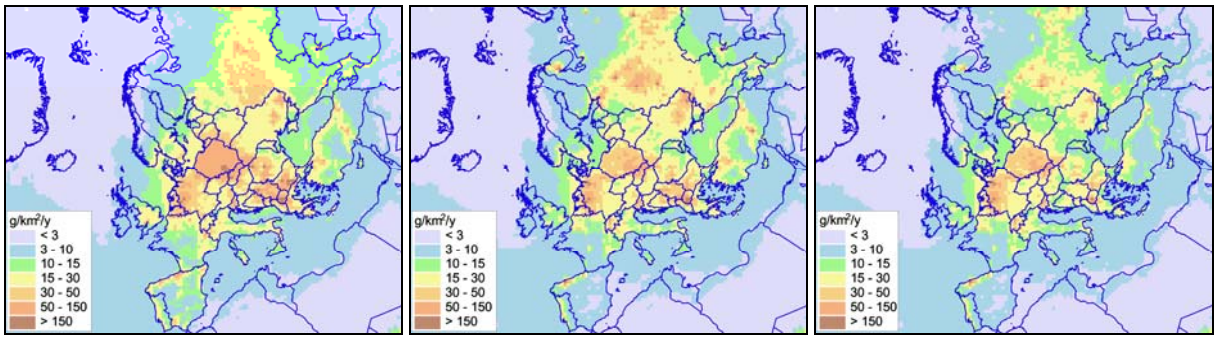


Fig. 5.2. *Cumulative distribution functions of cadmium depositions to coniferous forests and crops in 2000 calculated with official/TNO emissions*

Ecosystem-dependent depositions of nine heavy metals were calculated for 2000 and for three emission projection scenarios for 2020. Depending on emission scenario, deposition levels differ in regions of Europe. Example, showing total depositions of cadmium in Europe, calculated on the base of Official/TNO

emissions, and for two emission projection scenarios, is given in Fig. 5.3. Over most part of Europe depositions in 2020 are lower compared to those in 2000. Obviously, this is explained by the projected decrease of heavy metal emissions between 2000 and 2020. Nevertheless, in some countries, e.g. Russia or Ukraine, some increase of deposition was obtained. The reason for this is that in scenario for 2000 official emission were used for these countries, while for 2020 TNO estimates were applied. According to the available emission data, TNO emission estimates for these countries for 2020 are higher than official emissions for 2000.



a b c

Fig. 5.3*. Spatial distribution of total depositions of cadmium in 2000 and 2020. (a): in 2000, official and TNO emissions; (b): in 2020, scenario 2 (current legislation); and (c) in 2020, scenario 3 (Full implementation of the Protocol)*

Similar results for mercury are presented in terms of concentrations in precipitation, because this parameter is essential in evaluation of mercury critical levels [*UBA*, 2004]. Similar to depositions, the concentrations of mercury in precipitation exhibit decrease between 2000 and 2020 over major part of Europe (Fig. 5.4). However, similar to depositions of other metals, in Russia and the Ukraine concentrations of mercury in precipitation in 2020 are higher than in 2000.

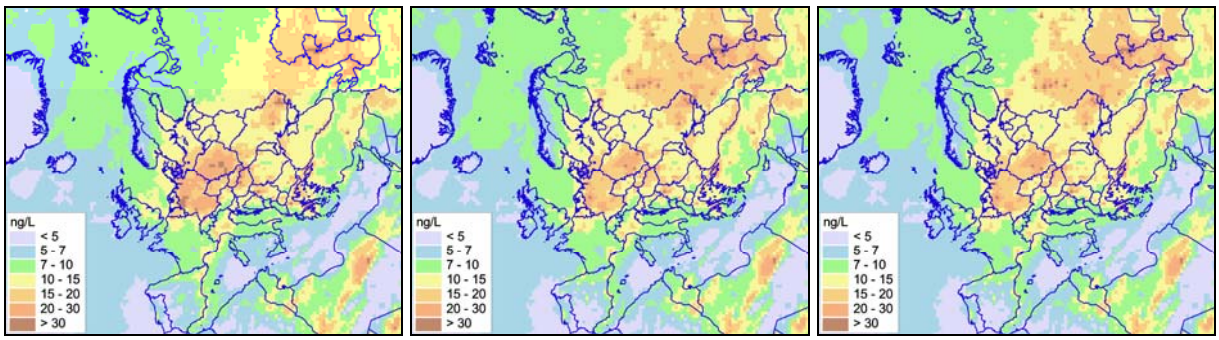


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Fig. 5.4. *Spatial distribution of mercury concentrations in precipitation in 2000 and 2020. (a): in 2000, official and TNO emissions; (b): in 2020, scenario 2 (current legislation); and (c) in 2020, scenario 3 (Full implementation of the Protocol)*

Depositions of all nine metals, based on the Official/TNO and the ESPREME emissions for 2000, and three emission projections for 2020 were transferred to CCE. The CCE experts have started evaluation of exceedances of atmospheric depositions over critical loads. An example demonstrating map of exceedance for nickel in 2000 is given in Fig. 5.5. The depositions to calculate the exeedance

are based on the Official/TNO emissions, and the exceedance was calculated including both ecotoxicological and human health effects. This map was kindly presented by CCE colleagues. More detailed information regarding deposition modelling results on the base of different emission scenarios and computation of exceedances will be available in joint CCE/MSC-E report.

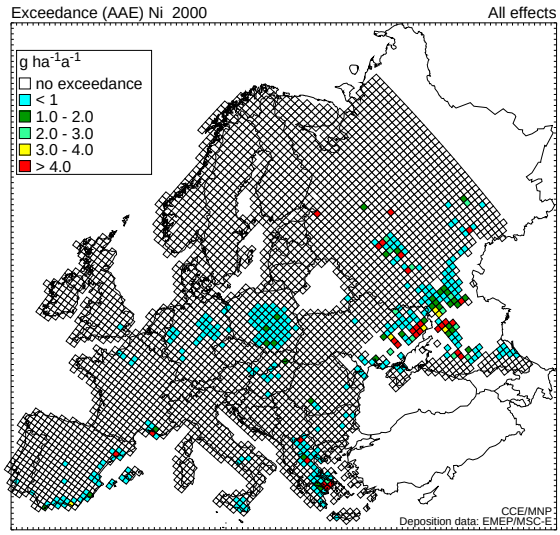


Fig. 5.5. *Exceedance of Ni depositions (official/TNO emissions) for 2000. Both ecotoxicological and human health effects are included*

5.3. Task Force on Heavy Metals

In 2006 MSC-E had a close cooperation with the Task Force on Heavy Metals and contributed to the preparatory work for the review of the Protocol. MSC-E attended two meetings of the Task Force (Dessau, Germany and Ottawa, Canada). The main aims of the meetings were to assist WGSR in preparation of the sufficiency and effectiveness review of the HM Protocol summarizing the best available scientific information about effects of depositions, emissions, technological development and changing economical conditions. MSC-E was responsible for the overview of emission data and contributed to the section on best available scientific information on the effects of the depositions of heavy metals. More detailed information, prepared by MSC-E for the review is available in MSC-E report [*Gusev et al*., 2006] and in the Internet at the website of TFHM [*http://www.unece.org/env/tfhm/*] or MSC-E [*http://www.msceast.org/*].

Emissions

Overview of emission covers period from 1990 to 2003. Background document deals with both metals targeted by the Protocol (lead, cadmium and mercury) and with other metals, which emissions are reported by Parties (arsenic, nickel, chromium, zinc, copper and selenium). The document included the information on official emission data reporting, emission trends in Europe and North America, emission reductions/changes in countries, emission data quality and their uncertainties, key sources of emissions.

Emission trends of lead, cadmium and mercury in EMEP region for 1990 – 2003 were estimated for 24 countries, which reported their data at least for both 1990 and 2003. As for Parties outside EMEP,

emissions of lead, cadmium and mercury were reported by Canada for all years in the considered period, and the USA reported the emissions for 1990, 1996, 1999, and 2002. Besides, changes of emissions in individual countries between 1990 and 2003 were described. It was shown that lead emissions have reduced in all countries, cadmium and mercury emissions – in majority of countries. In some countries emissions of cadmium and mercury have increased.

Information on emission key sources was reflected on the base of emission sector data, reported in NFR (Nomenclature For Reporting) codes. There were 8 countries, which reported their sector emissions of all three metals, targeted by the Protocol, in NFR codes both for 1990 and 2003. This information could be useful for the development of potential future strategies to further reduce emissions both in individual countries and in the EMEP region as a whole.

According to the Guidelines for estimating and reporting emission data [*EB.AIR/GE.1/2002/7*], some countries every year re-calculate previously reported emission data. In 2005, 15 countries recalculated previously reported data on lead, cadmium and mercury for all or for selected years in the period of 1990-2002. It was shown that in some cases current and previously reported emission data can differ by tens of percents or even by an order of magnitude.

Section, devoted to heavy metal emission uncertainties dealt with reported uncertainties of Danish emission estimates. According to [*Illerup et al*., 2005], the uncertainties of lead emissions were equal to 261%, cadmium - 263% and mercury - 229%. Another way to estimate the uncertainty of the emission data could be comparison of reported emission data with the data available through other sources, e.g. with emission expert estimates. Comparison of national reported emission totals with emission expert estimates [*Berdowski et al*., 1997; *Denier van der Gon et al.,* 2005] indicates that the difference between the emissions in some countries can exceed an order of magnitude. This fact points at necessity of more careful analysis of the data from viewpoint of uncertainties.

The changes in arsenic, chromium, copper, nickel and zinc emissions in the EMEP region for the period of 1990-2003 were estimated on the basis of total emissions of the 17 countries, and for selenium – of 13 countries. For the considered period they reported their national emission data, on a voluntary basis, for each of the above metals.

Total reported emissions of other metals in the considered countries have decreased between 1990 and 2003. The magnitude of the decrease ranges from 70% (chromium) to 7% (selenium). Finally, background document also describes changes of the emissions of other metals in individual countries and presents information about key sources of the emissions for 2003.

Atmospheric transport

The background document summarizes best available information about atmospheric transport and depositions of heavy metals, their levels and effects in the environment and biota. MSC-E contribution included brief assessment of monitored concentrations in air and in precipitation of lead, cadmium and mercury for 1990 – 2003, long-term trends of depositions to countries of Europe and information about source-receptor relationships. Besides, intercontinental transport of mercury was discussed.

According to EMEP monitoring data, the country-averaged concentrations of lead in air and in precipitation in Europe decreased 1.5 – 3 times, of cadmium – 2 – 4 times for 1990 - 2003. Concentrations of mercury in precipitation reduced about twice. Air concentrations of mercury do not demonstrate any noticeable long-term trend.

Information about long-term changes of modelled pollution levels in European countries was presented as ratio of country-averaged levels in 1990 to those in 2003. According to MSC-E modelling results [*Ilyin and Travnikov*, 2005], the overall decrease of modelled lead and cadmium in Europe made up 2.3 times, of mercury – 1.6 times.

The decrease of concentrations and depositions in individual countries may differ essentially from one country to another. For example, the largest decrease of air concentrations, averaged over country's territory, took place in Slovenia (7 times) (Fig. 5.6). In 18 countries the concentrations decreased at least 3 times, and in 16 countries – between 2 and 3 times. The highest decline of air concentrations of cadmium took place in Monaco (3.4 times) and the Netherlands (3.3 times). Germany, the Czech Republic, Republic of Moldova and Slovakia are characterized by the highest (up to 3 times) decrease of mercury depositions.

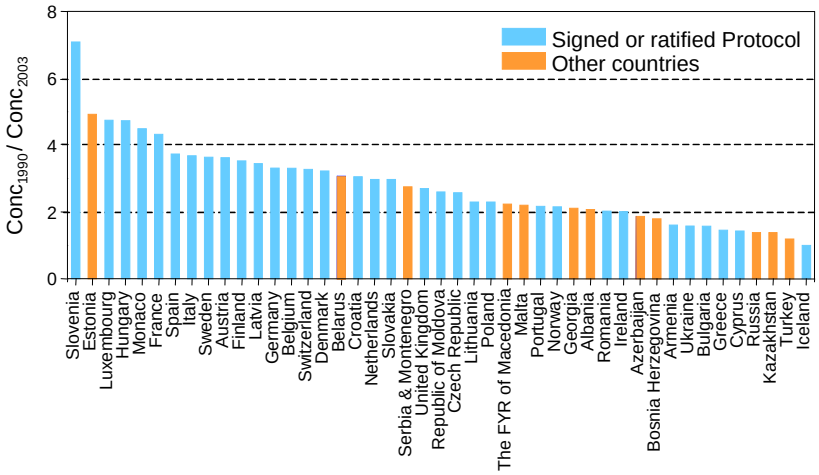


Fig. 5.6. *Ratio of the modelled country-averaged air concentrations of lead in 1990 to those in 2003 in countries of Europe*

Besides, the contribution of the transboundary transport to pollution of European countries with heavy metals was evaluated on the base of MSC-E modelling results. The intercontinental transport of mercury was discussed and its contribution to mercury pollution levels in Europe was demonstrated.

5.4. Task Force on Health

The Executive Body for the Convention requested the Joint WHO/UNECE Task Force on Health Aspects to assess and report on health effects of long-range transport of heavy metals (cadmium, lead and mercury). The report on health effects from long-range transboundary air pollution (LRTAP), prepared under supervision of Task Force on Health, deals with sources, atmospheric transport and depositions of heavy metals, their contents and budgets in environmental media (air, soil, waters), and reported health effects of the metals for humans. The main aim of the report was to establish links between concentrations of heavy metals in the environmental media and inputs to human bodies as well as contribution of LRTAP to pollution levels in the environment.

Contribution of MSC-E to the report dealt with the most recent information on sources, transport, modelled and measured concentrations/depositions of lead, cadmium and mercury. The information presented by MSC-E covered period of time between 1990 and 2003. Heavy metal emissions and their trends were described on the base of both officially reported data and emission expert estimates. Special attention was paid to evaluation of ability of the metals to transport over long distances.

The main conclusions of the report were formulated at the 9th meeting of the Joint Convention/WHO Task Force on Health Aspects of Long-range Transboundary Air Pollution held in Berlin, Germany, 30-31 May 2006. The main conclusions concerning importance of LRTAP on heavy metals health effects of humans presented below.

Cadmium

Inputs of LRTAP and fertilizers to top soil are roughly of the same magnitude. These inputs continue to contribute to already accumulated, relatively large present store of cadmium in the top soil. In spite of the decreasing cadmium emissions, and pollution levels in the environment, the decrease of cadmium body burdens in non-smokers in the last decade is not recorded, according to recently published data. Cadmium is accumulating in soils and catchments, thus increasing the risk of exposure through food. Therefore, every effort should be made to further reduce cadmium emissions and the direct input of cadmium to the soil.

Lead

Annual inputs of lead to top soils through LRTAP are similar to those from fertilizers. These inputs are relatively small compared to lead stores already accumulated in soils. However, the LRTAP may contribute to lead contents of crops significantly through deposition directly on the plants. Though uptake via roots is relatively small, on the long term the rise of lead level in soils is a matter of concern and should be avoided due to the possible health risk of low level lead exposure.

Mercury

Little information is available on the provenance of methylmercury in marine fish, and on the contribution of long-range transport in the process. Evidence exists on increasing levels in marine fish and mammals in the Arctic, indicating the impact of long range transport.

5.5. Task Force on Hemispheric Transport of Air Pollution

MSC-E and CCC continued cooperation with the EMEP Task Force on Hemispheric Transport of Air Pollution (TF HTAP). The Centres took part in the TF HTAP Workshop on organization of intercomparison of intercontinental models held in Washington, USA, in January 2006. MSC-E presented the results of intercomparison studies of mercury and POPs models, previously organized and supervised by MSC-E. In addition to this, MSC-E shared experience obtained through these studies and proposed its view on the forthcoming intercomparison of intercontinental transport models. CCC overviewed measurement data available for the intercomparison.

Besides, MSC-E supported the organization of the second meeting of the TF HTAP (Moscow, Russia 2006) and presented information on its regional and hemispheric mercury modelling activities. Particularly, the hemispheric estimates of mercury atmospheric transport demonstrate that intercontinental contributions to mercury deposition in the EMEP region, North America, and the Arctic are significant. For example, the intercontinental transport contributes 25-60% to mercury depositions in Europe.

5.6. European Commission and national experts

ESPREME project

MSC-E in 2006 participated in the EU ESPREME project (SSPI-CT-2003-502527) launched in 2004. The project aims to develop methods and tools to support European environmental policy making in the specific case of reducing the harmful impacts of heavy metals in a harmonised way across Europe. The role of MSC-E in the project is to assess by means of its chemical transport models the atmospheric dispersion of selected heavy metals (Hg, Pb, Cd, As, Ni and Cr) and their deposition to water and soil.

The main activities carried out by MSC-E during the last year were connected with extensive evaluation of the regional chemical transport model MSCE-HM, improvement of some model parameterizations and performing calculations for the base year and future scenarios both on regional and hemispheric scales. Particularly, the following activities were performed:

- Extensive evaluation of the chemical transport model MSCE-HM against available measurements was performed under joint support of EMEP and ESPREME. Modelled concentration of heavy metals in the ambient air and precipitation were compared with observations from the EMEP monitoring network for the period 1990-2003 and with calculations of other models. Results of the model evaluation were discussed at the EMEP/TFMM Workshop on the review of the EMEP models on HMs and POPs [*http://www.msceast.org/events/review.html*].

- Parameterization for the process of heavy metal re-suspension from soils and seawater was elaborated based on the state-of-the-art researches in the fields of mineral dust production and seasalt aerosol formation. Mobilization of previously deposited and natural heavy metals from soils and the ocean surface can considerably contribute to heavy metal pollution levels, particularly in regions remote from industrial sources. Developed parameterization was tested and incorporated to the chemical transport model.

- Hemispheric transport of Hg was evaluated using the hemispheric chemical transport model MSCE-HM-Hem for evaluation of boundary conditions for the regional modelling. Global character of Hg dispersion requires consideration of the contamination with this pollutant at scales wider than regional one since contribution because of significant contribution of the intercontinental transport. The most recent global Hg emission inventory for the year 2000 was utilized for the assessment [*http://www.amap.no/Resources/HgEmissions/*].

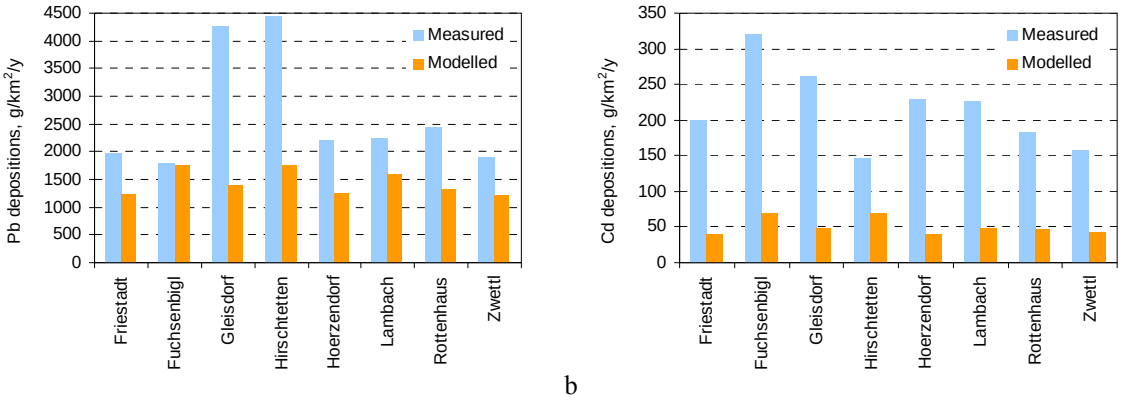
- Calculations of Pb, Cd, Hg, Cr, Ni, As atmospheric transport and depositions in Europe were performed for the base year 2000 and future scenarios for 2010. Obtained concentration and deposition fields were analyzed and compared with available measurements. Calculations of the source-receptor relationships were also performed for all European countries and all emission scenarios.

Mercury intercomparison study

Results of two last stages of the mercury models intercomparison study conducted with co-ordination of MSC-E in the period 2000-2005 were prepared for publication in a peer-reviewed literature. The first article contains description of comparison of modelling results obtained by different scientific groups with short-term measurements. The other one includes results of the intercomparison of long-term mercury calculations of mercury concentrations and depositions and evaluation of atmospheric balances for European countries. Both articles were submitted for publication in the journal "Science of the Total Environment".

External evaluation of modelling results

Depositions to croplands of Austria modelled by MSC-E were compared with the available measurements of depositions of lead, cadmium and mercury [*Spiegel et al*., 2003] and with depositions estimated from moss measurements. It was demonstrated that modelled data of lead somewhat underestimate, and cadmium – significantly underestimate measured levels (Fig. 5.7).



a b

Fig. 5.7. *Modelled and measured depositions of lead (a) and cadmium (b) over arable lands in Austria*

5.7. Helsinki Commission

During 2005 MSC-E in co-operation with EMEP Centres and Helsinki Commission prepared contribution to the joint annual report for HELCOM devoted to the evaluation of airborne pollution load to the Baltic Sea in 2003 [*Bartnicki et al*., 2005]. The Centre has prepared the information on atmospheric transport and depositions of lead, cadmium, and mercury to the Baltic Sea as well as contributions to depositions of particular HELCOM countries on the basis of officially submitted emission data.

MSC-E has provided updated environmental indicator reports with regard to temporal variations of heavy metals emissions to the atmosphere and their depositions over the Baltic Sea in period from 1990 to 2003. Prepared indicator reports are available in the Internet at the web site of Helsinki Commission [*www.helcom.fi*].

According to official data annual emissions of heavy metals from HELCOM countries have decreased during the period 1990-2002 by 47% for cadmium, 65% for mercury, and 62% for lead (Fig. 5.8). For individual countries, the most significant drop of cadmium emissions can be noted for Finland (80%). In case of lead and mercury emission, the most significant decrease can be seen for Sweden (97% and 83%). The reduction in heavy metal emission to the atmosphere is a consequence of increased use of lead-free fuels, use of cleaner production technologies as well as of economic contraction and industrial restructuring in Poland, Estonia, Latvia, Lithuania, and Russia in early 1990s.

The most significant drop in depositions over the Baltic Sea is obtained for lead (71%) (Fig. 5.9). The decrease of cadmium and mercury depositions is amounted to 41% and 43%, respectively. On the level of individual sub-basins the most significant drop in cadmium depositions can be noted for the Gulf of Finland (58%) and the Gulf of Bothnia (58%). In case of lead the most significant decrease can be noted for the Belt Sea (80%) and the Kattegat (79%). Largest decrease in mercury depositions is obtained for the Belt Sea (60%). In spatial distribution of heavy metals depositions on the Baltic Sea the highest levels can be noted for the southwestern part of the Baltic Sea (the Belt Sea and the Baltic Proper). Significant levels of lead and cadmium depositions can also be noted for the Gulf of Riga. Among the HELCOM countries the most significant contributions to depositions over the Baltic Sea belong to Poland, Germany, and Russia.

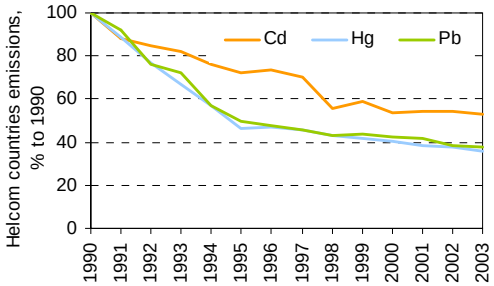


Fig. 5.8. *Trend of anthropogenic emissions of cadmium, mercury, and lead from HELCOM countries in 1990-2003 according to official emissions data*

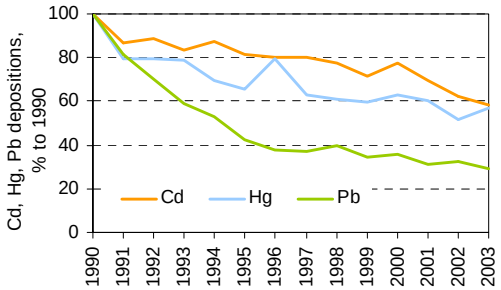


Fig. 5.9. *Temporal variations of cadmium, mercury, and lead depositions to the Baltic Sea in 1990-2003*

CONCLUSIONS

The activities of EMEP Centres CCC and MSC-E in the field of monitoring and modelling of heavy metals in 2006 were focused on assessment of the pollution levels in Europe in 2004, support of the Working Group on Effects in development of effect-based approach and contribution to the Task Force on Heavy Metals and the Task Force on Hemispheric Transport of Air Pollution. Special attention was paid to the response to recommendations formulated by EMEP TFMM Workshop aimed at further improvement of MSC-E model formulation and analysis of its input data. The main conclusions on the activities, undertaken by CCC and MSC-E, are presented below.

Monitoring of heavy metals

1. Measurements of heavy metal pollution levels in 2004 showed that the lowest concentrations of lead, cadmium and mercury were observed in Northern Scandinavia. In general, concentration levels increase towards the southeastern of Europe.

2. At present the EMEP monitoring network contains 63 stations measuring lead and cadmium, of which 26 stations measure concentrations of these metals both in air and precipitation. There are 18 stations where at least one mercury form is measured. The monitoring stations, however, are non-uniformly distributed over European territory: they are mainly located in Central and Northern Europe.

3. Annual analytical intercomparison of national laboratories treating measurements of heavy metals indicates an essential improvement of data quality during the period of 1995-2004, but still there is need for improvement for some countries.

Emissions

1. In 2006, 28 European countries (about 60% Parties to the CLRTAP located in the EMEP region) submitted national total and sector data on lead, cadmium and mercury emissions for 2004 reporting year. Among them 16 countries recalculated previously reported emission totals for all or selected years of the period of 1990-2003. In several countries emission changes after the recalculations exceed 100% for some years.

2. Completeness of reported national total emissions is not sufficed in many countries. It leads to underestimation of national emission totals.

3. Inconsistency in time series of sector emissions takes place in some countries. It makes total emissions for different years not commensurable and distorts emission trends.

4. In the EMEP region emissions of the priority metals (lead, cadmium, mercury) and other metals (in particular, arsenic, chromium and nickel) exhibit a tendency to reduction. However in some individual countries emissions of some metals increase.

Model review and development

1. MSCE-HM model has been reviewed at the EMEP/TFMM Workshop held in Moscow in October 2005. The Workshop concluded that the MSCE-HM model is suitable for the evaluation of the long-range transboundary transport and depositions of heavy metals in Europe. However, significant difficulties still remain with the official emissions for Pb and Cd. Besides, a number of recommendations, aimed at further improvement of heavy metal atmospheric modelling were drawn. The results of the workshop were adopted at the 7th Meeting of the Task Force on Measurements and Modelling (Helsinki, May 2006).

2. Following the recommendations of TFMM, a tentative parameterisation of wind re-suspension of particle-bound heavy metals (Pb, Cd, As, Ni and Cr) from natural surfaces (soil and seawater) was developed. Preliminary calculations demonstrate that wind re-suspension of some heavy metals can significantly contribute to airborne pollution in Europe.

3. Another activity aimed at improvement of the model formulation was connected with evaluation of input meteorological data. The procedure of the evaluation was initiated and preliminary results demonstrate that the meteorological fields used for the modelling process reasonably well agree with reference ECMWF data.

4. MSC-E has extended the model parameterisation for calculations of the atmospheric transport and depositions of the second priority metals (As, Cr, and Ni). Pilot calculations of concentrations and depositions of these metals demonstrated reasonable agreement with available measurements.

Model assessment of pollution levels

1. According to the modelling results the spatial distribution of environmental pollution levels of heavy metals in 2004 was highly non-uniform. The deposition flux in different parts of Europe can differ by more than an order of magnitude. High deposition levels are characteristic of Central and Southern Europe, the lowest levels – of Northern Europe.

2. The influence of the transboundary transport on heavy metal depositions in Europe is significant. More than a half of anthropogenic depositions of heavy metals is defined by the transboundary transport in 33, 30 and 23 European countries for lead, cadmium and mercury, respectively. Fraction of national emissions of lead and cadmium contributing to the transboundary transport in Europe ranges from 60% to 80% for different countries. In case of mercury, this fraction is commonly higher than 80%.

3. Modelled concentrations of lead and cadmium in air and in precipitation are well correlated with the observed data. The modelled levels of lead underestimate measurements by around 30%, of cadmium – by a factor of 2, which can be caused by underestimated anthropogenic or natural and historical emissions. Mercury concentrations in air were reproduced by the model with high accuracy. The difference between modelled and measured values does not exceed 10%. Modelled concentrations of mercury in precipitation satisfactories agree (within 25%) with measurements at most of the stations.

4. The modelling results for lead and cadmium were also extensively evaluated using different emission scenarios. Results of the analysis have shown that the addition of natural and

historical emissions of lead resulted to significant improvement of modelling results against measurements. In general, modelling results based on the unofficial expert emission estimates developed in the EU ESPREME project significantly better agree with measurements than those based on the official emissions data.

5. Mercury hemispheric transport and depositions were evaluated at the hemispheric scale. Comparison of pollution levels estimated for 1996 and 2000 showed that the most significant decrease of mercury depositions took place in Europe and North America because of reduction of anthropogenic emissions. Along with the estimates of the intercontinental transport, the hemispheric model was applied for implementation of the one-way nesting to the regional modelling of mercury pollution. For this purpose monthly mean concentrations of three mercury forms (elemental, reactive gaseous and particulate) were calculated with the hemispheric model at the EMEP domain boundaries and were assimilated the regional model.

Co-operation

1. In the framework of co-operation with the Working Group on Effects MSC-E performed calculations of atmospheric depositions second priorities of heavy metals to ecosystems for the development of the effects-based approach. The depositions were calculated for three emission scenarios: emissions for 2000 based on officially reported data with TNO expert estimates, and two TNO emission projections for 2020.

2. MSC-E contributed relevant information on modelled and measured pollution levels of lead, cadmium and mercury, as well as overview of heavy metal emissions within the UN ECE region to the Task Force on Heavy Metals. These contributions were included in TFHM background document prepared as a scientific basis for the review of the Protocol on Heavy Metals.

3. MSC-E supported the organization of the second meeting of the EMEP TF HTAP (Moscow, Russia, June 2006) and presented information on its activities in regional and hemispheric mercury modelling.

4. The EMEP Centres were also involved in cooperation with other subsidiary bodies to the Convention as well as international organizations and national programmes (HELCOM, EC, OSPAR, UNEP).

REFERENCES

Aas W. and K.Breivik [2004] Heavy metals and POP measurements, 2002. EMEP/CCC-Report 7/2004

Alcamo J., Bartnicki J., Olendrzynski K., and Pacyna J., [1992]. Computing heavy metals in Europe's atmosphere – I. Model development and testing. *Atmospheric Environment*, 26A, No 18, 3355-3369.

Bartnicki J. [1998] Heavy Metal Eulerian Transport Model – HMET. Model description and results. DNMI Research Report No 65, 286 p.

Bartnicki J., A. Gusev, T. Berg, H.Fagerli [2005] Atmospheric Supply of Nitrogen, Lead, Cadmium, Mercury and Lindane to the Baltic Sea in 2003. MSC-W Technical Report 3/2005.

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

Berdowski J.J.M., Pulles M.P.J., Visschedijk A.J.H. [1998]. Incremental cost and remaining emission in 2010 of Heavy Metals (HM) resulting from the implementation of the draft HM Protocol under the UN/ECE Convention on Long Range Transboundary Air Pollution, TNO-MEP – R 98/020, Apeldoorn, The Netherlands, 69 p.

Denier van der Gon D. H.A.C., van het Bolscher M., Visschedijk A.J.H. and Zandveld P.Y.J. [2005]. Study to the effectiveness of the UNECE Heavy Metals Protocol and costs of possible additional measures. Phase I: Estimation of emission reduction resulting from the implementation of the HM Protocol. TNO-report B&O-A R 2005/193.

Gomes L., Rajot J.L., Alfaro S.C. and A. Gaudichet [2003] Validation of a dust production model from measurements performed in semi-arid agricultural areas of Spain and Niger. Catena 52, 257 – 271

Gong S.L. [2003] A parameterization of sea-salt aerosol source function for sub- and super-micron particles. Global Biogeochemical Cycles 17(4), 1097-1103

Gong S.L., Zhang X.Y., Zhao T.L., McKendry I.G., Jaffe D.A., Lu N.M. [2003] Characterization of soil dust aerosol inChina and its transport and distribution during 2001 ACE-Asia: 2. Model simulation and validation. Journal of Geophysical Research, 108(D9), 4262

Grell G. A., J. Dudhia and D. R. Stauffer [1995] A description of the fifth-generation Penn State/NCAR Mesoscale Model (MM5). NCAR/TN-398+STR. NCAR Technical Note. Mesoscale and Microscale Meteorology Division. National Center for Atmospheric Research. Boulder, Colorado. pp. 122.

Gusev A., Ilyin I., Mantseva M., Rozovskaya O., Shatalov V., and Travnikov O. [2006]. Progress in further development of MSCE-HM and MSCE-POP models (implementation of the model review recommendations). EMEP/MSC-E Technical Report 4/2006

Illerup J.B., Nielsen M., Winther M., Mikkelsen M.H., Hoffman L., Gyldenkærne S. and P.Fauser [2005] Annual Danish Emissions Inventory Report to UNECE. Inventories from the base year of the protocols to year 2003. Ministry of Environment. National Environmental Research Institute. Available at http://cdr.eionet.eu.int/dk/Air_Emission_Inventories/Submission_EMEP_UNECE.

Ilyin I. and O. Travnikov [2005] Modelling of heavy metal airborne pollution in Europe: Evaluation of the model performance. EMEP/MSC-E Technical report 8/2005. June 2005

Milford J.B. and Davidson C.I. [1985] The Sizes of Particulate Trace Elements in the Atmosphere - A Review. Journal of Air Pollution Control Association, Vol. 35, No. 12, pp. 1249-1260.

Nijenhuis W.A.S., Van Pul W.A.J., and De Leeuw F.A.A.M., 2001. Deposition of heavy metals metals to the Convention waters of the Oslo and Paris Commissions. *Water, Air and Soil Pollution*, vol. 126, pp.121-14.

Pacyna E. G., Pacyna J. M., Steenhuisen F. and Wilson S. [2006] Global anthropogenic mercury emission inventory for 2000. *Atmos. Environ.* vol. 40, No. 22, pp.4048-4063.

Sofiev M., Petersen G., Kruger O., Schneider B., Hongisto M., and Jylha K., 2001. Model simulations of the atmospheric trace metals concentrations and deposition over the Baltic Sea. *Atmospheric Environment*, vol. 35, No. 8, pp.1395-1409.

Spiegel H., K.E. Böhm, K. Roth and M. Sager [2003]: Atmospheric Deposition of Trace Metals onto Arable Land in Austria. Proc. 7th Intern. Conf. On the Biogeochem. Of Trace Elements; Uppsala`03, p.90-91.

TFMM Workshop Minutes [2005] Minutes of Workshop on the review of the EMEP Models on Heavy Metals and Persistent Organic Pollutants, Moscow, 13 – 14 October 2005, www.msceast.org/events/review.html.

Travnikov O. and I.Ilyin [2005] Regional Model MSCE-HM of Heavy Metal Transboundary Air Pollution in Europe. EMEP/MSC-E Technical Report 6/2005, 59 pp.

UBA [2004] Manual on methodologies and criteria for modelling and mapping of critical loads and levels and air pollution effects, risks and trends. Umweltbundesamt, Berlin; www.icpmapping.org

Wilson S. J., Steenhuisen F., Pacyna J. M. and Pacyna E. G. [2006] Mapping the spatial distribution of global anthropogenic mercury atmospheric emission inventories. *Atmos. Environ.* vol. 40, No. 24, pp.4621-4632.

Zender C. S., Bian H. and D. Newman [2003] Mineral dust entrainment and deposition (DEAD) model: description and 1990s dust climatology. *Journal of Geophysical Research*, vol. 108, No. D14, pp. 4416.

Annex A

EMEP WORKPLAN FOR 2006

2.2. Atmospheric measurements and modelling

Description/objectives:

To support the implementation of the HM Protocol to the Convention; provide the measurement and modelling tools necessary for further abatement policies; compile and evaluate information on transboundary air pollution and implement the EMEP monitoring strategy adopted in 2004. The Task Force on Measurements and Modelling, led by the United Kingdom and co-chaired by WMO, reviews and assesses the scientific and operational activities of EMEP related to monitoring and modelling, evaluates their contribution to the effective implementation and further development of the Protocol and reviews national activities on measurements, modelling and data validation.

Main activities and time schedule for monitoring:

(b) Review, store and make available the 2005 monitoring data (CCC, MSC-W, MSC-E);

Main activities and time schedule for atmospheric modelling for HMs:

(a) Prepare information on lead, cadmium and mercury for 2004: deposition and air concentrations fields in the EMEP area (50 km x 50 km); country-to-country deposition matrices; deposition to the regional seas; comparing model results for air concentrations and precipitation as well as deposition fluxes with measurements; and present calculations for Hg dispersion at the hemispheric scale for the evaluation of European pollution from global sources and boundary conditions for regional EMEP modelling (MSC-E);

(b) Further develop the MSC-E models and the input databases of geophysical and meteorological data for regional and hemispherical modelling (MSC-E);

(c) Prepare maps of depositions (exceedances) for different emission scenarios for the development of effects-based approaches (MSC-E, CCE);

(d) Prepare an overview report on best available emission data, including 1990 and recent years, on atmospheric transport, ambient concentrations and deposition of heavy metals for the sufficiency and effectiveness review of the Protocol on Heavy Metals. Carry out pilot model calculations for atmospheric transport of arsenic, nickel, and chromium (MSC-E,CCC, MSC-W, TFEIP);

(e) Prepare a peer reviewed publication devoted to the third stage of the intercomparison study of Hg models (MSC-E, Bulgaria, Canada, Denmark, Germany, Sweden, and United States).

2.4. Hemispheric transport of air pollution

Description/objectives:

To develop a fuller scientific understanding of the hemispheric transport of air pollution and estimate such transport for specific air pollutants. The Task Force on the Hemispheric Transport of Air Pollution, led by the United States and the European Community, will coordinate activities, including collaboration with other international bodies and networks with related interests within and outside the UNECE region.

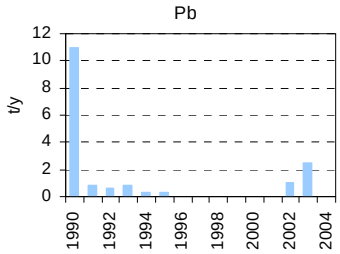
Main activities and time schedule:

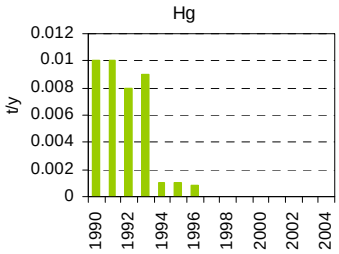
(c) Link measurements at regional and hemispheric scales; investigate further the intercontinental transport of air pollution and assess its impact on European surface pollution levels, using the EMEP monitoring data (CCC, MSC-E, MSC-W).

Annex B

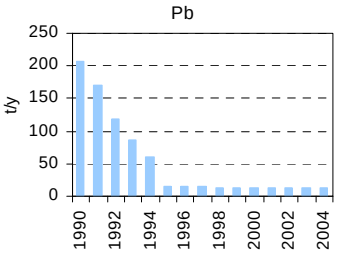
TEMPORAL VARIATIONS OF LEAD, CADMIUM AND MERCURY EMISSIONS FOR THE PERIOD 1990-2004 IN INDIVIDUAL COUNTRIES IN ACCORDANCE WITH REPORTED DATA

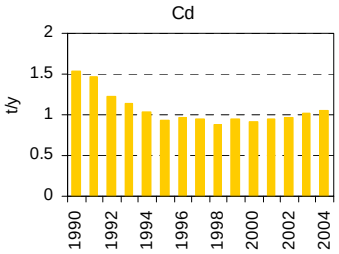
Armenia

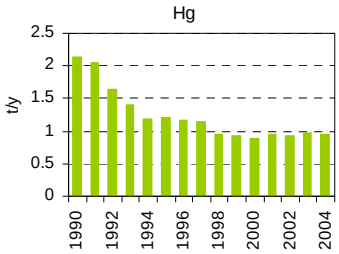




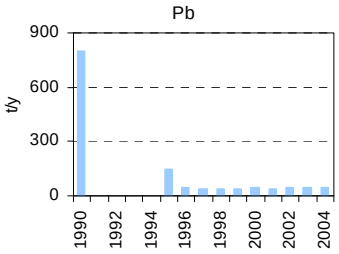
Austria

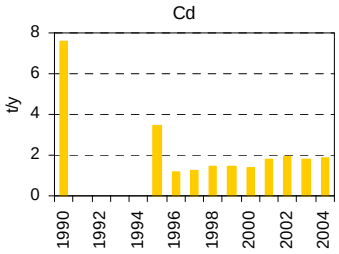


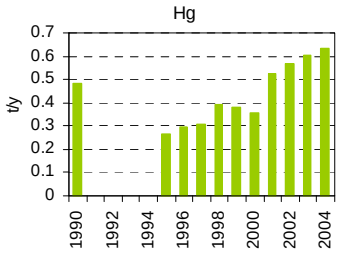




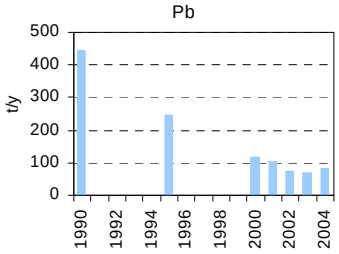
Belarus

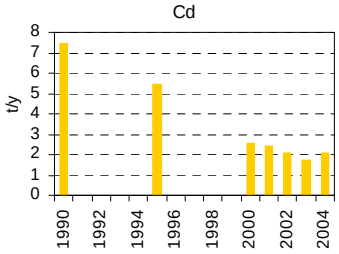


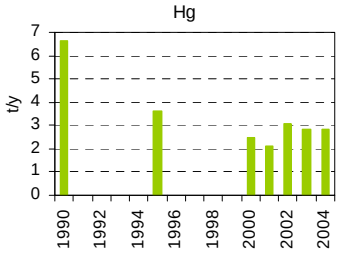




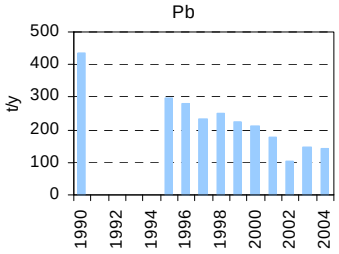
Belgium

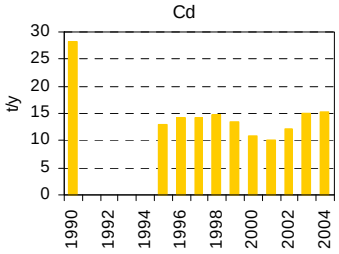


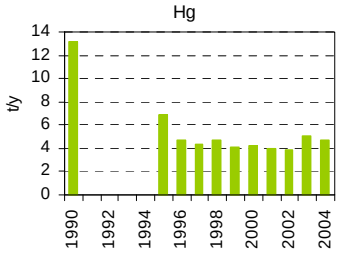




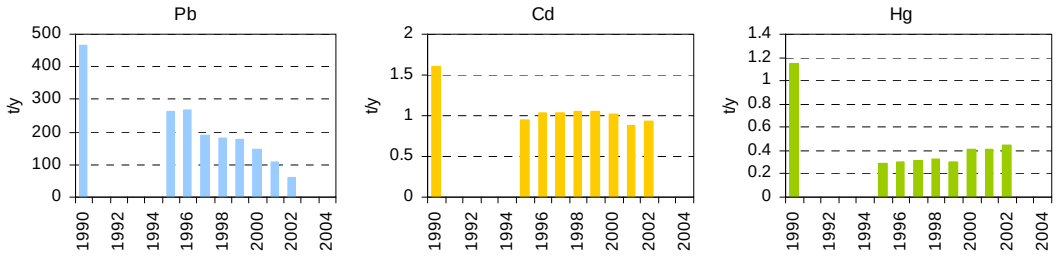
Bulgaria



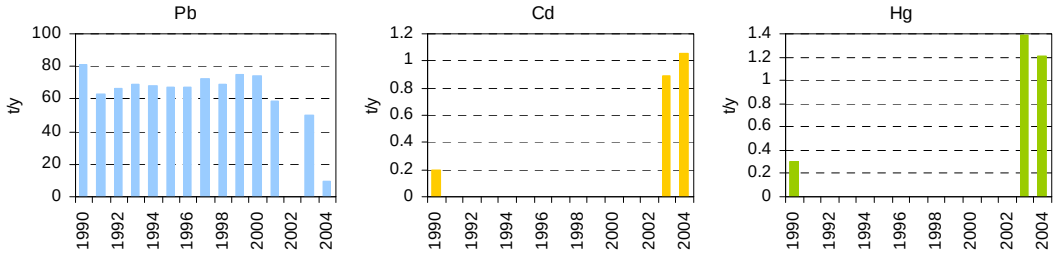




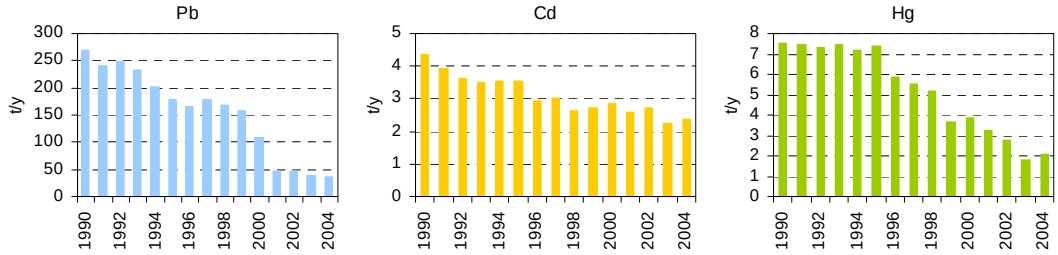
Croatia



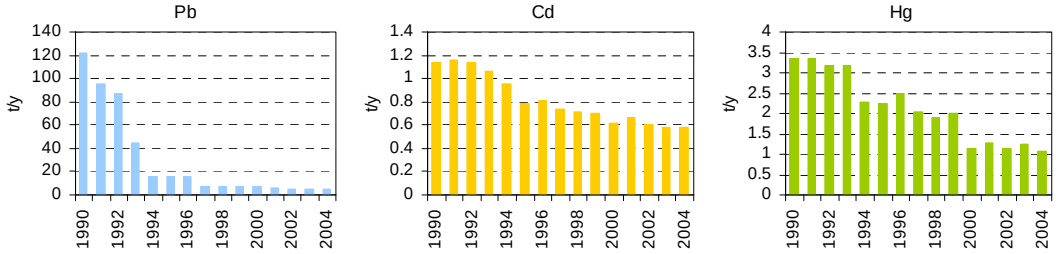
Cyprus



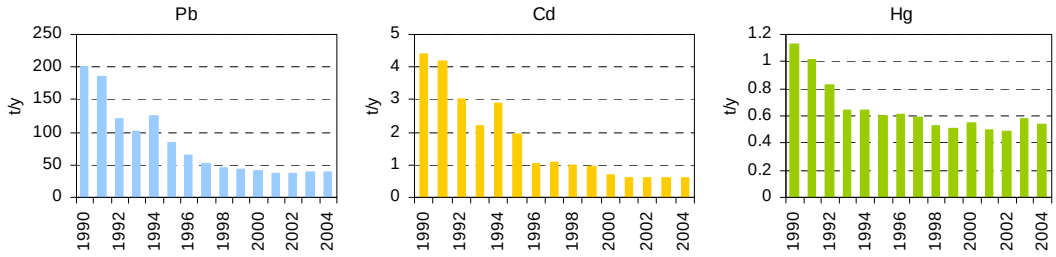
Czech Republic



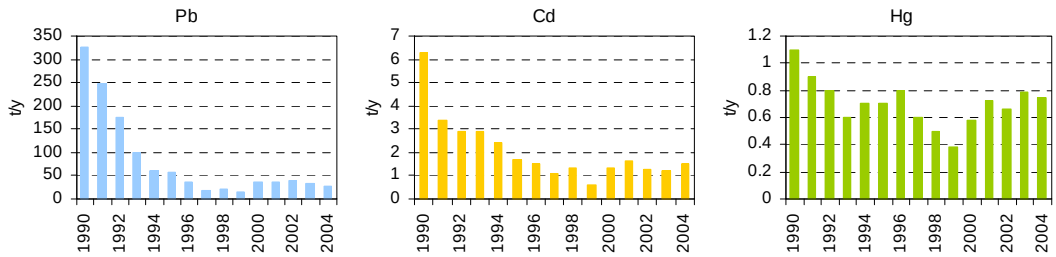
Denmark



Estonia



Finland



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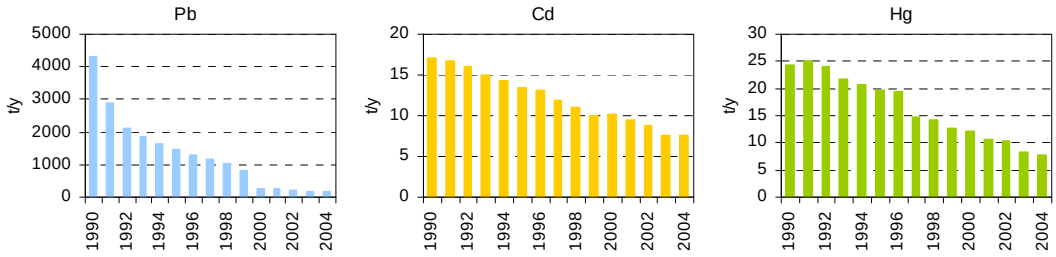

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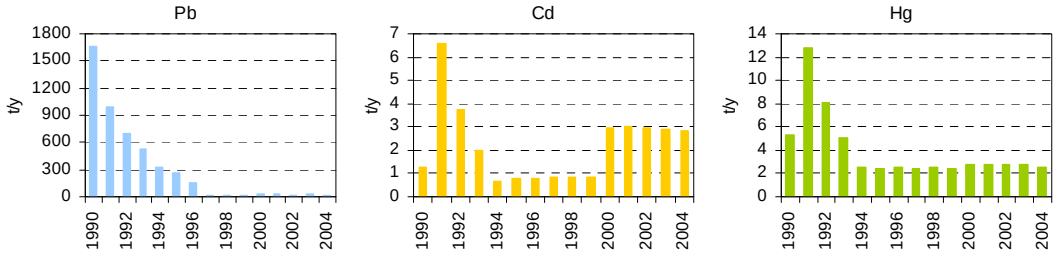
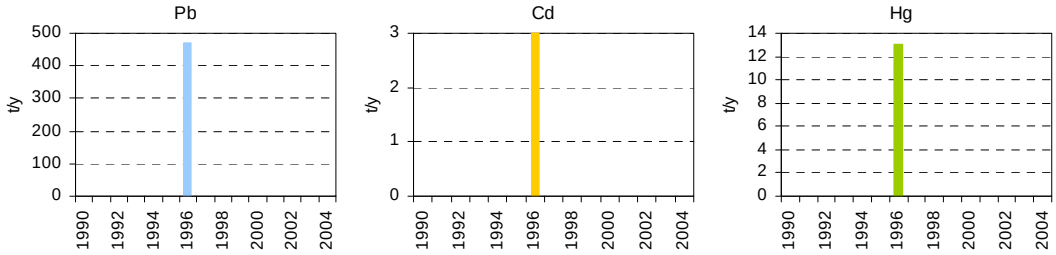
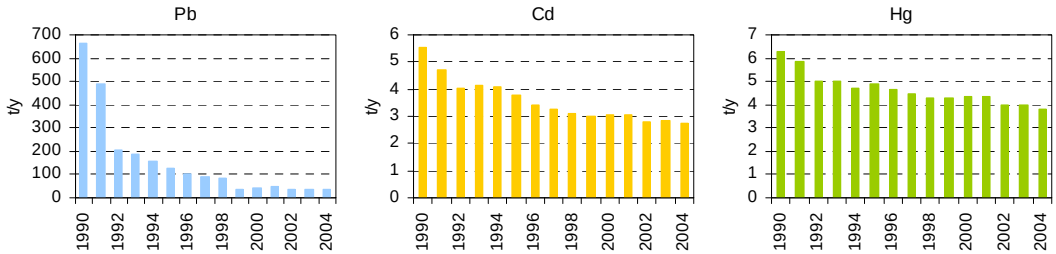


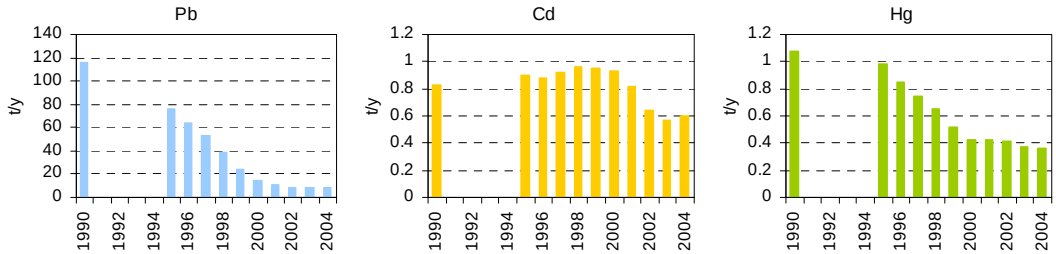
Greece



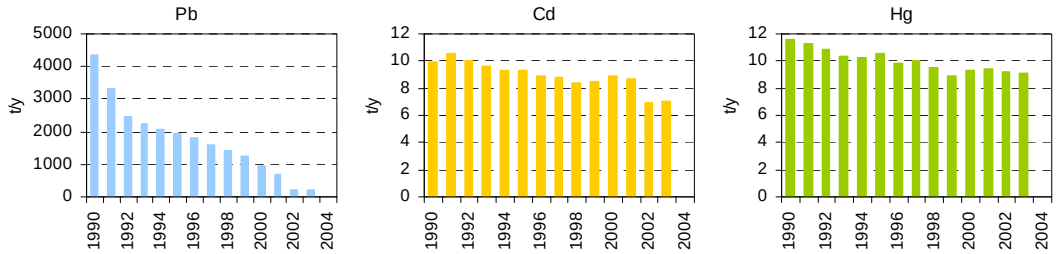
Hungary



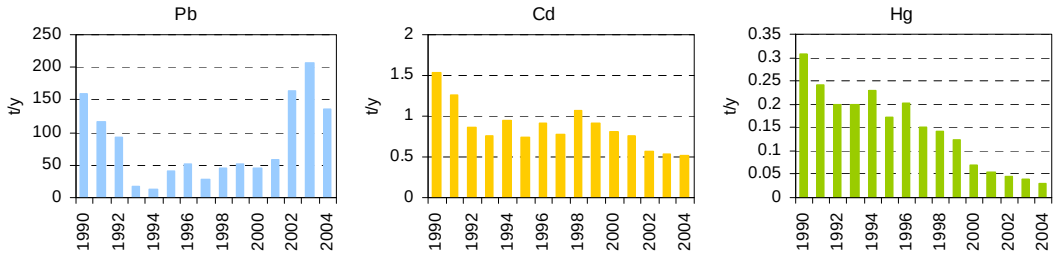
Ireland



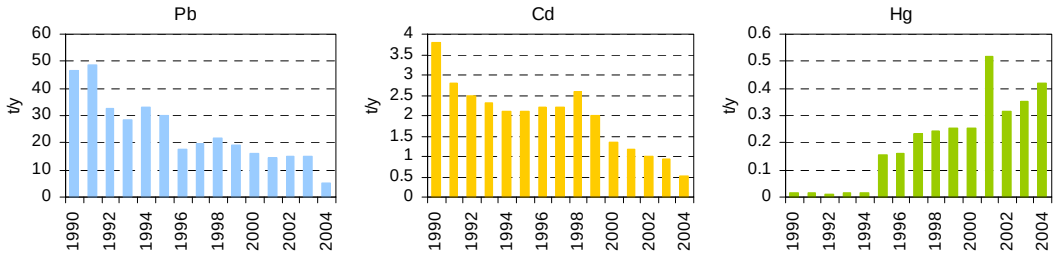
Italy



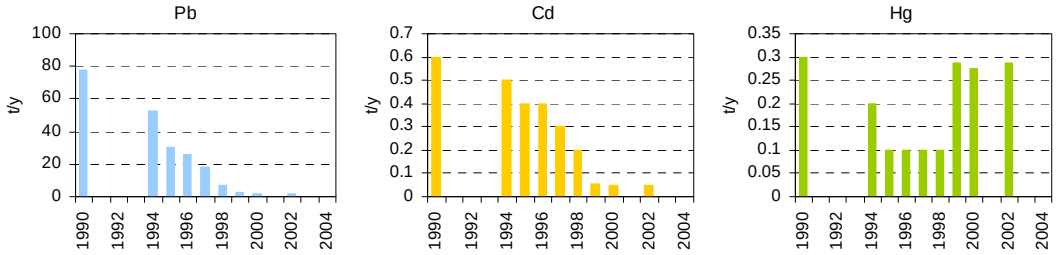
Latvia



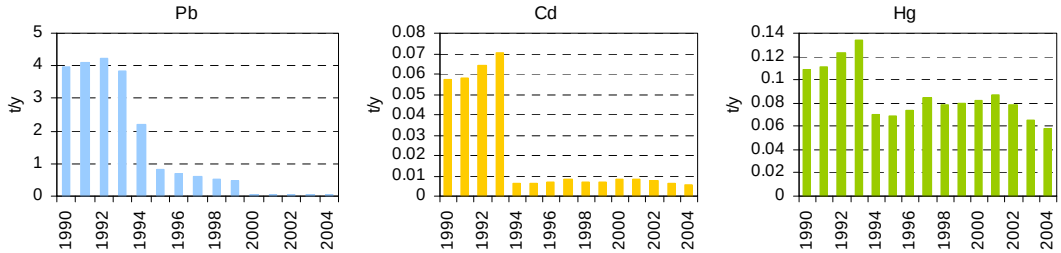
Lithuania



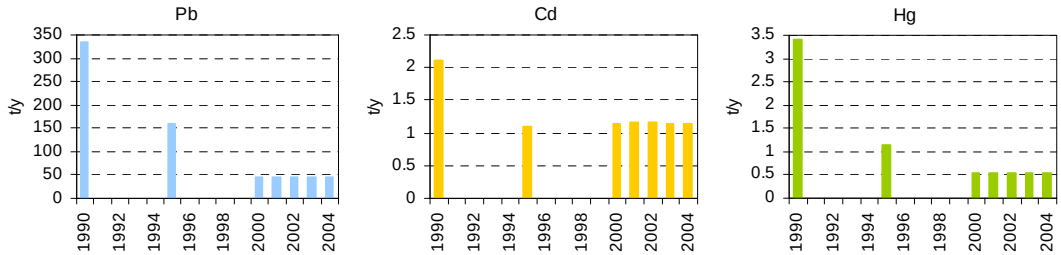
Luxembourg



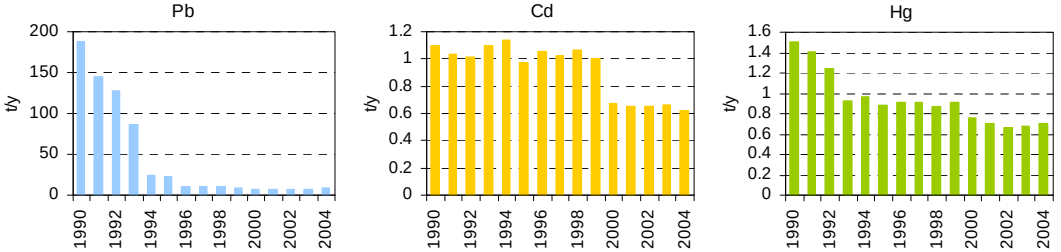
Monaco



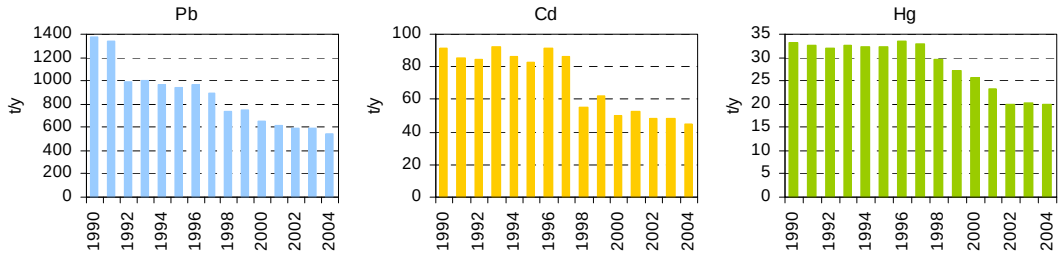
Netherlands



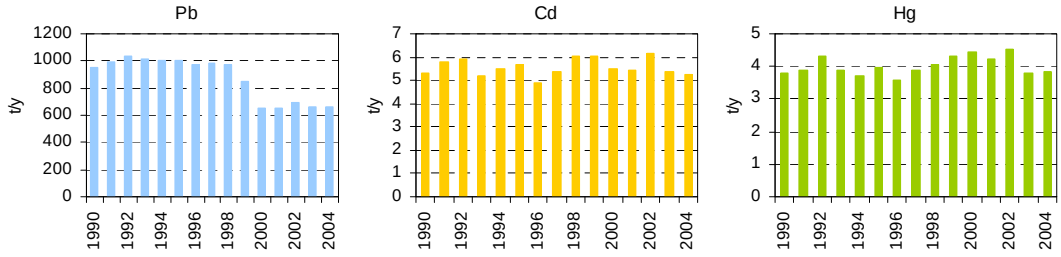
Norway



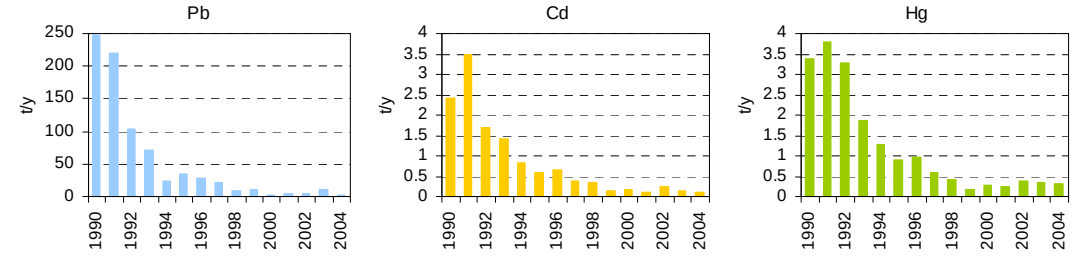
Poland



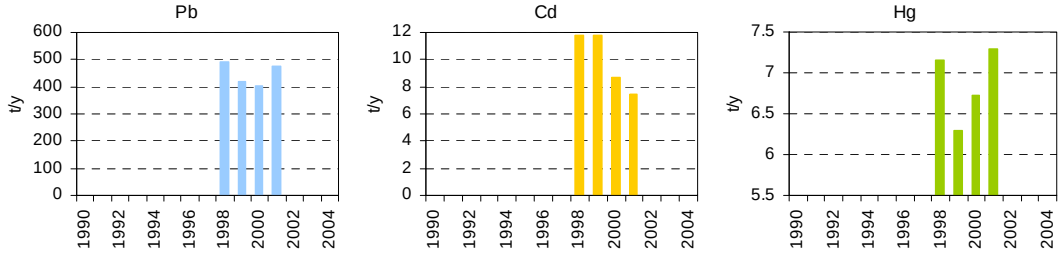
Portugal



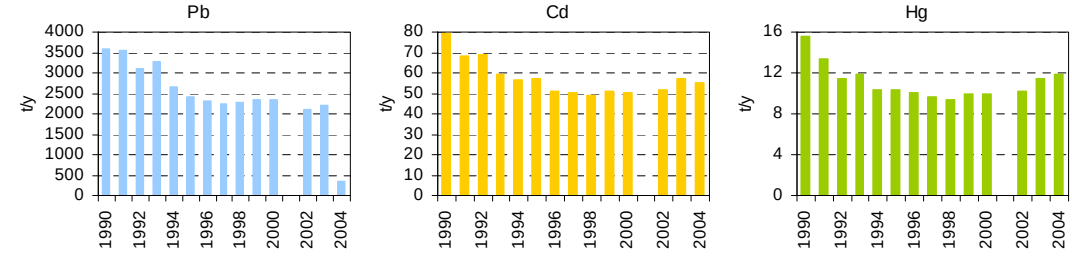
Republic of Moldova



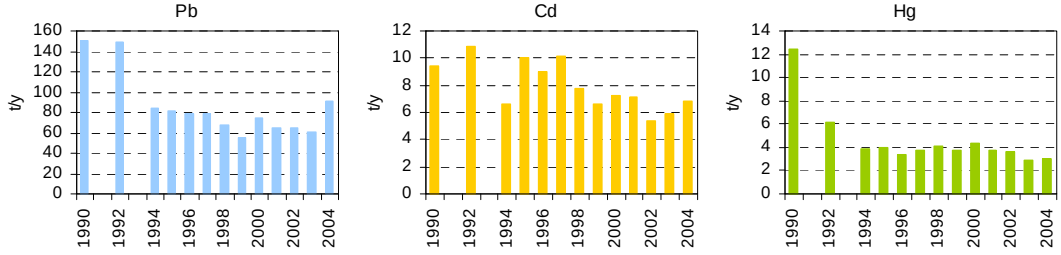
Romania



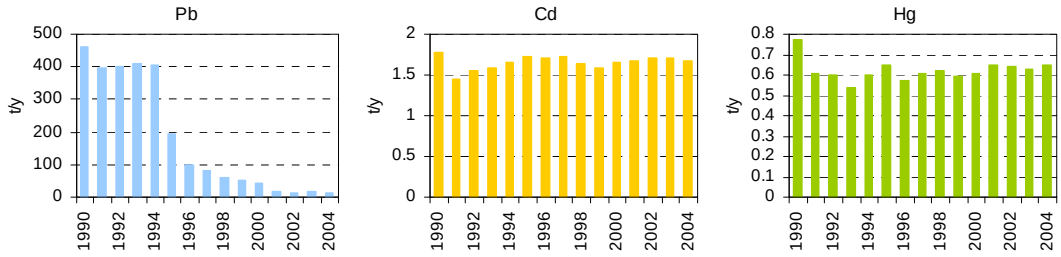
Russian Federation



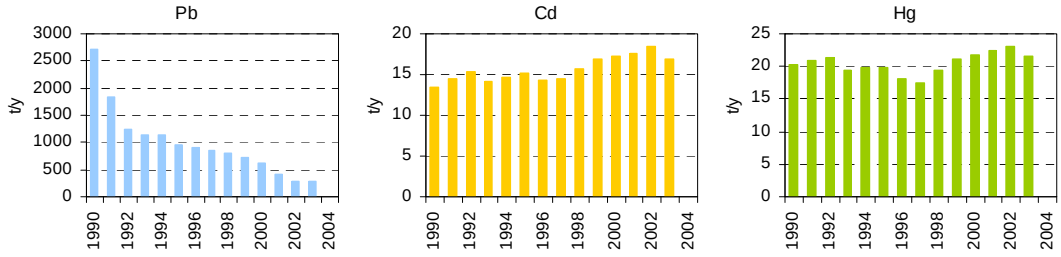
Slovakia



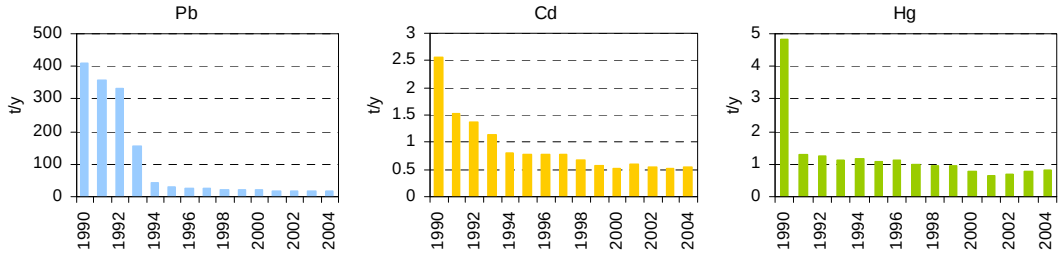
Slovenia



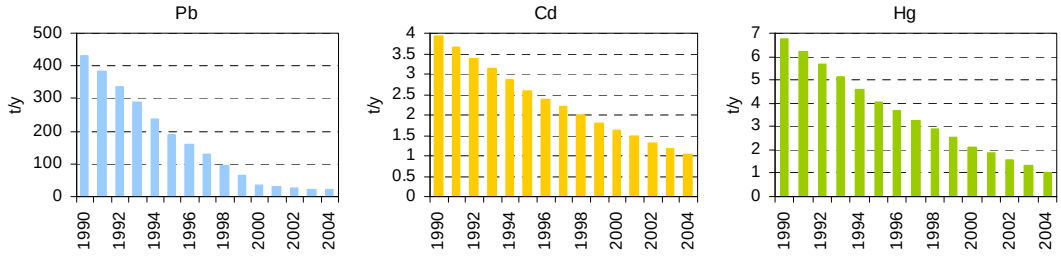
Spain



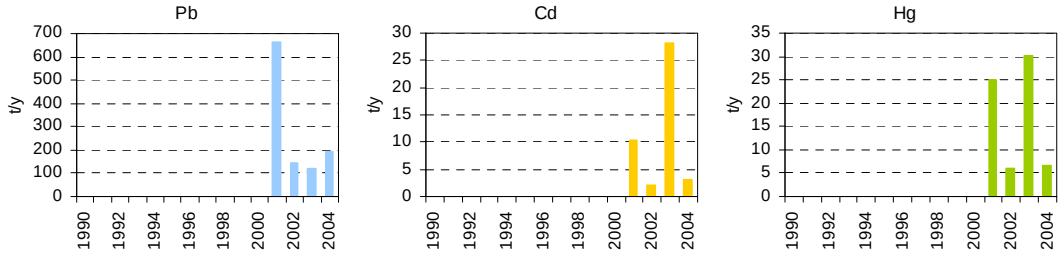
Sweden



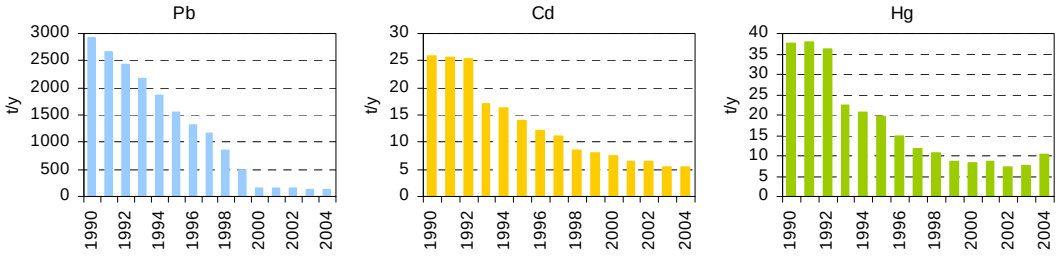
Switzerland



Ukraine



United Kingdom



Annex C

PERCENTAGE CHANGES IN THE TOTAL NATIONAL EMISSIONS OF Pb, Cd AND Hg AFTER THE RECALCULATIONS

[100*($E_{current}$-$E_{previous}$)/$E_{previous}$, %]

The highest changes are flagged.

		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003
Austria	Pb	-0.02	-0.4	-0.1	-0.01	0.1	-1.0	-1.3	-1.1	-1.3	-1.2	-0.3	-0.7	-2.4	-1.2
	Cd	1.5	1.5	-1.4	-1.6	-1.2	-1.5	-1.8	-1.7	-1.9	2.1	2.1	2.2	-2.8	-0.8
	Hg	-0.8	-0.2	-0.6	-1.0	-0.2	-0.6	-1.1	-0.7	-0.7	-0.7	0.8	-0.5	-2.6	-2.0
Belgium	Pb	-22					-32					4.3	3.3	2.1	4.6
	Cd	-4.9					6.3					6.6	2.2	-35	-45
	Hg	-0.2					-0.3					0.6	2.5	11	16
Denmark	Pb	-0.3	-0.2	-0.2	-2.1	-1.1	-0.2	0.2	-0.01	-0.001	-0.01	-0.002	0.02	-0.2	6.3
	Cd	-0.0002	0.3	0.4	0.3	0.3	0.5	0.5	-0.01	-0.02	-0.01	0.005	0.6	-2.0	0.6
	Hg	-0.01	-0.01	0.005	-0.004	0.03	-0.01	0.01	-0.02	0.01	-0.01	-0.002	0.3	-1.2	2.2
Estonia	Pb	-14	-11	-0.01	0.4	16	-3.7	-19	-29	-16	-2.8				
	Cd	173	181	169	150	207	117	11	8.9	21	22				
	Hg	-13	-15	-15	-15	-20	-20	-22	-24	-20	-17				
France	Pb	-0.02	-0.03	-0.05	-0.05	-0.1	-0.1	-0.1	-0.1	-0.2	-0.2	-0.3	-0.6	-0.6	-1.1
	Cd	-0.03	-0.1	-0.1	-0.1	-0.04	-0.1	-0.1	-0.3	-0.6	-0.7	-1.0	-1.4	-2.1	-3.1
	Hg	-0.03	-0.4	-0.1	-0.2	-0.3	-0.2	-0.2	0.3	0.9	2.2	3.3	4.9	4.5	5.3
Germany	Pb	2.0	9.0	8.2	6.9	6.5	10	18	1328						
	Cd														
	Hg														
Hungary	Pb	-2.5					2.5					13			
	Cd	-0.2					-0.6					11			
	Hg	-0.3					0.7					3.6			
Ireland	Pb													-33	-40
	Cd													144	117
	Hg													-73	-76
Latvia	Pb	1451	1518	1754	212	119	714	920	312	548	643	242	768	2184	2826
	Cd	-16	-4	-11	-23	-21	-18	11	-6	35	22	34	37	6	-2
	Hg	-56	-57	-47	-34	-42	-42	-32	-37	-45	-43	-55	-60	-67	-75
Norway	Pb	0.1	0.1	0.1	0.1	0.3	0.3	0.6	0.6	0.5	0.5	0.5	-0.1	-0.1	0.9
	Cd	-33	-34	-35	-33	-3.2	-3.5	1.1	-3.7	-5.5	3.6	-5.2	-5.9	-0.2	-1.2
	Hg	1.0	1.1	1.2	1.7	1.6	1.8	1.7	1.7	1.5	1.3	1.4	1.5	1.7	1.5
Portugal	Pb	-27	-27	-25	-25	-25	-26	-26	-26	-26	-30	-36	-37	-21	
	Cd	113	121	86	107	150	131	159	150	129	126	131	125	134	
	Hg	-1.2	-1.2	-1.0	-1.1	-1.2	-1.2	-1.3	-1.8	-1.4	-2.1	-2.8	-2.2	0.5	
Republic of Moldova	Pb	-1.8													
	Cd	-21													
	Hg	-21													
Slovenia	Pb	0.4	3.2	3.2	2.8	0.04	0.3	-0.6	-0.5	-1.2	-1.0	5.0	9.2	16	0.6
	Cd	6.0					1.2	-3.8	-1.7	-1.8	-1.9	7.1	7.0	7.6	6.2
	Hg	1.3				-1.6		-3.4		-1.6	-1.7	5.2	4.8	3.2	
Sweden	Pb	-14	-10	-6.6	-44	-3.2	23	24	24	25	28	33	34	40	31
	Cd	1.4	1.0	1.0	1.5	3.5	3.5	1.4	2.5	-0.1	2.0	-0.5	1.4	2.5	-8.4
	Hg	3.2	0.8	0.5	0.5	0.6	0.5	0.3	0.7	0.5	0.7	1.3	1.7	1.6	1.1
Switzer-land	Pb	-17	-17	-16	-16	-17	-16	-20	-27	-35	-51	-68	-71	-73	-75
	Cd	-6.5	-6.2	-5.8	0.8	5.8	3.5	4.0	-0.2	-8.3	-17	6.8	-30	-35	-18
	Hg	-0.4	2.1	5.3	9.4	15	23	18	13	10	-4.3	-15	-27	-37	-47
United Kingdom	Pb	-0.02	-0.01	-0.04	0.001	-0.01	-0.05	-0.04	0.4	-0.6	-0.03	-0.6	0.3	1.0	-2.1
	Cd	0.5	0.5	0.5	0.8	0.9	1.0	1.4	2.0	3.1	3.9	2.5	6.5	2.7	-1.4
	Hg	-0.2	-0.2	-0.8	-1.6	-2.4	-3.6	-4.2	-3.3	-5.0	-2.2	-1.9	2.4	2.6	4.4

Annex D

COUNTRY-TO-COUNTRY DEPOSITION MATRICES FOR 2004

Table C.1. *Codes of countries, regions and seas*

Country/Region/Sea	Code	Country/Region/Sea	Code
Albania	AL	Malta	MT
Armenia	AM	Monaco	MC
Austria	AT	Netherlands	NL
Azerbaijan	AZ	Norway	NO
Belarus	BY	Poland	PL
Belgium	BE	Portugal	PT
Bosnia and Herzegovina	BA	Republic of Moldova	MD
Bulgaria	BG	Romania	RO
Croatia	HR	Russian Federation (European part)	RU
Cyprus	CY	Serbia and Montenegro	CS
Czech Republic	CZ	Slovakia	SK
Denmark	DK	Slovenia	SI
Estonia	EE	Spain	ES
Finland	FI	Sweden	SE
France	FR	Switzerland	CH
Georgia	GE	The Former Yugoslav Republic of Macedonia	MK
Germany	DE	Turkey	TR
Greece	GR	Ukraine	UA
Hungary	HU	United Kingdom	GB
Iceland	IS		
Ireland	IE		
Italy	IT	Baltic Sea	BAS
Kazakhstan	KZ	Black Sea	BLS
Latvia	LV	Caspian Sea	CAS
Lithuania	LT	North Sea	NOS
Luxembourg	LU	Mediterranean Sea	MDT

***Table C.2**. Matrix of lead country-to-country depositions from anthropogenic sources in 2004, kg/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	ES	
AL	5203	0.1	13	0.4	326	19	641	6.1	17	3089	1.7	29	7.4	0.8	1.9	309	**AL**
AM	3.7	426	0.6	313	5.5	1.5	16	4.6	1.0	17	18	1.5	0.6	0.2	0.9	12	**AM**
AT	50	0.05	4243	0.3	558	580	266	38	908	1192	0.3	1354	441	22	24	541	**AT**
AZ	7.8	134	1.5	2436	13	3.2	40	16	2.4	38	41	3.7	1.3	0.5	3.8	27	**AZ**
BA	543	0.1	150	0.4	23446	81	748	33	68	7868	1.7	382	53	6.2	12	405	**BA**
BE	1.0	0.003	19	0.03	4.6	13024	2.6	2.8	85	7.1	0.02	33	166	7.5	2.0	513	**BE**
BG	595	1.3	62	3.5	792	72	48576	128	41	9891	22	215	44	7.1	29	299	**BG**
BY	99	3.6	138	13	465	421	922	16327	109	1503	2.5	815	213	75	558	339	**BY**
CH	9.2	0.01	64	0.03	42	246	19	3.4	4974	54	0.3	38	118	3.3	1.9	632	**CH**
CS	1985	0.3	146	1.1	5485	105	5569	64	68	61913	7.0	440	66	7.8	22	430	**CS**
CY	5.0	0.2	0.7	0.2	6.4	0.8	22	0.7	1.0	22	758	1.4	0.5	0.1	0.2	13	**CY**
CZ	30	0.1	730	0.4	305	705	249	58	301	890	0.2	7716	717	35	38	288	**CZ**
DE	24	0.1	966	0.9	136	18469	110	158	3376	321	0.5	1796	8277	258	104	2903	**DE**
DK	2.8	0.03	20	0.2	15	592	21	23	38	51	0.1	88	166	568	24	194	**DK**
EE	3.3	0.1	15	0.4	22	138	41	258	17	63	0.2	83	63	32	4648	74	**EE**
ES	41	0.03	37	0.2	174	701	73	9.2	168	220	0.7	60	69	7.6	7.1	96722	**ES**
FI	8.1	0.4	37	2.1	52	476	101	544	40	152	0.5	204	154	98	4000	164	**FI**
FR	71	0.1	161	0.3	371	7073	134	21	2341	432	2.1	237	922	41	17	21162	**FR**
GB	5.2	0.1	46	0.7	24	2544	22	27	114	52	0.2	116	264	39	24	1619	**GB**
GE	33	166	5.8	570	63	8.2	194	39	8.3	188	34	11	3.8	1.1	5.6	71	**GE**
GR	1328	1.0	35	3.1	528	52	6770	56	40	3570	46	102	24	3.3	13	559	**GR**
HR	366	0.05	253	0.3	4724	78	551	33	81	4493	1.2	429	53	8.2	11	385	**HR**
HU	195	0.1	461	0.5	2546	168	1317	74	118	9062	1.4	871	104	13	21	248	**HU**
IE	1.1	0.004	3.1	0.04	5.1	150	1.9	1.5	15	8.3	0.1	6.6	13	2.7	1.8	310	**IE**
IS	0.5	0.01	5.6	0.1	3.9	132	4.6	4.3	10	7.7	0.01	12	20	4.2	1.9	102	**IS**
IT	992	0.3	487	1.0	3121	306	979	42	1123	3307	24	515	147	13	19	3396	**IT**
KZ	93	86	37	712	245	65	931	326	45	761	34	84	27	6.4	124	255	**KZ**
LT	19	0.3	60	1.5	96	266	137	1315	55	273	0.5	325	138	66	257	218	**LT**
LU	0.1	0.0	2.5	0.002	0.6	292	0.3	0.3	12	0.9	0.002	3.1	28	0.5	0.2	46	**LU**
LV	10	0.2	35	1.0	55	245	92	687	36	163	0.3	192	117	65	799	170	**LV**
MC	0.01	0.0	0.02	0.0	0.04	0.02	0.01	0.001	0.1	0.05	0.0	0.02	0.008	0.001	0.00	0.202	**MC**
MD	36	1.0	10	1.9	104	22	681	128	7.6	455	4.5	40	11	3.0	15	38	**MD**
MK	1198	0.2	14	0.5	246	16	2171	10	12	4229	3.4	41	8.4	0.9	3.1	124	**MK**
MT	0.5	0.0	0.1	0.0	0.8	0.1	0.6	0.01	0.2	1.2	0.03	0.1	0.1	0.003	0.004	2.2	**MT**
NL	1.3	0.01	20	0.1	6.1	5158	4.0	6.7	50	11	0.03	53	252	11	4.1	473	**NL**
NO	14	0.2	56	1.8	81	703	95	138	89	237	0.4	231	205	224	175	293	**NO**
PL	119	0.7	580	3.3	936	2350	947	1505	420	2738	2.1	7608	1974	325	405	1070	**PL**
PT	0.8	0.001	1.9	0.01	3.9	46	1.7	0.7	8.8	4.4	0.04	3.3	4.4	0.5	0.7	6101	**PT**
RO	662	3.0	276	7.6	3314	281	12571	455	155	17785	30	759	152	26	91	542	**RO**
RU	850	379	486	2463	2347	1526	8469	12168	506	7859	221	1837	707	302	17949	2533	**RU**
SE	26	0.3	70	2.1	156	1500	276	579	82	499	0.8	403	411	759	1019	453	**SE**
SI	63	0.02	307	0.1	627	49	152	11	67	831	0.3	172	30	3.3	4.3	146	**SI**
SK	80	0.2	307	0.7	836	189	634	97	92	2843	0.5	1940	122	15	32	157	**SK**
TR	465	205	60	168	544	97	4219	271	68	2419	1015	163	51	12	48	811	**TR**
UA	469	41	347	178	1943	687	7039	4020	244	7381	54	1515	362	82	382	805	**UA**
BAS	25	0.5	127	2.1	143	2404	239	701	142	473	0.7	728	855	850	4247	794	**BAS**
BLS	416	32	80	90	819	118	7162	627	70	3651	119	238	59	18	91	417	**BLS**
CAS	27	124	11	1764	71	17	228	87	15	210	54	25	8.0	1.9	27	87	**CAS**
MDT	5096	6.8	513	11	6735	786	8463	169	1044	11301	2012	785	321	27	56	19644	**MDT**
NOS	27	0.2	202	1.9	132	8923	161	171	454	388	0.6	719	1612	651	167	3627	**NOS**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CS**	**CY**	**CZ**	**DE**	**DK**	**EE**	**ES**	

***Table C.2**. Matrix of lead country-to-country depositions from anthropogenic sources in 2004, kg/y (continued)*

Receptors ↓ Emitters →

	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	MC	
AL	0.9	156	23	0.8	5563	178	61	1.3	0.01	1705	17	0.6	0.7	10.1	0.1	**AL**
AM	0.3	5.2	1.8	139	91	4.0	2.1	0.1	0.001	21	389	0.3	0.04	5.3	0.003	**AM**
AT	9.8	1318	393	0.2	342	1171	728	15	0.05	4142	9.0	7.7	22	164	0.6	**AT**
AZ	1.5	11	4.5	270	197	8.8	5.0	0.2	0.003	47	2971	0.9	0.1	19	0.01	**AZ**
BA	5.9	346	68	0.5	1520	5387	1040	3.3	0.02	3589	13	5.2	2.4	85	0.4	**BA**
BE	1.2	6266	1809	0.01	8.6	8.3	3.4	42.9	0.05	80	1.01	0.9	121	16	0.04	**BE**
BG	11.6	208	78	6.2	17079	485	533	3.6	0.02	1280	150	8.8	2.1	152	0.1	**BG**
BY	167	556	461	12.7	1171	450	483	20	0.12	762	419	562	8.9	7167	0.1	**BY**
CH	1.0	2087	330	0.02	132	93	18	13	0.03	3796	1.0	0.6	10.3	14	0.8	**CH**
CS	11.1	348	85	1.8	6713	2219	1681	3.9	0.03	3110	43	8.1	3.3	128	0.3	**CS**
CY	0.1	5.1	1.3	0.2	218	4.6	2.2	0.1	0.00	29	2.0	0.1	0.03	1.08	0.004	**CY**
CZ	17.7	946	501	0.4	278	478	704	19	0.06	672	15	11.6	22	240	0.1	**CZ**
DE	50.8	15682	7250	0.6	241	205	204	226	0.54	1747	32	32.7	533	714	0.6	**DE**
DK	13.3	658	1060	0.2	24	19	28	35	0.16	71	5.7	6.2	6.9	137	0.02	**DK**
EE	404	152	157	0.6	62	28	33	7.1	0.04	60	8.8	100	2.7	8117	0.01	**EE**
ES	3.2	4470	1131	0.1	267	236	58	64	0.22	1947	7.4	1.9	16	41	0.6	**ES**
FI	11976	476	499	1.7	109	69	75	24	0.30	148	54	136	8	5280	0.0	**FI**
FR	8.9	59974	7990	0.2	638	604	134	345	0.60	7206	11	5.5	338	119	10.2	**FR**
GB	12.5	4210	31479	0.2	36	31	24	1131	1.24	187	22	5.9	35	154	0.1	**GB**
GE	2.3	36	12	1986	720	41	15	0.6	0.00	181	685	2.3	0.2	38	0.04	**GE**
GR	5.6	261	55	6.1	97713	325	216	2.8	0.02	1828	125	4.1	1.6	65	0.2	**GR**
HR	4.8	356	60	0.3	1305	12673	1327	2.7	0.02	4129	7.2	4.4	2.4	73	0.5	**HR**
HU	10.2	376	129	0.9	1130	3801	8961	5.2	0.02	2266	16	8.9	5.3	134	0.3	**HU**
IE	1.1	400	830	0.02	4.6	5.4	2.3	1230	0.37	33	1.3	0.4	2.5	10	0.01	**IE**
IS	2.0	176	425	0.1	5.7	4.8	3.8	31	39	27	2.8	0.7	2.4	14	0.01	**IS**
IT	8.7	3450	336	1.9	7226	4706	667	17.2	0.08	77054	38	7.0	10.5	134	8.1	**IT**
KZ	52.9	162	75	261	1971	172	94	3.8	0.03	570	45385	18.0	1.6	507	0.1	**KZ**
LT	83.9	334	315	1.7	259	134	130	14	0.06	257	47	1345	5.3	6631	0.05	**LT**
LU	0.1	612	85	0.001	0.8	1.1	0.4	2.5	0.004	10	0.1	0.1	121	1.4	0.005	**LU**
LV	184	285	286	1.4	160	70	75	12.8	0.07	156	26.8	524	4.8	36532	0.03	**LV**
MC	0.0	0.8	0.02	0.0	0.1	0.1	0.01	0.001	0.00	2.4	0.0	0.0	0.001	0.003	0.03	**MC**
MD	5.3	38.8	25.4	3.9	590	66	55	1.1	0.01	156	65.2	5.3	0.5	92	0.02	**MD**
MK	1.4	72.1	15.9	1.0	10880	136	100	0.8	0.005	572	19.4	0.9	0.5	15	0.1	**MK**
MT	0.003	1.1	0.2	0.001	5.6	0.6	0.1	0.01	0.0	6.6	0.02	0.002	0.003	0.03	0.001	**MT**
NL	2.0	2953	2057	0.04	10.9	8.7	5.9	49.0	0.08	68	2.6	1.8	21.4	35	0.03	**NL**
NO	220	873	2268	1.2	112	85	98	111	1.55	224	43.3	33	11.8	820	0.1	**NO**
PL	169	2486	1963	4.6	1308	1295	1577	74	0.31	1694	107	256	48	3343	0.3	**PL**
PT	0.3	220	101	0.01	8.9	7.4	1.8	7.9	0.03	68	0.4	0.1	1.0	3.1	0.03	**PT**
RO	36.5	625	277	24	7839	2287	2513	12.2	0.06	3354	251	31	8.0	536	0.5	**RO**
RU	5816	2494	2123	1794	15915	1849	1414	102	1.14	5171	75136	961	32	34506	0.9	**RU**
SE	1998	1460	1834	1.8	249	160	181	72	0.61	302	55	162	18	4165	0.1	**SE**
SI	1.7	197	34	0.1	326	2465	353	1.5	0.01	2533	4.1	1.6	1.6	30	0.3	**SI**
SK	14.2	301	167	0.9	603	1051	2600	6.9	0.03	892	25	13.4	5.6	212	0.1	**SK**
TR	21.0	360	117	297	21164	399	267	6.3	0.05	1935	1409	16.0	2.8	267	0.3	**TR**
UA	150	1050	739	191	7545	1716	2159	33.0	0.15	2910	6979	172	16.6	2594	0.4	**UA**
BAS	3235	2189	2346	2.5	259	189	231	89.9	0.38	442	56	328	34.1	10855	0.1	**BAS**
BLS	39.2	290	137	430	12899	511	343	6.3	0.05	1580	3031	32.2	3.0	555	0.2	**BLS**
CAS	9.3	49.1	20.4	274	615	49.9	28.6	1.1	0.01	192	45227	4.9	0.5	128	0.03	**CAS**
MDT	32.3	9550	1150	20	104120	7061	1127	55.7	0.24	52300	291	19.2	26.1	335	7.4	**MDT**
NOS	88.6	12807	29254	1.4	179	138	202	855	4.39	709	50	49.0	104	1078	0.2	**NOS**
	FI	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KZ**	**LT**	**LU**	**LV**	**MC**	

***Table C.2**. Matrix of lead country-to-country depositions from anthropogenic sources in 2004, kg/y (continued)*

Receptors ↓ Emitters →

	MD	MK	NL	NO	PL	PT	RO	RU	SE	SI	SK	TR	UA	Total, t/y	
AL	1.5	2467	8.5	0.5	253	235	717	27	1.2	20	115	322	81	22	**AL**
AM	0.4	10	0.7	0.1	26	11	51	60	0.4	0.8	5.5	1687	46	3.4	**AM**
AT	1.6	119	260	9.2	5596	340	1229	64	25	1184	2037	98	122	30	**AT**
AZ	1.0	21	1.7	0.5	71	23	131	434	1.3	1.9	13.7	2519	172	10	**AZ**
BA	2.2	492	43	3.1	3058	270	2748	42	10.9	236	1665	276	222	55	**BA**
BE	0.03	1.9	1041	4.0	223	551	10	4.4	6.1	4.3	11.5	3.4	3.6	24	**BE**
BG	48	4655	41	4.3	2475	202	23881	384	13	72	1154	6342	1512	122	**BG**
BY	64	310	241	47	24294	295	6500	2813	198	110	2050	1106	7294	80	**BY**
CH	0.1	15	88	1.9	245	380	50	5.1	2.9	46	39	46	7.9	14	**CH**
CS	8.5	5551	54	4.1	3847	295	12186	117	14	165	2455	1170	511	117	**CS**
CY	0.1	13	0.4	0.1	13	7.2	37	3.9	0.1	0.9	4.8	747	9.3	1.9	**CY**
CZ	2.2	85	387	15	15857	253	1236	93	41	174	2470	103	171	37	**CZ**
DE	1.9	51	8610	82	13023	2778	635	206	198	101	647	123	204	90	**DE**
DK	0.4	7.6	536	29	1676	156	168	36	102	6.2	88	14	49	6.7	**DK**
EE	1.7	11	89	36	2174	50	298	503	184	10	133	80	223	18	**EE**
ES	0.4	65	274	7.0	545	59626	215	25	7.6	79	129	96	32	168	**ES**
FI	5.8	29	286	225	4805	133	837	2217	1856	22	317	113	617	36	**FI**
FR	0.8	119	2008	23	1772	10445	444	43	37	220	298	257	57	126	**FR**
GB	0.4	10	1466	34	1111	2276	110	74	41	13	79	23	59	48	**GB**
GE	4.7	100	4.3	0.9	201	51	534	791	2.4	8.9	39	6229	515	14	**GE**
GR	17	5397	25	2.4	1144	423	5342	234	6.6	47	473	8046	736	136	**GR**
HR	1.8	349	40	3.6	3191	238	1851	32	12	1036	1646	218	176	40	**HR**
HU	4.2	573	89	5.8	6442	175	10766	66	18	617	12321	346	508	64	**HU**
IE	0.03	1.5	76	2.6	66	433	12	4.6	2.9	2.0	6.3	1.8	4.6	3.7	**IE**
IS	0.1	1.3	84	10.5	121	197	21	7.8	4.1	2.0	10.3	6.6	9.5	1.5	**IS**
IT	3.6	1078	131	7.7	4143	2337	2281	80	21	1610	1296	1909	248	123	**IT**
KZ	24	305	35	9.3	1410	188	3327	9518	27	45	277	6284	4147	79	**KZ**
LT	6.5	48	169	36	10288	163	949	644	179	43	486	166	688	27	**LT**
LU	0.003	0.2	40	0.3	18	35	1.2	0.4	0.5	0.6	1.3	0.3	0.3	1.3	**LU**
LV	4.1	33	152	50	5368	131	654	588	256	23	284	135	488	49	**LV**
MC	0.0	0.01	0.01	0.0	0.1	0.1	0.03	0.001	0.0	0.04	0.03	0.05	0.003	0.004	**MC**
MD	345	126	14	2.0	943	28	7478	244	5.6	12	157	1001	1761	15	**MD**
MK	2.6	16417	8.2	0.6	374	91	1235	41	1.7	16	191	783	121	39	**MK**
MT	0.002	0.8	0.04	0.002	0.7	0.8	0.9	0.0	0.01	0.1	0.2	1.1	0.1	0.03	**MT**
NL	0.1	2.5	6398	7.2	460	564	18	10	8.7	4.0	23	5.8	6.9	19	**NL**
NO	3.3	34	500	2667	4329	380	699	360	540	25	298	97	307	18	**NO**
PL	29	354	1451	95	222645	1099	7556	916	405	395	7982	767	2775	282	**PL**
PT	0.01	1.3	20	0.6	29	155081	4.4	2.3	0.5	3.3	4.0	5.1	1.8	162	**PT**
RO	245	2524	153	15	9672	385	180942	945	41	354	5676	6941	5077	268	**RO**
RU	271	2877	979	463	40419	1964	35217	228170	1885	466	4965	43665	47209	616	**RU**
SE	11.0	74	1063	1036	11032	405	2003	888	7334	41	663	234	948	43	**SE**
SI	0.6	89	22	1.6	1215	102	573	14	4.8	3413	535	88	51	15	**SI**
SK	4.5	244	111	6.5	15254	133	5488	97	23	266	18523	229	460	54	**SK**
TR	75	1468	50	9.3	2594	657	11678	2263	24	78	658	185601	4218	246	**TR**
UA	586	1696	419	51	37454	668	54642	11686	168	370	8182	14833	76633	259	**UA**
BAS	8.7	68	1634	248	20774	594	1800	1372	2679	64	915	232	878	63	**BAS**
BLS	226	1695	64	15	4633	289	24487	5015	46	102	1040	39886	13464	125	**BLS**
CAS	6.1	84	8.9	2.5	405	74	823	3409	6.4	13	86	4601	1225	60	**CAS**
MDT	41	6434	331	20	6737	9195	13081	527	46	1353	2400	45545	1651	320	**MDT**
NOS	2.5	68	8634	633	10351	4301	1094	313	452	52	680	94	286	90	**NOS**
	MD	**MK**	**NL**	**NO**	**PL**	**PT**	**RO**	**RU**	**SE**	**SI**	**SK**	**TR**	**UA**	**Total, t/y**	

***Table C.3**. Matrix of cadmium country-to-country depositions from anthropogenic sources in 2004, kg/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	ES	
AL	41.2	0.01	1.1	0.1	7.4	0.4	64.5	0.2	0.8	128.5	0.2	1.7	0.9	0.1	0.03	17.6	**AL**
AM	0.02	26.3	0.05	41.2	0.1	0.03	1.7	0.2	0.1	0.6	1.8	0.1	0.1	0.02	0.01	0.8	**AM**
AT	0.3	0.0	342.7	0.1	11.3	13.6	22.5	1.4	46.4	53.0	0.0	72.2	36.2	2.4	0.3	29.8	**AT**
AZ	0.1	8.1	0.1	471.9	0.3	0.1	4.2	0.6	0.1	1.4	4.1	0.2	0.2	0.1	0.05	1.7	**AZ**
BA	3.8	0.004	13.1	0.1	602.0	1.8	71.3	1.2	3.3	397.5	0.1	21.2	7.4	0.7	0.2	23.1	**BA**
BE	0.01	0.0	1.2	0.01	0.1	360.8	0.3	0.1	4.1	0.3	0.0	2.6	16.3	0.9	0.03	25.3	**BE**
BG	4.3	0.1	5.4	0.7	18.8	1.6	5854.4	4.9	2.1	427.6	1.5	12.4	6.4	0.8	0.4	17.3	**BG**
BY	0.6	0.1	10.2	2.5	9.2	9.6	80.8	651.7	5.1	57.9	0.2	45.7	34.3	8.4	8.5	18.0	**BY**
CH	0.1	0.0	6.4	0.01	0.8	6.2	1.8	0.1	272.6	2.3	0.03	2.7	6.6	0.3	0.02	35.8	**CH**
CS	14.6	0.01	13.3	0.2	133.2	2.4	449.3	2.5	3.4	2996.9	0.4	25.6	9.8	0.9	0.3	24.7	**CS**
CY	0.0	0.01	0.1	0.03	0.1	0.02	2.4	0.03	0.05	0.9	91.8	0.1	0.05	0.01	0.003	0.8	**CY**
CZ	0.2	0.004	58.2	0.1	6.4	16.0	22.3	2.4	14.5	40.2	0.02	580.3	120.5	4.2	0.5	14.9	**CZ**
DE	0.2	0.006	70.7	0.2	2.7	447.4	9.7	5.6	177.2	13.0	0.1	194.7	1084.7	31.8	1.4	143.6	**DE**
DK	0.0	0.002	1.4	0.0	0.3	13.6	1.5	0.8	1.8	1.8	0.01	6.2	22.6	65.0	0.3	9.6	**DK**
EE	0.0	0.005	1.1	0.1	0.4	3.1	3.9	7.4	0.8	2.4	0.02	5.4	9.5	3.3	76.4	3.7	**EE**
ES	0.3	0.002	2.9	0.0	3.3	17.2	6.7	0.3	8.1	8.3	0.1	3.4	6.1	0.8	0.1	5724.8	**ES**
FI	0.1	0.02	2.7	0.4	1.0	10.6	8.9	17.7	1.9	5.7	0.05	12.5	21.7	10.0	56.6	8.6	**FI**
FR	0.5	0.003	12.3	0.1	6.8	198.5	12.2	0.7	124.7	16.6	0.2	16.1	66.8	4.4	0.2	1134.1	**FR**
GB	0.0	0.003	3.0	0.1	0.5	63.4	2.0	0.9	5.4	2.0	0.02	8.5	26.8	4.0	0.3	82.2	**GB**
GE	0.2	10.0	0.4	112.0	1.4	0.2	21.5	1.5	0.4	7.3	3.4	0.6	0.4	0.1	0.1	4.2	**GE**
GR	10.1	0.05	2.8	0.5	11.9	1.2	933.0	2.1	2.0	129.2	4.3	5.7	3.0	0.3	0.2	32.8	**GR**
HR	2.5	0.002	23.3	0.1	86.5	1.7	50.7	1.3	4.0	237.9	0.1	23.3	7.3	0.9	0.1	21.7	**HR**
HU	1.3	0.01	46.3	0.1	54.3	3.8	108.0	2.9	5.7	396.4	0.1	49.6	14.4	1.4	0.3	13.8	**HU**
IE	0.01	0.0	0.2	0.01	0.1	3.7	0.2	0.1	0.8	0.3	0.005	0.5	1.5	0.3	0.03	15.5	**IE**
IS	0.003	0.001	0.3	0.03	0.1	3.1	0.4	0.2	0.5	0.3	0.001	0.9	2.3	0.4	0.02	5.7	**IS**
IT	6.9	0.02	44.2	0.2	61.2	6.9	95.4	1.5	55.5	130.3	2.4	27.8	14.3	1.3	0.2	204.0	**IT**
KZ	0.6	4.6	2.8	143.6	5.0	1.4	93.6	13.9	2.1	28.4	3.3	4.5	3.4	0.7	1.7	14.3	**KZ**
LT	0.1	0.02	4.6	0.3	1.8	6.1	12.9	37.3	2.6	11.0	0.04	20.6	22.4	7.1	3.4	11.2	**LT**
LU	0.0	0.0	0.2	0.0	0.0	9.7	0.0	0.0	0.6	0.0	0.0	0.2	3.0	0.1	0.002	2.3	**LU**
LV	0.1	0.01	2.7	0.2	1.1	5.5	9.5	20.1	1.7	6.5	0.03	12.5	18.3	6.7	12.0	8.6	**LV**
MC	0.0	0.0	0.001	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.01	**MC**
MD	0.2	0.03	0.8	0.4	2.2	0.5	59.3	5.7	0.4	17.1	0.3	2.2	1.7	0.3	0.2	2.1	**MD**
MK	9.5	0.01	1.2	0.1	5.6	0.4	188.4	0.4	0.6	127.4	0.3	2.4	1.1	0.1	0.04	7.1	**MK**
MT	0.004	0.0	0.01	0.00	0.02	0.002	0.07	0.00	0.01	0.04	0.003	0.004	0.003	0.0	0.0	0.1	**MT**
NL	0.0	0.0	1.3	0.0	0.1	120.0	0.4	0.2	2.4	0.5	0.003	4.1	26.0	1.3	0.1	22.7	**NL**
NO	0.1	0.01	4.1	0.4	1.6	16.3	8.5	4.8	4.3	9.6	0.04	14.8	27.6	23.4	2.3	15.0	**NO**
PL	0.8	0.04	47.4	0.7	19.5	54.2	83.6	64.7	20.6	120.8	0.2	467.0	409.5	38.8	5.7	54.5	**PL**
PT	0.0	0.0	0.1	0.0	0.1	1.1	0.2	0.0	0.4	0.2	0.004	0.2	0.4	0.1	0.0	372.1	**PT**
RO	4.7	0.1	22.2	1.4	75.3	6.1	1116.8	17.7	7.3	735.7	2.1	41.9	20.5	2.7	1.2	30.5	**RO**
RU	5.8	19.4	35.5	502.3	48.2	33.5	855.6	559.9	24.0	297.9	21.1	102.5	99.7	30.4	291.1	139.3	**RU**
SE	0.2	0.02	5.5	0.4	3.3	33.5	24.9	19.3	4.0	20.8	0.1	26.2	57.3	82.1	13.6	23.2	**SE**
SI	0.4	0.001	31.1	0.0	12.2	1.1	13.0	0.4	3.3	39.7	0.03	9.2	3.3	0.4	0.1	8.2	**SI**
SK	0.5	0.01	29.6	0.1	18.2	4.4	51.5	3.7	4.7	126.8	0.04	111.1	18.8	1.8	0.5	8.8	**SK**
TR	3.2	11.7	4.6	22.0	11.8	2.2	421.1	11.3	3.3	95.4	110.0	9.0	6.2	1.2	0.7	49.1	**TR**
UA	3.2	1.7	26.6	34.6	40.7	15.2	647.8	185.7	11.5	292.2	4.8	83.8	53.6	9.0	5.6	43.8	**UA**
BAS	0.2	0.02	9.5	0.4	2.9	52.8	21.1	23.3	6.5	18.6	0.1	46.1	121.6	97.5	63.3	39.1	**BAS**
BLS	2.8	1.6	5.8	16.4	17.3	2.4	712.7	26.3	3.1	138.5	11.7	11.7	7.6	1.8	1.3	22.9	**BLS**
CAS	0.2	6.9	0.8	384.6	1.5	0.4	22.2	3.5	0.7	7.7	5.3	1.3	0.9	0.2	0.3	5.1	**CAS**
MDT	35.3	0.3	40.2	1.7	136.9	16.8	940.3	5.9	46.2	405.5	226.0	40.8	24.7	2.6	0.7	1303.7	**MDT**
NOS	0.2	0.0	13.6	0.4	2.6	216.5	12.3	5.7	21.6	13.5	0.1	51.2	159.7	70.4	2.1	180.3	**NOS**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CS**	**CY**	**CZ**	**DE**	**DK**	**EE**	**ES**	

***Table C.3**. Matrix of cadmium country-to-country depositions from anthropogenic sources in 2004, kg/y (continued)*

Receptors ↓ Emitters →

	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	MC	
AL	0.05	6.4	0.9	0.02	33.1	2.3	5.0	0.1	0.003	64.4	0.3	0.1	0.02	0.04	0.01	**AL**
AM	0.02	0.3	0.1	4.8	0.6	0.1	0.2	0.01	0.0	0.6	8.2	0.02	0.001	0.02	0.0	**AM**
AT	0.5	66.1	15.1	0.01	2.0	19.9	55.2	1.1	0.02	103.8	0.2	0.8	0.6	0.6	0.1	**AT**
AZ	0.1	0.6	0.2	10.8	1.2	0.1	0.4	0.0	0.001	1.5	67.3	0.1	0.002	0.1	0.001	**AZ**
BA	0.3	17.0	2.6	0.01	8.7	73.9	80.1	0.2	0.01	85.3	0.3	0.5	0.1	0.3	0.1	**BA**
BE	0.1	250.0	74.2	0.0	0.1	0.1	0.2	3.2	0.02	2.2	0.02	0.1	2.9	0.1	0.004	**BE**
BG	0.6	9.7	3.0	0.2	92.0	6.9	42.9	0.2	0.01	37.1	3.2	0.7	0.1	0.5	0.02	**BG**
BY	8.6	24.0	17.7	0.4	6.4	6.4	37.3	1.4	0.05	18.9	8.9	43.5	0.2	24.6	0.02	**BY**
CH	0.0	166.5	13.0	0.001	0.8	1.6	1.3	0.9	0.01	92.6	0.02	0.1	0.3	0.0	0.1	**CH**
CS	0.6	16.5	3.4	0.05	41.1	27.4	130.8	0.3	0.01	84.3	0.9	0.7	0.1	0.5	0.04	**CS**
CY	0.0	0.2	0.1	0.01	1.5	0.1	0.2	0.005	0.0	1.0	0.05	0.005	0.001	0.004	0.001	**CY**
CZ	1.0	42.2	19.2	0.01	1.6	7.2	55.6	1.3	0.02	17.0	0.3	1.3	0.5	0.9	0.02	**CZ**
DE	2.7	664.7	284.2	0.02	1.4	3.2	15.2	15.7	0.2	45.2	0.8	3.8	13.7	2.8	0.1	**DE**
DK	0.7	25.2	39.4	0.01	0.1	0.3	1.9	2.2	0.1	1.8	0.1	0.7	0.2	0.5	0.002	**DK**
EE	20.1	6.1	5.8	0.02	0.4	0.4	2.4	0.5	0.02	1.6	0.2	12.1	0.1	27.2	0.002	**EE**
ES	0.2	150.6	46.7	0.004	1.6	3.6	4.0	4.7	0.1	78.8	0.1	0.2	0.4	0.1	0.1	**ES**
FI	686.3	18.4	18.6	0.1	0.6	1.0	5.8	1.6	0.1	3.9	1.1	14.4	0.2	18.4	0.004	**FI**
FR	0.5	2851.8	348.2	0.01	3.6	10.0	9.8	25.9	0.2	226.4	0.2	0.6	10.0	0.4	1.4	**FR**
GB	0.7	161.3	1411.4	0.01	0.2	0.4	1.6	77.1	0.5	4.6	0.4	0.6	0.9	0.5	0.01	**GB**
GE	0.1	1.8	0.4	66.1	4.5	0.6	1.1	0.04	0.002	4.9	14.9	0.2	0.01	0.1	0.004	**GE**
GR	0.3	11.7	2.2	0.2	625.7	4.3	16.4	0.2	0.01	69.7	2.5	0.3	0.04	0.2	0.02	**GR**
HR	0.3	17.6	2.3	0.01	7.2	224.1	103.7	0.2	0.01	102.1	0.2	0.4	0.1	0.3	0.1	**HR**
HU	0.6	18.4	5.0	0.02	6.2	55.9	787.0	0.4	0.01	58.3	0.3	0.8	0.1	0.5	0.04	**HU**
IE	0.1	15.4	35.3	0.001	0.03	0.1	0.2	95.4	0.2	0.8	0.03	0.04	0.1	0.0	0.002	**IE**
IS	0.1	7.0	16.6	0.002	0.03	0.1	0.2	2.2	15.1	0.7	0.1	0.1	0.1	0.05	0.001	**IS**
IT	0.4	204.4	13.0	0.05	43.5	79.1	50.2	1.2	0.03	2023	0.7	0.6	0.3	0.5	1.0	**IT**
KZ	2.5	7.7	2.8	8.1	12.1	2.4	7.1	0.3	0.02	15.3	1052.0	1.5	0.04	1.6	0.01	**KZ**
LT	4.4	14.3	11.6	0.05	1.4	2.0	10.0	0.9	0.02	6.6	0.9	114.2	0.1	27.2	0.006	**LT**
LU	0.0	18.6	3.5	0.0	0.005	0.02	0.03	0.2	0.001	0.3	0.002	0.01	3.4	0.0	0.001	**LU**
LV	9.7	11.6	10.4	0.04	1.0	1.0	5.7	0.8	0.02	4.1	0.6	93.9	0.1	164.9	0.004	**LV**
MC	0.0	0.04	0.001	0.0	0.001	0.002	0.001	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.005	**MC**
MD	0.3	1.7	0.9	0.1	3.4	0.9	4.4	0.1	0.003	4.0	1.4	0.4	0.01	0.3	0.003	**MD**
MK	0.1	3.2	0.6	0.03	66.3	1.7	7.7	0.1	0.002	18.0	0.4	0.1	0.01	0.1	0.007	**MK**
MT	0.0	0.05	0.01	0.0	0.04	0.01	0.01	0.001	0.0	0.3	0.0	0.0	0.0	0.0	0.0	**MT**
NL	0.1	112.7	81.2	0.001	0.1	0.1	0.4	3.5	0.03	1.7	0.1	0.2	0.5	0.1	0.003	**NL**
NO	11.9	35.7	85.7	0.04	0.6	1.2	6.9	7.2	0.6	5.6	0.9	3.5	0.3	2.9	0.007	**NO**
PL	9.4	106.4	76.6	0.1	7.4	19.9	128.2	5.1	0.1	44.2	2.3	25.5	1.2	13.8	0.04	**PL**
PT	0.0	7.7	4.2	0.0	0.1	0.1	0.1	0.6	0.01	2.2	0.01	0.01	0.02	0.01	0.003	**PT**
RO	1.8	28.9	10.5	0.6	45.7	31.9	212.3	0.8	0.02	85.2	5.2	2.4	0.2	1.8	0.1	**RO**
RU	291.1	110.1	79.5	55.1	95.8	25.8	106.3	6.8	0.5	137.0	1644.5	83.2	0.7	110.7	0.1	**RU**
SE	111.2	55.7	68.8	0.1	1.3	2.4	14.4	4.7	0.2	7.9	1.1	18.8	0.4	15.6	0.01	**SE**
SI	0.1	10.5	1.3	0.004	1.8	50.5	26.9	0.1	0.003	75.0	0.1	0.2	0.04	0.1	0.03	**SI**
SK	0.8	14.2	6.5	0.02	3.3	16.2	224.4	0.5	0.01	23.4	0.5	1.3	0.1	0.8	0.01	**SK**
TR	1.0	17.0	4.6	6.5	142.7	5.5	20.2	0.4	0.02	60.2	28.7	1.2	0.1	0.9	0.03	**TR**
UA	7.6	46.8	27.5	5.0	43.4	25.2	185.1	2.2	0.1	75.3	143.8	14.0	0.4	9.1	0.1	**UA**
BAS	195.0	81.7	85.2	0.1	1.4	2.8	17.3	5.8	0.1	11.4	1.1	43.0	0.8	43.4	0.01	**BAS**
BLS	1.9	13.1	5.0	10.0	79.4	6.7	24.4	0.4	0.02	41.5	61.3	2.5	0.1	1.8	0.03	**BLS**
CAS	0.4	2.5	0.8	9.6	3.8	0.7	2.1	0.1	0.01	5.2	1020.0	0.4	0.01	0.4	0.004	**CAS**
MDT	1.6	395.2	43.8	0.5	710.0	104.9	80.4	3.7	0.1	1906.6	5.9	1.5	0.6	1.1	0.9	**MDT**
NOS	4.4	483.8	1114.5	0.04	1.0	1.9	14.0	57.4	1.7	17.2	1.0	4.9	2.4	3.7	0.03	**NOS**
	FI	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KZ**	**LT**	**LU**	**LV**	**MC**	

***Table C.3**. Matrix of cadmium country-to-country depositions from anthropogenic sources in 2004, kg/y (continued)*

Receptors ↓ Emitters →

	MD	MK	NL	NO	PL	PT	RO	RU	SE	SI	SK	TR	UA	Total, kg/y	
AL	0.1	367.9	0.2	0.04	19.2	1.4	10.9	3.1	0.0	2.0	8.3	6.7	1.2	798	**AL**
AM	0.02	1.3	0.02	0.01	2.0	0.1	0.7	6.6	0.0	0.1	0.3	46.6	0.7	146	**AM**
AT	0.1	13.4	6.6	0.8	388.2	2.2	22.1	11.4	0.7	135.9	195.9	2.2	1.4	1679	**AT**
AZ	0.05	2.8	0.04	0.04	5.5	0.2	1.8	36.2	0.0	0.2	0.7	76.2	2.8	702	**AZ**
BA	0.1	59.6	1.0	0.2	223.2	1.7	51.8	6.2	0.3	24.7	115.2	5.5	2.4	1908	**BA**
BE	0.001	0.2	32.1	0.4	15.9	3.6	0.2	1.0	0.2	0.4	1.4	0.1	0.0	801	**BE**
BG	2.4	433.1	1.0	0.3	184.3	1.3	296.9	56.2	0.4	8.6	64.3	127.0	22.1	7753	**BG**
BY	2.6	35.8	5.9	3.7	2138.3	1.9	93.4	630.5	5.8	12.1	115.7	27.1	94.5	4309	**BY**
CH	0.004	2.0	2.2	0.2	16.7	2.4	0.8	1.0	0.1	4.6	3.6	1.0	0.1	647	**CH**
CS	0.4	641.9	1.3	0.3	288.4	1.9	175.4	16.8	0.4	18.9	148.0	23.2	6.3	5307	**CS**
CY	0.01	1.7	0.01	0.01	1.0	0.05	0.5	0.4	0.0	0.1	0.3	24.0	0.2	128	**CY**
CZ	0.1	9.7	9.6	1.3	1185.9	1.6	23.5	18.5	1.2	20.4	890.8	2.3	1.9	3196	**CZ**
DE	0.1	6.1	216.4	7.3	1030.6	17.7	11.3	45.8	5.6	10.8	77.3	2.8	2.5	4671	**DE**
DK	0.01	0.7	13.3	2.5	135.4	1.0	2.7	7.6	3.1	0.6	8.9	0.3	0.6	375	**DK**
EE	0.1	1.3	2.1	2.9	179.8	0.3	4.8	94.2	6.7	1.1	10.3	2.0	3.0	503	**EE**
ES	0.02	8.3	7.4	0.6	37.5	404.9	3.4	5.1	0.2	7.8	11.1	2.2	0.4	6563	**ES**
FI	0.3	3.1	6.8	13.9	394.2	0.9	12.8	333.8	49.5	2.4	25.6	3.0	8.1	1783	**FI**
FR	0.03	14.5	58.2	2.0	124.7	67.7	6.8	8.7	1.0	22.6	29.1	5.5	0.7	5424	**FR**
GB	0.02	1.2	40.2	2.8	76.1	14.5	1.7	14.0	1.1	1.3	9.0	0.5	0.8	2023	**GB**
GE	0.2	13.6	0.1	0.1	15.4	0.4	6.6	103.2	0.1	0.9	2.0	208.7	8.2	618	**GE**
GR	0.8	741.6	0.6	0.2	82.7	2.8	71.3	29.2	0.2	5.0	27.2	169.5	11.0	3015	**GR**
HR	0.1	41.5	1.0	0.3	238.5	1.5	32.5	5.4	0.3	96.2	137.4	4.2	1.9	1481	**HR**
HU	0.2	63.3	2.2	0.5	508.0	1.2	233.5	12.2	0.5	74.6	426.6	7.6	5.4	2968	**HU**
IE	0.001	0.2	2.1	0.2	4.9	2.7	0.2	1.0	0.1	0.2	0.6	0.05	0.1	183	**IE**
IS	0.01	0.1	2.1	0.7	8.9	1.2	0.3	1.5	0.1	0.2	1.0	0.2	0.1	73	**IS**
IT	0.2	141.1	3.1	0.6	291.4	15.3	36.2	11.3	0.6	154.4	116.5	42.8	3.0	3887	**IT**
KZ	1.2	38.6	0.8	0.7	105.6	1.3	40.4	1085.1	0.8	4.7	14.3	184.6	71.3	2987	**KZ**
LT	0.3	5.6	4.1	2.8	953.7	1.0	15.8	222.4	5.1	5.0	42.9	4.0	8.6	1607	**LT**
LU	0.0	0.03	1.1	0.0	1.3	0.2	0.02	0.1	0.01	0.1	0.2	0.01	0.004	45	**LU**
LV	0.2	4.2	3.6	3.9	471.4	0.8	11.1	136.7	7.7	2.6	24.2	3.7	6.6	1086	**LV**
MC	0.0	0.002	0.0	0.0	0.01	0.001	0.001	0.0	0.0	0.005	0.003	0.001	0.0	0.2	**MC**
MD	20.3	15.1	0.3	0.2	77.6	0.2	70.2	34.8	0.2	1.3	7.6	20.5	24.4	384	**MD**
MK	0.1	2955.7	0.2	0.04	27.2	0.6	17.9	4.9	0.0	1.7	11.9	14.9	1.7	3480	**MK**
MT	0.0	0.1	0.001	0.0	0.05	0.006	0.01	0.005	0.0	0.01	0.02	0.03	0.001	0.9	**MT**
NL	0.0	0.3	189.2	0.6	32.9	3.8	0.3	2.1	0.3	0.4	3.1	0.1	0.1	613	**NL**
NO	0.1	3.8	12.3	237.6	339.0	2.5	11.4	68.0	16.6	2.5	28.6	2.4	4.1	1025	**NO**
PL	1.3	42.2	35.8	8.1	19383.9	7.0	146.5	277.8	11.7	47.2	778.8	17.8	31.0	22618	**PL**
PT	0.001	0.2	0.5	0.04	1.9	1152.8	0.1	0.5	0.02	0.3	0.5	0.1	0.03	1547	**PT**
RO	11.4	307.9	3.6	1.2	734.7	2.6	3249.5	125.3	1.1	40.3	243.9	152.0	61.2	7449	**RO**
RU	12.9	366.2	22.5	30.3	3216.6	12.9	465.8	39672.4	51.3	49.7	286.8	1310.3	792.7	52104	**RU**
SE	0.5	7.9	25.9	72.9	952.3	2.6	33.2	179.2	216.0	4.6	58.8	5.7	11.9	2189	**SE**
SI	0.03	10.4	0.6	0.1	89.6	0.7	9.6	2.1	0.1	509.4	49.8	1.9	0.6	964	**SI**
SK	0.2	27.0	2.8	0.6	1233.2	0.9	118.9	18.2	0.7	32.8	1630.3	5.1	5.0	3748	**SK**
TR	3.8	199.8	1.2	0.7	196.7	4.6	142.5	188.4	0.6	8.4	35.7	6357.5	68.2	8260	**TR**
UA	29.0	207.1	9.9	4.0	3146.8	4.4	842.8	1856.2	4.7	42.3	284.2	377.3	1266.7	10121	**UA**
BAS	0.4	7.0	38.5	19.1	1809.0	3.7	30.7	278.0	79.8	7.2	91.1	5.7	10.8	3373	**BAS**
BLS	11.4	222.2	1.4	1.1	337.5	1.9	254.6	416.2	1.2	10.6	43.8	1007.4	217.6	3759	**BLS**
CAS	0.3	10.6	0.2	0.2	28.7	0.5	10.3	262.4	0.2	1.3	4.2	139.0	20.7	1966	**CAS**
MDT	1.9	823.3	7.5	1.5	453.4	64.9	167.1	59.8	1.2	132.1	174.3	1163.9	22.5	9558	**MDT**
NOS	0.1	7.2	227.1	54.6	775.3	28.6	17.9	59.5	13.3	5.0	68.6	2.2	3.4	3721	**NOS**
	MD	**MK**	**NL**	**NO**	**PL**	**PT**	**RO**	**RU**	**SE**	**SI**	**SK**	**TR**	**UA**	**Total**	

***Table C.4**. Matrix of mercury country-to-country depositions from anthropogenic sources in 2004, kg/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	ES	
AL	27.4	0.002	0.3	0.01	2.9	0.2	5.2	0.03	0.3	31.7	0.05	0.5	0.2	0.1	0.01	4.9	**AL**
AM	0.01	23.1	0.03	9.2	0.1	0.03	0.3	0.02	0.03	0.2	0.6	0.05	0.04	0.02	0.01	0.4	**AM**
AT	0.1	0.0	162.9	0.02	4.8	6.5	1.5	0.2	19.9	10.5	0.01	34.9	13.6	1.9	0.1	14.5	**AT**
AZ	0.03	5.9	0.1	134.1	0.2	0.1	0.7	0.1	0.1	0.5	1.6	0.1	0.1	0.1	0.02	1.0	**AZ**
BA	1.4	0.002	3.9	0.02	366.3	0.8	5.1	0.2	1.1	81.3	0.04	6.4	1.8	0.4	0.1	8.7	**BA**
BE	0.003	0.0	0.3	0.001	0.04	361	0.04	0.02	1.4	0.1	0.001	0.7	3.9	0.6	0.01	6.9	**BE**
BG	1.4	0.02	1.4	0.1	6.8	0.7	851	0.5	0.7	70.4	0.5	3.6	1.5	0.5	0.1	6.1	**BG**
BY	0.2	0.05	3.0	0.3	3.4	4.0	6.6	124.7	1.8	10.5	0.1	14.2	8.7	6.4	3.2	7.7	**BY**
CH	0.02	0.0	1.9	0.003	0.3	3.3	0.2	0.02	147.1	0.4	0.01	1.0	2.4	0.2	0.01	14.5	**CH**
CS	5.8	0.005	3.5	0.03	59.2	1.0	39.3	0.3	1.1	993	0.1	7.2	2.3	0.5	0.1	8.8	**CS**
CY	0.01	0.007	0.02	0.01	0.05	0.01	0.2	0.004	0.02	0.1	98.1	0.03	0.02	0.01	0.001	0.2	**CY**
CZ	0.1	0.002	21.1	0.02	2.4	6.8	1.3	0.3	5.5	7.3	0.01	340	46.6	3.3	0.1	5.8	**CZ**
DE	0.1	0.005	27.9	0.05	1.2	175.6	0.9	0.7	81.0	2.6	0.03	75.6	547.6	29.7	0.4	43.3	**DE**
DK	0.01	0.001	0.4	0.01	0.1	5.3	0.1	0.1	0.6	0.3	0.004	1.9	7.7	112.6	0.1	3.0	**DK**
EE	0.01	0.002	0.3	0.01	0.2	1.1	0.4	0.9	0.3	0.5	0.01	1.4	2.0	2.5	35.2	1.4	**EE**
ES	0.1	0.001	0.6	0.01	0.7	6.3	0.4	0.1	2.1	1.0	0.02	1.0	1.5	0.5	0.04	3149	**ES**
FI	0.04	0.02	0.9	0.1	0.6	4.7	1.3	2.3	0.8	1.8	0.03	3.8	5.4	7.9	21.2	4.4	**FI**
FR	0.1	0.003	3.4	0.02	1.9	122.8	0.9	0.1	55.2	2.4	0.1	4.9	18.2	3.0	0.1	395	**FR**
GB	0.01	0.002	0.8	0.03	0.2	28.2	0.3	0.1	1.9	0.5	0.01	2.3	5.9	2.7	0.1	27.3	**GB**
GE	0.1	5.3	0.2	20.6	0.6	0.1	2.4	0.2	0.2	1.4	1.2	0.3	0.2	0.1	0.03	1.9	**GE**
GR	3.9	0.02	0.8	0.1	4.1	0.5	109.5	0.2	0.6	22.5	1.3	1.6	0.8	0.2	0.1	10.4	**GR**
HR	0.9	0.001	7.5	0.01	37.7	0.8	2.8	0.1	1.2	54.5	0.03	8.0	1.9	0.6	0.04	8.2	**HR**
HU	0.4	0.002	16.3	0.02	22.9	1.7	6.4	0.4	1.9	88.9	0.04	19.7	4.4	1.0	0.1	6.1	**HU**
IE	0.002	0.0	0.1	0.002	0.0	1.7	0.03	0.01	0.3	0.1	0.002	0.2	0.4	0.2	0.01	4.5	**IE**
IS	0.003	0.001	0.1	0.01	0.1	1.1	0.1	0.03	0.2	0.1	0.002	0.3	0.6	0.3	0.01	1.7	**IS**
IT	2.5	0.01	12.9	0.03	21.3	3.3	6.2	0.2	22.0	19.4	0.4	8.9	4.3	1.0	0.1	69.1	**IT**
KZ	0.2	2.3	1.1	25.3	2.3	1.1	8.9	1.5	1.0	5.3	1.3	2.0	1.5	0.7	0.6	7.6	**KZ**
LT	0.05	0.01	1.2	0.04	0.7	2.4	1.2	7.5	0.9	1.9	0.03	5.6	5.5	5.8	1.3	4.3	**LT**
LU	0.0	0.0	0.05	0.0	0.005	5.0	0.003	0.002	0.2	0.01	0.0	0.1	0.5	0.04	0.0	0.7	**LU**
LV	0.03	0.004	0.7	0.03	0.4	2.1	0.9	2.7	0.5	1.2	0.02	3.2	4.2	5.1	4.0	3.3	**LV**
MC	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.001	0.0	0.0	0.0	0.0	0.0	0.0	0.006	**MC**
MD	0.1	0.01	0.3	0.04	0.8	0.3	6.6	0.7	0.1	3.1	0.1	0.8	0.5	0.3	0.1	0.9	**MD**
MK	4.3	0.002	0.3	0.0	2.0	0.2	20.5	0.04	0.2	38.7	0.1	0.6	0.3	0.1	0.01	2.5	**MK**
MT	0.001	0.0	0.001	0.0	0.005	0.001	0.003	0.0	0.001	0.006	0.0	0.001	0.001	0.0	0.0	0.03	**MT**
NL	0.003	0.0	0.3	0.0	0.1	85.8	0.04	0.03	0.8	0.1	0.001	1.1	8.0	1.2	0.02	5.7	**NL**
NO	0.1	0.01	1.3	0.1	0.8	7.3	1.1	0.7	1.8	2.2	0.02	4.9	7.8	20.6	0.9	6.9	**NO**
PL	0.3	0.01	12.7	0.1	6.7	20.1	6.6	15.5	6.9	22.2	0.1	199	115.6	29.6	1.7	19.0	**PL**
PT	0.002	0.0	0.0	0.001	0.02	0.4	0.02	0.005	0.1	0.0	0.001	0.1	0.1	0.04	0.01	98.5	**PT**
RO	1.5	0.03	6.9	0.1	29.8	2.8	115.3	2.3	2.4	158.7	0.8	14.2	5.7	2.0	0.4	12.2	**RO**
RU	2.2	9.3	12.6	79.2	20.9	20.3	78.7	91.4	10.5	57.1	7.9	35.8	31.3	26.3	142.7	70.3	**RU**
SE	0.1	0.01	1.8	0.1	1.5	13.3	2.6	2.7	1.7	4.6	0.1	8.5	14.9	82.9	4.3	10.0	**SE**
SI	0.1	0.001	12.7	0.01	3.8	0.5	0.7	0.05	0.9	6.4	0.01	3.7	1.0	0.3	0.02	3.2	**SI**
SK	0.2	0.003	9.8	0.02	6.4	1.7	3.3	0.5	1.5	24.2	0.02	45.4	4.8	1.2	0.1	3.4	**SK**
TR	1.1	7.7	1.6	3.7	5.1	1.5	64.4	1.2	1.5	18.2	48.2	3.6	2.4	0.9	0.3	19.3	**TR**
UA	1.0	0.6	7.6	4.0	14.3	6.5	61.9	25.3	3.9	51.1	1.8	26.1	13.8	6.6	2.1	17.7	**UA**
BAS	0.1	0.01	2.9	0.1	1.4	19.3	2.2	3.7	2.3	4.0	0.05	14.6	40.6	128.7	33.6	13.8	**BAS**
BLS	0.8	0.6	1.4	1.7	5.1	1.1	86.2	2.3	0.9	18.8	3.9	3.2	1.9	1.1	0.4	8.3	**BLS**
CAS	0.1	3.9	0.4	98.5	0.7	0.4	2.3	0.4	0.3	1.6	2.2	0.6	0.5	0.2	0.1	2.8	**CAS**
MDT	17.1	0.1	10.5	0.3	48.1	5.5	102.4	0.7	10.8	64.3	142.9	11.9	6.3	1.7	0.2	635	**MDT**
NOS	0.1	0.01	3.7	0.1	1.0	103.2	1.1	0.8	7.9	2.3	0.03	13.9	41.0	66.5	0.6	56.7	**NOS**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CS**	**CY**	**CZ**	**DE**	**DK**	**EE**	**ES**	

***Table C.4**. Matrix of mercury country-to-country depositions from anthropogenic sources in 2004, kg/y (continued)*

Receptors ↓ Emitters →

	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	MC	
AL	0.01	1.8	0.6	0.0	93.4	0.4	2.1	0.02	0.001	15.7	0.1	0.02	0.03	0.001	0.0	**AL**
AM	0.005	0.1	0.1	2.6	0.9	0.01	0.1	0.003	0.001	0.3	4.5	0.01	0.004	0.0	0.0	**AM**
AT	0.1	25.0	11.4	0.004	3.2	3.2	27.7	0.3	0.01	53.7	0.2	0.2	1.4	0.01	0.3	**AT**
AZ	0.02	0.3	0.3	7.4	2.2	0.04	0.3	0.01	0.002	0.8	48.1	0.03	0.01	0.002	0.0	**AZ**
BA	0.04	5.2	1.9	0.01	13.5	17.3	43.3	0.1	0.004	32.7	0.2	0.1	0.1	0.01	0.2	**BA**
BE	0.01	157.4	48.7	0.0	0.1	0.02	0.1	0.6	0.003	0.7	0.02	0.02	12.2	0.001	0.01	**BE**
BG	0.1	3.1	2.0	0.04	157.8	1.0	17.2	0.1	0.01	11.1	1.1	0.2	0.1	0.0	0.1	**BG**
BY	1.0	8.9	12.5	0.1	9.8	0.9	16.0	0.4	0.02	8.2	4.7	13.9	0.5	0.8	0.1	**BY**
CH	0.01	45.1	9.9	0.001	1.2	0.2	0.6	0.3	0.004	50.0	0.04	0.02	0.7	0.001	0.3	**CH**
CS	0.1	5.1	2.3	0.01	66.9	6.6	72.8	0.1	0.01	27.7	0.4	0.2	0.2	0.01	0.1	**CS**
CY	0.002	0.1	0.04	0.003	1.5	0.01	0.1	0.003	0.0	0.2	0.03	0.002	0.002	0.0	0.001	**CY**
CZ	0.1	15.8	12.6	0.005	2.4	1.0	27.1	0.3	0.01	7.0	0.3	0.4	1.3	0.02	0.0	**CZ**
DE	0.3	279.1	187.5	0.01	2.5	0.5	7.3	3.5	0.03	19.1	0.7	1.0	38.7	0.1	0.2	**DE**
DK	0.1	9.3	26.6	0.002	0.2	0.04	0.8	0.5	0.005	0.7	0.1	0.2	0.3	0.01	0.01	**DK**
EE	2.4	2.1	4.0	0.01	0.6	0.1	1.1	0.1	0.004	0.7	0.1	4.4	0.1	0.8	0.0	**EE**
ES	0.05	50.5	28.6	0.002	1.5	0.3	1.4	1.0	0.02	13.7	0.1	0.05	0.7	0.003	0.2	**ES**
FI	104	8.4	16.0	0.03	1.7	0.2	3.2	0.5	0.03	2.3	1.3	4.5	0.4	0.4	0.02	**FI**
FR	0.1	1843	235	0.01	4.1	1.1	3.6	5.9	0.04	84.1	0.3	0.2	34.2	0.01	7.2	**FR**
GB	0.1	76.2	1913	0.005	0.5	0.1	0.8	20.4	0.03	1.8	0.3	0.2	1.8	0.01	0.02	**GB**
GE	0.02	0.7	0.5	49.0	7.2	0.1	0.6	0.02	0.002	2.0	8.5	0.05	0.02	0.003	0.02	**GE**
GR	0.05	3.4	1.5	0.05	2089	0.7	6.5	0.1	0.01	16.6	0.9	0.1	0.1	0.005	0.1	**GR**
HR	0.03	5.3	1.7	0.003	8.8	57.0	57.6	0.1	0.004	51.9	0.1	0.1	0.1	0.01	0.2	**HR**
HU	0.1	6.3	3.8	0.01	8.3	13.0	793	0.1	0.01	24.9	0.3	0.2	0.3	0.01	0.1	**HU**
IE	0.01	6.4	30.3	0.0	0.1	0.01	0.1	48.7	0.01	0.3	0.03	0.01	0.1	0.001	0.004	**IE**
IS	0.03	2.6	9.3	0.002	0.1	0.02	0.2	0.5	0.1	0.4	0.1	0.0	0.1	0.001	0.004	**IS**
IT	0.1	53.7	9.1	0.02	51.4	12.1	19.0	0.3	0.02	1456	0.4	0.2	0.6	0.01	4.1	**IT**
KZ	0.4	3.7	3.4	3.8	17.4	0.4	3.5	0.1	0.01	7.4	1144	0.5	0.1	0.04	0.1	**KZ**
LT	0.4	5.1	8.1	0.02	2.0	0.3	3.8	0.2	0.01	2.6	0.5	51.6	0.3	1.0	0.02	**LT**
LU	0.001	7.3	2.2	0.0	0.01	0.003	0.01	0.04	0.0	0.1	0.002	0.002	17.7	0.0	0.001	**LU**
LV	0.9	4.2	7.0	0.01	1.4	0.1	2.3	0.2	0.01	1.7	0.3	50.0	0.2	5.8	0.01	**LV**
MC	0.0	0.02	0.001	0.0	0.001	0.0	0.0	0.0	0.0	0.04	0.0	0.0	0.0	0.0	0.03	**MC**
MD	0.04	0.7	0.8	0.02	4.3	0.1	2.2	0.02	0.002	1.5	0.7	0.1	0.04	0.01	0.01	**MD**
MK	0.01	1.0	0.4	0.01	177.7	0.3	3.3	0.02	0.001	5.4	0.2	0.02	0.03	0.001	0.02	**MK**
MT	0.0	0.01	0.003	0.0	0.04	0.001	0.003	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	**MT**
NL	0.02	53.1	53.6	0.001	0.1	0.0	0.2	0.7	0.004	0.5	0.05	0.05	1.1	0.003	0.01	**NL**
NO	1.6	15.1	61.6	0.02	1.8	0.2	3.2	1.8	0.05	2.7	0.8	1.2	0.7	0.1	0.0	**NO**
PL	0.8	36.7	52.2	0.04	12.0	2.5	55.8	1.3	0.03	16.8	1.5	7.9	2.4	0.3	0.1	**PL**
PT	0.01	2.4	2.2	0.0	0.1	0.01	0.1	0.1	0.003	0.6	0.01	0.004	0.05	0.0	0.01	**PT**
RO	0.2	9.8	7.6	0.1	68.9	5.4	112.5	0.2	0.01	33.7	2.5	0.7	0.5	0.0	0.2	**RO**
RU	34.1	51.8	74.1	24.9	141.9	4.4	49.9	2.2	0.2	62.7	1194	26.8	2.3	3.0	0.4	**RU**
SE	15.2	22.8	50.9	0.03	3.2	0.4	6.9	1.3	0.04	4.2	1.1	6.0	0.9	0.3	0.0	**SE**
SI	0.01	2.8	0.9	0.002	2.0	8.0	11.3	0.0	0.002	39.6	0.1	0.04	0.1	0.003	0.1	**SI**
SK	0.1	4.7	4.1	0.01	4.8	2.6	167.0	0.1	0.004	8.8	0.4	0.3	0.3	0.02	0.0	**SK**
TR	0.2	6.7	4.5	3.0	192.9	1.0	10.0	0.2	0.02	19.3	14.7	0.4	0.2	0.02	0.1	**TR**
UA	1.0	16.3	19.7	1.7	59.5	3.4	76.8	0.6	0.03	27.6	73.1	4.2	0.9	0.2	0.2	**UA**
BAS	26.1	30.0	61.9	0.04	2.7	0.5	8.1	1.5	0.03	5.3	1.0	18.8	1.5	1.4	0.04	**BAS**
BLS	0.2	3.8	3.5	5.1	109.2	0.8	8.0	0.1	0.01	12.4	25.6	0.6	0.1	0.03	0.1	**BLS**
CAS	0.1	1.2	1.0	5.4	5.9	0.1	1.2	0.04	0.01	2.7	1429	0.1	0.04	0.01	0.02	**CAS**
MDT	0.3	139	22.4	0.2	1109	19.9	28.5	0.8	0.04	645	2.5	0.4	0.9	0.03	3.1	**MDT**
NOS	0.5	228	937	0.02	2.1	0.3	5.3	13.3	0.1	6.8	0.9	1.3	5.0	0.1	0.1	**NOS**
	FI	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KZ**	**LT**	**LU**	**LV**	**MC**	

***Table C.4**. Matrix of mercury country-to-country depositions from anthropogenic sources in 2004, kg/y (continued)*

Receptors ↓ Emitters →

	MD	MK	NL	NO	PL	PT	RO	RU	SE	SI	SK	TR	UA	Total, kg/y	
AL	0.1	31.1	0.04	0.01	3.1	0.2	3.7	0.2	0.02	0.3	1.3	2.6	0.7	231	**AL**
AM	0.0	0.1	0.01	0.01	0.4	0.02	0.4	0.5	0.01	0.02	0.1	26.9	0.5	72	**AM**
AT	0.1	0.8	1.7	0.2	74.3	0.6	5.1	1.0	0.2	21.9	16.7	0.9	1.4	523	**AT**
AZ	0.1	0.3	0.02	0.02	1.3	0.1	1.0	4.0	0.03	0.1	0.2	35.1	2.0	248	**AZ**
BA	0.1	3.4	0.2	0.1	36.2	0.3	10.7	0.6	0.1	3.5	17.5	2.0	2.0	669	**BA**
BE	0.003	0.02	11.7	0.1	2.4	0.6	0.1	0.1	0.1	0.1	0.1	0.04	0.1	610	**BE**
BG	2.1	29.9	0.2	0.1	28.3	0.3	199.0	3.3	0.1	1.0	13.0	58.9	13.3	1489	**BG**
BY	2.2	2.0	1.3	1.1	320.3	0.5	24.3	45.7	1.7	1.5	22.3	9.4	69.4	774	**BY**
CH	0.01	0.1	0.6	0.05	3.4	0.5	0.3	0.1	0.03	0.7	0.4	0.5	0.1	287	**CH**
CS	0.3	49.1	0.3	0.1	49.4	0.4	66.1	1.2	0.1	2.7	31.1	7.7	4.3	1518	**CS**
CY	0.01	0.1	0.003	0.002	0.2	0.0	0.2	0.05	0.002	0.01	0.1	14.7	0.1	116	**CY**
CZ	0.1	0.5	2.2	0.3	321.3	0.4	5.4	1.2	0.4	2.4	25.0	1.0	1.5	871	**CZ**
DE	0.1	0.4	84.7	1.9	193.2	3.1	3.0	3.0	1.8	1.5	6.1	1.5	2.3	1830	**DE**
DK	0.01	0.05	2.9	0.9	21.6	0.2	0.6	0.5	1.6	0.1	0.8	0.2	0.4	201	**DK**
EE	0.1	0.1	0.4	0.7	22.2	0.1	1.3	6.0	1.6	0.1	1.3	0.8	2.0	99	**EE**
ES	0.02	0.3	1.2	0.1	5.6	105.6	0.8	0.4	0.1	0.6	0.9	0.6	0.3	3378	**ES**
FI	0.3	0.4	1.6	4.3	53.0	0.3	4.5	23.4	16.9	0.4	3.6	1.7	6.4	315	**FI**
FR	0.04	0.6	14.0	0.6	19.9	12.4	1.9	0.8	0.4	2.7	2.4	2.1	0.7	2885	**FR**
GB	0.03	0.1	8.8	0.6	10.9	2.9	0.6	1.1	0.3	0.2	0.7	0.4	0.8	2113	**GB**
GE	0.2	0.9	0.03	0.0	2.9	0.1	3.1	4.9	0.04	0.1	0.5	85.1	5.8	207	**GE**
GR	0.6	71.8	0.1	0.1	11.7	0.5	27.3	1.8	0.1	0.6	4.4	86.9	5.7	2487	**GR**
HR	0.1	2.0	0.2	0.1	39.0	0.3	6.9	0.5	0.1	21.3	17.1	1.3	1.6	397	**HR**
HU	0.2	3.1	0.5	0.1	100.4	0.3	53.0	1.0	0.2	12.3	320.6	2.6	6.2	1521	**HU**
IE	0.002	0.01	0.4	0.1	0.8	0.5	0.1	0.1	0.03	0.03	0.1	0.04	0.1	96	**IE**
IS	0.01	0.03	0.3	0.2	1.9	0.2	0.2	0.2	0.1	0.04	0.1	0.2	0.2	22	**IS**
IT	0.1	6.5	0.8	0.2	45.9	2.2	7.8	0.9	0.2	25.6	10.7	12.0	1.9	1893	**IT**
KZ	1.0	2.5	0.3	0.3	19.8	0.5	14.6	176.2	0.4	0.7	3.4	71.4	48.0	1586	**KZ**
LT	0.3	0.3	0.9	0.7	116.9	0.3	3.8	11.1	1.3	0.5	4.7	1.6	5.8	262	**LT**
LU	0.0	0.002	0.3	0.01	0.2	0.04	0.01	0.01	0.004	0.01	0.01	0.004	0.01	35	**LU**
LV	0.2	0.3	0.8	1.0	58.4	0.2	2.7	7.5	1.8	0.3	2.8	1.3	4.2	184	**LV**
MC	0.0	0.0	0.0	0.0	0.002	0.0	0.0	0.0	0.0	0.001	0.0	0.0	0.0	0.112	**MC**
MD	41.2	0.8	0.1	0.05	15.7	0.1	56.6	2.6	0.1	0.2	2.3	7.7	24.6	177	**MD**
MK	0.1	273.5	0.04	0.02	4.3	0.1	6.1	0.3	0.02	0.2	2.0	6.0	1.0	552	**MK**
MT	0.0	0.005	0.0	0.0	0.01	0.001	0.003	0.0	0.0	0.001	0.002	0.01	0.001	0.2	**MT**
NL	0.004	0.0	87.0	0.2	5.1	0.6	0.1	0.1	0.1	0.05	0.2	0.1	0.1	306	**NL**
NO	0.2	0.4	2.8	95.2	52.0	0.7	3.5	4.9	6.2	0.4	2.9	1.6	3.5	321	**NO**
PL	1.1	2.4	8.0	2.1	5281.1	1.4	31.1	14.6	3.4	5.2	103.5	8.2	29.5	6138	**PL**
PT	0.001	0.01	0.1	0.02	0.4	462.6	0.04	0.1	0.01	0.04	0.04	0.1	0.04	568	**PT**
RO	14.3	17.0	0.8	0.3	136.1	0.6	1741.3	9.4	0.4	5.6	84.5	57.1	55.1	2720	**RO**
RU	11.6	22.8	6.2	10.1	483.1	4.7	154.0	4371.3	17.5	7.2	54.0	491.1	587.2	8591	**RU**
SE	0.5	0.7	5.3	26.2	136.0	0.8	9.0	11.1	103.9	0.7	7.6	3.2	9.1	577	**SE**
SI	0.0	0.4	0.1	0.04	15.6	0.1	2.1	0.2	0.0	103.1	5.1	0.6	0.5	226	**SI**
SK	0.2	1.4	0.6	0.1	228.4	0.2	21.9	1.3	0.2	4.3	409.7	2.1	5.2	971	**SK**
TR	3.7	13.3	0.4	0.3	35.1	1.1	71.4	19.6	0.3	1.3	8.3	4221.8	45.7	4856	**TR**
UA	41.2	11.0	2.1	1.2	581.3	1.2	244.1	180.8	1.6	5.2	119.6	143.9	1678.0	3541	**UA**
BAS	0.4	0.6	8.6	6.3	283.6	1.0	8.1	18.4	36.9	1.0	10.0	2.9	8.1	812	**BAS**
BLS	13.0	11.5	0.3	0.3	43.1	0.5	120.9	46.9	0.4	1.1	8.5	569.1	163.8	1287	**BLS**
CAS	0.3	0.8	0.1	0.1	5.6	0.2	4.1	37.2	0.1	0.2	1.1	59.0	15.1	1686	**CAS**
MDT	1.5	45.9	1.3	0.4	67.0	9.2	63.6	5.0	0.4	18.0	18.8	689.4	14.0	3964	**MDT**
NOS	0.1	0.4	54.9	19.1	112.7	5.9	3.5	3.8	4.9	0.7	5.1	1.4	2.5	1714	**NOS**
	MD	**MK**	**NL**	**NO**	**PL**	**PT**	**RO**	**RU**	**SE**	**SI**	**SK**	**TR**	**UA**	**Total, kg/y**	