Convention on Long-Range Transboundary Air Pollution

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STATUS REPORT
2/2012 June 2012

Long-term Changes of Heavy Metal Transboundary Pollution of the Environment (1990-2010)

EMEP contribution to the revision of the Heavy Metal Protocol

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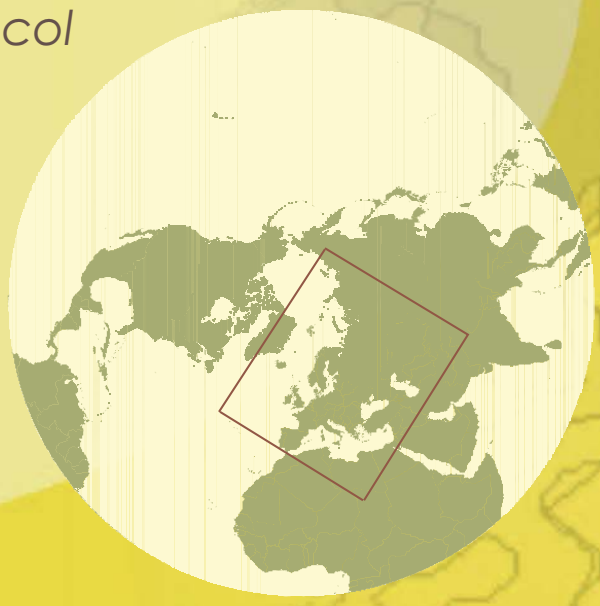

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METEOROLOGICAL SYNTHESIZING CENTRE - EAST

O.Travnikov, I.Ilyin, O.Rozovskaya, M.Varygina

CHEMICAL CO-ORDINATING CENTRE

W.Aas, H.T.Uggerud

CENTRE ON EMISSION INVENTORIES AND PROJECTIONS

K. Mareckova and R. Wankmueller

MCS-E

Meteorological Synthesizing Centre - East

Krasina pereulok, 16/1
123056 Moscow
Russia
Phone.: +7 495 981 15 67
Fax: +7 495 981 15 66
E-mail: msce@msceast.org
Internet: www.msceast.org

CCC

Norwegian Institute for Air Research (NILU)
P.O. Box 100
NO-2027 Kjeller
Norway
Phone: +47 63 89 80 00
Fax: +47 63 89 80 50
E-mail: kjetil.torseth@nilu.no
Internet: www.nilu.no

CEIP

Umweltbundesamt GmbH
Spittelauer Lande 5
1090 Vienna
Austria
Phone: +43-(0)1-313 04
Fax: +43-(0)1-313 04/5400
E-mail:emep.emissions@umweltbundesamt.at
Internet: www.umweltbundesamt.at/

EXECUTIVE SUMMARY

Scientific information presented in this report has been prepared jointly by Meteorological Synthesizing Centre – East (MSC-E), Chemical Co-ordinating Centre (CCC) and Centre on Emission Inventories and Projections (CEIP) of the Cooperative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) with contribution from the Coordination Centre for Effects (CCE) of the Working Group on Effects (WGE). The objective of the report is to support the revision of the 1998 Protocol on Heavy Metals (Protocol). This Protocol is one of the eight protocols of the UN ECE Convention on Long-range Transboundary Air Pollution (Convention) that identifies specific measures to be taken by Parties to cut harmful effects of heavy metal emissions on the environment and human health. Heavy metals targeted by the Protocol are lead (Pb), cadmium (Cd) and mercury (Hg).

Lead is a pollutant that is toxic at very low exposure levels and has acute and chronic effects on human health. It is a multi-organ system toxicant that can cause neurological, cardiovascular, renal, gastrointestinal, hematological and reproductive effects. It also bioaccumulates and adversely impacts both terrestrial and aquatic ecosystems. Cadmium is a non-essential and toxic element for humans mainly affecting kidneys and the skeleton. It is also a carcinogen by inhalation. Important endpoints of cadmium include kidney and bone damage and cancer. In the environment, cadmium is toxic to plants, animals and micro-organisms. Mercury is toxic in multiple forms but the main concern is associated with the organic compounds, especially methylmercury. Mercury can damage the liver, kidneys and the digestive and respiratory systems. It also causes brain and neurological damage and impairs growth. It affects animals in the same way as humans and is very toxic to aquatic life.

The concern regarding harmful effects of heavy metals on human health and the environment has led to the initiation of monitoring, assessment, regulation, and control activities on international and national levels. Currently, activities of AMAP, European Commission, HELCOM, OSPAR, UNEP, WHO, Basel and Rotterdam Conventions, various national programmes etc. are focused on gradual reduction and prevention of air pollution, including long-range transboundary transport of heavy metals.

According to the Article 7 of the Protocol each Party shall report to EMEP information on the levels of emissions of Cd, Pb, Hg, using methodologies and temporal and spatial resolution, specified by the EMEP Steering Body. Since signing of the Protocol in 1998 the number of Parties reported emission data increased from 30 to 46. On the other hand, data on spatial distribution of emissions are reported only by 28 Parties.

The EMEP monitoring network for heavy metals has been developing continuously since 1999. The number of monitoring sites measuring lead, cadmium and mercury increased from 44 in 1990 to 66 in 2010. The monitoring network covers a significant part of the EMEP countries. However, large territories in Eastern and Southern Europe as well as in Central Asia remain uncovered.

In order to support implementation of the Protocol EMEP provides the Executive Body for the Convention with information on deposition and transboundary transport of heavy metals within the geographical scope of EMEP (Article 8).

This year assessment performed by the EMEP Centres is focussed on evaluation of heavy metal pollution levels in the EMEP region and their trends over the period from 1990 to 2010. Information on emissions, monitoring data and model estimates has been generated within EMEP, WGE and other Bodies to the Convention.

Anthropogenic emissions were significantly reduced in the EMEP countries over the last two decades. Lead emissions dropped by 90% since 1990, whereas emissions of cadmium and mercury decreased approximately by 60%. Change of heavy metal deposition varied over the EMEP countries. Both modelling results and observations showed that deposition fluxes decreased on average by 75% for lead, 50% for cadmium, and 30% for mercury

In spite of deposition reduction of pollution levels in Europe transboundary transport continues to play an important role in heavy metal pollution of the EMEP countries. Change in the emission pattern led to the redistribution of transboundary fluxes between the countries. Contribution of foreign sources to heavy metal anthropogenic deposition has changed substantially in some countries but still remains significant in most of them.

Lead deposition dramatically decreased in the EMEP countries mostly due to the phase out of leaded gasoline from use in road transport. However, reduction of lead emissions from other sectors was less significant. Human health and the environment continue to be at risk in many EMEP countries despite important reductions of lead deposition.

Since 1990 cadmium deposition was reduced by 50% on average and the changes ranged from about 60% reduction in some countries of Western and Central Europe to moderate increase in the countries of Caucasus and Central Asia. High deposition levels still remain in a number of ‘hot spots’ close to industrial regions, which require more detailed analysis on national or local scales.

Mercury is dispersed globally in the atmosphere. Its deposition in the EMEP countries decreased slightly (by 30%) since 1990 due to large contribution of emissions from other continents. Nowadays, intercontinental transport contributes more than 65% to total mercury deposition in the EMEP countries. Therefore, both regional and global efforts are needed to reduce mercury pollution. Mercury levels in many EMEP countries still pose significant risk to human health and the environment. It accumulates in the food chain, for example, in predatory fish in lakes and seas, and reaches humans.

Reduction of heavy metal pollution levels was accompanied by changes in the key source categories of both emissions and deposition. Prevailing contribution of road transport for lead and metal production for cadmium in 1990 were replaced by industrial and non-industrial combustion in 2010. Changes in sectoral composition of mercury emissions were less significant. Nowadays, the prevailing sectors in deposition of all three metals include *stationary combustion in industry* (1A2), *non-industrial combustion* (1A4), *metal production* (2C) and *public electricity and heat production* (1A1a). These source categories will require priority mitigation efforts to reduce heavy metal pollution in future.

Pollution reduction in the Eastern Europe, Caucasus and Central Asia (EECCA) countries currently is of high priority within the LRTAP Convention (*Action Plan for EECCA*). However, assessment of heavy metal pollution in these countries is restricted by the lack of national emissions and monitoring data. Official data on anthropogenic emissions are reported only by 5 of 12 EECCA countries. Among them, two countries report information on spatial distribution of emissions. Besides, no monitoring data on heavy metal concentration in air and precipitation are reported so far. Therefore, additional attention should be paid to the development of national emission inventories and monitoring networks in these countries.

In summary, although heavy metal pollution levels have been reduced considerably in the EMEP countries, they are still high enough to pose a significant risk to human health and the environment at present and in future.

CONTENTS

ACKNOLEDGEMENTS

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INTRODUCTION

Pollution of the environment by heavy metals and their compounds can cause harmful effects on human health and ecosystems. The atmosphere is one of the major pathways of heavy metal dispersion in the environment. Heavy metals emitted to the atmosphere from combustion of fossil fuels, industrial processes and other sources both contribute to pollution levels nearby emission sources and can be transported over long distances (from hundreds to thousands of kilometers) with atmospheric flows reaching remote regions.

The concern regarding harmful effects of heavy metals on human health and the environment has led to the initiation of monitoring, assessment, regulation, and control activities on international and national levels. Currently, activities of AMAP, European Commission, HELCOM, OSPAR, UNEP, WHO, Basel and Rotterdam Conventions, various national programmes etc. are focused on gradual reduction and prevention of air pollution, including long-range transboundary transport of heavy metals [*WHO/CLRTAP*, 2009; *UNEP*, 2010a; *UNEP*, 2010b; *TF HTAP*, 2010; *AMAP*, 2011; *EEA*, 2011].

In order to identify specific measures to be taken to cut adverse effects of heavy metal emissions on the environment 41 Parties to the Convention on Long-Range Transboundary Air Pollution (Convention) signed and/or ratified the 1998 Protocol on Heavy Metals (Protocol). According to the Protocol, the Cooperative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) shall provide the Executive Body for the Convention with information on deposition and transboundary transport of heavy metals within the geographical scope of EMEP. The Centre of Emission Inventories and Projections (CEIP) prepares data on atmospheric emissions based on information reported by the Parties to the Convention. Measurements of heavy metal concentrations in air and precipitation are carried out at the EMEP monitoring network under the methodological guidance of the Chemical Coordinating Centre (CCC). Along with that the Meteorological Synthesizing Centre – East (MSC-E) performs the model assessment of deposition and air concentrations of heavy metals over the EMEP region as well as the transboundary fluxes between the EMEP countries.

Heavy metals targeted by the Protocol include lead (Pb), cadmium (Cd) and mercury (Hg). These pollutants pose a significant risk to human health and the environment [e.g. *UNEP*, 2010a; *UNEP*, 2010b; *EEA*, 2011]:

- Lead is a pollutant that is toxic at very low exposure levels and has acute and chronic effects on human health. It is a multi-organ system toxicant that can cause neurological, cardiovascular, renal, gastrointestinal, hematological and reproductive effects. It also bioaccumulates and adversely impacts both terrestrial and aquatic ecosystems.

- Cadmium is a non-essential and toxic element for humans mainly affecting kidneys and the skeleton. It is also a carcinogen by inhalation. Important endpoints of cadmium include kidney and bone damage and cancer. In the environment, cadmium is toxic to plants, animals and micro-organisms.

- Mercury is toxic in multiple forms but the main concern is associated with the organic compounds, especially methylmercury. Mercury can damage the liver, kidneys and the digestive and respiratory systems. It also causes brain and neurological damage and impairs growth. It affects animals in the same way as humans and is very toxic to aquatic life.

The objective of this report is to support the revision of the 1998 Protocol on Heavy Metals with scientific information on long-term changes of heavy metal pollution levels within the scope of the EMEP region. The report has been prepared jointly by the Meteorological Synthesizing Centre – East

(MSC-E), the Chemical Co-ordinating Centre (CCC) and the Centre on Emission Inventories and Projections (CEIP) with contribution from the Coordination Centre for Effects (CCE).

Chapter 1 of the report was prepared jointly by CEIP and MSC-E and includes information on heavy metal emissions to the atmosphere and their changes over the period 1990-2010. Detailed data on emissions reporting by the Parties to the Convention, long-term emission trends in the EMEP countries based on the officially reported data, the key category analysis and emission uncertainties were prepared by CEIP. In addition, MSC-E characterized non-Party expert estimates used to fill gaps in the official data in preparation of heavy metal emission datasets for modelling. It also described emission data for mercury available on a global scale.

In Chapter 2 prepared by CCC the current state of development of the EMEP monitoring network for heavy metals is characterized along with its changes during the last two decades. Current levels and long-term trends of heavy metal concentrations in the ambient air and precipitation measured at the EMEP monitoring sites are described.

Chapter 3 written by MSC-E with contribution from CCE is focused on evaluation of long-term changes of heavy metal pollution levels in the EMEP countries. Trends of heavy metal deposition over the period 1990-2010 are analysed along with changes in transboundary fluxes between the countries. The key source categories of both emissions and deposition of heavy metals are evaluated. Besides, adverse effects of heavy metals on human health and ecosystems are characterised on the base of critical load exceedancies.

Other MSC-E on-going and research activities carried out in accordance with the EMEP Work-plan [ECE/EB.AIR/109/Add.2] are described in detail in the technical reports prepared by the Centre. In particular, progress in development of the GLEMOS multi-scale modelling system is reported in the joint MSC-E/MSC-W Technical Report on global modelling [*Jonson and Travnikov*, 2012]. Results of the heavy metal pollution assessment of the Czech Republic performed within the framework of the country-specific Case Study are presented in [*Ilyin et al.*, 2012]. Other aspects of model development including improvement of the heavy metal re-suspension scheme, elaboration of the adjoint modelling approach etc. are described in the EMEP/MSC-E Technical Report [*Shatalov et al,* 2012].

1. ATMOSPHERIC EMISSIONS OF HEAVY METALS

Parties to the LRTAP Convention should annually submit air pollution emission data (SOx, NOx, NMVOCs, NH_3, CO, HMs, POPs and PM) to the EMEP Centre on Emission Inventories and Projections (CEIP) and notify the LRTAP Convention Secretariat thereof. Parties are requested to report emission inventory data before 15 February using standard formats in accordance with the EMEP Reporting Guidelines [*UNECE*, 2009]. Original submissions from the Parties can be accessed via the CEIP homepage at http://www.ceip.at/overview-of-submissions-under-clrtap/2012-submissions.

1.1. Reporting of heavy metal emissions

Eight heavy metals are part of the annual reporting of emissions under CLRTAP, of which three (Cd, Hg and Pb) are covered by the Heavy Metal Protocol[1]. Completeness and consistency of heavy metal data submitted to UNECE/CEIP has improved slightly in recent years, and the number of Parties providing heavy metal emissions to UNECE/EMEP has increased since 2007 by about 12% (Fig.1.1).

In the year 2012, 45 out of the 51 Parties to the LRTAP Convention submitted inventories, of which seven (Georgia, Greece, Iceland, Liechtenstein, Luxembourg, Russian Federation and Turkey) did not report heavy metal emissions. Thirty-eight Parties to the CLRTAP provided information on the emissions and 21 Parties submitted the full time series from 1990 to 2010 for all 3 heavy metals. Of 29 Parties to the Protocol, three (Liechtenstein, Luxembourg and Romania) did not submit their base year (BY) emissions. Heavy metal emissions for 2010 were reported by 25 Parties to the Protocol whereas 4 Parties (Liechtenstein, Luxembourg, the Republic of Moldova and the USA) did not provide the required information to the UNECE secretariat (see Table 1.1). More information on the completeness and consistency of submitted data can be found in EEA & CEIP Inventory review report 2012.

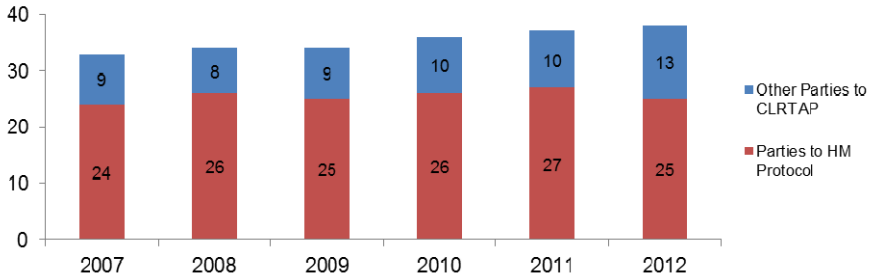


Fig. 1.1. *Number of Parties reporting heavy metal emission data to EMEP since 2007, status as of 31 May 2012*

[1]The Protocol on Heavy Metals (HM) was signed in 1998 and came into force in 2003.

Table 1.1. *Overview of emission reporting by Parties to the Protocol and differences between BY and 2010 emissions, status as of 31 May 2012*

			Cadmium (Mg)			Lead (Gg)			Mercury (Mg)		
Parties	Entry into force	Base year (BY)	Base year emissions	2010 emissions	Reduction to BY [%]	Base year emissions	2010 emissions	Reduction to BY [%]	Base year emissions	2010 emissions	Reduction to BY [%]
Austria (1985)	2004	1985	3.10	1.10	-65%	0.33	0.015	-95%	3.70	1.00	-73%
Belgium	2005	1990	7.20	2.70	-63%	0.49	0.043	-91%	6.80	2.10	-69%
Bulgaria	2003	1990	5.20	1.90	-63%	0.32	0.11	-67%	2.40	0.88	-63%
Canada	2003	1990	91.00	16.00	-82%	1.23	0.19	-85%	35.00	5.20	-85%
Croatia	2007	1990	1.30	0.58	-55%	0.54	0.028	-95%	1.50	0.75	-50%
Cyprus	2004	1990	0.05	0.07	45%	0.025	0.0026	-90%	0.15	0.13	-13%
Czech Republic	2003	1990	4.30	0.88	-80%	0.27	0.03	-90%	7.50	3.50	-53%
Denmark	2003	1990	1.00	0.19	-81%	0.13	0.01	-91%	3.10	0.44	-86%
Estonia	2006	1990	4.40	0.67	-85%	0.21	0.039	-81%	1.10	0.63	-43%
Finland	2003	1990	6.30	1.40	-78%	0.34	0.023	-93%	1.10	0.90	-18%
France	2003	1990	21.00	2.90	-86%	4.26	0.083	-98%	24.00	4.20	-83%
Germany	2003	1990	17.00	5.30	-69%	2.08	0.19	-91%	28.00	9.30	-67%
Hungary	2005	1990	5.50	0.71	-87%	0.66	0.017	-97%	6.30	0.78	-88%
Latvia	2005	1990	0.32	0.23	-28%	0.092	0.0082	-91%	0.24	0.08	-68%
Liechtenstein	2004	1990	not reproted	not reported		not reported	not reported		not reported	not reported	
Lithuania	2005	1990	3.80	0.43	-89%	0.047	0.0026	-94%	0.02	0.39	2067%
Luxembourg	2003	1990	NR	not reported		NR	not reported		NR	not reported	
Monaco (1992)	2004	1992	0.06	0.00	-93%	0.0041	0.000035	-99%	0.12	0.05	-60%
Netherlands	2003	1990	2.10	2.50	19%	0.34	0.044	-87%	3.50	0.69	-80%
Norway	2003	1990	1.20	0.59	-51%	0.19	0.0046	-98%	1.50	0.58	-61%
Republic of Moldova	2003	1990	2.40	not reported		0.25	not reported		3.40	not reported	
Romania (1989)	2003	1989	NE	2.20		NE	0.061		NE	5.30	
Slovakia	2003	1990	9.40	1.20	-87%	0.15	0.056	-63%	12.00	1.20	-90%
Slovenia	2004	1990	0.59	0.40	-32%	0.36	0.015	-96%	0.62	0.42	-32%
Sweden	2003	1990	2.30	0.58	-75%	0.36	0.013	-96%	1.60	0.55	-66%
Switzerland	2003	1990	3.90	1.30	-67%	0.35	0.023	-93%	6.70	1.10	-84%
United Kingdom	2005	1990	23.00	2.40	-90%	2.89	0.059	-98%	38.00	6.30	-83%
United States	2003	1990	180.00	not reported		2.996	not reported		187.00	not reported	
EU	2003	1990	258.00	103.00	-60%	23.156	2.55	-89%	230.00	87.00	-62%

Reporting of gridded heavy metal emissions

Irrespective of the reporting year, only 19 out of the 48 countries which are part of the extended EMEP area reported sectoral gridded emissions of heavy metals for 2000 and 2010. A slightly higher number of countries (21) submitted sectoral gridded heavy metal emissions for the year 2005. In 2012, 18 Parties submitted gridded sectoral emissions for 2010 (Fig. 1.2).

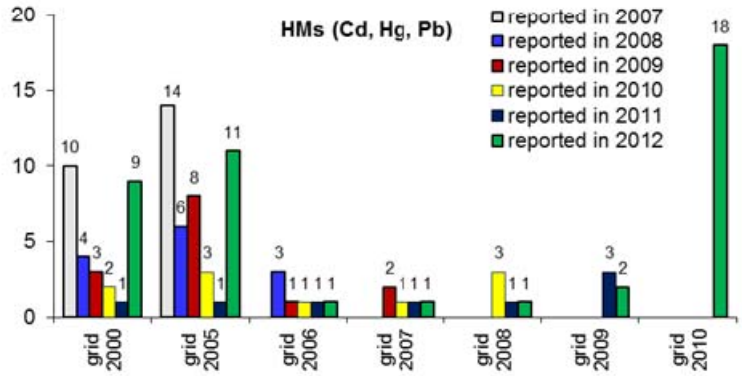


Fig. 1.2. *Number of Parties reporting gridded sectoral data to EMEP, status as of 31 May 2012*

Notes:

- 23 countries (out of 48[2]) did not report gridded sectoral heavy metal data, neither for 2005 nor for 2010 (Albania, Armenia, Azerbaijan, Bosnia and Herzegovina, Georgia, Greece, Iceland, Italy, Kazakhstan, Kyrgyzstan, Liechtenstein, Luxembourg, TFY Republic of Macedonia, Malta, Republic of Moldova, Monaco, Montenegro, Norway, Romania, Russian Federation, Serbia, Slovakia and Turkey).

- France, Hungary, Latvia, Lithuania, the United Kingdom and Switzerland did not provide gridded sectoral data for 2010.

- Belgium, Bulgaria, the Czech Republic and Poland did not provide gridded sectoral emissions for 2005.

1.2. Heavy metal emission trends

According to Article 3 of the Protocol, Parties should reduce their annual emissions of each substance from the levels in reference year (base year).

Complete information on trends for all countries and pollutants is available in the EMEP/CEIP public database WebDab[3] http://webdab1.umweltbundesamt.at/official_country_trend.html.

[2] The Parties EU, Canada and USA are not considered in this overview, as their emissions are not used in EMEP models.

[3] WebDab contains all emission data officially submitted to the secretariat of the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) by Parties to the Convention.

Cadmium

Compared with 1990, 28 countries reported lower cadmium emissions for 2010, while 3 countries reported higher 2010 emissions. In the EU and the USA, reported emissions declined by about 60% and in Canada by more than 80% (Fig. 1.3).

The highest emission reductions (around 90%) are reported by Monaco, United Kingdom and Lithuania (see Table A1 in Annex A). Increases in emissions were reported by Belarus (53%), Cyprus (45%) and the Netherlands (20%), although Cyprus and Netherlands are Parties to the Protocol.

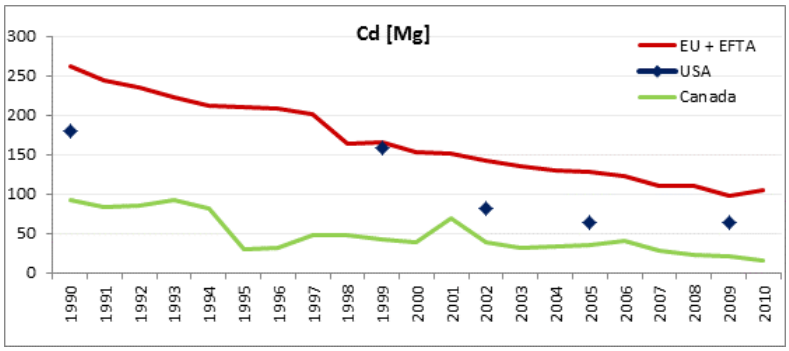


Fig. 1.3. *Cd emission trends of the EU+EFTA, USA and Canada, 1990-2010*

Lead

During the period 1990 and 2010, lead emissions decreased in all 31 countries which reported data for both years. Significant reductions of lead emissions were reported by the EU, Canada (more than 80%) and also the USA (almost 60%) (Fig. 1.4)).

In general, the reported lead reductions between 1990 and 2010 are significant in all countries. The most substantial emission decreases (more than 95 %) were reported by Monaco, France, the United Kingdom, Norway, Hungary, Sweden and Slovenia (see Table A2 in Annex A).

An opposite trend has been observed only in Serbia, where emissions increased from 44.3 Mg in 2000 to 147.7 Mg in 2010.

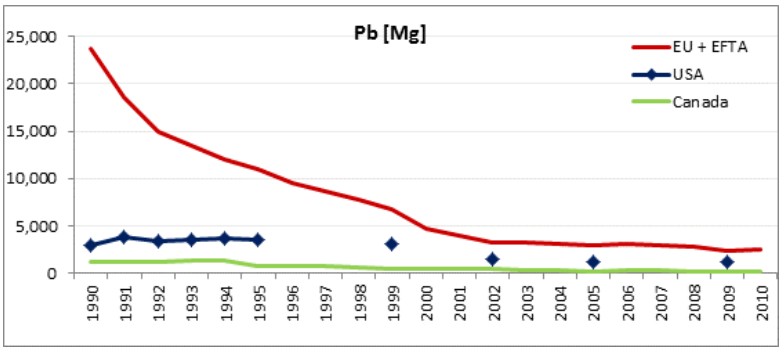


Fig. 1.4. *Pb emission trends for the EU, EFTA, USA and Canada, 1990-2010*

Mercury

Compared with 1990, mercury emissions decreased in 2010 in 29 countries and increased in 2 countries. The most substantial decreases were reported by Slovakia (91%), Hungary (88%) and Denmark (86%). Higher emissions (compared to 1990) in 2010 were reported by the FYR of Macedonia (65%) and Lithuania (2050%) (see Table A3 in Annex A). The low emissions reported by Lithuania for the years 1990 to 1995 might indicate an error in the estimates and should be checked by the Party.

The observed decline of mercury emissions in Canada from 1990 to 2010 is 85%, in the EU+EFTA 63% and in the USA about 45% (see Fig. 1.5).

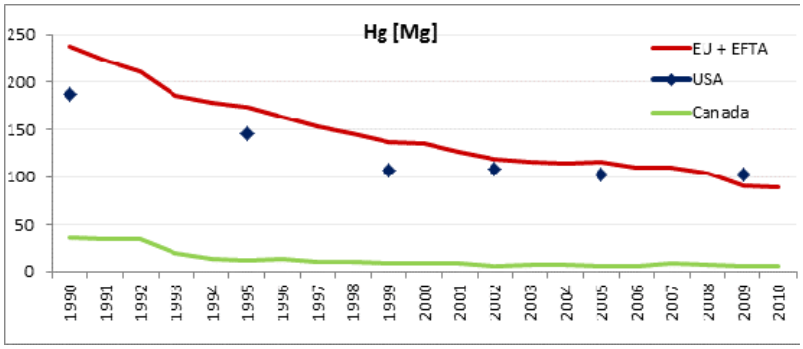


Fig. 1.5. *Hg emission trends in the EU+ EFTA, USA and Canada, 1990-2010*

1.3. Key category analysis

Key category analysis (KCA) can help to identify most important emission sources for individual air pollutants[4]. Following the revised EMEP/EEA Air Pollutant Emission Inventory Guidebook [*EEA/EMEP*, 2009], key categories are those which, when summed up in descending order of magnitude, cumulatively add up to 80% of the total level.

For the KCA Parties are divided into 2 groups: a) “EU-27, EFTA, Croatia and Macedonia” and b) “Other” countries.

Canada and the USA cannot be included in the KCA while their emissions are not provided in NFR categories.

Three emission categories were identified as being significant for both groups and for all three heavy metals (Cd, Pb and Hg), namely:

“1A1a Public Electricity and Heat Production”,

“1A fi Stationary Combustion in Manufacturing Industries and Construction: Other” and

“2C1 Iron and Steel Production”.

[4] A key category is one that has a significant influence on a country’s total inventory in terms of absolute levels of emissions, the trend in emissions, or both.

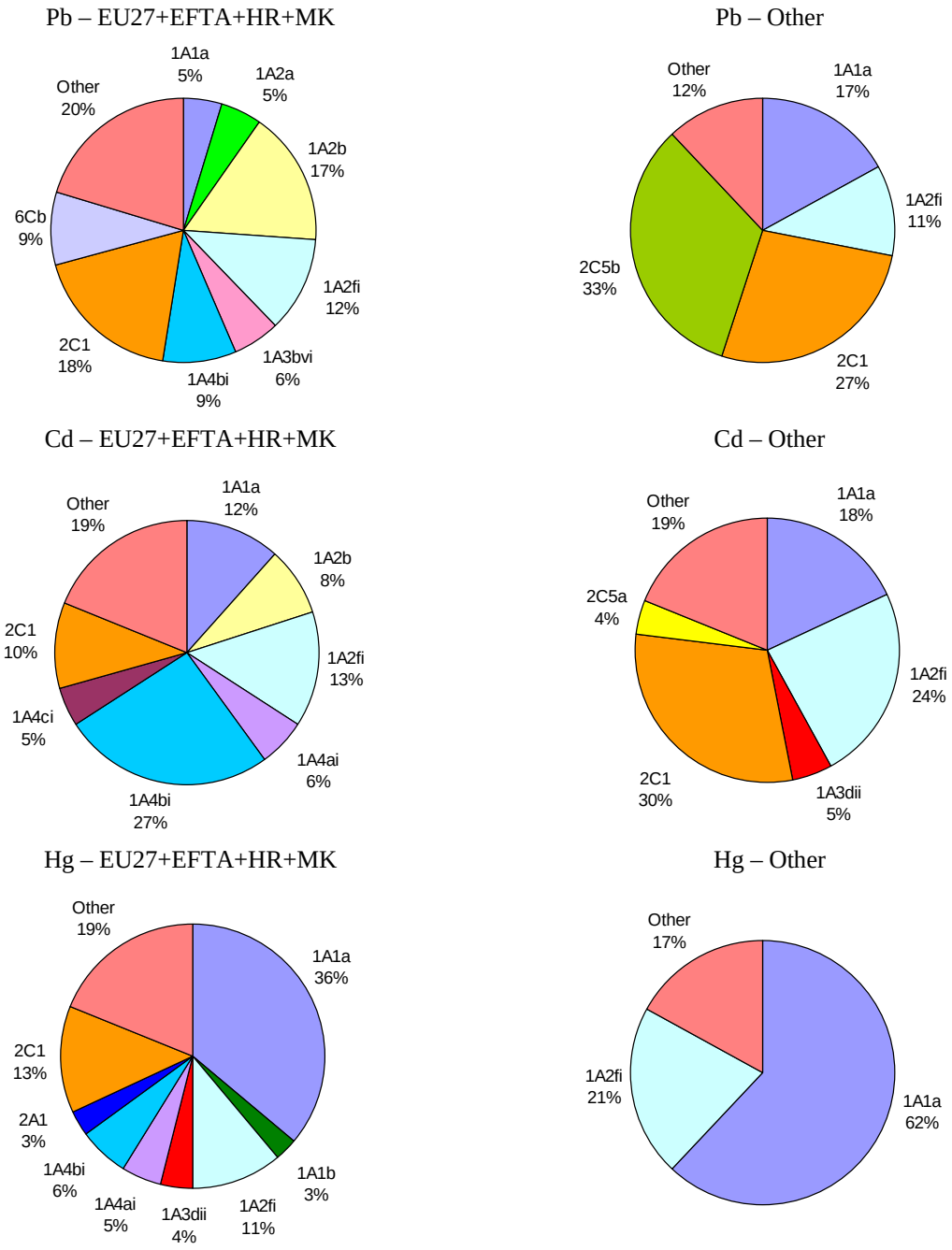


***Fig. 1.6**: Key category analysis; heavy metals (Cd, Pb, Hg) 2010*

Notes:

The "Other Cat" in Fig.1.6 is the sum of the remaining (non-key) categories.

Parties might allocate emissions to NFR categories in a different way. Some Parties make use of the emission inventory notation key IE (included elsewhere) or allocate emissions to the "Other" (sub)category, which means that emissions occurring in one NFR sector are reported in emission estimates of a different sector. Frequent use of IE notation key can influence the results of KCA.

In general, more key categories have been identified for the first group of countries "EU-27, EFTA, Croatia and Macedonia". A limited number of identified key categories in the "Other" group seem to

indicate incomplete reporting of heavy metals and frequent reporting of notation keys like IE, NA, NE[5] instead of numbers. Particularly significant sources of the emissions like “1A2b Stationary Combustion in Manufacturing Industries and Construction: Non-ferrous Metals” and “1A4bi Residential: Stationary plants” are - surprisingly - no key categories in “Other” countries.

The most significant differences between these 2 groups are observed in the reporting of mercury emissions, where more than 80% of the emissions in “Other” countries are reported only in 2 categories “1A1a Public Electricity and Heat Production” and “1A2fi Stationary Combustion in Manufacturing Industries and Construction”, compared to 8 key categories identified in the group “EU-27, EFTA, Croatia and Macedonia”.

1.4. Emission uncertainties

It is very difficult to quantify the uncertainty of reported emissions, as countries do not usually provide information on the uncertainties of estimates. Changes in the reporting of the 2005 emissions in subsequent years are therefore regarded as an indicator of uncertainty. The tables in Annex B show variations of 2005 heavy metal emissions as reported between 2007 and 2012. The last column shows the range of reported values. The fluctuations of reported 2005 data during the last six years indicate a relatively high level of uncertainty for all heavy metal emissions.

Fig. 1.7, 1.8 and 1.9 illustrate the variations observed in the 2005 emissions reported for individual countries, with 0% corresponding to the latest available 2005 emissions as reported in 2012, and the red bars indicating the difference to emissions reported in previous years. Minus values indicate that the 2005 emissions reported in 2012 are higher than the value reported in previous years. Reported 2005 emissions show variations exceeding a few orders of magnitude for all 3 heavy metals (Cd, Pb and Hg). Such major differences may indicate errors or incomplete data in some submissions.

No deviation from the value reported in 2012 does not necessarily mean accurate 2005 emissions; this rather implicates that there is only one submission for 2005 data from this Party, i.e. that the Party has not updated its historical emissions as recommended by the EMEP Reporting Guidelines.

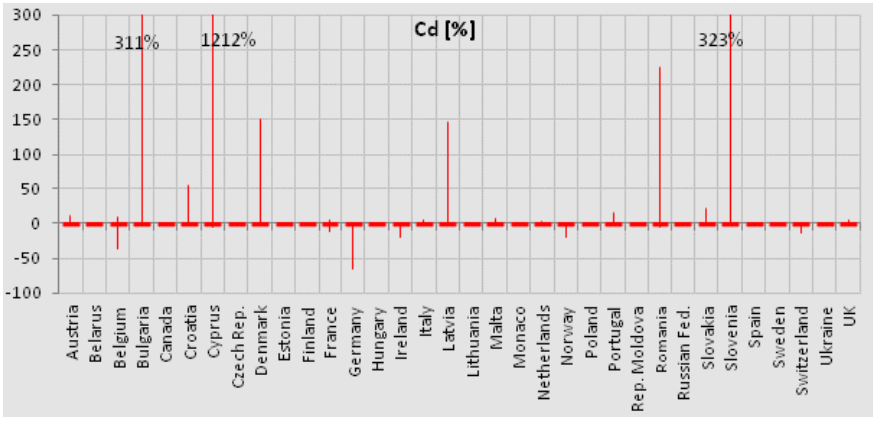


Fig. 1.7. *Fluctuations in 2005 Cd emissions as reported between 2007 and 2012 (0% corresponds to data reported in 2012)*

[5] Notation keys IE -included elsewhere, NA -not applicable, and NE -not estimated are defined in the EMEP reporting guidelines 2009

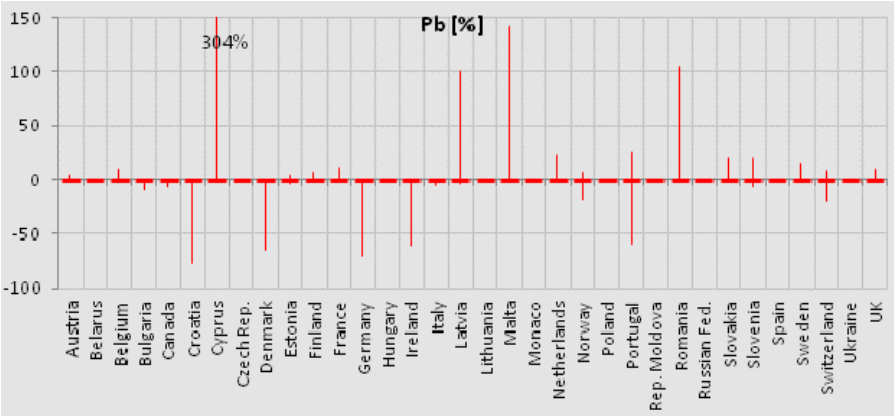


Fig. 1.8. *Fluctuations in 2005 Pb emissions as reported between 2007 and 2012 (0% corresponds to data reported in 2012)*

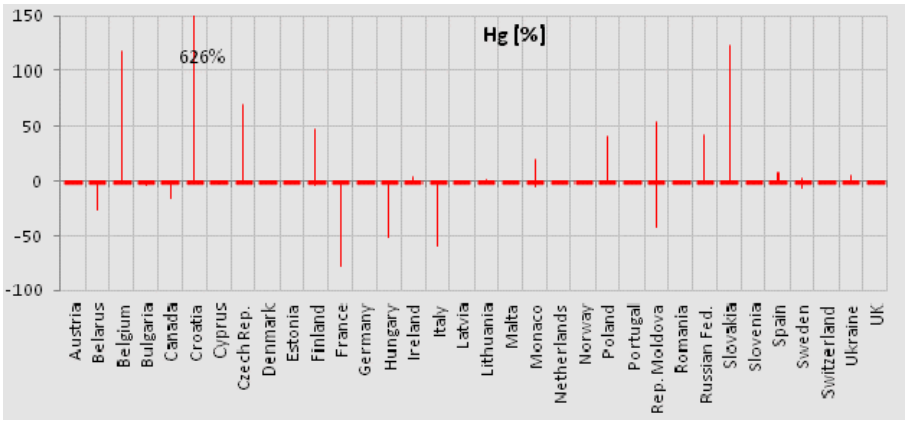


Fig. 1.9. *Fluctuations of 2005 Hg emissions as reported between 2007 and 2012 (0% corresponds to data reported in 2012)*

Notes:

- The figures include only Parties which reported 2005 heavy metal emissions for at least one year.
- Minus values in figures indicate that 2005 emissions reported in 2012 are higher than the value reported in previous years; plus values indicate that the 2005 emissions reported in 2012 are lower than the ones reported in previous years.
- No variations indicate that emissions have not been updated.

1.5. Emission data for model assessment

The data on emission totals from the EMEP countries for the period from 1990 to 2010 used for modelling were based on the official data received from the EMEP Centre on Emission Inventories and Projections (CEIP) [http://www.emep-emissions.at/ceip/]. If countries did not report their national emission data, emission totals were taken from non-Party emission estimates made by TNO [*Berdowski* et al. 1997, *Denier van der Gon et al.*, 2005]. Information about spatial distribution of heavy metal emissions at least for one year of the period 1990-2010 was provided by 27 countries (Austria,

Belarus, Belgium, Bulgaria, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Ireland, Italy, Latvia, Lithuania, the Netherlands, Norway, Poland, Portugal, Slovakia, Slovenia, Spain, Sweden, Switzerland and the United Kingdom). Gridded emissions for the period from 1990 to 2010 were prepared by CEIP and MSC-E for the EMEP countries with spatial resolution 50×50 km^2.

The official information on emissions for the Asian part of the EMEP domain was not available. Therefore, emission data for this region were based on non-Party emission estimates. Lead emissions in Kazakhstan, Kyrgyzstan, Turkmenistan, Tajikistan and Uzbekistan for 1990 were taken from the global inventory for 1990 [*Pacyna et al.*, 1995; *http://www.ortech.ca/cgeic/index.html*] and for 1995, 2000, 2005 and 2010 were estimated expecting the same emission reduction in these countries as in the Russian Federation according to the recent EMEP official data. Total emissions of lead from the Asian part of Russia were assessed using the official emission data for the European part of the country in 1990, 1995, 2000, 2005 and 2010 and keeping the ratio between the European and the Asian parts obtained from the global lead inventory. Besides, the global emission data were also used for the other Asian and African countries, falling fully or partly into the EMEP domain, assuming the same emission change between 1990 and 2010 as for Turkey. Turkey was selected for this purpose because it was the only country for which the non-Party estimates of lead emission changes were available. Spatial distribution of lead emissions from all these countries was obtained by interpolation of the global gridded emissions with 1°×1° spatial resolution into the model grid.

Mercury emissions for the Asian part of the EMEP domain and for the northern African countries were derived from global mercury inventories for 1990 [*AMAP*, 1998], 1995 and 2000 [*Pacyna and Pacyna*, 2002; *AMAP*, 2005] and 2005 [*AMAP/UNEP*, 2008; *AMAP*, 2011]. It was assumed that the emissions were not changed significantly between 2005 and 2010.

Global emission inventories for cadmium are currently not available. That is why the cadmium emission data for the Asian part of the EMEP domain and for the north of Africa were obtained on the basis of the global mercury inventories [*AMAP*, 1998; *Pacyna and Pacyna*, 2002; *AMAP*, 2005; *AMAP/UNEP*, 2008; *AMAP*, 2011]. For this purpose, cadmium emission was assumed to be proportional to emission of mercury with a coefficient depending on a region: $E_{Cd} = \alpha \cdot E_{Hg}$. For the eastern part of Russia the proportionality coefficient (α) was taken the same as for the European part of the country (1.14). The coefficient for the remaining Central Asian countries was assumed to be the same as that for Kyrgyzstan (0.56). For the other Asian countries and Africa the coefficient was taken equal to that for Turkey (0.91). All coefficients were estimated on the basis of the TNO inventory [*Denier van der Gon et al.*, 2005].

In 2010 total anthropogenic emission of lead from the EMEP countries made up around 2965 tonnes (Fig. 1.10a). Lead emissions decreased by 31601 tonnes over the period 1990-2010 that corresponded to around 90% of total emission in 1990. This decrease is attributed to the transport sector (1A3), where Pb emissions dropped down due to the removal of lead from gasoline.

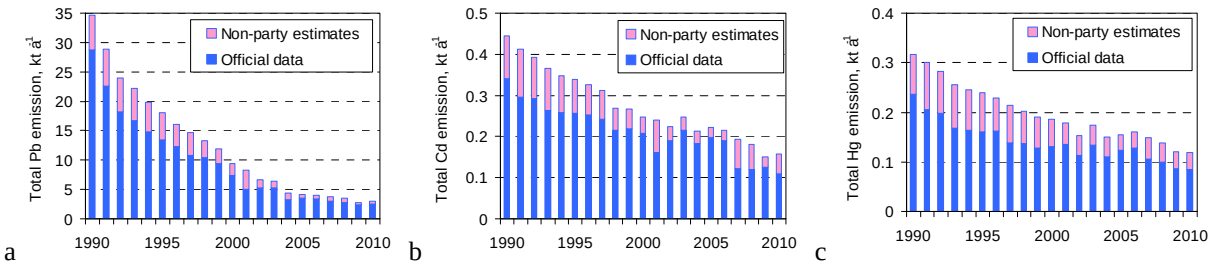


Fig. 1.10. *Trend emissions of lead (a), cadmium (b) and mercury (c) in the EMEP countries, 1990-2010.*

Total emission of cadmium in the EMEP countries in 2010 was 157 tonnes, which is 287 tonnes (65%) lower than in 1990, as a result of emission decrease in Metal production (2C), Stationary combustion in industry (1A2) and Public electricity and heat production (1A1a) (Fig. 1.10b).

Emissions of mercury in the EMEP countries decreased from 317 tonnes in 1990 to 120 tonnes in 2010, with reduction of 62% (Fig. 1.10c). It was occurred mainly due to the reductions in the Public electricity and heat production (1A1a) and Stationary combustion in industry (1A2), and also Metal production (2C) and Waste incineration (6C).

According to the non-Party emission estimates, heavy metal emissions in Kazakhstan, Kyrgyzstan, Turkmenistan, Tajikistan, Uzbekistan and the eastern part of Russia decreased by 90% (Pb), 56% (Cd) and 48% (Hg) in the period 1990-2010 and made up around 1000 tonnes (Pb), 40 tonnes (Cd) and 50 tonnes (Hg) in 2010.

Spatial distributions of heavy metal emissions used in the modelling in 1990 and 2010 are shown in Fig. 1.11.

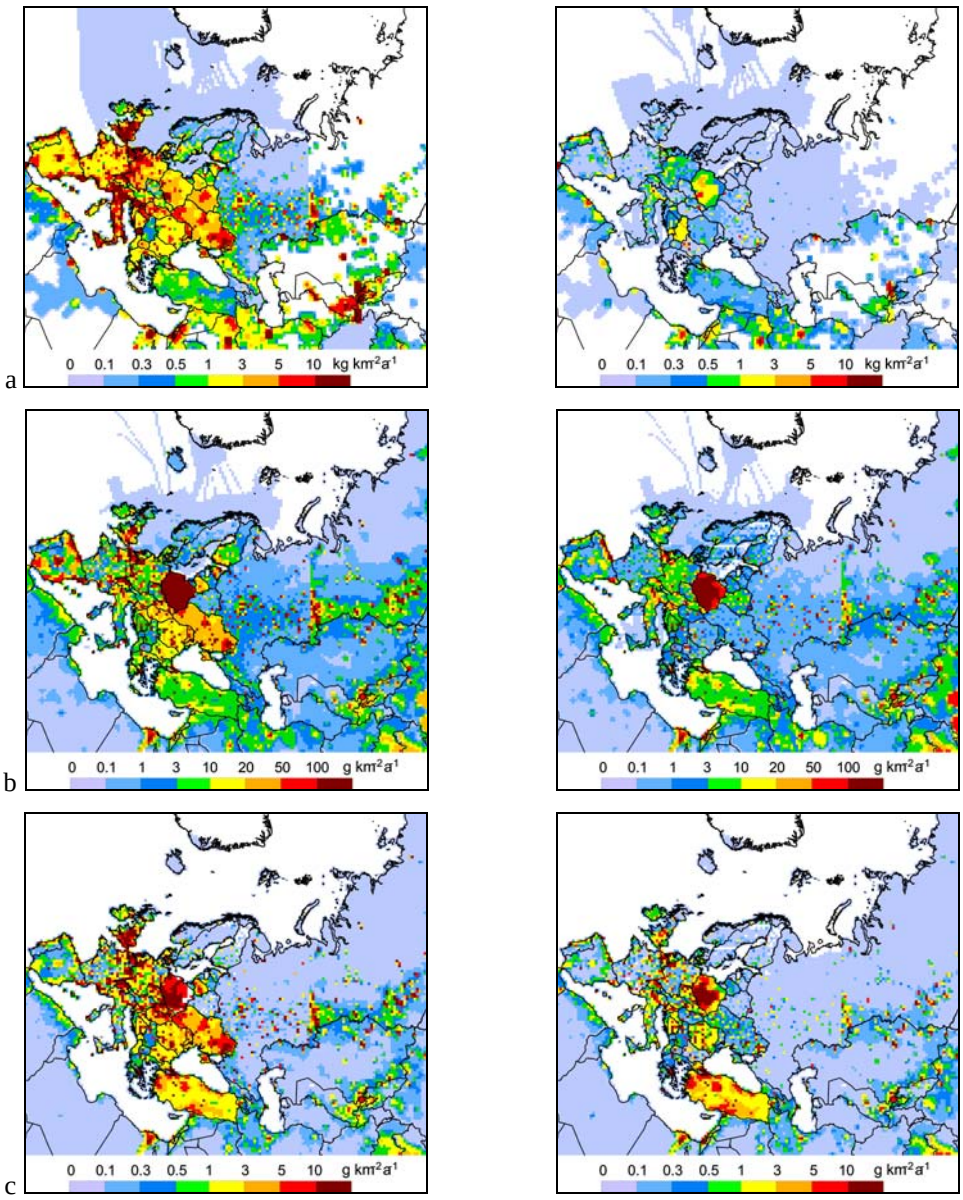


Fig. 1.11. *Spatial distribution of lead (a), cadmium (b) and mercury (c) emissions over the EMEP domain in 1990 (left) and 2010 (right).*

1.6. Mercury emissions on a global scale

The global inventories of mercury anthropogenic emissions to the atmosphere were produced for the years 1990 [*AMAP*, 1998] and 1995 and 2000 [*Pacyna and Pacyna*, 2002; *AMAP*, 2005]. Most recently, an inventory of the global anthropogenic Hg emissions for 2005 (the '2005 v5' inventory) was prepared in a joint AMAP/UNEP project in 2008. Details on the methods, data sources and other information are reported by *AMAP/UNEP* [2008] and *Pacyna et al.* [2010]. Further work on the 2005 inventory was undertaken as part of the AMAP assessment [*AMAP*, 2011], resulting in the '2005 v6' inventory with revised estimates of total emissions of mercury to air in 1990, 1995, 2000 and 2005. Figure 1.12 presents the global distribution of anthropogenic emissions of mercury in 2005. Areas with elevated mercury emissions correspond to highly industrialized regions: Europe, Eastern United States, South-eastern China, Japan, India and South Africa.

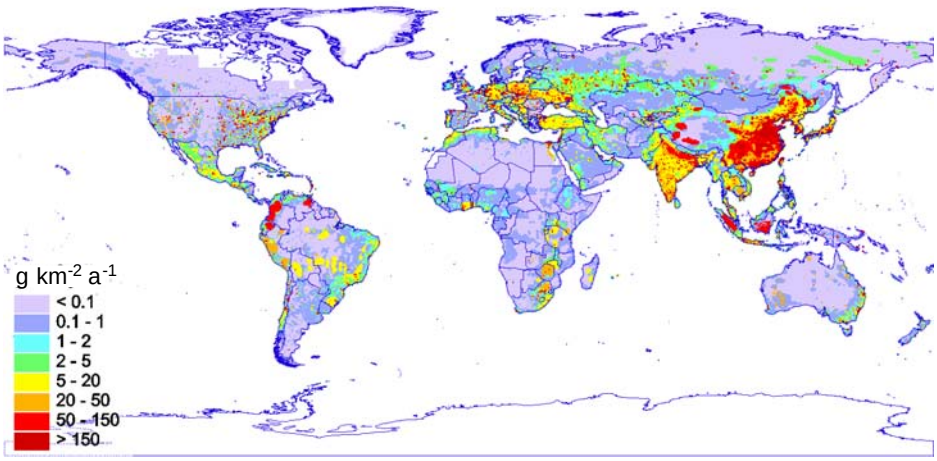


Fig. 1.12. *Global distribution of anthropogenic emissions of mercury in 2005*

The global mercury inventory in 2005 comprises of atmospheric emissions from by-product sectors, product use, cremation and artisanal mining and amounts to 1920 tonnes in total. The largest emissions of mercury to the global atmosphere from the by-product sectors occur from combustion of fossil fuels (mainly coal) in power plants, industrial and residential boilers (880 tonnes), metal production (200 tonnes) and cement production (190 tonnes). Figure 1.13 summarizes contribution of various anthropogenic activities to the global mercury emission.

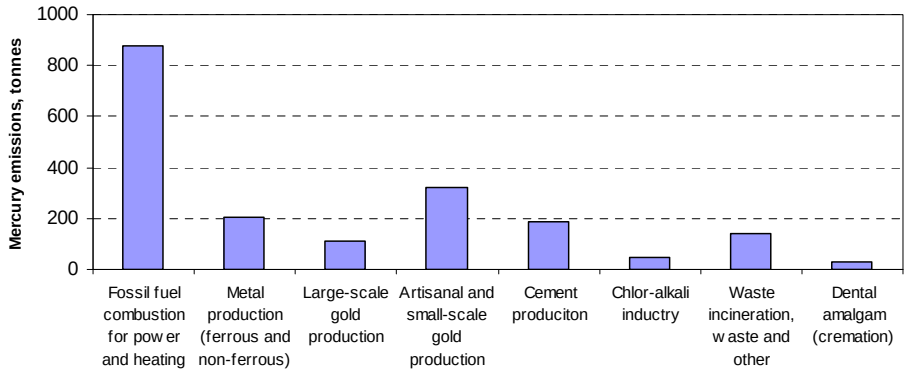


Fig. 1.13. *Global anthropogenic emissions of mercury to air in 2005 from various sectors*

Contribution of different continents and regions to mercury emission in 2005 on a global scale is shown in Fig. 1.14. As seen Asian emission sources contribute about 65% to the global mercury emissions whereas the contribution of European and North American sources does not exceeded 20% in sum.

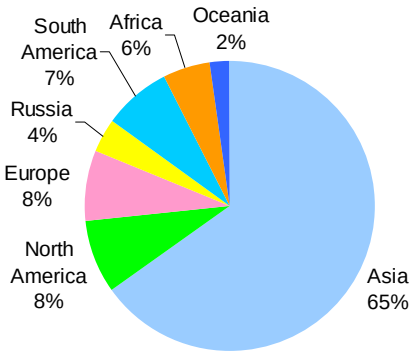


Fig. 1.14. *Contribution of different continents and regions to the global anthropogenic emissions of Hg in 2005*

Combustion of fuels for production of electricity and heat is the largest source of anthropogenic mercury emission in Europe, North America, Asia and Russia, and is responsible for about 35-50% of the anthropogenic emissions in Oceania and Africa. However, in South America, the artisanal and small-scale gold mining (ASGM) is responsible for the largest part of the continent emission (about 60%). The ASGM emissions in some Asian countries as well as in several countries in South America account for the fact that such countries as Indonesia, Brazil and Colombia appear in the top ten ranked mercury emitting countries. China, with its more than 2000 coal-fired power plants, is the largest single emitter of mercury worldwide. Together, three countries, China, India and the United States are responsible for about 60% of the global mercury emission in 2005 (1095 out of 1920 tonnes).

Comparison of total anthropogenic mercury emissions in different continents and for different reference years is presented in Fig. 1.15. As seen total mercury emission in Asia in 1990 was 2-3 times higher than that in Europe, Russia and North America. During the period 1990-2005 Hg emissions decreased in Europe, Russia and North America but significantly increased in Asia. Some increase also took place in Africa and South America.

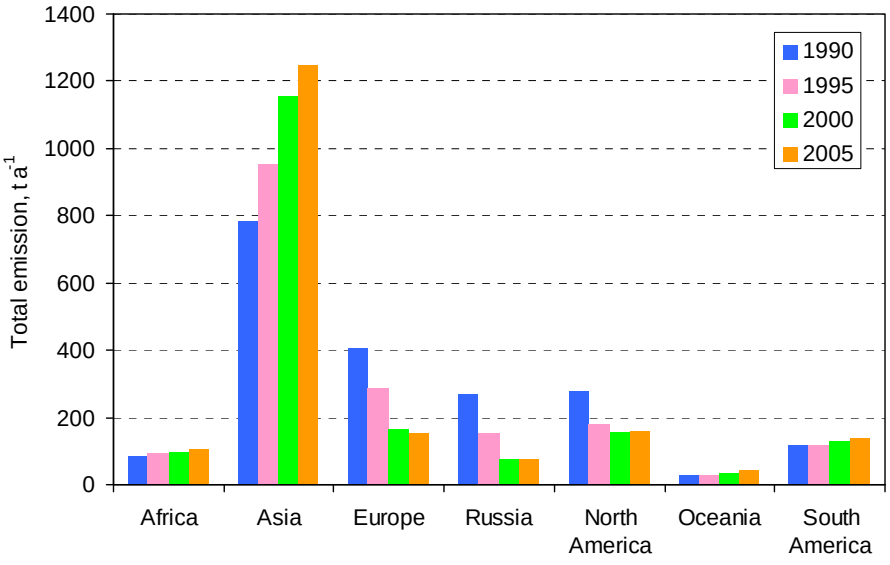


Fig. 1.15. *Change of global anthropogenic emissions of total mercury to the atmosphere from 1990-2005*

2. EMEP MONITORING NETWORK FOR HEAVY METALS

2.1. 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 [*http://ebas.nilu.no*] 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.

Detailed information about the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [*Aas and Breivik*, 2012]. The EMEP monitoring network has evolved substantially during the period 1990-2010. In 1990 there were 17 sites measuring heavy metals, though no mercury measurements. In 2010, there were 66 measurement sites among which 31 sites did measurements of heavy metals both in air and precipitation. Monitoring of mercury was performed at 31 sites at least for one form of mercury, but not all of the sites do have complete datasets for the whole year. 11 sites were measuring mercury both in air and precipitation, though only 9 of these sites measured gaseous mercury (in difference from particulate mercury) and fulfilled the monitoring obligations.

Heavy metals didn't become part of the official EMEP monitoring programme before 1999. There was no any EMEP reference method the first years and one should therefore expect poorer comparability between the measurements in the first decade. The first laboratory intercomparison was conducted in 1995 and from 1999 it has been performed annually. One can expect that the laboratories have improved during the years due to the various laboratory and field intercomparisons which have been initiated under EMEP. The increased number of laboratories/sites does have an effect on the overall uncertainty in the heavy metals measurements in EMEP. There are commonly higher uncertainties in the measurements of the first years. Furthermore, since the concentration level of heavy metals have decreased substantially these two decades there is a need for more sensitive methods to measure now compared to 1990. There seems to be an increase of measurements below detection limit and this induce a higher uncertainty in the estimated average concentrations.

There is a need for better coverage of especially mercury in large part of the EMEP domain. It should be noted however that the measurement obligations set by the EMEP monitoring strategy [*UNECE,* 2009] and the EU air quality directives [*EU,* 2004, 2008] have clearly improved the site coverage during the last years.

2.2. Observed concentration levels of lead, cadmium and mercury in 2010

Annual averages of Pb, Cd and Hg concentrations in precipitation and in air in 2010 are presented in Fig. 2.1-2.6. Note that Cyprus with measurements of heavy metals in air is outside the map domain so included as a dislocated point south of Turkey. The lowest concentrations for all elements in air as well as precipitation are generally 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, i.e. in the Benelux countries for lead and cadmium in air. The new site in Romania show high concentrations indicating the importance of getting more sites with continuous measurements in this region The spatial distribution of elemental mercury in air does not follow a general pattern; with almost the same concentration in Germany compared to the Arctic. In precipitation there are several sites (in Portugal, Latvia and Ireland) with high detection limits and these are not included in the map, for those included the highest level is seen in Sweden and Belgium.

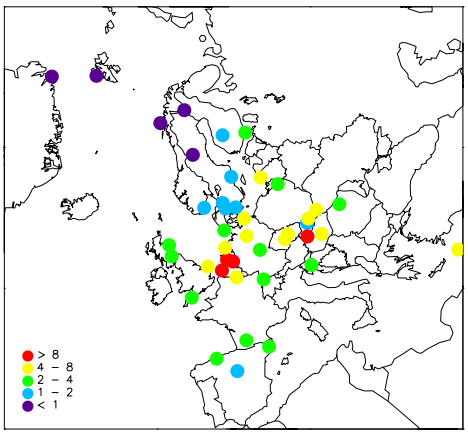


Fig. 2.1. *Pb in aerosol, ng/m^3*

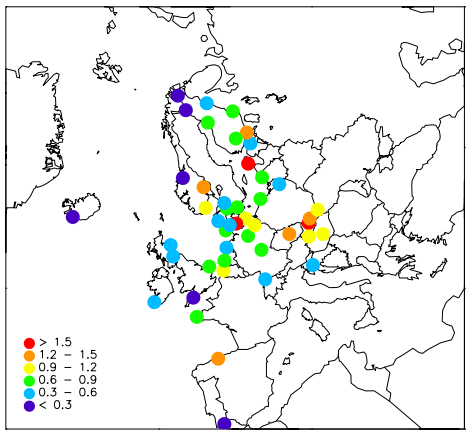


Fig. 2.2. *Pb in precipitation*

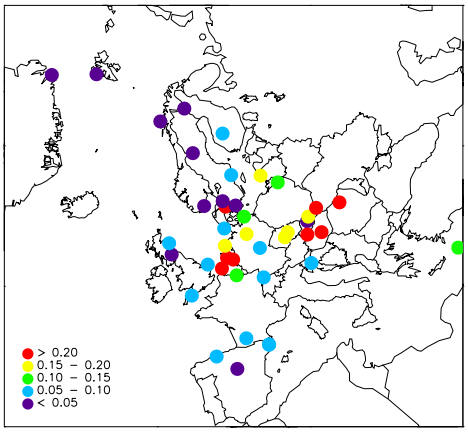


Fig. 2.3*. Cd in aerosol, ng/m^3*

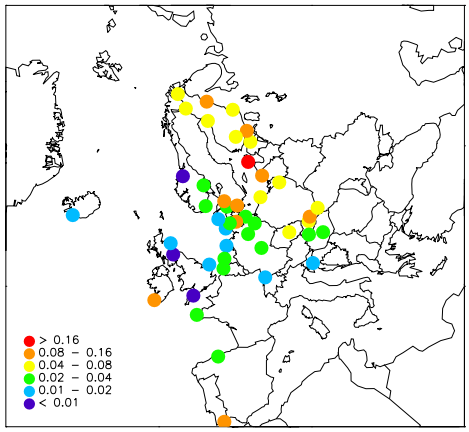


Fig. 2.4. *Cd in precipitation, µg/L*

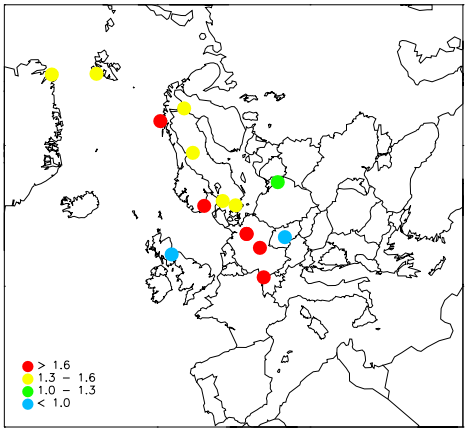


Fig. 2.5*. Hg (g) in air, ng/m^3*

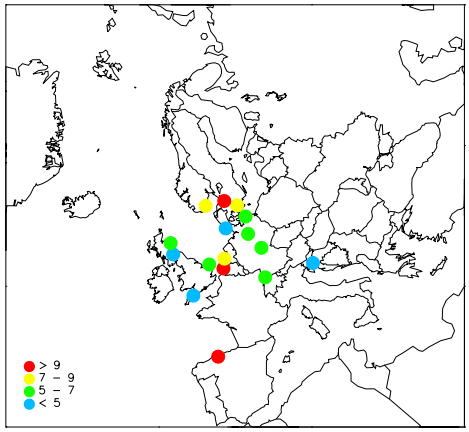


Fig. 2.6. *Hg in precipitation, ng/L*

3.3. Long-term observations of heavy metals in Europe

The time series of lead and cadmium from 1990 and 2010 at selected EMEP sites are presented in Fig. 2.7, and these show clearly a reduction in both elements during these two decades. Trend analysis for 1990-2009 for the EMEP sites have been recently done by *Tørseth et al.* [2012]. The reductions in measured lead are between 80% (in precipitation) and 90% (in aerosols) at the 11 (aerosols) and 7 (precipitation) sites with sufficient data capture for this period. The emission reduction

is in the same range (84%), mainly due to the use of unleaded gasoline and the introduction of efficient dust-removal installations in industrial plants (Pacyna et al. 2009). The reduction in measured cadmium is also significant, between 64 % (in precipitation) and 84% (in aerosols) at 8 and 9 sites respectively. This is also in line with the reported emission reductions (54%). One should keep in mind that the sites with long term measurements are situated in central and Northern Europe, and that their average decrease may be higher than for the EMEP domain as a whole. A major decline of the European Hg emissions occurred at the end of the 1980s and around 1990 [*Pacyna et al.,* 2009], but there are no major changes since 1990 [*Tørseth et al.* 2012].

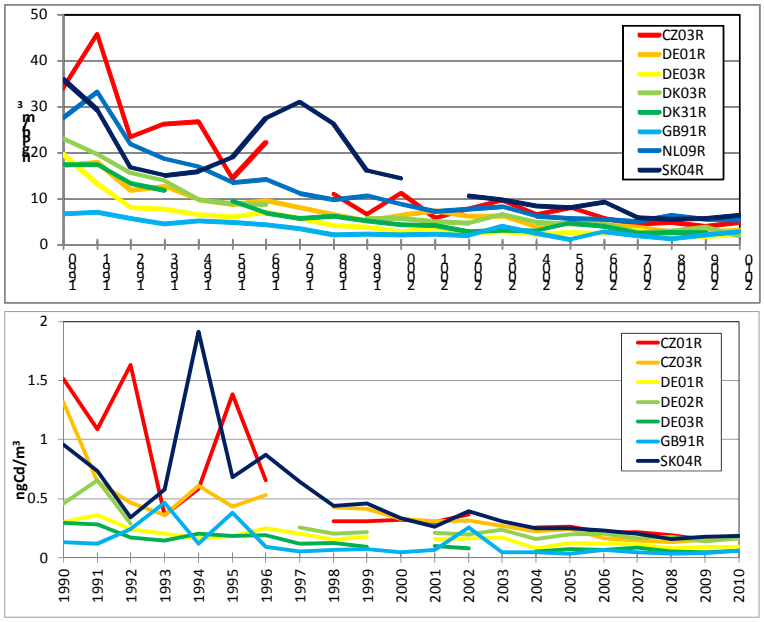


Fig. 2.7*. Annual average concentration of lead (top) and cadmium (bottom) in aerosols from 1990-2010 at selected EMEP sites.*

3. LONG-TERM CHANGES OF HEAVY METAL POLLUTION (1990-2010)

Assessment of long-term changes of heavy metal pollution levels in the EMEP countries over the period from 1990 to 2010 has been done based on both measurement data from the EMEP monitoring network and simulation results of the MSCE-HM model. Integrated analysis of temporal trends includes comparative evaluation of changes in atmospheric deposition of lead, cadmium and mercury during the considered period with regard to emission changes in different parts of Europe.

3.1. Trends of heavy metal deposition in the EMEP countries

Atmospheric deposition is one of the major routes of human and ecosystems exposure to heavy metals through contamination of soils, crops and aquatic organisms. Total deposition from the atmosphere consists of wet and dry components due to precipitation scavenging and uptake by the surface, respectively. Since monitoring data characterize only wet component of deposition, total atmospheric load can only be evaluated with modelling results. Long-term changes of lead, cadmium and mercury deposition in the EMEP countries are discussed below.

Lead

Total emission of lead in the EMEP countries was reduced by more than 90% since 1990 until 2010 (Fig. 3.1a) and the largest reduction took place in the first half of the period. Atmospheric deposition followed these changes to some extent and decreased on average by 75% (Fig. 3.1b). Area mean deposition flux over the territory of the EMEP countries changed from 3.3 kg km^{-2} a^{-1} in 1990 to 0.8 kg km^{-2} a^{-1} in 2010. However, lead deposition exhibits large variation altering within the range 0.2-8.2 kg km^{-2} a^{-1} in 1990 and within 0.1-1.8 kg km^{-2} a^{-1} in 2010. Thus, the upper deposition levels have decreased more significantly (by 78%) as they commonly take place in the vicinity of emission sources and they are largely affected by reduction of anthropogenic emissions.

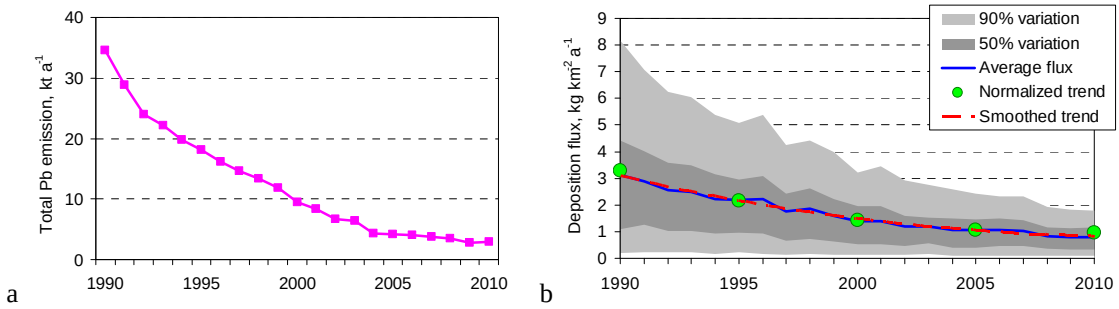


Fig. 3.1. *Temporal trends of Pb anthropogenic emissions (a) and deposition flux (b) in the EMEP countries. Deposition flux: blue solid line presents area weighted average deposition flux; dashed red line is smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the whole territory, respectively.*

Simulated reduction of lead pollution levels is confirmed by long-term measurements at the EMEP monitoring sites. Figure 3.2 shows simulated and observed trends of lead air concentration and wet deposition over the period 1990-2010 along with changes of anthropogenic emissions. The emission trends present sum of anthropogenic emissions from countries mostly influencing lead concentration and deposition at the considered sites. As seen the model successfully reproduces both the observed

levels and long-tern changes of these parameters but does not follow the emission changes particularly in the second half of the period. Model and observations either exhibit smaller reduction over the whole period or demonstrate significant delay in comparison with the emission change.

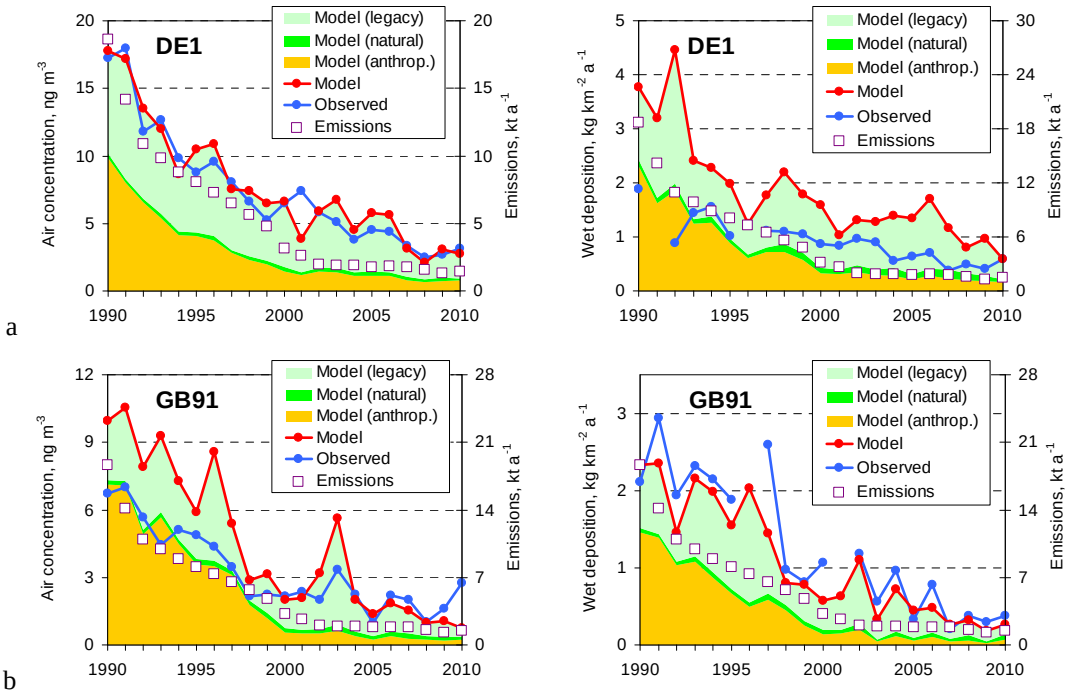


Fig. 3.2. *Simulated and observed trends of air concentration and wet deposition of lead at some EMEP monitoring sites: (a) - DE1, Germany; (b) – GB01, the United Kingdom. Modelling results show contribution of anthropogenic emissions as well as natural and legacy component of wind re-suspension. The emission trends present sum of national emissions over countries contributing more than 90% of deposition at the considered sites.*

These inconsistencies are likely explained by missed emission sources. These can include unaccounted or underestimated anthropogenic sources or emissions due to natural processes such as wind re-suspension of previous atmospheric deposition, or both. An additional argument supporting this assumption is significant underestimation (by a factor of 2) of measured air concentrations and deposition fluxes by modelling results obtained by different models when only anthropogenic emission data are used for simulations [*Ilyin et al.*, 2007a]. To account for possible contribution of natural processes to lead air concentration and deposition a model of wind re-suspension of heavy metals accumulated in soil and roadside dust as well as in seawater was developed and used for simulation of heavy metal pollution [*Gusev et al.*, 2006; *Ilyin et al.*, 2007b]. It is largely based on approaches applied in wind erosion and dust suspension modelling [*Marticorena and Bergametti*, 1995; *Alfaro and Gomes*, 2001; *Gomes et al.*, 2003].

As seen from Fig. 3.2 contribution of anthropogenic emissions to lead air concentration and deposition drops rapidly in the first half of the period following the emission trend. Contribution of wind re-suspension is divided in two components. The first component presents re-suspension of mineral dust with natural content of lead that corresponds to average lead concentration in the Earth' crust. The second component accounts for the legacy contribution of lead accumulated in soil and roadside dust due to previous atmospheric deposition. The legacy component significantly exceeds the natural one that is in general agreement with measurements of topsoil and dust enrichment with heavy metals. Relative contribution of re-suspension increases by the end of the period due to rapid reduction of anthropogenic emissions. It should be noted that due to large uncertainties of the top soil and dust

enrichment factors estimates of the legacy contribution also contain significant uncertainties. Therefore, taking into account that the model successfully reproduces observed levels, this component can indirectly fill the gap in emissions from unaccounted anthropogenic sources.

Spatial changes of lead deposition over the EMEP domain between 1990 and 2010 are shown in Fig. 3.3. The largest deposition decrease occurred over the British Isles, southern Spain, countries of Central Europe, southern Scandinavia and the western part of the Russian Federation. Elevated deposition levels remain in northern France, the Benelux countries, western Germany, northern Italy, southern Poland and eastern Ukraine. Considerable deposition fluxes are also over the Balkan countries. Relatively high deposition over the Caspian Sea and southern Russia was caused by wind re-suspension from desert areas of Central Asia.

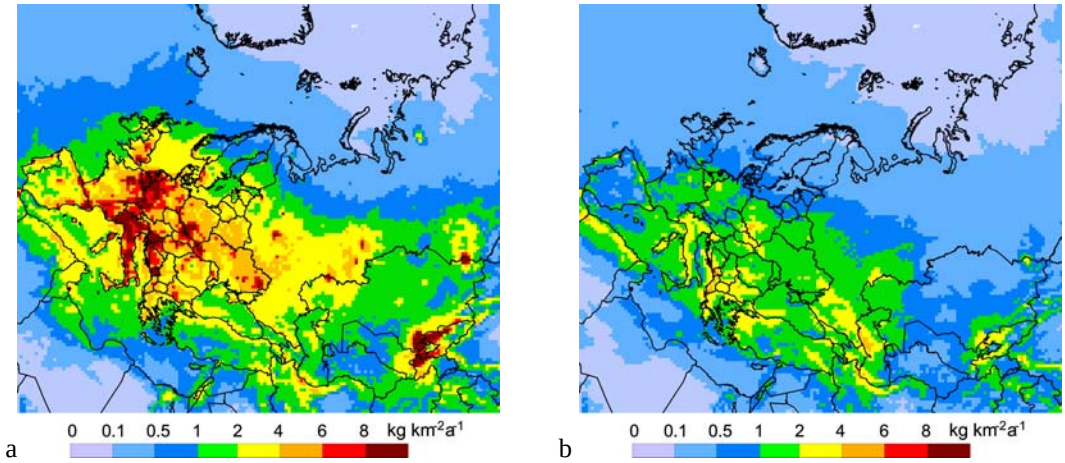


Fig. 3.3. *Spatial distribution of annual Pb deposition flux over Europe in 1990 (a) and 2010 (b)*

Relative changes of lead deposition in individual countries of Europe and Central Asia over the period from 1990 to 2010 are shown in Fig. 3.4. The whole period is divided into four 5-years long sub-periods to analyse dynamics of the changes. This diagram (as well as similar diagrams for other metals) is based on the normalized temporal trend estimates instead of real deposition levels for each particular year. The trend normalizing procedure is aimed at exclusion of the meteorological variability from estimates of deposition changes and revealing the primary influence of emission reduction. Description of the applied normalizing procedure is available in the EMEP/MSC-E Technical report [*Shatalov et al*., 2012].

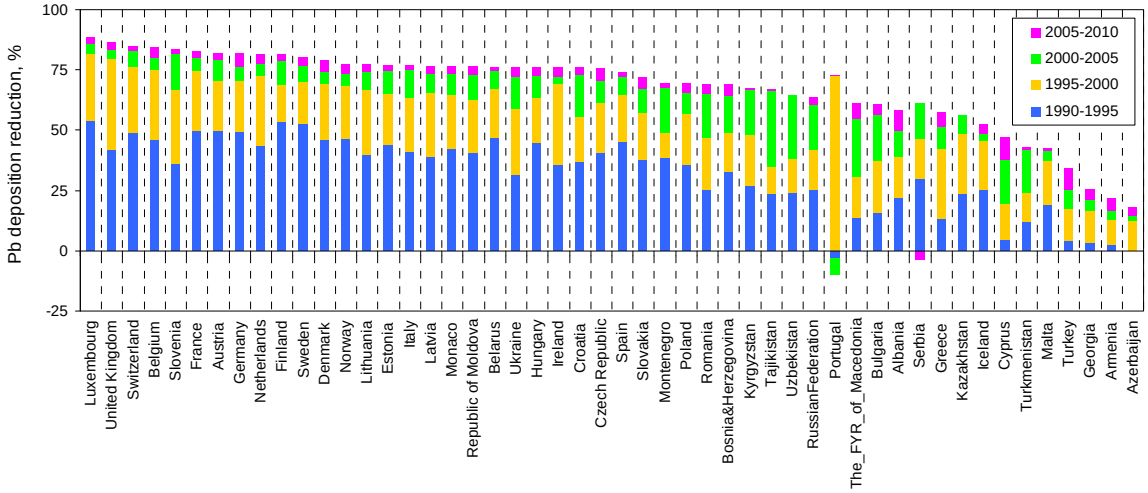


Fig. 3.4. *Relative reduction of Pb deposition over the period 1990-2010 in different countries of Europe and Central Asia*

As seen total reduction varies between 18% and 88% in different countries and exceeds 75% in more than half of the EMEP countries. In majority of the countries the largest changes occurred during the first 5-10 years of the period. However, considerable reduction also took place during the third sub-period (2000-2005) in some countries of Central and Eastern Europe. The lowest reduction occurred in the Caucasus countries. However, it should be noted that the model estimates for these countries are partly based on non-official expert estimates of anthropogenic emissions because of the lack of national data.

Long-term trends of lead deposition are exemplified by some countries located in different parts of Europe (Fig 3.5). The most noticeable deposition changes are characteristics of Western Europe presented in the figure by Switzerland and the United Kingdom (Figs. 3.5a and 3.5b). The largest decrease in these countries fell on the period 1990-2000 and after that the changes slowed down. Deposition flux in the United Kingdom varied significantly in the beginning of the period (1.3-8 kg $km^{-2}a^{-1}$) due to large north-to-south gradient within the country (see Fig. 3.3a). The variability of deposition fluxes is even higher in the Scandinavian countries (Figs. 3.5c and 3.5d), where atmospheric loads of lead drops essentially from south to north (Fig. 3.3). The southern parts of the countries are largely affected by emission sources from Central Europe, whereas the northern parts are relatively clean due to remoteness from industrial regions. As seen the upper deposition levels in the countries decreased more rapidly following emission reduction of major European sources. Lead deposition in the Caucasus countries such as Armenia and Georgia (Figs. 3.5e and 3.5f) did not change significantly over the whole period due to smaller reduction or even growth of emissions in these or neighbouring countries.

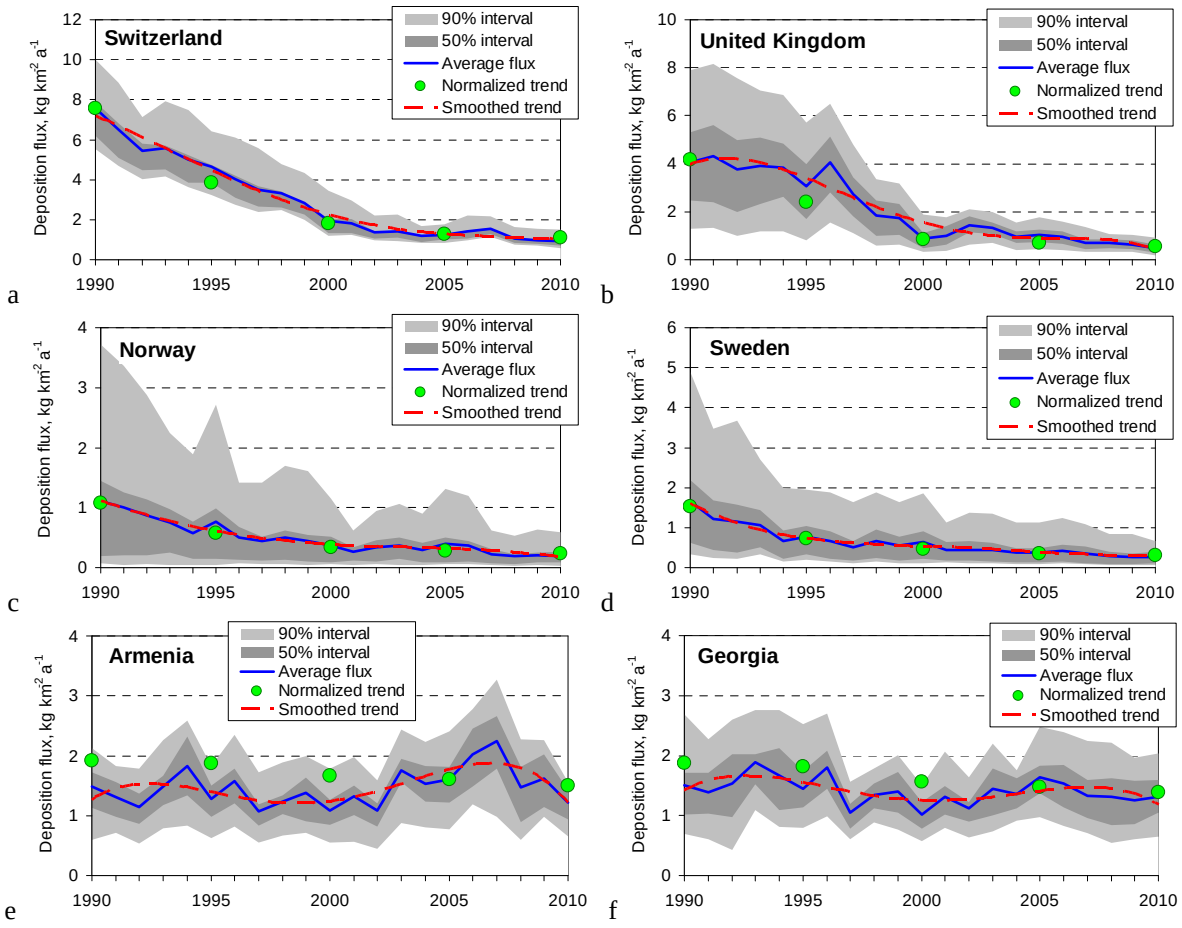


Fig. 3.5. *Temporal trends of Pb deposition flux in individual EMEP countries. Blue solid line presents area weighted average deposition flux; dashed red line is a smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the countries' territories, respectively.*

Cadmium

Total cadmium emission in the EMEP countries was reduced by 65% between 1990 and 2010 (Fig. 3.6a). It led to considerable decrease of cadmium atmospheric deposition (Fig. 3.6b). On average, deposition fluxes decreased approximately by 50% (from 60 to 33 g $km^{-2}a^{-1}$). On the other hand, deposition flux significantly varied over the region during the whole period ranging between 5 and 153 g $km^{-2}a^{-1}$ in 1990 and between 4 and 29 g $km^{-2}a^{-1}$ in 2010. Thus, the upper levels of deposition decreased ultimately by 80%.

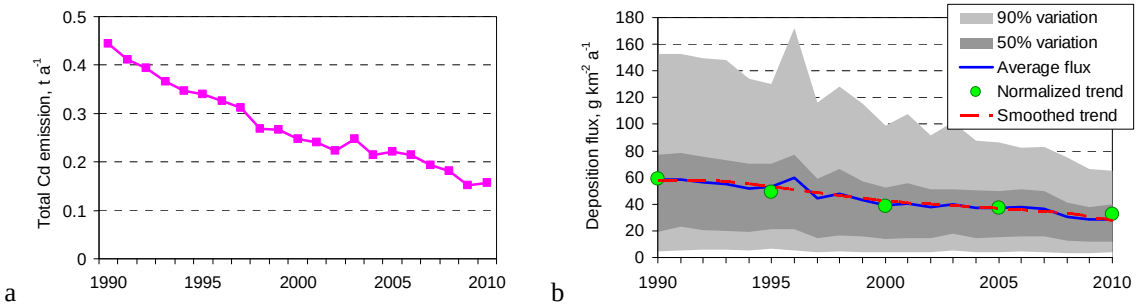


Fig. 3.6. *Temporal trends of Cd anthropogenic emissions (a) and deposition flux (b) in the EMEP countries. Deposition flux: blue solid line presents area weighted average deposition flux; dashed red line is a smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the whole territory, respectively.*

Estimates of cadmium deposition decrease are in a good agreement with observations performed at the EMEP monitoring sites. Figure 3.7 shows comparison of simulated temporal changes of cadmium air concentrations and wet deposition with observations at two EMEP sites. Long-term changes of anthropogenic emissions affecting cadmium levels at these sites are also shown in the figure. All three components of the analysis demonstrate similar levels of reduction between 1990 and 2010. However, it should be noted that the sites are located in Central and Northern Europe and do not reflect deposition decrease in the whole EMEP region. Wind re-suspension of cadmium accumulated in soil (legacy component) significantly contributes to cadmium deposition over the whole period.

The highest deposition levels in 1990 were characteristics of Western, Central and, partly, Eastern Europe decreasing both in the north and south directions (Fig. 3.8a). The most significant deposition reduction also took place in the same regions leading to relative levelling of the deposition pattern by the end of the period (Fig. 3.8b). An exception is high cadmium deposition fluxes remaining in some areas of Europe including western Germany, southern Poland with the neighbouring Czech Republic and Slovakia, as well as the Caucasus and some others. Elevated deposition levels of cadmium in these ‘hot spots’ can still present significant problem for the environment pollution and human health.

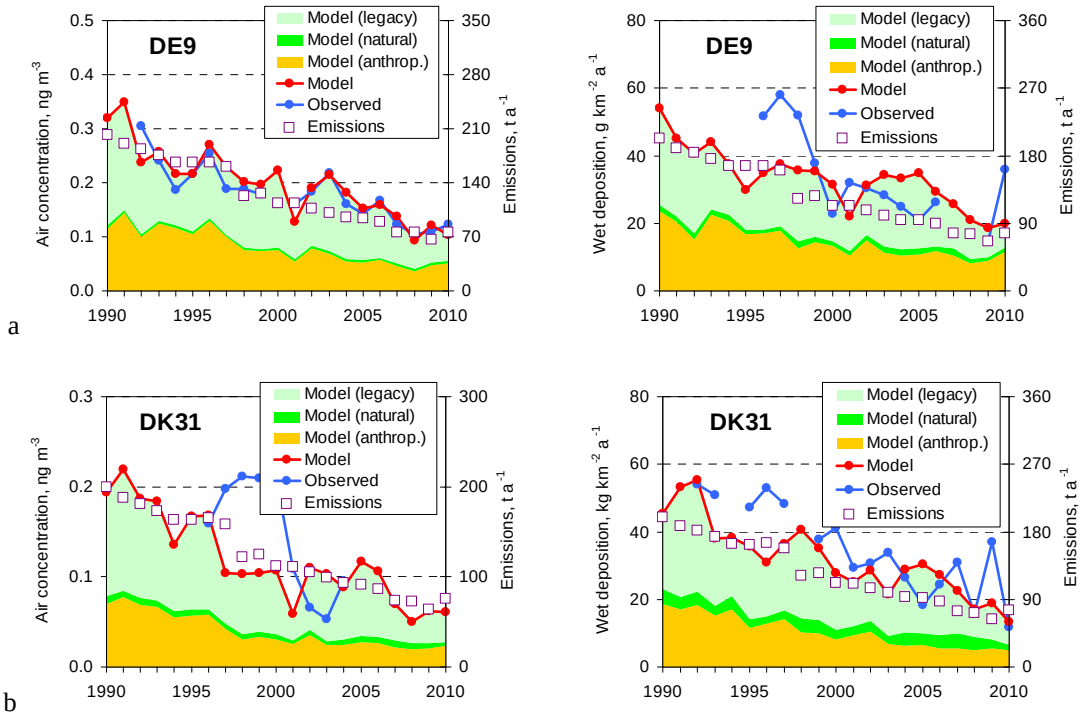


Fig. 3.7. *Simulated and observed trends of air concentration and wet deposition of cadmium at some EMEP monitoring sites: (a) – DE9, Germany; (b) – DK31, Denmark. Modelling results show contribution of anthropogenic emissions as well as natural and legacy component of wind re-suspension. The emission trends present sum of national emissions over countries contributing more than 90% of deposition at the considered sites.*

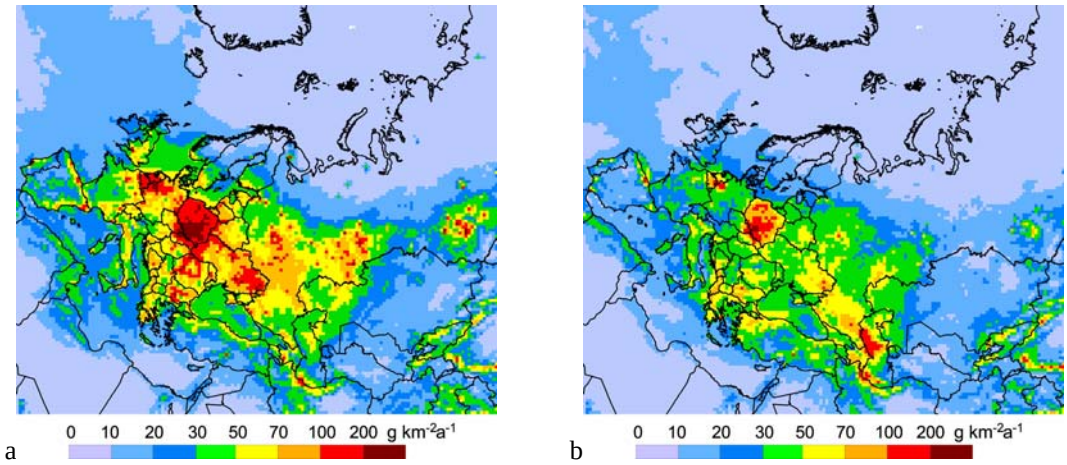


Fig. 3.8. *Spatial distribution of annual Cd deposition flux over Europe in 1990 (a), and 2010 (b)*

Dynamics of cadmium deposition changes in different countries of Europe and Central Asia is illustrated in Fig. 3.9. The overall changes over the period 1990-2010 varied from about 60% reduction in some countries of Western and Central Europe to moderate increase in countries of the Caucasus and Central Asia. In almost half of the countries decrease of deposition exceeded 50% of their levels in the year 1990. As that of lead, deposition decrease of cadmium was somewhat larger during the first 10 years of the period in many countries. But in contrast to lead the deposition reduction commonly continued up to 2010. It should be noted that increase of cadmium deposition in some countries mainly took place in the second half of the period.

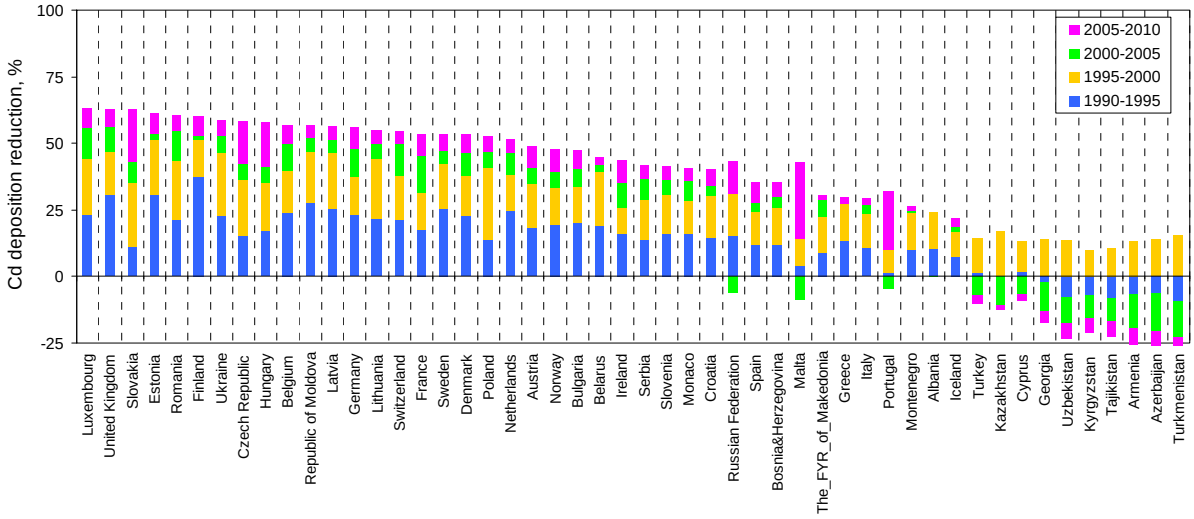


Fig. 3.9. *Relative reduction of Cd deposition over the period 1990-2010 in different countries of Europe and Central Asia*

An example of cadmium deposition trends in different parts of the region is given in Fig. 3.10. In some, particularly, small countries levels of cadmium deposition insignificantly varied over the countries' territory and gradually changed over the period (Fig. 3.10a) or remained almost unchanged (Fig. 3.10b). In contrast, in bigger countries or in countries with large emission source deposition fluxes substantially varied over the territory (Figs. 3.10c and 3.10d). For instance, cadmium deposition levels in the western part of Germany (North Rhine-Westphalia) are several times higher than those in the south-eastern part (Bavaria) due to significant sources located in western Germany as well as in neighbouring Belgium and the Netherlands (Fig. 3.8). Similarly, there is a strong increasing west-to-east gradient of cadmium deposition in the Czech Republic. Particularly, deposition levels in its most eastern regions are greatly larger than those in the western part because of emissions from national sources and sources located in southern Poland. Therefore, decrease of elevated deposition in such areas is largely defined by local emission reduction, whereas changes of lower deposition levels are more affected by distant sources. In the countries of Caucasus and Central Asia no significant decreasing trends (or even increasing of levels) are detected that indicates remaining problem of cadmium pollution. These estimates, however, require refinement with use of national emission data and observations from these countries.

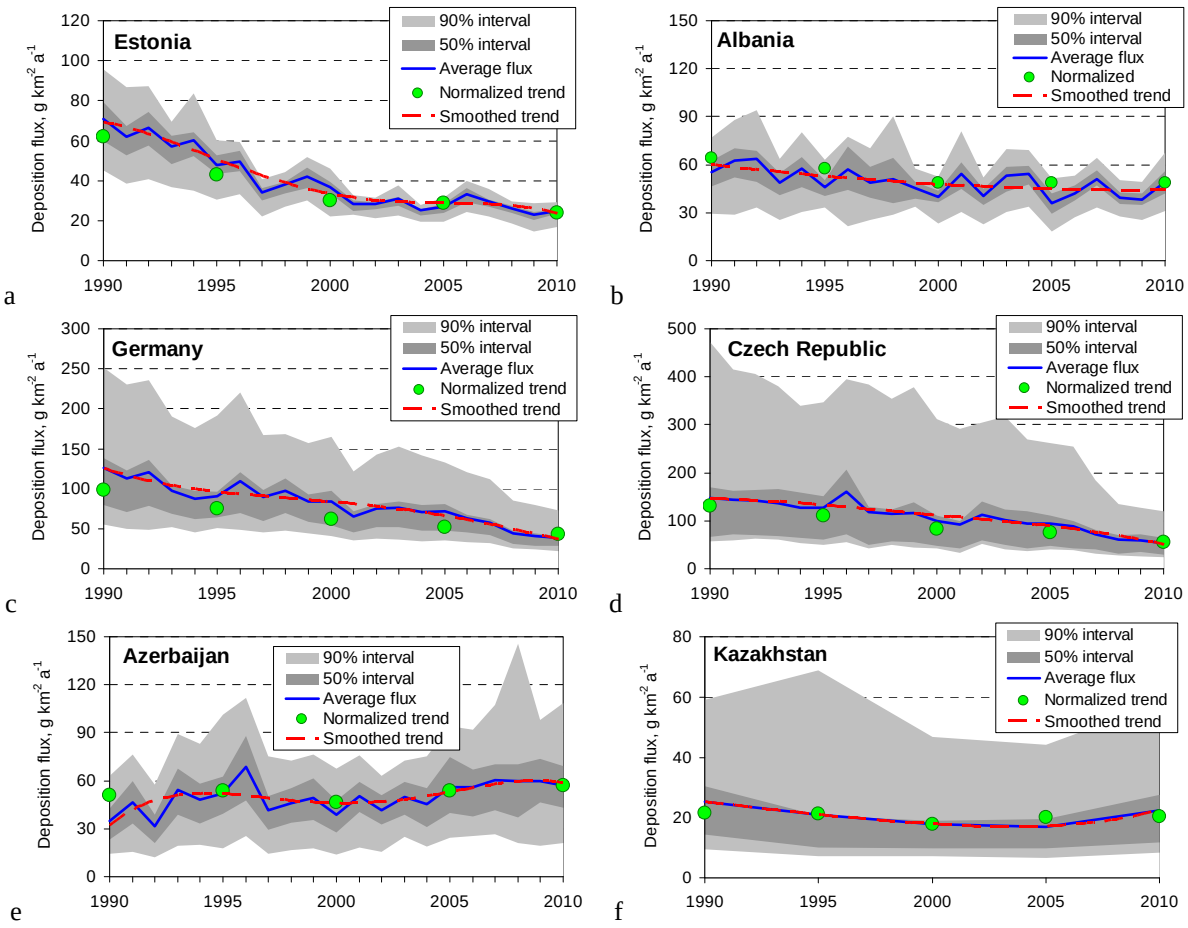


Fig. 3.10. *Temporal trends of Cd deposition flux in individual EMEP countries. Blue solid line presents area weighted average deposition flux; dashed red line is a smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the countries' territories, respectively.*

Reduction of cadmium emissions in many EMEP countries led to noticeable decrease of European background deposition levels. However, elevated levels still remain in numerous 'hot spots', i.e. industrial regions or areas around strong emission sources. Evaluation of cadmium pollution in such areas requires more detailed analysis with finer spatial resolution. This sort of analysis has been performed within a framework of a Case Study on heavy metal pollution of the Czech Republic [*Ilyin et al.*, 2012]. Variety of national-scale data has been collected and analysed to characterize different aspects of the country pollution involving detailed emission data, observations and model simulations with high spatial resolution. Simulated levels of cadmium deposition over the Czech Republic and the neighbouring countries are shown in Fig. 3.11. As seen the deposition pattern is very heterogeneous. High deposition fluxes are characteristics of the eastern region of the country (Moravian-Silesian and Zlin Regions) as well as in areas close to the Czech-Polish and Czech-Slovak border. Somewhat elevated fluxes are also typical for large urban areas such as the Prague district. Therefore, these areas are subject of particular attention in future.

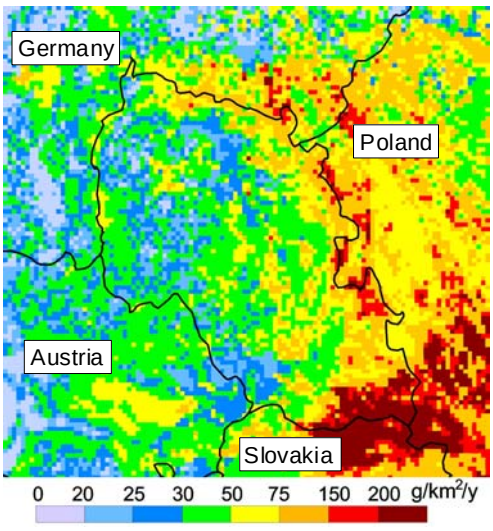


Fig. 3.11. *Cadmium deposition fluxes in the Czech Republic in 2007 estimated with fine spatial resolution (5x5 km^2)*

Mercury

Anthropogenic emissions of mercury in the EMEP countries were considerably reduced (by 60%) over the period 1990-2010, whereas corresponding decrease of atmospheric deposition did not exceed 30% (Fig. 3.12). Average deposition fluxes in the region have changed from 13 g $km^{-2}a^{-1}$ in 1990 to 9 g $km^{-2}a^{-1}$ in 2010. Moreover, relatively high deposition levels typical for industrial or populated areas decreased more noticeably (from 34 to 18 g $km^{-2}a^{-1}$). On the other hand, lower levels characterizing remote areas remained practically unchanged (around 3 g $km^{-2}a^{-1}$).

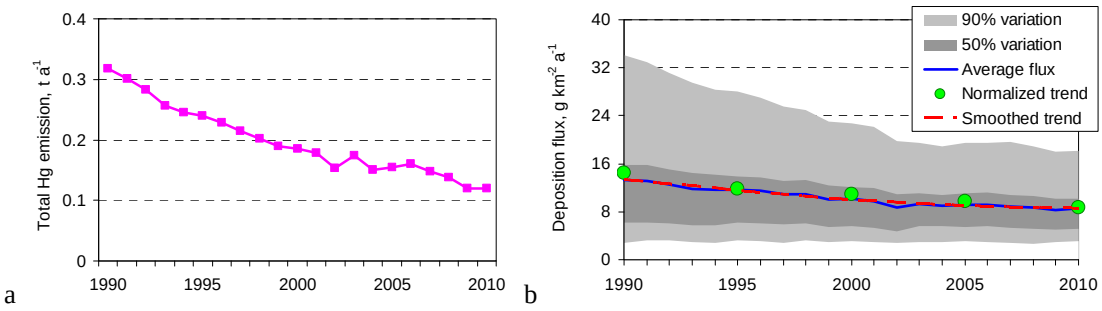


Fig. 3.12. *Temporal trends of Hg anthropogenic emissions (a) and deposition flux (b) in the EMEP countries. Deposition flux: blue solid line presents area weighted average deposition flux; dashed red line is a fourth-order polynomial smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the whole territory, respectively.*

The reason for the low deposition sensitivity to emission reduction is in a large contribution of mercury transport from other continents. Indeed, due to very long residence time of mercury in the atmosphere it is easily transported between the continents and can travel over thousands of kilometres reaching remote regions. Figure 3.13 shows spatial distribution of mercury deposition on a global scale. As seen elevated deposition levels occur not only over the major industrial regions in Europe, North America, and Asia etc. but also over the oceans. Therefore, changes of mercury atmospheric deposition in Europe are largely affected by emission changes in other continents. According to available estimates, global emission of mercury was not significantly changed during last 20 years [*AMAP*, 2011]. Emission reduction in developed countries of Europe and North America is compensated by emission increase in countries with growing economies (mostly in Asia). In addition, emissions from natural and legacy sources (volcanoes, geogenic sources, re-emission of mercury accumulated in soil and seawater etc.), which contribute about two thirds of global mercury deposition, are insignificantly changed on a scale of decades. The slowly changed contribution of the global background essentially hampers deposition reduction in the EMEP countries (Fig. 3.14a). Nowadays, current anthropogenic emissions in the EMEP countries make up about one third of total deposition to the region (Fig. 3.14b), whereas the rest is defined by anthropogenic emissions from other continents (17%) and contribution of natural and legacy sources (about 50%). Therefore, both regional and global abatement efforts are needed to reduce mercury pollution in the region.

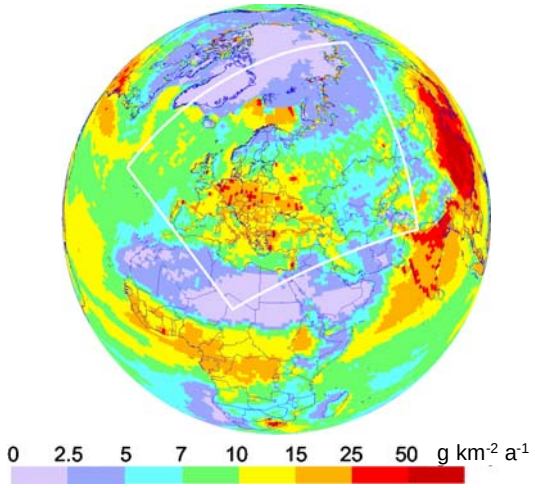


Fig. 3.13. *Spatial distribution of mercury deposition flux on a global scale in 2010. White rectangle shows location of the EMEP domain.*

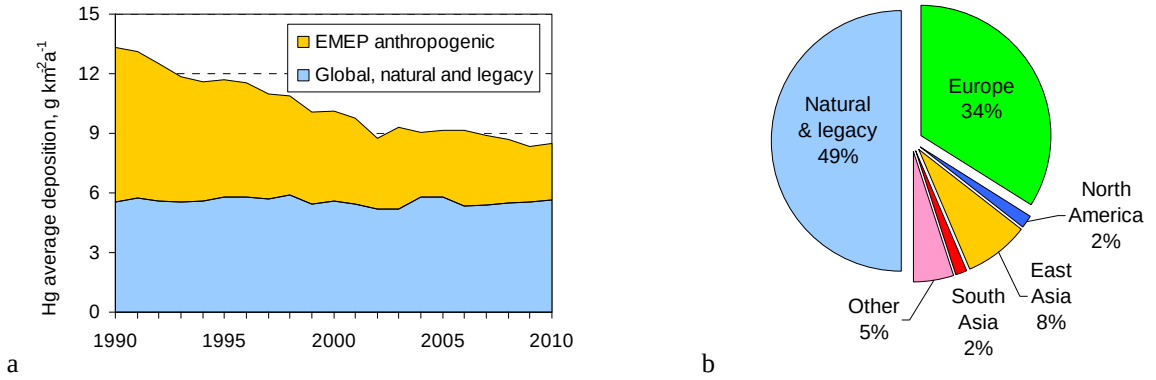


Fig. 3.14. *Long-term changes of average mercury deposition flux (a) and contribution of different source types to mercury deposition in the EMEP countries in 2005 (b)*

Large contribution of the global background to mercury pollution levels in Europe is clearly seen from comparison of simulated and observed trends of mercury concentration and deposition with trends of anthropogenic emissions (Fig. 3.15). As seen, concentration of elemental mercury is largely (up to 90%) defined by the global background. However, it should be mentioned that the global component indirectly includes emissions from European sources, which were transported out of the region and mixed up in the global pool of atmospheric mercury. Long-term changes of mercury concentration rather reflect the global emission trend than the regional emission reduction. Wet deposition is more affected by anthropogenic emissions from European sources because they are more determined by removal of short-lived mercury forms emitted from local and regional sources.

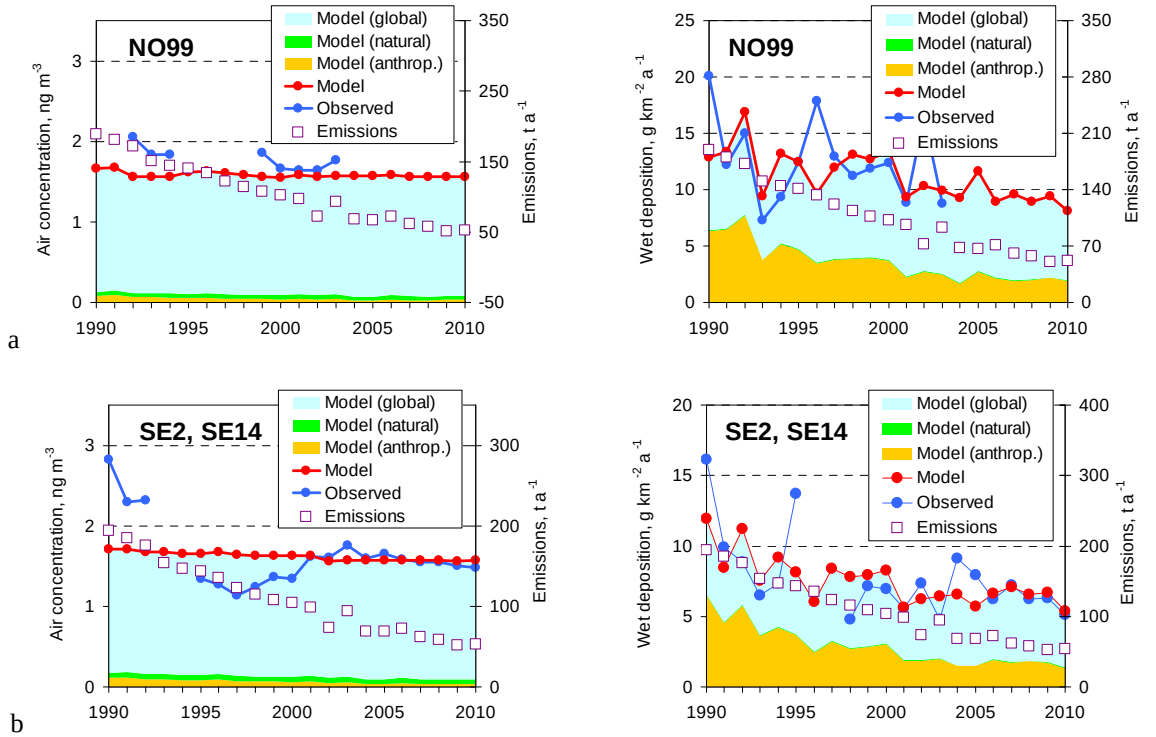


Fig. 3.15. *Simulated and observed trends of air concentration and wet deposition of mercury at some EMEP monitoring sites: (a) – NO99, Norway; (b) – SE2, SE14, Sweden. Modelling results show contribution of anthropogenic and natural emissions in Europe as well as contribution of the global background. The emission trends present sum of national emissions over countries contributing more than 90% of anthropogenic deposition at the considered sites.*

Spatial changes of mercury deposition levels in the EMEP countries between 1990 and 2010 are shown in Fig. 3.16. Similar to other metals, the most significant changes took place in Western and Central Europe in areas with the largest deposition loads. Mercury deposition noticeably decreased over the United Kingdom, the Benelux countries, western Germany, Poland and Ukraine. Some reduction also occurred over the southern part of the Russian Federation. On the other hand, deposition levels in Northern and Southern Europe less affected by regional sources remain practically unchanged.

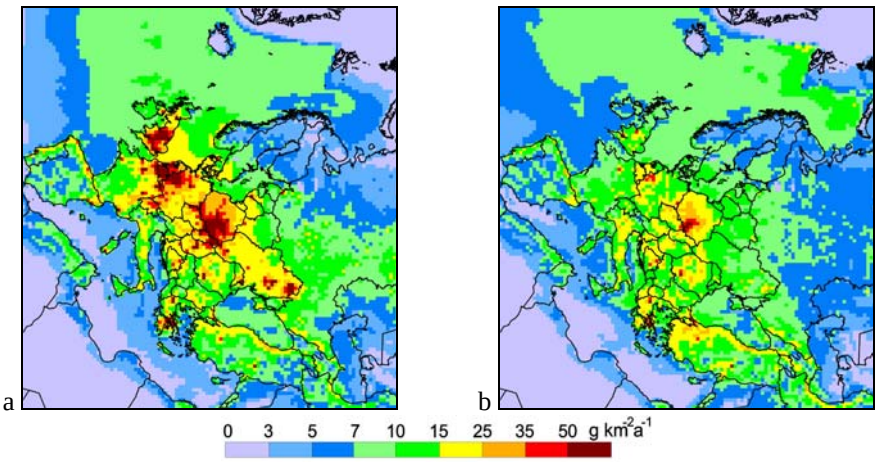


Fig. 3.16. *Spatial distribution of annual Hg deposition flux over Europe in 1990 (a), and 2010 (b)*

Relative changes of mercury deposition in different countries of Europe and Central Asia over the period 1990-2010 are shown in Fig. 3.17. The changes differ from about 70% decline in Western and Central Europe to slight growth in Central Asia and the Caucasus. Deposition decrease exceeded 50% of their value in 1990 in one third of the countries and mostly fell on the first half of the period. The smallest deposition reduction took place in countries with low national emissions and countries located far from the major emission sources and largely affected by the global background.

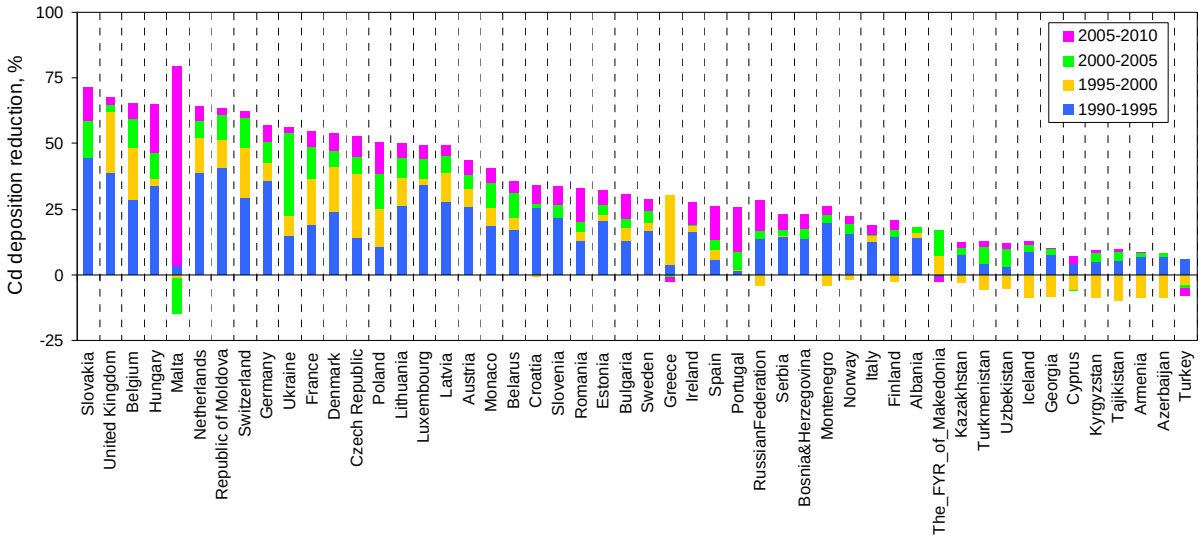


Fig. 3.17. *Relative reduction of Hg deposition over the period 1990-2010 in different countries of Europe and Central Asia*

Figure 3.18 shows a few examples of long-term changes of mercury deposition in some EMEP countries. In countries of Western Europe, presented by Belgium and the Netherlands (Figs. 3.18a and 3.18b), significant deposition reduction occurred over the first 5-10 years of the period and then deposition reduction slowly continued up to the present time. No significant trends were found for average deposition in the Scandinavian countries (e.g. Norway, Fig. 3.18c) and in the Mediterranean region (e.g. Italy, Fig. 3.18d). However, some deposition reduction occurred in these countries in areas around emission sources. Deposition levels in Central Asia changed slightly ranging from moderate decrease in Kazakhstan (Fig. 3.18e) to small increase in Tajikistan (Fig. 3.18f).

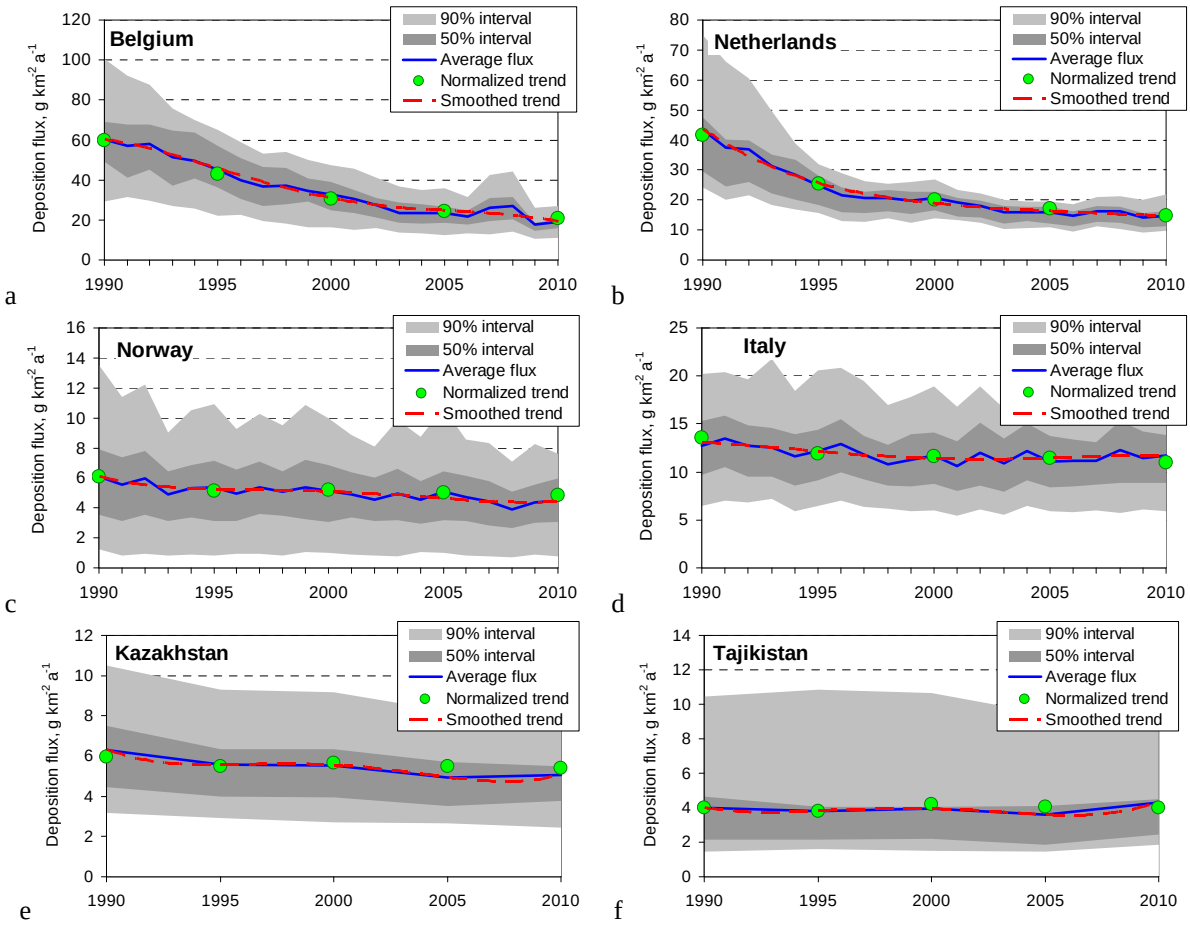


Fig. 3.18. *Temporal trends of Hg deposition flux in individual EMEP countries. Blue solid line presents area weighted average deposition flux; dashed red line is a smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the countries' territories, respectively.*

3.2. Changes in transboundary pollution over 20 years

Transboundary transport plays an important role in heavy metal pollution of the EMEP countries. Given the residence time of the considered pollutants ranging from days (Pb and Cd) to months (Hg) they can be transported over hundreds of kilometres from one country to another and contribute to local pollution. Emission reduction in different parts of the EMEP region during last 20 years resulted in redistribution of transboundary fluxes between the countries. Changes in transboundary pollution over the period 1990-2010 are considered below. Only anthropogenic component of deposition is analysed hereafter in terms of transboundary transport as an object of current and future abatement measures.

Lead

As it was shown above both anthropogenic emissions and deposition of lead were dramatically reduced in the EMEP countries during last two decades. As the rate of emission reduction varied among the countries transboundary fluxes characterizing countries' mutual pollution were changed. Figure 3.19 illustrates relative contribution of domestic and foreign sources to anthropogenic deposition of lead in the EMEP countries and its changes between 1990 and 2010. In 1990 contribution of foreign sources exceeded the impact of domestic sources in more than 80% of the EMEP countries. This fraction somewhat decreased down to 70% in 2010 but still remained significant. It means that pollution in the EMEP countries by lead continues to be largely affected by transboundary pollution. The largest changes of transboundary pollution took place in a few countries due to more (or less) intensive changes of their national emissions with regard to the average changes in Europe. The largest increase of the role of transboundary pollution occurred in the Russian Federation, Lithuania, Republic of Moldova and Greece. On the other hand, contribution of transboundary transport considerably decreased in Serbia, Malta, Azerbaijan and Georgia.

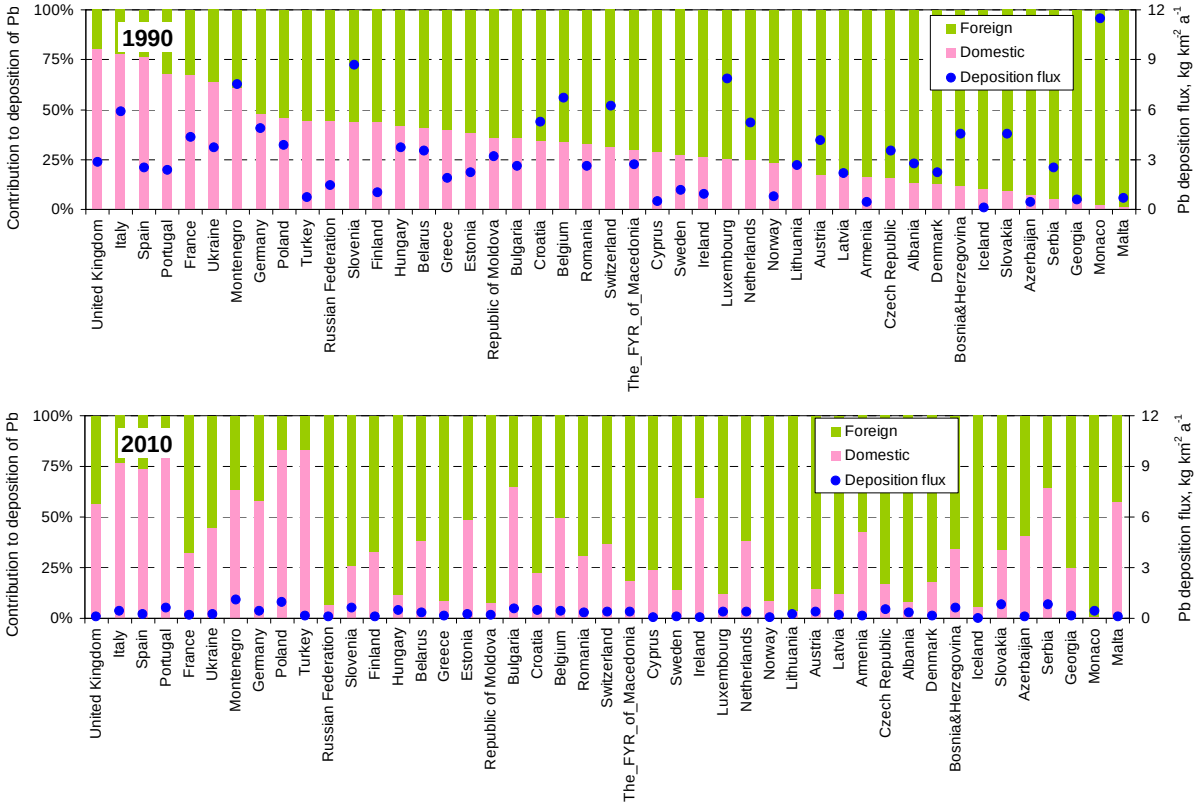


Fig. 3.19. *Relative contribution of domestic and foreign emission sources to* ***lead*** *anthropogenic deposition in the EMEP countries. Blue dots show average levels of anthropogenic deposition of* ***lead*** *in the countries.*

Changes in contribution of transboundary transport to lead deposition are illustrated in Fig. 3.20 for two countries – the Russian Federation and Serbia. As seen relative contribution of domestic sources to lead deposition in the Russian Federation drastically decreased between 1990 and 2010. And it was accompanied by appropriate increase of contributions from other countries. The reason of these changes is in sharp reduction of lead anthropogenic emissions in the country. According to the official emission data reported by the country total lead emission was reduced by a factor of 7 between 2003 and 2004 and continued decreasing gradually up to 2010. As a result, lead deposition in the Russian

Federation in 2010 is largely defined by transboundary transport from the neighbours. However, it should be noted that such rapid emission dynamics could be connected with uncertainties of the emission estimates and requires additional validation.

The other example demonstrates the opposite tendency when contribution of domestic sources in Serbia increased from 5% to 63% over the period. It considerably reduced influence of transboundary pollution in the country. This effect was caused by noticeable growth of anthropogenic emissions in the country during last few years.

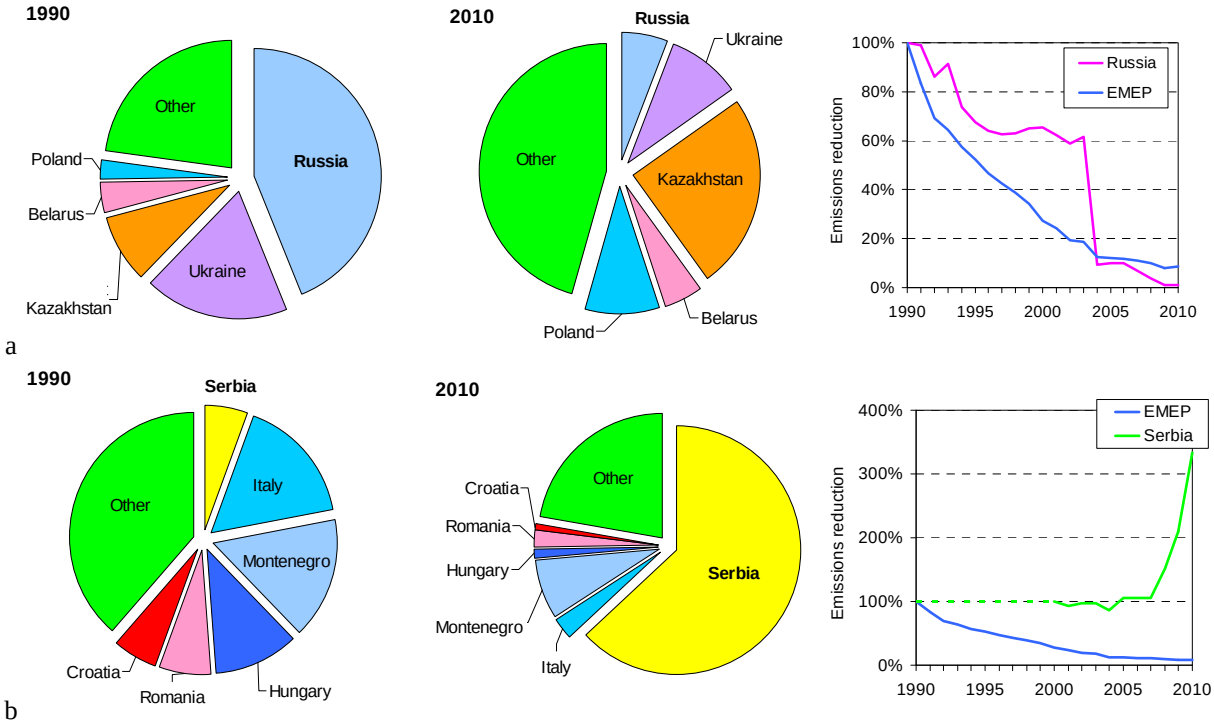


Fig. 3.20. *Changes in contribution of different EMEP countries to lead deposition in the Russian Federation (a) and Serbia (b) between 1990 and 2010. Lead emission changes in these countries are given in comparison with changes of total emissions of the EMEP countries.*

Cadmium

In general, cadmium emission and deposition reduction was less substantial than those of lead. As a result changes in contribution of cadmium transboundary pollution in the EMEP countries during the period 1990-2010 were lower as well. Contribution of foreign sources to cadmium deposition dominated over domestic emissions in about 75% of the EMEP countries in 1990. This proportion was not considerably changed over 20 years (Fig. 3.21). On the other hand some redistribution of transboundary fluxes took place in a number of countries due to inhomogeneous emission reduction in the region. Contribution of transboundary transport to cadmium deposition increased the most significantly in Slovakia, Ukraine, Republic of Moldova and Lithuania, whereas its role decreased considerably in the Netherlands, Ireland, Belarus and Latvia.

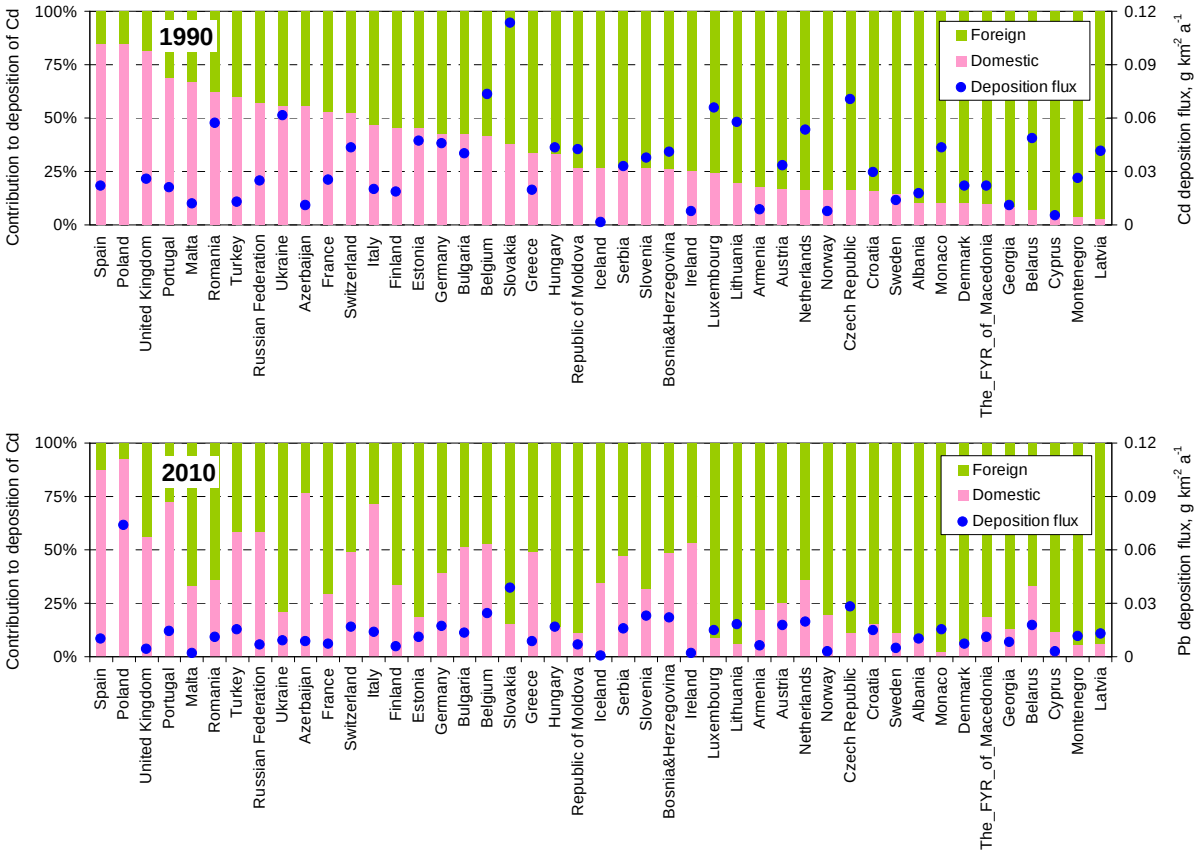


Fig. 3.21. *Relative contribution of domestic and foreign emission sources to* ***cadmium*** *anthropogenic deposition in the EMEP countries. Blue dots show average levels of anthropogenic deposition of* ***cadmium*** *in the countries.*

Different changes of cadmium transboundary transport between the EMEP countries are exemplified by two countries – Slovakia and the Netherlands (Fig. 3.22). In 1990 cadmium deposition in Slovakia consisted of approximately equal contributions of Slovak domestic sources, emissions from neighbouring Poland and all other countries in total. National Slovak emissions were largely reduced by 2010, in particular, in the second half of the period. According to the official emission data, the national total dropped by a factor of 5 between 2006 and 2007. In contrast, anthropogenic emissions in Poland decreased moderately over the period and even increased somewhat in the last years. These factors led to considerable decrease of contribution of domestic sources (down to 15%) and increase of contribution of Polish emissions (up to 64%) by 2010.

Increase of the role of domestic sources along with decrease of contribution of transboundary transport between 1990 and 2010 was found for the Netherlands. Growth of national cadmium emissions in the country in the second half of the period was accompanied by substantial emission reduction in France and the United Kingdom – of two major contributors of cadmium deposition in the Netherlands. It resulted in the decrease of relative contribution of foreign sources to cadmium pollution in the country.

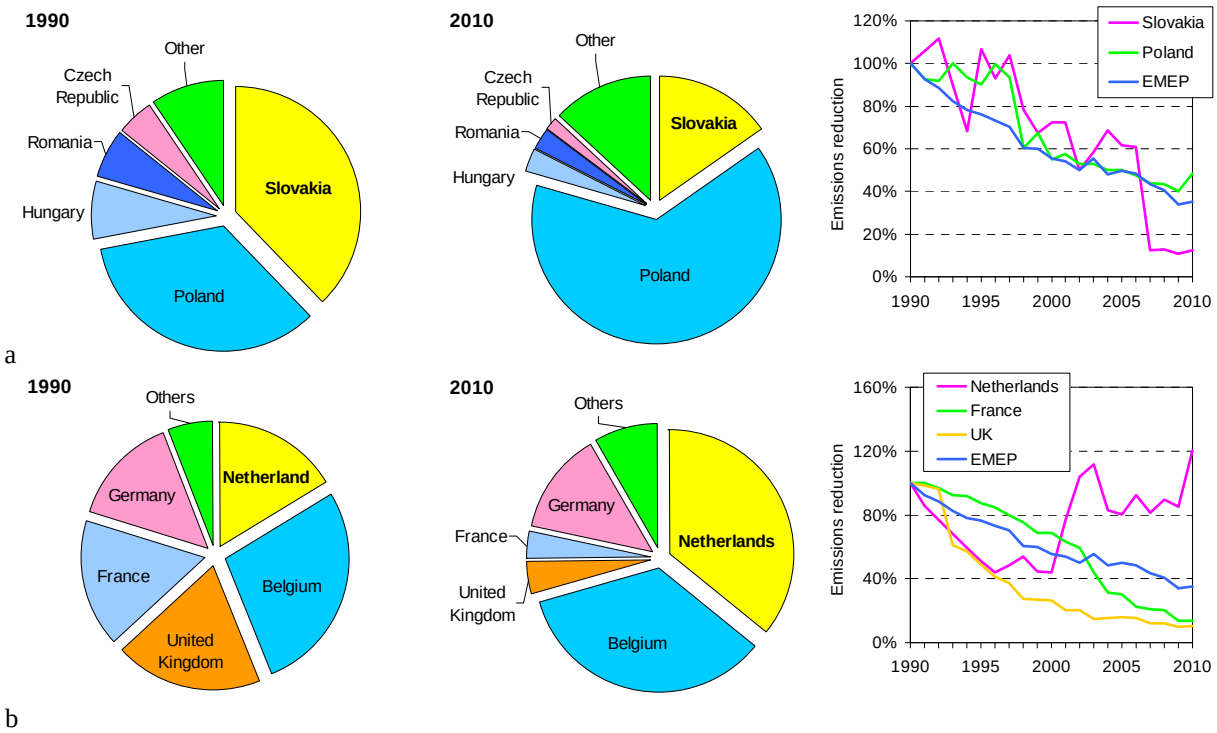


Fig. 3.22. *Changes in contribution of different EMEP countries to cadmium deposition in Slovakia (a) and the Netherlands (b) between 1990 and 2010. Cadmium emission changes in these countries are given in comparison with changes of total emissions of the EMEP countries*

Mercury

Mercury differs from other two metals by more complicated character of atmospheric transport involving chemical transformations. It occurs in the atmosphere in different physical and chemical forms with diverse properties. Long-lived elemental form prevailing in the free atmosphere allows intercontinental transport of mercury. On the other hand, short-lived oxidized forms emitted by anthropogenic sources are quickly (from hours to days) deposited defining local pollution.

Figure 3.23 shows proportion of domestic and foreign sources in anthropogenic deposition of mercury in the EMEP countries. Deposition from foreign sources dominated in 64% of the countries in 1990 and this fraction decreased to 60% in 2010. This value is even lower than similar proportions for lead and cadmium indicating the fact that mercury behaves on a regional scale as a regional or local pollutant. However, this diagram presents only contribution of emission sources from the EMEP countries and does not reflect considerable impact of intercontinental transport mentioned above. Contribution of transboundary transport to mercury deposition has changed in some EMEP countries because of different rates of emission reduction. In particular, its role increased noticeably in the Republic of Moldova, Slovakia, Lithuania and the Russian Federation, and decreased in Ireland, Serbia, Luxemburg and Belarus.

Redistribution of transboundary fluxes of mercury between the EMEP countries is illustrated with particular cases of the Republic of Moldova and Ireland (Fig. 3.24). Drastic reduction of mercury emissions in the Republic of Moldova between 1990 and 2000 caused substantial decrease of deposition to the country from domestic sources and increase of relative contribution of mercury deposition from the neighbouring countries. Thus, transboundary transport became to play the prevailing role in mercury pollution of the country. In contrast, contribution of foreign sources decreased considerably in Ireland due to insignificant changes of national emissions and large emissions reduction in the United Kingdom – the biggest contributor to mercury deposition in the country.

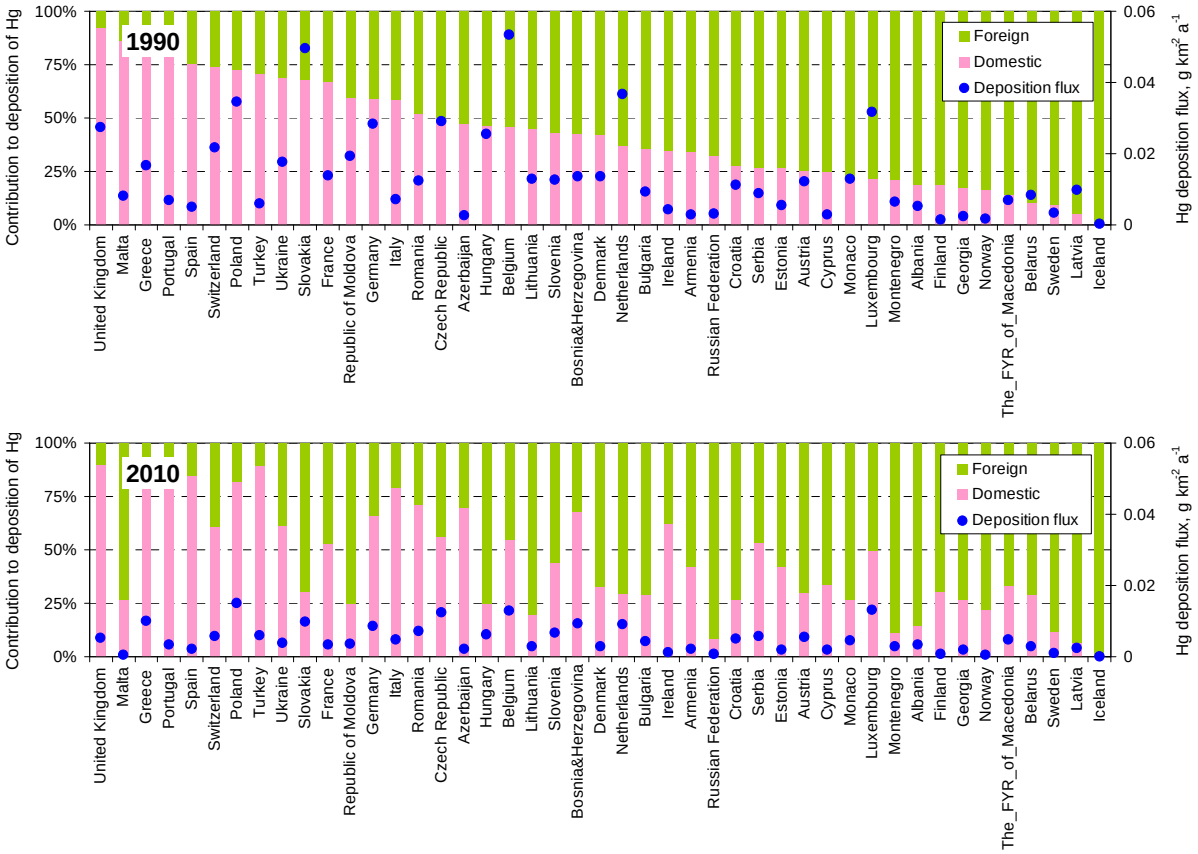


Fig. 3.23. *Relative contribution of domestic and foreign emission sources to* ***mercury*** *anthropogenic deposition in the EMEP countries. Blue dots show average levels of anthropogenic deposition of* ***mercury*** *in the countries.*

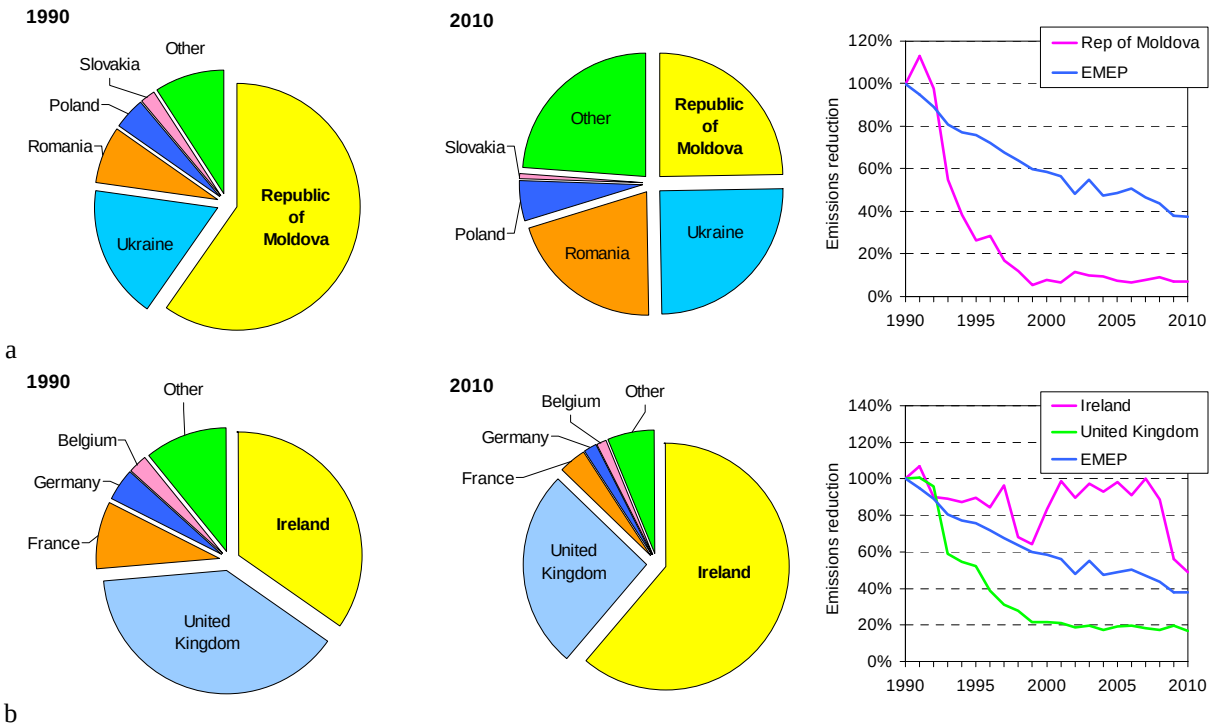


Fig. 3.24. *Changes in contribution of different EMEP countries to mercury deposition in Republic of Moldova (a) and Ireland (b) between 1990 and 2010. Mercury emission changes in these countries are given in comparison with changes of total emissions of the EMEP countries.*

3.3. Key source categories of heavy metal pollution

Reduction of heavy metal pollution levels was accompanied by changes in the key source categories of both emissions and deposition. Atmospheric emissions of heavy metals from some emission sectors were reduced more substantially comparing to other sectors. It resulted in the change in the sectoral composition of heavy metal pollution in the EMEP countries.

Lead deposition dramatically decreased in the EMEP countries from 1990 to 2010 mostly due to the phase out of leaded gasoline from the use in road transport. In 1990 contribution of road transport to lead deposition exceeded 75% in total over the countries (Fig. 3.25). The remainder consisted from metal production (9%), stationary combustion in industry (4%), public electricity and heat production (4%), non-industrial combustion (3%) and others (4%). Due to wide abatement efforts emissions of lead from road transport were drastically reduced (by a factor of 65) over last 20 years in most of the countries. Reduction of emission from other sectors was less significant and varied from 1.6 to 4.1 times. As a result, contribution of road transport decreased down to 11% of total lead deposition in the EMEP countries in 2010. Metal production and stationary combustion in industry became the dominant emission sectors in 2010 with relative contributions to total deposition of 29% and 26%, respectively. The other two key sectors (public electricity and heat production and non-industrial combustion) made up 26% of total deposition in sum.

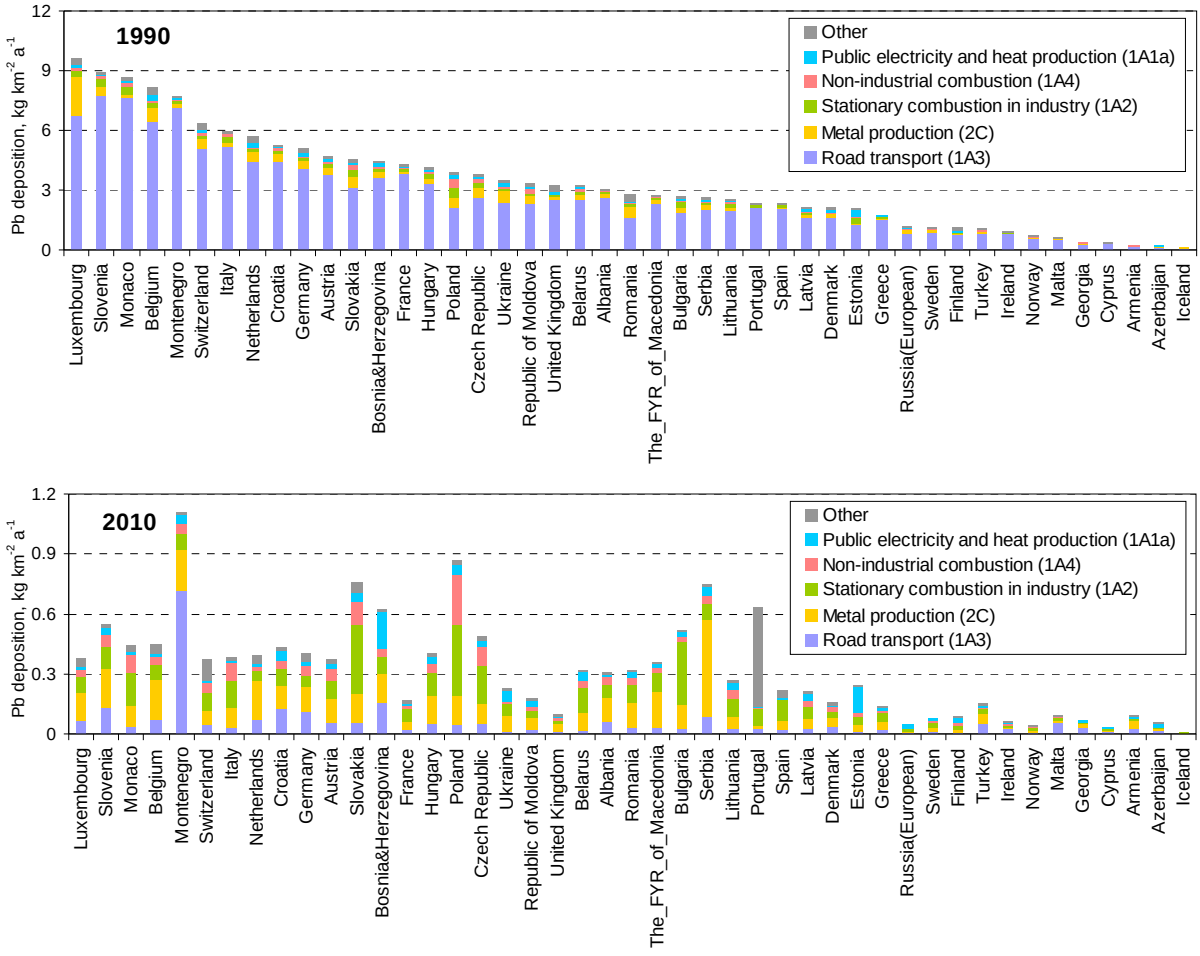


Fig. 3.25. *Contribution of the key source categories to anthropogenic deposition of* ***lead*** *in the EMEP countries in 1990 and 2010.*

Decrease of cadmium deposition in the EMEP countries from 1990 to 2010 was less significant than deposition reduction of lead and accompanied by smaller change in the sectoral composition. The four major source categories – metal production (23%), stationary combustion in industry (22%), non-industrial combustion (20%), and public electricity and heat production (17%) – made up more than 80% of cadmium deposition in 1990 (Fig. 3.26). Due to more or less homogeneous emission reduction in all these sectors they maintained the prevailing role in cadmium pollution but their relative importance changed. The highest reduction occurred for cadmium emissions from metal production (by a factor of 4.7). It led to reduction of relative contribution of this sector to deposition in the EMEP countries from 23% in 1990 to 12% in 2010. On the other hand, relatively small reduction of emissions from non-industrial combustion resulted in the dominant role of this sector in 2010. Nowadays, it contributes about one third of total cadmium deposition in the region. However, it should be noted that there is significant inconsistency in emission data reported by the EMEP countries for the non-industrial combustion sector. About 90% of total cadmium emissions from this key sector in 2010 were contributed only by 5 countries (Poland, Italy, Turkey, Germany and Belgium), of which 75% were made up by Poland. These essential differences in the sectoral composition of cadmium emissions raised a question of comparability and completeness of the reported data.

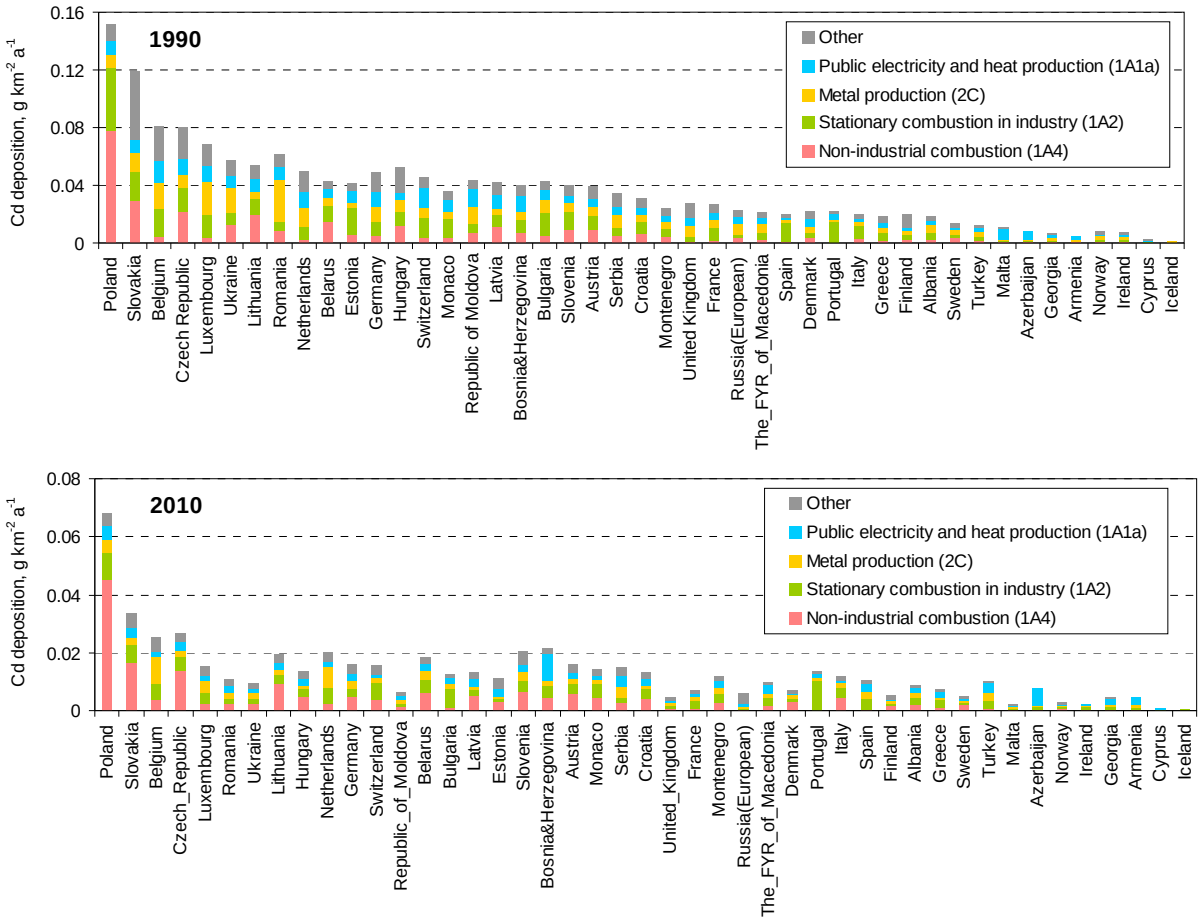


Fig. 3.26. *Contribution of the key source categories to anthropogenic deposition of* ***cadmium*** *in the EMEP countries in 1990 and 2010.*

No substantial changes in the key source categories of mercury deposition occurred between 1990 and 2010 in the EMEP countries (Fig. 3.27). Combustion of fossil fuels for public electricity and heat production made up 35% of total mercury deposition in the EMEP countries in 1990. Other key categories included stationary combustion in industry (18%), metal production (11%), non-industrial

combustion (10%), and waste incineration (6%). Mercury emissions from all these sectors were reduced over the period 1990-2010 by factors of 2.2-3.1. An exception was the waste incineration sector, which emissions decreased by a factor of 5 leading to change of its relative contribution to total deposition from 6% in 1990 to 3% in 2010. In contrast, contribution of public electricity and heat production increased up to 40%.

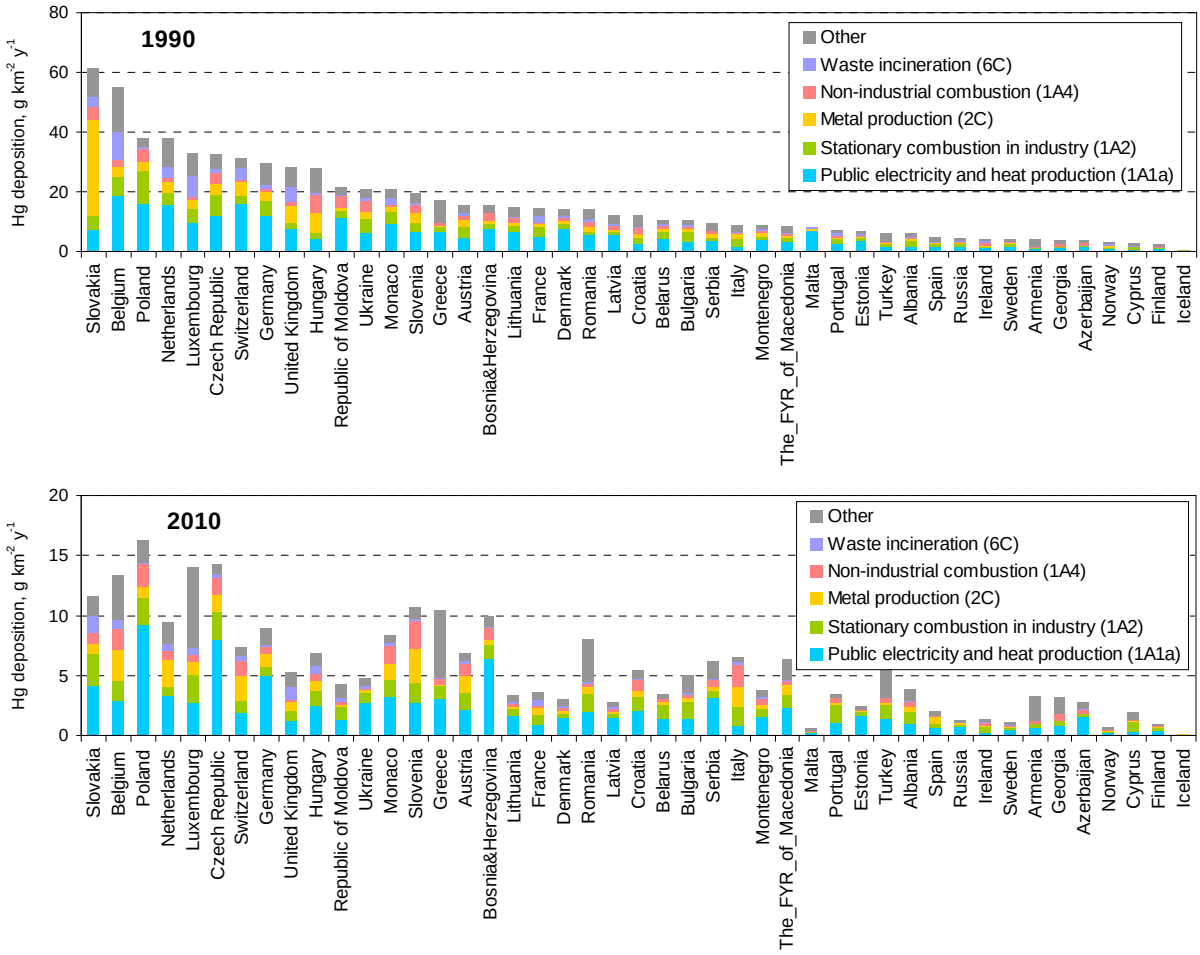


Fig. 3.27. *Contribution of the key source categories to anthropogenic deposition of* ***mercury*** *in the EMEP countries in 1990 and 2010.*

Thus, changes in the sectoral composition of heavy metal emissions have led to unification of the key source categories the most significantly affecting deposition of lead, cadmium and mercury in the EMEP countries at present. Nowadays, the prevailing sectors in deposition of all three metals include stationary combustion in industry (1A2), non-industrial combustion (1A4), metal production (2C) and public electricity and heat production (1A1a) (Fig. 3.28). These source categories will require priority mitigation efforts to reduce heavy metal pollution in future.

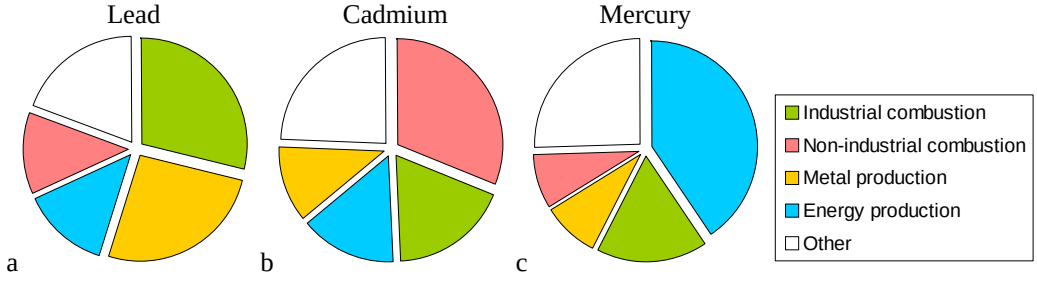


Fig. 3.28. *Relative contribution of the key source categories to total deposition of lead (a), cadmium (b) and mercury (c) in the EMEP countries in 2010.*

3.4. Exceedances of critical loads

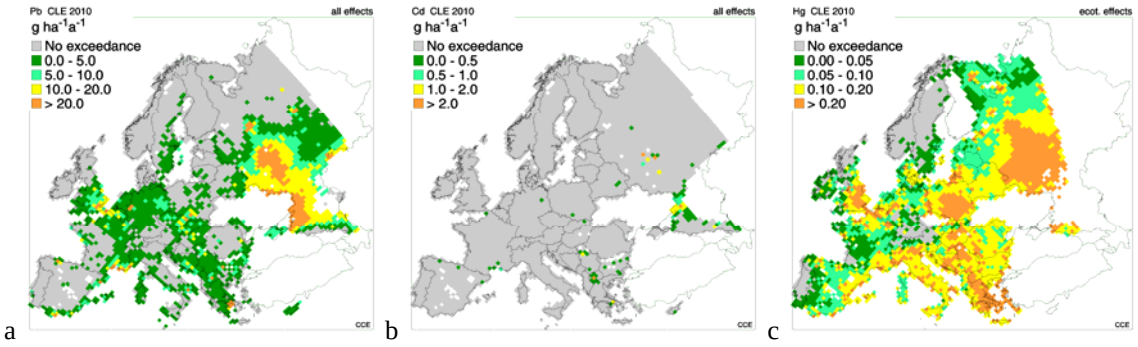
Assessment of long-term adverse effects caused by heavy metal pollution on ecosystems and human health was carried out by the Coordinating Centre for Effects (CCE) of the Working Group on Effects (WGE) using the critical load approach [*Slootweg et al,* 2010]. The most recent evaluation of heavy metal deposition exceedances of critical loads was performed in the framework of the project initiated by the Dutch Ministry of Housing, Spatial Planning and the Environment. The aim of the project was to support negotiations on the revision of the Heavy Metal Protocol [*Vischedijk et al,* 2010, *Slootweg et al,* 2010]. A short summary of the results of this project is given below.

Critical loads of heavy metals have been modelled and mapped with respect to the following effect-end points: (1) human health effect (drinking water) via terrestrial ecosystems; (2) human health effect (food quality) via terrestrial ecosystems; (3) eco-toxicological effect on terrestrial ecosystems; (4) eco-toxicological effect on aquatic ecosystems; (5) human health effect (food quality) via aquatic ecosystems.

The first four effects are based on critical concentrations of the metal in the soil solution. Using a mass balance for the root layer, this concentration is related to the deposition. Fertilisation of agricultural areas also causes cadmium and lead to enter soil systems, but this is not taken into account in this assessment. For each ecosystem the minimum of the critical loads for all effects is taken. The last effect is directly related to the concentration in rainwater. More on the calculations of critical loads can be found in the Mapping Manual [*UBA*, 2004].

Critical loads for heavy metals were reported by 18 countries: Austria, Belarus, Belgium, Bulgaria, Cyprus, the Czech Republic, Finland, France, Germany, Italy, the Netherlands, Poland, Russia, Slovakia, Sweden, Switzerland, Ukraine and the United Kingdom. Critical loads for other countries were calculated with the CCE background database [*Slootweg*, 2005].

Model simulations of lead, cadmium and mercury deposition fluxes were carried out by MSC-E [*Vischedijk et al*, 2010]. In order to evaluate areas where atmospheric metal deposition fluxes are higher than critical loads, the values of the average accumulated exceedances (AAE) were computed as the ecosystem area-weighted sum of the individual exceedances (deposition minus critical load, with zero for non-exceedance) of all ecosystems in a grid cell. Spatial distribution of the critical load exceedances over the EMEP countries in 2010 is shown in Fig. 3.29 for all three heavy metals. As seen atmospheric deposition fluxes of lead and mercury exceed estimated critical loads over large areas in Europe. The most significant exceedances occur in the southern part of the Russian Federation. It means that human health and/or ecosystems are at risk in these areas due to elevated deposition of heavy metals. In contrast, the critical loads for cadmium are not exceeded anywhere except for some pollution “hot spots”. However, it should be mentioned that these estimates do not take into account possible input of cadmium from agricultural areas with fertilisation and, therefore, they are probably underestimated.



a b c

Fig. 3.29*. Exceedances (AAE) of critical loads of lead (a), cadmium (b) and mercury (c) in 2010.*

As it follows from the description of the modelling results for 1990–2010, re-suspension is an important part of heavy metal deposition. However, in the calculation of the critical loads this outflux of re-suspension from soil is not taken into account. Besides, in the context of this study, measures aimed at reducing re-suspension from agricultural sources have not been considered. Within an effects-based approach three solutions are conceivable:

A. Since critical loads do not take re-suspension into account, it should be deducted from the deposition before calculating an exceedance.

B. Add the re-emission (at critical level) to the critical load.

C. Another way to assess scenarios all-together is to model concentrations of heavy metals in the soil and the soil solution dynamically. This would result in violations rather than exceedances of the critical load, i.e. areas where the concentration in a particular year exceeds the critical limit.

Two sources of the metal in the soil can be distinguished, from the parent material (as a mineral), and anthropogenic from either historic deposition or otherwise, for example by fertilizer input. For all options more knowledge on re-suspension is needed to assess the need for measures.

3.5. Uncertainties of pollution assessment

Assessment of pollution levels includes evaluation of emission inventories, monitoring data and modelling results of air concentrations, deposition and transboundary fluxes. Quality of each of the component comprises quality of the assessment. Uncertainties of the emission data of lead, cadmium and mercury are described in Chapter 1 of this report. This section is focused on the uncertainties of the monitoring data and model estimates.

Monitoring

Uncertainty of monitoring data depends on a number of factors such as sampling procedure, storing and analysis of samples in laboratories, possible sample contamination etc. Estimation of the uncertainty caused by analytical methods is performed by regular intercomparisons of national laboratories involved in the analysis of lead and cadmium measurements sampled at the EMEP stations. According to the results of these intercomparisons for several recent years, accuracy for majority of laboratories participated in these exercises match the data quality objective criteria (DQO). The DQO accepted 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. However, it should be noted that

annual intercomparison studies started only in 1999. Hence, quality of measurements before this year is unknown, and likely, is lower compared to the second decade of the considered period.

Laboratory intercomparisons provide evaluation of the accuracy of analytical methods. Overall measurement accuracy can be estimated by field campaigns. Field comparison of measurements of total gaseous mercury concentrations held in May, 2005, demonstrated that the results of most of the laboratories, participated in the comparison, fell within ±30% range, and for concentrations in precipitation - within ±40% range [*Aas*, 2006]. Uncertainty of wet deposition of lead and cadmium, estimated on the base of the results of 2006-2007 field campaign, was around 20% [*Aas et al.*, 2009]. However, these estimates do not take into account the effect of representativeness of station location.

Modelling

Uncertainty of the modelling results with regard to inaccuracies of input parameters was evaluated [*Travnikov and Ilyin*, 2005]. Intrinsic model uncertainties for lead, cadmium and mercury are summarized in Table 3.1. These uncertainties include effect of inaccuracies of input parameters except for anthropogenic and natural emissions, re-emission and wind re-suspension of particulate components. The range indicates 90% confidence interval of the uncertainty variation over the model domain. The intrinsic model uncertainty of particle-bound heavy metals (Pb, Cd) varies from 20% to 65% over the domain with average values 43%, 40% and 33% for concentration in air, concentration in precipitation and total deposition, respectively. The intrinsic model uncertainty of Hg differs for different outputs. It does not exceed 20% on average for TGM concentration (the range 16-22%) but reaches 40% for total deposition and 50% for concentration in precipitation (the ranges 20-57% and 29-74%, respectively).

Table 3.1. *Model intrinsic uncertainty of the main model output parameters*

Output parameter	Uncertainty, %	
	Pb, Cd	Hg
Air concentration	43 (22-64)	19 (16-22)
Concentration in precipitation	40 (20-57)	53 (29-74)
Total deposition	33 (19-49)	39 (20-57)

Another part of evaluation of the model uncertainties is comparison of modelled values with field observations. Concentrations in air and wet deposition fluxes measured at the EMEP stations were utilised for the comparison. Quality control of measurement data was undertaken. The procedure of the control is described in [*Ilyin and Travnikov*, 2005]. In particular, values below detection limit were omitted and unrealistically high outliers were removed form the comparison.

Results of the comparison of modelled and observed values differ for lead, cadmium and mercury. As seen from Fig. 3.29, most of modelled annual concentrations of lead in air and wet deposition match the observed parameters with a factor of two. There are some low observed values which were underestimated by the model. These are stations mostly located in Scandinavia (Finland, Norway).

Most of the modelled cadmium levels agree with the observed levels within a factor of two as well (Fig. 3.30b). However, overall underestimation of the observed concentrations and wet deposition is obvious. Relatively good agreement between modelled and measured values is noted for stations in Germany, Denmark, Sweden, Belgium, the United Kingdom and the Netherlands. Underestimation of the observed levels often takes place at stations in Norway, Finland, Lithuania, and Latvia. Besides,

significant underestimation of cadmium levels is noted for the first half of the considered period, which may be explained by uncertainties of measurements.

Modelled and observed mercury background concentrations in air exhibit low spatial and temporal variability compared to lead and cadmium. As a result, both modelled and measured values range mostly within 1.5 -2 ng/m^3 limits (Fig. 3.30c). Modelled wet deposition fluxes of mercury reasonably agree with observations (Fig. 3.30c). The model tends to somewhat overestimate low deposition levels measured at stations in the United Kingdom.

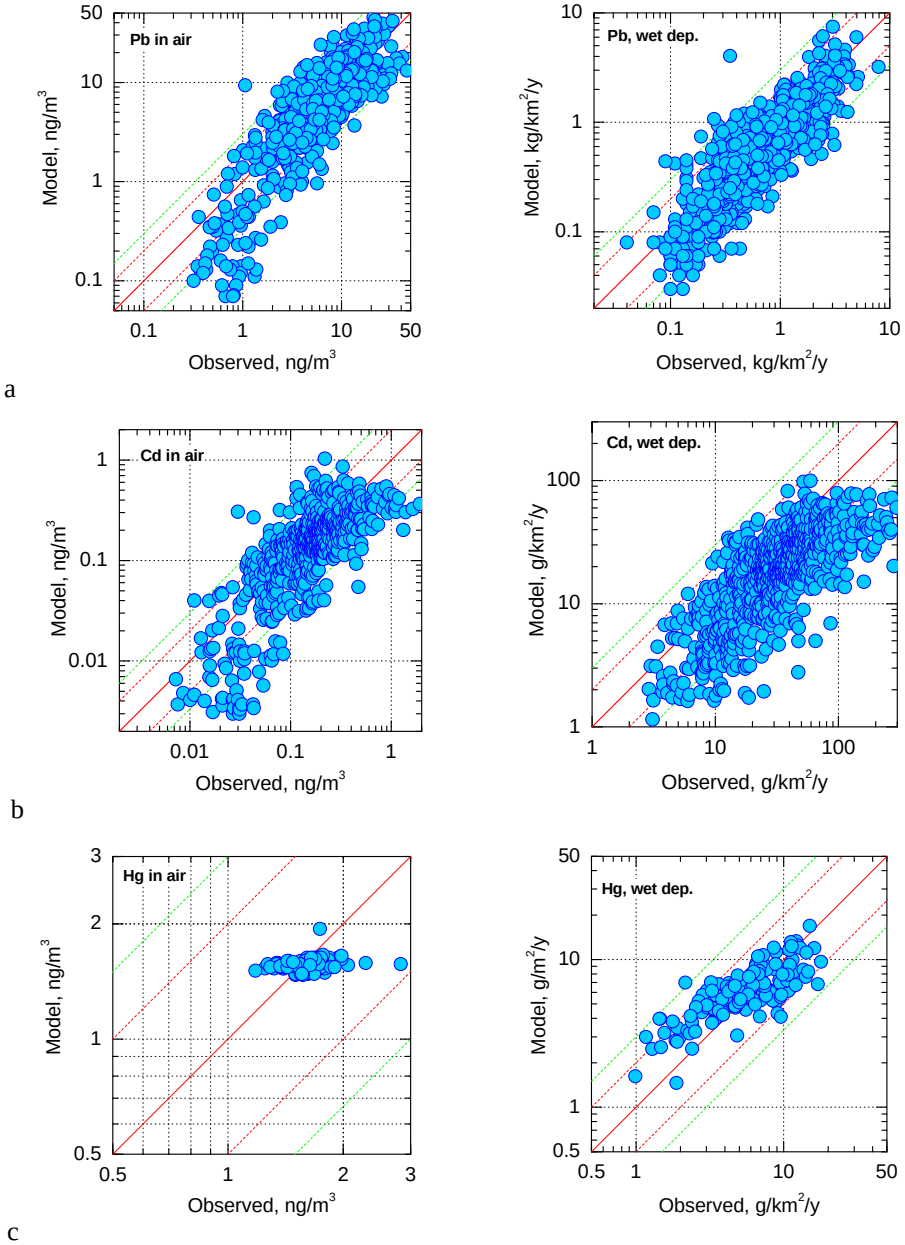


***Fig. 3.30**. Modelled vs. observed concentrations of lead (a), cadmium (b) and mercury (c) in air (left) and wet deposition fluxes (right). Solid red line depicts 1:1 ratio; dashed lines: deviation within factor 2 (red) and factor of 3(green)*

Degree of agreement between modelled and observed parameters was characterized by statistical indicators (Table 3.2). These are mean normalized bias (MNB), normalized root mean square error (NRMSE), correlation coefficient (R_{corr}) and a fraction of stations for which the ratio between modelled and measured value falls within a certain factor. For example, F2 relates to two-fold deviation, F3 – three-fold deviation.

Modelled levels of lead satisfactory agree with the observed concentrations and deposition. The bias is below than the uncertainty of the model, and comparable with the data quality objective criteria for analytical methods. Besides, correlation coefficients are significant, assuming that the model is able to reproduce temporal and spatial trends of lead pollution levels.

Table 3.2. *Statistical parameters of the model-to-observation comparison for concentrations in air and wet deposition fluxes*

	Pb, wet dep.	Pb, air conc.	Cd, wet dep.	Cd, air conc.	Hg, wet dep.	Hg, air conc.
NRMSE*	0.62	0.69	0.96	0.99	0.39	0.14
MNB**:	-14.8	-3.6	-48.0	-18.2	2.7	-3.4
R_{corr}:	0.76	0.69	0.59	0.51	0.73	0.24
F2:	75	75	55	69	92	100
F3:	95	92	81	87	99	100
N	796	642	772	586	163	99

* $\mathrm{NRMSE} = \frac{1}{\overline{O}} \cdot \sqrt{\frac{\sum_{i}^{N} (M_i - O_i)^2}{N}}$ ** $\mathrm{MNB} = \frac{(\overline{M} - \overline{O})}{\overline{O}} \cdot 100\%$

M_i, O_i –modelled and observed values at i^{th} station. $\overline{M}$, $\overline{O}$ - averaged modelled and observed values, *N* – number of model-measurement pairs

Modelling uncertainties of cadmium levels are higher compared to those for lead. The bias for wet deposition fluxes is around -50% and only 55% of model-observation pairs satisfy criterion of a factor of two. It means that the model tends to underestimate cadmium wet deposition. This overall underestimation is mainly conditioned by high (3 times or ever more) discrepancies for certain stations located mostly in Scandinavia, in Baltic countries and the Czech Republic. It likely caused by uncertainties of the emission data. Another reason could be uncertainties of measurements. Concentrations of cadmium in precipitation are much lower than those of lead, hence their monitoring is more challenging task. For concentrations in air the agreement between modelled and observed levels is better than that for wet deposition. The bias is around -18% means only little underestimation of the observed concentrations taken into account uncertainties of the model, emissions and monitoring.

Modelled mercury wet deposition agree with the observed levels with considerable accuracy. The bias and NRMSE are small compared to other considered metals. The bias and NRMSE are also low for concentrations of mercury in air, which indicates good agreement between modelled and observed levels. However, correlation coefficient is not high (0.24). It is explained by the fact that spatial and temporal variability of mercury background concentrations in air is low and comparable with accuracy of its measurements and uncertainty of the model.

The considered statistical parameters were evaluated for each year in the period from 1990 to 2010 (Annex C). Variability of the statistical indicators within this period caused by evolution of monitoring network, changes of quality of measurement data as well as ability of the model to reproduce conditions of each particular year.

Mean bias for wet deposition of lead ranges from about -34% to 9%. For air concentrations the range is -25% - 17%. Therefore, the levels of lead in each year were captured reasonably well by the model. Almost in all years at least 2/3 of model-observation pairs for concentrations and deposition agree with a factor of two.

For wet deposition of cadmium the observed levels are underestimated by the model in all years of the considered period: the bias ranges from -69% to -26%. As a rule, the highest underestimation takes place in the beginning of the period, tending to decline in the end. Similar trend is noted for cadmium concentrations in air: underestimation of about -50% in nineties is replaced by some (33%) overestimation in the end of the period.

Mean normalized bias for mercury wet deposition flux varies with ±35%. Unlike cadmium, the bias does not exhibit any distinct tendency to rise or decline within the considered period. The bias for concentration in air stays within ±20% over most of the period 1990-2010. The exception is early nineties. It is worth mentioning that in early nineties the number of stations reporting measured wet deposition of mercury was very low (2-3 stations). Besides, it is difficult to assess quality of mercury monitoring data in this period. Hence the statistical indexes for these years are not rather reliable.

To summarize the results of the comparison described above it is possible to conclude that the model is capable of reproducing levels and their long-term trends of lead and mercury pollution as well as cadmium concentrations in air with satisfactory accuracy, keeping in mind uncertainties of the model as such, emissions and monitoring data. The uncertainties of modeled wet deposition of cadmium are higher compared to that for lead and mercury. Possible reasons of this, such as uncertainties of model parameterizations, completeness and accuracy of emission data and uncertainties of measurements require further detailed investigation.

CONCLUSIONS

Long-term changes of heavy metal pollution in the EMEP countries have been assessed on the base of the integrated analysis of information on atmospheric emissions, ambient measurements and model simulations. Temporal trends and changes in transboundary fluxes of lead, cadmium and mercury over the period 1990-2010 were calculated. Contribution of the key source categories to heavy metal pollution in the EMEP countries were evaluated. The main conclusions of the analysis are formulated below.

- Forty one countries signed or ratified the Protocol on heavy metals (as of May 2012). Since then the number of Parties that reported emission data increased from 30 to 46. Gridded emission data are reported only by 28 Parties.

- Anthropogenic emissions were significantly reduced in the EMEP countries over the last two decades. Lead emissions dropped by 90% since 1990, whereas emissions of cadmium decreased approximately by 65%, and mercury – by 60%, respectively.

- The EMEP monitoring network for heavy metals has been developing continuously since 1999. The number of monitoring sites measuring lead, cadmium and mercury increased from 44 in 1990 to 66 in 2010. The monitoring network covers significant part of the EMEP countries. However, large territories in Eastern and Southern Europe as well as in Central Asia remain uncovered.

- Measurements of lead and cadmium in air and precipitation demonstrate substantial decrease of pollution levels in Europe since 1990. In contrary, long-term observations of mercury available at few sites show less significant changes.

- Atmospheric deposition of lead decreased on average by 75% over the period 1990-2010 mainly due to the phase-out of leaded gasoline from use in road transport. The reduction varies from 18% to 88% in different countries. Human health and the environment continue to be at risk in many EMEP countries despite important reductions of lead deposition.

- Since 1990 cadmium deposition levels have reduced by 50% on average and the changes ranged from about 60% reduction in some countries of Western and Central Europe to moderate increase in the countries of Caucasus and Central Asia. Nevertheless, cadmium remains to be an unresolved problem in many ‘hot spots’ located close to industrial regions.

- Decrease of mercury atmospheric deposition during the last two decades did not exceed 30% on average (varying from reduction by 70% to slight growth in different countries) due to large contribution of emissions from other continents. Nowadays, intercontinental transport contributes up to 65% of total mercury deposition in the EMEP countries. Therefore, both regional and global efforts are needed to reduce mercury pollution.

- In spite of deposition reduction of pollution levels in Europe transboundary transport continues to play an important role in heavy metal pollution of the EMEP countries. Change in the emission pattern led to redistribution of transboundary fluxes between the countries. Contribution of foreign sources to heavy metal anthropogenic deposition has changed substantially in some countries but still remains significant in most of them.

- Reduction of heavy metal pollution levels was accompanied by changes in the key source categories of both emissions and deposition. Atmospheric emissions of heavy metals from

some emission sectors were reduced more substantially than releases from other sectors. It resulted in change of the sectoral composition of heavy metal pollution in the EMEP countries.

- Four key source categories – *stationary combustion in industry*, *non-industrial combustion*, *metal production* and *public electricity and heat production* – make up the largest contribution to current pollution with all three metals and require priority mitigation measures in future.

- Heavy metal pollution in the countries of the Eastern Europe, Caucasus and Central Asia (EECCA) is not characterized adequately due to the lack of observations and official information on emissions. Therefore, additional attention should be paid to the development of national emission inventories and monitoring networks in these countries.

Thus, although heavy metal pollution levels have been reduced considerably in the EMEP countries, they are still high enough to pose a significant risk to human health and the environment at present and in future.

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ANNEX A

HM EMISSION TREND TABLES

Complete emission trend tables of reported pollutants can be downloaded from the CEIP website http://www.ceip.at/overview-of-submissions-under-clrtap/2012-submissions.

Table A1. *Cd emission trend table, Mg*

Country	1990	1995	2000	2005	2006	2007	2008	2009	2010	Change 1990 - 2010
Albania	0.31	0.08	0.04	0.06	0.06	0.06	0.08	0.12		-62%
Armenia						0.21				
Austria	1.58	0.98	0.92	1.06	1.06	1.09	1.12	1.04	1.14	-28%
Azerbaian										
Belarus	2.11	1.11	1.38	2.07	2.49	2.58	2.75	3.02	3.23	53%
Belgium	7.20	5.27	3.05	2.68	2.75	2.51	2.78	2.05	2.65	-63%
Bosnia & Herzegovina										
Bulgaria	5.22	3.64	3.46	2.94	2.82	2.52	3.22	2.18	1.90	-64%
Canada	91.44	29.25	38.06	34.94	40.47	28.17	22.50	20.36	15.88	-83%
Croatia	1.32	0.92	0.65	0.60	0.58	0.51	0.47	0.46	0.58	-56%
Cyprus	0.05	0.06	0.08	0.09	0.08	0.08	0.09	0.08	0.07	45%
Czech Republic	4.34	3.55	2.85	3.11	3.18	2.91	3.76	3.37	0.88	-80%
Denmark	0.99	0.50	0.35	0.26	0.22	0.23	0.23	0.19	0.19	-80%
Estonia	4.40	1.96	0.56	0.58	0.55	0.68	0.61	0.48	0.67	-85%
European Union	257.53	205.89	149.78	126.69	120.91	108.64	107.76	95.84	103.07	-60%
Finland	6.34	1.65	1.29	1.31	1.29	0.96	1.23	1.29	1.41	-78%
France	20.57	17.96	14.12	6.23	4.62	4.27	4.22	2.87	2.85	-86%
FYR of Macedonia	0.20	0.38	0.40	0.15	0.16	0.14	0.17	0.17	0.18	-10%
Georgia										
Germany	17.26	11.40	10.23	7.32	6.92	6.39	5.49	4.73	5.31	-69%
Greece										
Hungary	5.51	3.76	3.05	1.53	1.74	1.48	1.64	3.42	0.71	-87%
Iceland	NR	NR	NR	NR	NR	NR	NR	NR	NR	
Ireland	0.84	0.95	1.06	0.72	0.64	0.63	0.57	0.42	0.42	-50%
Italy	10.06	9.33	8.77	8.13	8.27	8.92	8.73	7.14	8.19	-19%
Kazakhstan										
Kyrgyzstan										
Latvia	0.32	0.19	0.19	0.22	0.22	0.22	0.21	0.20	0.23	-29%
Liechtenstein										
Lithuania	3.80	2.10	1.35	0.37	0.37	0.40	0.31	0.45	0.43	-89%
Luxembourg	NR	NR	NR	NR	NR	NR				
Malta			0.48	0.59	0.60	0.62	0.56	0.56	0.04	
Monaco	0.06	0.01	0.01	0.01	0.00	0.01	0.01	0.00	0.00	-92%
Montenegro	0.08	0.02	0.06	0.06	0.07	0.07	0.09	0.05		-38%
Netherlands	2.09	1.07	0.92	1.67	1.93	1.70	1.87	1.79	2.51	20%
Norway	1.17	1.05	0.78	0.64	0.69	0.63	0.61	0.48	0.59	-50%
Poland	91.60	82.60	50.40	46.02	43.45	40.35	39.81	36.84	44.26	-52%
Portugal	5.42	5.75	5.53	6.17	5.17	5.41	5.03	3.41	3.76	-31%
Republic of Moldova	2.42	0.59	0.17	0.15	0.16		0.17	0.13		-95%
Romania	NE	NE	NE	3.10	3.14	3.68	3.16	2.82	2.17	-30%
Russian Federation	79.40	57.40	50.50	59.40	59.40			22.60	NE	-72%
Serbia			2.56	1.85	1.85	1.85	1.98	1.99	1.94	-24%
Slovakia	9.44	9.98	7.05	5.99	5.92	1.22	1.23	1.03	1.21	-87%
Slovenia	0.87	0.71	0.59	0.53	0.55	0.56	0.64	0.59	0.58	-33%
Spain	26.78	23.20	20.37	18.89	18.31	15.39	14.97	13.03	15.51	-42%
Sweden	2.27	0.73	0.51	0.53	0.55	0.55	0.51	0.54	0.58	-74%
Switzerland	3.87	2.64	1.80	1.28	1.34	1.26	1.26	1.23	1.26	-67%
Turkey										
Ukraine				6.84	5.10	8.95	8.32	4.44	2.81	-59%
United Kingdom	23.05	11.28	6.03	3.65	3.57	2.85	2.77	2.31	2.39	-90%
USA	180.00			63.00				63.00		-65%

Note: For countries which did not report 2010 or 1990 emissions, the closest values have been used to calculate the difference in last column.

Table A2. *Pb emission trend table, Mg*

Country	1990	1995	2000	2005	2006	2007	2008	2009	2010	Change 1990 - 2010
Albania	62	67	76	60	4	4	4	4		-94%
Armenia	11	0.3				9				
Austria	219	16	12	14	14	14	15	13	15	-93%
Azerbaian										
Belarus	794	147	46	50	57	59	63	66	70	-91%
Belgium	491	259	106	78	73	64	73	34	43	-91%
Bosnia & Herzegovina										
Bulgaria	321	333	267	127	128	118	225	112	107	-67%
Canada	1,227	754	548	249	271	266	239	221	188	-85%
Croatia	536	323	273	51	46	42	38	33	28	-95%
Cyprus	25	26	21	2	2	3	3	3	3	-90%
Czech Republic	269	180	108	47	43	44	39	40	26	-90%
Denmark	125	23	17	16	14	12	12	11	11	-91%
Estonia	205	85	36	35	31	40	35	28	39	-81%
European Union	23,156	10,835	4,615	2,987	3,000	2,904	2,813	2,336	2,549	-89%
Finland	338	67	45	22	25	22	20	18	23	-93%
France	4,258	1,434	239	125	116	112	99	74	83	-98%
FYR of Macedonia	96	98	105	23	7	6	7	7	7	-92%
Georgia										
Germany	2,075	693	433	352	348	339	199	174	191	-91%
Greece										
Hungary	663	130	42	38	37	35	36	32	17	-97%
Iceland	NR	NR	NR	NR	NR	NR	NR	NR	NR	
Ireland	125	80	18	20	19	19	19	15	14	-89%
Italy	4,414	2,028	944	280	288	312	301	232	270	-94%
Kazakhstan										
Kyrgyzstan										
Latvia	92	63	10	8	9	9	8	8	8	-91%
Liechtenstein										
Lithuania	47	30	16	6	6	7	4	3	3	-94%
Luxembourg	NR	NR	NR	NR	NR	NR				
Malta			1	1	1	1	1	1	3	
Monaco	4	0.8	0.06	0.04	0.03	0.04	0.04	0.04	0.04	-99%
Montenegro	309	141	142	48	46	52	47	45		-85%
Netherlands	336	159	33	35	36	41	36	37	44	-87%
Norway	187	23	8	7	7	8	7	4	5	-98%
Poland	1,372	937	648	536	589	553	510	459	524	-62%
Portugal	566	785	73	189	180	139	186	151	205	-64%
Republic of Moldova	249	34	3	5	5		4	3		-99%
Romania	NE	NE	NE	107	105	106	92	54	61	
Russian Federation	3,591	2,426	2,352	355	355			32	NE	-99%
Serbia			44	47	47	47	67	93	148	
Slovakia	150	71	67	70	72	60	61	42	56	-63%
Slovenia	357	260	63	13	15	15	15	14	14	-96%
Spain	2,788	967	627	274	277	277	271	237	247	-91%
Sweden	355	37	26	14	14	14	9	13	13	-96%
Switzerland	352	168	37	25	25	25	25	23	23	-93%
Turkey										
Ukraine				304	297	309	213	171	159	
United Kingdom	2,887	1,529	149	107	88	79	73	63	59	-98%
USA	2,996	3,577		1,230				1,230		-59%

Note: For countries which did not report 2010 or 1990 emissions, the closest values have been used to calculate the difference in the last column.

Table A3. *Hg emission trend table, Mg*

Country	1990	1995	2000	2005	2006	2007	2008	2009	2010	Change 1990 - 2010
Albania	0.27	0.04	0.02	0.05	0.06	0.10	0.12	0.12		-57%
Armenia	0.01	0.00				0.35				
Austria	2.14	1.20	0.89	0.99	1.01	1.01	1.03	0.89	0.99	-54%
Azerbaian										
Belarus	1.07	0.51	0.36	0.65	0.72	0.74	0.81	0.91	0.85	-21%
Belgium	6.79	3.51	3.30	2.49	2.32	3.40	3.84	2.03	2.05	-70%
Bosnia & Herzegovina										
Bulgaria	2.44	1.94	1.48	1.55	1.70	1.53	1.39	1.00	0.88	-64%
Canada	35.14	12.12	8.00	6.21	5.48	7.85	7.03	6.35	5.22	-85%
Croatia	1.45	0.40	0.55	0.83	0.71	0.76	0.76	0.62	0.75	-48%
Cyprus	0.15	0.17	0.17	0.18	0.18	0.18	0.18	0.15	0.13	-15%
Czech Republic	7.52	7.40	3.84	3.77	3.85	3.92	4.11	4.30	3.48	-54%
Denmark	3.06	2.35	1.05	0.81	0.70	0.68	0.68	0.52	0.44	-86%
Estonia	1.12	0.60	0.51	0.52	0.52	0.65	0.57	0.44	0.63	-44%
European Union	229.59	169.08	131.36	113.24	107.73	107.55	102.02	88.70	86.60	-62%
Finland	1.15	0.73	0.59	0.85	1.00	0.83	0.78	0.77	0.90	-22%
France	23.97	19.61	11.01	6.22	6.25	4.47	4.30	3.86	4.18	-83%
FYR of Macedonia	0.24	0.55	0.55	0.28	0.28	0.24	0.39	0.30	0.39	65%
Georgia										
Germany	28.26	13.85	13.88	11.68	11.38	11.01	9.80	8.88	9.29	-67%
Greece										
Hungary	6.26	4.86	4.36	4.15	3.16	2.83	3.01	2.82	0.78	-88%
Iceland	NR	NR	NR	NR	NR	NR	NR	NR	NR	
Ireland	0.87	0.77	0.72	0.85	0.79	0.87	0.77	0.49	0.42	-51%
Italy	11.49	10.29	9.15	9.92	10.18	10.79	10.38	8.61	9.52	-17%
Kazakhstan										
Kyrgyzstan										
Latvia	0.24	0.09	0.06	0.07	0.07	0.08	0.07	0.07	0.08	-69%
Liechtenstein										
Lithuania	0.02	0.15	0.25	0.41	0.42	0.43	0.27	0.62	0.39	2050%
Luxembourg	NR	NR	NR	NR	NR	NR				
Malta			0.49	0.61	0.61	0.63	0.57	0.57	0.01	
Monaco	0.11	0.07	0.08	0.06	0.04	0.06	0.05	0.05	0.05	-55%
Montenegro	0.07	0.02	0.07	0.06	0.08	0.06	0.09	0.05		-32%
Netherlands	3.51	1.41	1.01	0.85	0.79	0.77	0.66	0.63	0.69	-80%
Norway	1.46	0.84	0.73	0.69	0.64	0.65	0.60	0.51	0.58	-60%
Poland	33.30	32.30	25.60	20.10	16.08	16.12	15.65	14.22	14.85	-55%
Portugal	3.80	4.00	3.73	3.31	2.89	2.70	2.56	2.46	2.06	-46%
Republic of Moldova	3.37	0.89	0.26	0.24	0.22		0.31	0.24		-93%
Romania	NE	NE	NE	7.41	7.62	10.64	8.28	4.51	5.34	-28%
Russian Federation	15.60	10.40	10.00	14.00	14.00			0.98	NE	
Serbia			1.37	1.57	1.57	1.57	1.64	1.64	1.62	18%
Slovakia	12.47	3.93	5.20	2.79	3.31	2.39	2.65	1.04	1.18	-91%
Slovenia	1.17	0.91	0.93	0.79	0.80	0.84	0.90	0.83	0.66	-44%
Spain	14.57	14.70	12.67	12.06	11.24	10.37	9.50	8.00	7.82	-46%
Sweden	1.58	1.04	0.73	0.69	0.55	0.59	0.53	0.59	0.55	-65%
Switzerland	6.71	4.13	2.19	1.10	1.14	1.10	1.12	0.97	1.05	-84%
Turkey										
Ukraine				5.96	15.65	7.56	6.79	5.62	6.79	14%
United Kingdom	37.62	19.69	8.12	7.19	7.31	6.84	6.52	7.41	6.29	-83%
USA	187.00	146.00		103.00				103.00		-45%

Note: For countries which did not report 2010 or 1990 emissions, the closest values have been used to calculate the difference in the last column.

Annex B

INDICATOR OF UNCERTAINTY

Table B1. *2005 Cd emissions as reported between 2007 and 2012, Mg*

Party	2005 as reported in						Difference
	2012	2011	2010	2009	2008	2007	Max-Min
Austria	1.06	1.08	1.08	1.17	1.11	1.08	10.1%
Belarus	2.07	2.07	2.07	2.07	2.07	2.07	0.0%
Belgium	2.68	2.92	1.72	1.71	1.68	1.99	46.4%
Bulgaria	2.95	12.12	12.12	12.12	12.12	12.12	311.5%
Canada	34.94	34.94	34.94	35.00	34.92	34.92	0.2%
Croatia	0.60	0.83	0.92	0.83	0.83	0.83	53.7%
Cyprus	0.09	0.08	0.08	1.13	1.12	1.12	1216.3%
Czech Republic	3.11	3.11	3.11	3.11	3.11	3.11	0.0%
Denmark	0.26	0.26	0.43	0.65	0.65	0.62	151.9%
Estonia	0.58	0.58	0.58	0.58	0.58	0.58	1.0%
Finland	1.31	1.31	1.30	1.30	1.30	1.30	0.2%
France	6.23	5.77	5.78	5.53	6.57	5.94	16.6%
Germany	7.32	6.48	2.57	2.50	2.69	2.71	65.8%
Hungary	1.53	1.53	1.53	1.53	1.53	1.53	0.0%
Ireland	0.72	0.72	0.72	0.72	0.58	0.58	20.1%
Italy	8.13	8.10	8.52	8.46	8.17	8.16	5.2%
Latvia	0.22	0.22	0.24	0.50	0.50	0.54	146.4%
Lithuania	0.37	0.37	0.37	0.37	0.37	0.37	0.0%
Malta	0.59	0.59	0.59	0.59	0.59	0.63	5.7%
Monaco	0.01	0.01	0.01	0.01	0.01	0.01	0.0%
Netherlands	1.67	1.67	1.68	1.68	1.68	1.73	3.2%
Norway	0.64	0.64	0.54	0.54	0.54	0.51	19.6%
Poland	46.02	46.02	46.02	46.02	46.02	46.02	0.0%
Portugal	5.44	6.17	6.24	6.20	5.88	5.67	14.8%
Republic of Moldova	0.15	0.15	0.15	0.15	0.15	0.15	0.0%
Romania	3.10	3.10	10.05	10.05	10.05	2.95	229.1%
Russian Federation	59.40	59.40	59.40	59.40	59.40	59.40	0.0%
Slovakia	5.99	7.17	7.22	6.10	6.10	6.10	20.5%
Slovenia	0.40	0.53	0.53	0.44	1.16	1.68	323.2%
Spain	18.89	18.86	18.49	18.48	18.47	18.37	2.8%
Sweden	0.53	0.53	0.53	0.53	0.51	0.53	3.0%
Switzerland	1.28	1.29	1.29	1.12	1.12	1.11	13.5%
Ukraine	6.84	6.84	6.84	6.84	6.84	6.84	0.0%
United Kingdom	3.65	3.59	3.53	3.53	3.74	3.82	8.1%

Notes: The table includes only Parties which reported HM emissions at least for one year
Red shaded cells indicate deviations of more than 50%

Table B2. *2005 Pb emissions as reported between 2007 and 2012, Mg*

Party	2005 as reported in						Difference
	2012	2011	2010	2009	2008	2007	Max-Min
Austria	13.56	13.68	13.71	14.14	13.71	13.57	4.3%
Belarus	50.14	50.14	50.14	50.14	50.14	50.14	0.0%
Belgium	78.36	86.24	75.86	75.93	77.18	78.22	13.2%
Bulgaria	127.45	114.81	114.81	114.81	114.81	114.81	9.9%
Canada	248.87	248.87	248.87	233.00	232.63	232.63	6.5%
Croatia	51.02	50.70	49.61	11.67	11.67	11.67	77.1%
Cyprus	2.26	2.24	2.23	9.14	3.76	3.76	305.6%
Czech Republic	47.08	47.08	47.08	47.08	47.08	47.08	0.0%
Denmark	15.66	15.47	8.93	5.67	5.72	5.58	64.4%
Estonia	35.03	34.55	33.76	36.68	36.68	36.68	8.3%
Finland	22.00	22.13	23.51	23.51	23.51	23.51	6.8%
France	124.57	122.31	122.36	123.09	138.16	134.42	12.7%
Germany	352.47	349.25	112.25	103.57	107.22	106.76	70.6%
Hungary	37.53	37.53	37.53	37.53	37.53	37.53	0.0%
Ireland	19.84	16.73	16.63	15.05	16.52	7.90	60.2%
Italy	280.02	279.37	277.60	265.73	265.70	263.44	5.9%
Latvia	8.37	8.37	8.04	15.20	14.28	16.73	103.8%
Lithuania	5.66	5.66	5.66	5.66	5.66	5.66	0.0%
Malta	0.79	0.79	0.79	0.82	0.82	1.91	142.3%
Monaco	0.04	0.04	0.04	0.04	0.04	0.04	0.0%
Netherlands	35.48	35.92	38.63	38.60	38.65	43.78	23.4%
Norway	7.09	6.87	6.31	6.31	7.57	5.85	24.3%
Poland	536.48	536.48	536.48	536.48	536.48	536.48	0.0%
Portugal	192.32	119.15	78.62	157.63	163.14	243.75	85.9%
Republic of Moldova	5.06	5.06	5.06	5.06	5.06	5.06	0.0%
Romania	106.71	106.71	162.48	162.48	162.48	218.39	104.7%
Russian Federation	355.00	355.00	355.00	355.00	355.00	355.00	0.0%
Slovakia	69.76	83.85	83.10	70.59	70.59	70.59	20.2%
Slovenia	14.12	13.08	13.08	13.49	16.99	14.19	27.7%
Spain	274.48	273.91	273.52	273.59	273.46	272.34	0.8%
Sweden	14.38	14.56	14.56	14.59	14.55	16.54	15.0%
Switzerland	24.99	25.93	27.04	20.75	24.49	20.17	27.5%
Ukraine	304.38	304.38	304.38	304.38	304.38	304.38	0.0%
United Kingdom	107.48	109.54	108.85	109.09	116.95	117.53	9.4%

Notes: The table includes only Parties which reported HM emissions at least for one year
Red shaded cells indicate deviations of more than 50%

Table B3. *2005 Hg emissions as reported between 2007 and 2012, Mg*

Party	2005 as reported in						Difference
	2012	2011	2010	2009	2008	2007	Max-Min
Austria	0.99	1.00	1.00	1.03	1.00	0.98	5.2%
Belarus	0.65	0.65	0.65	0.65	0.65	0.65	0.0%
Belgium	2.49	2.46	1.82	1.82	1.82	1.92	27.0%
Bulgaria	1.55	3.38	3.38	3.38	3.38	3.38	117.7%
Canada	6.21	6.21	6.21	6.00	6.16	6.16	3.3%
Croatia	0.83	0.83	0.73	0.69	0.69	0.69	16.4%
Cyprus	0.18	0.18	0.18	1.29	1.28	1.28	627.1%
Czech Republic	3.77	3.77	3.77	3.77	3.77	3.77	0.0%
Denmark	0.82	0.81	1.01	1.38	1.35	1.29	70.3%
Estonia	0.52	0.52	0.52	0.52	0.52	0.52	0.0%
Finland	0.85	0.85	0.85	0.85	0.85	0.85	0.0%
France	6.22	6.20	6.00	8.65	9.15	8.61	50.7%
Germany	11.68	8.18	3.80	3.77	2.72	2.65	77.3%
Hungary	4.15	4.15	4.15	4.15	4.15	4.15	0.0%
Ireland	0.85	0.85	0.82	0.85	0.42	0.41	51.4%
Italy	9.92	9.91	10.39	10.39	10.38	10.35	4.8%
Latvia	0.07	0.07	0.07	0.03	0.03	0.06	59.7%
Lithuania	0.41	0.41	0.41	0.41	0.41	0.41	0.0%
Malta	0.61	0.61	0.61	0.60	0.60	0.62	2.6%
Monaco	0.06	0.06	0.06	0.06	0.06	0.06	0.0%
Netherlands	0.85	0.81	0.81	0.81	0.81	1.02	23.8%
Norway	0.69	0.69	0.69	0.69	0.69	0.69	1.2%
Poland	20.10	20.10	20.10	20.10	20.10	20.10	0.0%
Portugal	2.92	3.32	3.36	3.39	3.25	4.12	41.2%
Republic of Moldova	0.24	0.24	0.24	0.24	0.24	0.24	0.0%
Romania	7.41	7.41	11.46	11.46	11.46	4.33	96.4%
Russian Federation	14.00	14.00	14.00	14.00	14.00	14.00	0.0%
Slovakia	2.79	3.96	3.96	2.90	2.90	2.90	41.9%
Slovenia	0.48	0.79	0.79	1.06	0.64	0.64	123.3%
Spain	12.06	11.99	11.89	11.89	11.83	11.70	3.0%
Sweden	0.69	0.69	0.69	0.73	0.73	0.75	8.1%
Switzerland	1.10	1.14	1.14	1.05	1.07	1.03	10.4%
Ukraine	5.96	5.96	5.96	5.96	5.96	5.96	0.0%
United Kingdom	7.19	7.10	7.13	7.09	7.18	7.57	6.7%

Notes: The table includes only Parties which reported HM emissions at least for one year
Red shaded cells indicate deviations of more than 50%

Annex C

STATISTICS OF THE MODEL EVALUATION

Table C.1. *Statistical parameters of the model-to-observation comparison for* ***wet deposition of lead*** *in 1990 - 2010*

Year	NRMSE	MNB, %	R_{corr}	F2	F3	N
1990	0.79	7.70	0.62	81.0	100.0	21
1991	0.79	8.82	0.60	81.0	95.2	21
1992	0.58	0.62	0.77	76.0	100.0	25
1993	0.41	-4.51	0.84	90.0	93.3	30
1994	0.47	-26.92	0.84	75.8	93.9	33
1995	0.46	-15.94	0.80	74.3	91.4	35
1996	0.63	-33.16	0.70	67.6	97.3	37
1997	0.57	-34.34	0.80	57.1	80.0	35
1998	0.47	-20.53	0.79	79.4	100.0	34
1999	0.53	-25.26	0.70	65.8	97.4	38
2000	0.55	-28.32	0.72	61.0	92.7	41
2001	0.55	-32.76	0.86	73.0	94.6	37
2002	0.46	-16.60	0.78	87.8	100.0	41
2003	0.50	-17.32	0.69	75.6	97.8	45
2004	0.41	-14.96	0.82	82.2	91.1	45
2005	0.56	-16.68	0.70	71.4	89.8	49
2006	0.64	0.62	0.58	83.3	95.8	48
2007	0.66	4.42	0.46	72.3	93.6	47
2008	0.58	-8.15	0.62	77.6	91.8	49
2009	0.62	0.89	0.56	72.0	98.0	50
2010	0.49	-13.52	0.70	77.1	97.1	35

Table C.2. *Statistical parameters of the model-to-observation comparison for* ***air concentrations of lead*** *in 1990 - 2010*

Year	NRMSE	MNB, %	Rcorr	F2:	F3:	N
1990	0.55	-6.77	0.21	71.4	100.0	14
1991	0.63	-1.73	0.31	62.5	93.8	16
1992	0.53	2.75	0.51	90.0	100.0	20
1993	0.58	3.91	0.37	76.2	100.0	21
1994	0.65	-7.23	0.34	78.3	100.0	23
1995	0.69	0.88	0.37	79.2	95.8	24
1996	0.73	0.28	0.45	71.4	92.9	28
1997	0.69	-6.66	0.53	76.0	96.0	25
1998	0.62	3.17	0.60	75.0	92.9	28
1999	0.47	-12.55	0.75	78.6	96.4	28
2000	0.41	-14.63	0.80	87.5	93.8	32
2001	0.42	-25.30	0.80	80.0	94.3	35
2002	0.43	-0.95	0.78	88.9	94.4	36
2003	0.47	-1.13	0.71	88.2	94.1	34
2004	0.41	-10.87	0.80	73.5	97.1	34
2005	0.63	0.06	0.61	66.7	94.9	39
2006	0.74	3.90	0.54	75.6	90.2	41
2007	0.93	17.54	0.57	65.1	86.0	43
2008	0.61	-16.67	0.62	63.6	84.1	44
2009	0.62	-0.78	0.78	75.0	88.6	44
2010	0.53	-10.97	0.76	66.7	75.8	33

Table C.3. *Statistical parameters of the model-to-observation comparison for* ***wet deposition of cadmium*** *in 1990 - 2010*

Year	NRMSE	MNB, %	R_{corr}	F2	F3	N
1990	1.01	-68.70	0.43	45.0	60.0	20
1991	1.13	-59.37	0.44	63.2	78.9	19
1992	1.10	-46.66	0.32	63.6	77.3	22
1993	0.90	-50.95	0.64	58.3	83.3	24
1994	0.91	-59.72	0.74	46.7	66.7	30
1995	0.71	-29.83	0.55	63.3	90.0	30
1996	0.87	-52.70	0.66	50.0	76.3	38
1997	0.95	-52.58	0.50	48.6	74.3	35
1998	0.81	-54.75	0.61	38.9	77.8	36
1999	0.91	-44.97	0.45	56.4	82.1	39
2000	0.60	-36.66	0.74	51.2	80.5	41
2001	1.06	-52.56	0.77	50.0	80.6	36
2002	0.93	-51.13	0.66	60.0	82.5	40
2003	1.06	-55.08	0.80	51.2	79.1	43
2004	0.83	-39.16	0.74	60.9	89.1	46
2005	0.73	-25.64	0.60	58.0	92.0	50
2006	0.58	-33.56	0.66	60.4	93.8	48
2007	0.86	-46.88	0.65	54.3	78.3	46
2008	0.70	-42.23	0.48	54.2	81.3	48
2009	0.91	-48.43	0.31	53.1	73.5	49
2010	0.65	-40.18	0.61	75.0	84.4	32

Table C44. *Statistical parameters of the model-to-observation comparison for* ***air concentrations of cadmium*** *in 1990 - 2010*

Year	NRMSE	MNB, %	R_{corr}	F2:	F3:	N
1990	0.77	-45.43	0.12	54.55	81.82	11
1991	0.83	-50.83	0.55	78.57	78.57	14
1992	1.13	-54.04	0.58	63.16	78.95	19
1993	0.59	-28.35	0.65	82.35	94.12	17
1994	0.60	-29.73	0.53	76.19	95.24	21
1995	0.96	-24.37	0.26	59.09	81.82	22
1996	0.78	-24.20	0.51	56.00	92.00	25
1997	0.52	-16.60	0.76	85.71	85.71	21
1998	0.46	-12.93	0.73	80.77	84.62	26
1999	0.55	-25.34	0.71	74.07	92.59	27
2000	0.56	-27.44	0.65	68.18	81.82	22
2001	0.59	-26.84	0.63	75.76	90.91	33
2002	0.50	-17.39	0.73	73.68	86.84	38
2003	0.50	-0.51	0.72	82.86	94.29	35
2004	0.58	-5.71	0.69	69.70	90.91	33
2005	0.93	17.50	0.53	57.14	85.71	35
2006	1.12	23.19	0.42	77.14	85.71	35
2007	1.20	33.51	0.60	65.79	86.84	38
2008	1.01	21.15	0.61	65.85	85.37	41
2009	1.16	33.21	0.71	67.50	90.00	40
2010	0.78	-8.65	0.51	48.48	78.79	33

Table C.5. *Statistical parameters of the model-to-observation comparison for* ***wet deposition of mercury*** *in 1990 - 2010*

Year	NRMSE	MNB, %	R_{corr}	F2:	F3:	N
1990	0.30	-21.51	0.83	100.0	100.0	3
1991	0.12	-1.44	-	100.0	100.0	2
1992	0.18	17.74	-	100.0	100.0	2
1993	0.25	4.34	-0.86	100.0	100.0	3
1994	0.34	5.98	-0.51	100.0	100.0	3
1995	0.28	-22.28	0.89	100.0	100.0	3
1996	0.47	-34.56	0.98	100.0	100.0	5
1997	0.13	-2.27	0.99	100.0	100.0	5
1998	0.37	22.02	0.86	88.9	88.9	9
1999	0.29	10.66	0.84	88.9	100.0	9
2000	0.34	15.85	0.82	87.5	100.0	8
2001	0.33	22.60	0.83	87.5	100.0	8
2002	0.34	1.56	0.82	88.9	100.0	9
2003	0.25	22.64	0.96	100.0	100.0	9
2004	0.40	-20.82	0.80	100.0	100.0	8
2005	0.33	-4.57	0.72	91.7	100.0	12
2006	0.52	-3.82	0.49	77.8	100.0	9
2007	0.55	-15.29	0.63	91.7	100.0	12
2008	0.40	-3.46	0.69	92.9	100.0	14
2009	0.53	30.85	0.71	81.3	93.8	16
2010	0.37	21.12	0.70	100.0	100.0	14

Table C.6. *Statistical parameters of the model-to-observation comparison for* ***air concentrations of mercury*** *in 1990 - 2010*

Year	NRMSE	MNB, %	R_{corr}	F2:	F3:	N
1990	0.45	-44.53	-	100	100	1
1991	0.31	-30.94	-	100	100	1
1992	0.24	-24.11	-	100	100	1
1993	0.15	-14.84	-	100	100	1
1994	0.16	-15.69	-	100	100	2
1995	0.10	1.67	-0.99	100	100	3
1996	0.16	5.16	-0.40	100	100	4
1997	0.20	19.85	0.87	100	100	3
1998	0.07	-2.95	-0.09	100	100	4
1999	0.13	-2.67	0.02	100	100	4
2000	0.11	0.75	-0.10	100	100	6
2001	0.08	-0.94	0.30	100	100	5
2002	0.10	-7.27	0.66	100	100	6
2003	0.07	0.46	0.65	100	100	7
2004	0.13	-11.77	0.63	100	100	4
2005	0.10	-3.03	0.79	100	100	7
2006	0.09	-4.75	0.19	100	100	9
2007	0.09	-4.75	0.19	100	100	9
2008	0.07	-3.32	0.67	100	100	8
2009	0.11	0.24	0.05	100	100	9
2010	0.09	6.68	-0.53	100	100	5