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Assessment of heavy metal transboundary pollution, progress in model development and mercury research

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EXECUTIVE SUMMARY

Heavy metals are toxic pollutants causing adverse health effects on humans and wildlife. Released to the atmosphere from combustion of fossil fuels and industrial activities they can be transported over long distances and deposited to the ground leading to contamination of agricultural lands, terrestrial and aquatic ecosystems. Selected heavy metals are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) since 1998 when the Protocol on Heavy Metals (hereafter, Protocol on HMs or the Protocol) came into force. Heavy metals targeted by the Protocol include lead, cadmium and mercury. Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) provides the Convention with information on deposition and transboundary transport of heavy metals within the geographical scope of EMEP.

This report outlines recent activities of the EMEP Centres in the field of heavy metal pollution assessment performed in accordance with the 2016-2017 Workplan for the implementation of the Convention [*ECE/EB.AIR/133/Add.1*]. Particular attention of the report is paid to the problem of mercury pollution both in the EMEP region and on a global scale. For this purpose, a digest of current MSC-E work in this area is given in a separate chapter to support the forthcoming discussion at the Second Joint Session of the Steering Body to EMEP and the Working Group on Effects (Geneva, September 2016).

Total anthropogenic emission in the EMEP region in 2014, officially reported by the Centre of Emission Inventories and Projections (CEIP), was 3467 tonnes for lead, 204 tonnes for cadmium and 167 tonnes for mercury. It is higher than the emissions in 2013 by 318, 34 and 8 tonnes, respectively. Completeness and uncertainties of emission data can significantly affect quality of the model estimates. Executive Body of CLRTAP initiated activity aimed at improvement of heavy metal emission inventories in the EMEP region. Following this initiative, the key parameters of heavy metal emission which affect quality of the model assessment were analysed and ranked in terms of their priority. It was indicated that quality of gridded emission data as well as chemical speciation of mercury emissions were at the top of the priority list. Other important parameters include data on temporal variability and vertical distribution of emissions for lead and cadmium, and global scale inventories as well as historical emissions for mercury. This ranking is a basis for further improvement of emission inventories under EMEP.

Measurements of lead and cadmium in the EMEP region in 2014 were carried out at 62 sites, and mercury – at 29 sites. In total, 22 Parties to the Convention reported heavy metal data to EMEP, but only 6 of them fulfilled their monitoring obligations as defined in the EMEP monitoring strategy. The lowest concentrations for all elements were generally found in northern Scandinavia, and the highest ones were noted for central and south-eastern Europe. Nowadays heavy metal monitoring data are available mainly from the western, central and northern parts of Europe, whereas in the eastern and southern parts of Europe and Central Asia the monitoring network is scarce. Therefore, more sites with continuous measurements in these regions are needed. Analysis of measurement data shows existence of low quality data at some monitoring sites. Therefore, laboratories in these countries need to evaluate their methodology.

Assessment of lead, cadmium and mercury pollution levels in the EMEP region has been performed for 2014. The highest heavy metal pollution levels are noted for the northern part of Germany, the Benelux region, the eastern part of Ukraine and the northern part of Italy. Besides, relatively high cadmium concentrations and deposition take place in the central part of Russia, and mercury levels – in the Balkan region. Country-averaged deposition fluxes of lead, cadmium and mercury from anthropogenic, secondary and non-EMEP sources in 2014 were calculated for each EMEP country. In 2014 main contribution of lead and cadmium deposition in the EMEP countries was made by secondary sources. Their contribution to total country-averaged deposition exceeded 50% in 45 countries (lead) and 41 countries (cadmium). In most of the countries the main contribution to mercury deposition was made by non-EMEP sources. In 48 countries the contribution exceeded 50%, and in 28 countries – exceeded 75%. Contribution of transboundary deposition of lead exceeded that of national sources in 24 countries. The corresponding value for cadmium was 38 countries, and for mercury – 17 countries. In most of the EMEP countries from 60% to more than 90% of emitted heavy metals were transported outside territories of the countries.

Special attention was paid to comparison of pollution levels in the EMEP domain in 2014 (year of the current assessment) and in 2013 (year of the previous assessment) and analysis of the differences. It was revealed that the changes of meteorological conditions (precipitation amounts, atmospheric transport patterns etc.) in two successive years could substantially affect pollution levels. For example, changes in atmospheric transport patterns led to growth of cadmium deposition by 75% in Sweden in 2014, whereas contributions of transboundary transport from major source countries increased more than two-fold. Therefore, the effect of year-to-year meteorological variability on source-receptor relationships in particular countries can be as strong as or even stronger than inter-annual changes in country's emission data. It means that simulation of source-receptor matrices should be done regularly in order to take into account all peculiarities (emissions changes, meteorological variability) of a particular year, which affect pollution levels in the EMEP countries.

Model evaluation of future emission scenarios for 2035 predicts general decrease of mercury deposition in Europe by 2035 from moderate reduction according to the Current Policy scenario to strong reduction for the Maximum Feasible Reduction scenario. Changes of mercury deposition levels as well as source apportionment differ in different parts of the EMEP region. The most significant decrease of mercury deposition is expected in Western and Central Europe. At the same time some of the scenarios predict deposition increase in limited number of 'hot spots' located in Eastern Europe and Central Asia. Decrease of mercury deposition in the EMEP region by 2035 is largely determined by reduction of domestic emissions and, to less extent, by emission change in other regions (first of all, in East Asia).

Further development of the Global EMEP Multi-media Modeling System (GLEMOS) is aimed at reduction of the model assessment uncertainties in the EMEP region. Following decisions of the Executive Body for CLRTAP [*ECE/EB.AIR/113/Add.1]*, MSC-E continues the work on transition of the EMEP operational modeling to the new EMEP grid. Pilot simulations performed with GLEMOS on the new EMEP grid demonstrate better model performance in comparison with measurements. Simulations with finer resolution provide more accurate pollution assessment of smaller EMEP countries as well as countries with complex relief.

A multi-model analysis of mercury processes in the atmosphere was performed to evaluate the key oxidation mechanisms in the atmosphere and identify future directions of model development. Four global scale models took part in the study. The simulation results were evaluated against observations from several monitoring networks. The models predicted similar spatial patterns of mercury concentration with pronounced gradient between the Southern and the Northern Hemispheres and elevated concentrations in the major industrial regions – East and South Asia, Europe and North America. Both absolute and relative contributions of anthropogenic emissions were higher in the Northern Hemisphere, where most of emission sources were located. Contribution of natural/legacy sources also increased northward but the gradient was considerably smaller. It was demonstrated that oxidation chemistry of mercury in the atmosphere contained significant uncertainties. There was possibility of multiple oxidation mechanisms governing transformation and removal of mercury from the atmosphere. The study supports improvement of model parameterizations and, ultimately, quality of the model assessment in the EMEP domain.

Co-operation with Parties to the Convention in the framework of the country-specific case studies of heavy metal pollution with fine spatial resolution is among of priority tasks of EMEP. This year the work, focused on assessment of lead pollution levels in Belarus, has been completed. The study shows that anthropogenic emissions of lead in Belarus and some neighbouring countries are likely incomplete. For the analysis of heavy metal emission data in the EECCA countries joint efforts of national emission experts of these countries together with CEIP and TFEIP are required. Besides, in order to improve quality of measurements in the EECCA region, participation of national laboratories in the regular intercomparisons under the Chemical Coordinating Centre (CCC) supervision is appreciated. Further country-specific activity is planned to be focused on Poland and the United Kingdom. The investigation of emission data effect on a country-scale assessment will be further continued in a case study for Poland, where possible uncertainties of temporal variation of emissions are identified. In addition, a case study for the United Kingdom has been also initiated. One of the important aims of this study is investigation of role of secondary sources in heavy metal pollution in the country. Finally, German national experts from Karlsruhe Institute of Technology (KIT) have been provided with modeling results on deposition of lead, cadmium and mercury. These data are used in development of national model of heavy metal and nutrient emissions to surface water bodies.

Another important aspect of MSC-E activities is collaboration with the subsidiary bodies to the Convention and other international organisations and programs. Progress in the model development and assessment of heavy metal pollution in the EMEP region and selected countries was presented at the EMEP Task Force on Measurements and Modeling (TFMM). MSC-E also continues co-operation with the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). An overview of ongoing activities within EMEP and relevant research projects focused on assessment of mercury pollution was given at the annual TF HTAP meeting. In addition, the Centre actively participates in joint work with CEIP and the Task Force on Emission Inventories and Projections (TFEIP) aimed at improvement of available inventories of heavy metal emissions.

MSC-E continues co-operation with the Working Group on Effects (WGE). Regular surveys of heavy metal concentrations in mosses, coordinated by International Cooperative Programme on Vegetation (ICP-Vegetation) are valuable supplementary source of monitoring information, used for assessment

of heavy metal pollution and its trends in the EMEP countries as well as verification of modeling results. It was shown that reduction of observed concentrations in mosses in Europe between 1990 and 2010 agreed well with the decline of total deposition. Information on ecosystem-dependent deposition is used for evaluation of negative effects of heavy metals on human health and the environment, in particular, to calculate critical load exceedances. To support this activity MSC-E performs regularly calculations of deposition fluxes of lead, cadmium and mercury to different land-cover categories. Country-specific information about ecosystem-dependent deposition is available at the MSC-E web-site for all EMEP countries [www.msceast.org].

EMEP closely collaborates with the United Nations Environment Programme (UNEP) supporting development of the Minamata Convention on Mercury. MSC-E has participated in a series of the UNEP Global Mercury Assessments (GMA) sharing information on mercury pollution and coordinating activities on global scale modeling. In cooperation with other EMEP Centres, MSC-E also performs regular model assessment of atmospheric pollution of the Baltic Sea by various pollutants including heavy metals. This work is carried out in accordance with the Memorandum of Understanding between CLRTAP and the Baltic Marine Environment Protection Commission (HELCOM). Besides, the Centre has long and successful experience of co-operation with the Arctic Monitoring and Assessment Programme (AMAP), which includes participation in joint assessments and projects. Possible future collaboration between AMAP and CLRTAP has been recently discussed at the joint meeting and can include evaluation of the Arctic pollution by mercury and other heavy metals.

Future directions of MSC-E activities will be aimed at improvement of heavy metal pollution assessment in the EMEP region. Particular attention will be paid to the problem of mercury pollution both in the EMEP region and on a global scale. National sale assessment of heavy metal pollution with high spatial resolution will be continued in the framework of country-specific case studies in close cooperation with national experts from Poland and the United Kingdom. Further development and evaluation of the Global EMEP Multi-media Modeling System (GLEMOS) will include transition of heavy metal operational modeling to the new EMEP grid, update of parameterizations related to mercury chemistry, further implementation of the multi-media approach to mercury simulations and distribution of GLEMOS for public use as an open code. Finally, MSC-E will continue co-operation with subsidiary bodies of the Convention (TFMM, TF HTAP, TFEIP, and WGE), international organizations (AMAP, UNEP, Minamata Convention, HELCOM etc.) and national experts.

CONTENTS

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The authors are grateful to Snezhina Toshovski from Karlsruhe Institute of Technology (KIT) in Germany for submitting contribution to the chapter 4 of the report. The contribution describes usage of MSC-E calculated heavy metal deposition data in modeling of heavy metal and nutrient emissions to surface water bodies.

INTRODUCTION

Heavy metals are known as toxic pollutants causing adverse health effects on humans and wildlife. Released to the atmosphere from combustion of fossil fuels and industrial activities they can be transported over long distances and deposited to the ground leading to contamination of agricultural lands, terrestrial and aquatic ecosystems. Selected heavy metals are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) since 1998 when the Protocol on Heavy Metals (hereafter, Protocol on HMs or the Protocol) came into force. Heavy metals targeted by the Protocol include lead, cadmium and mercury.

Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) provides 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 and expert estimates. 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). 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.

This report outlines recent activities of the EMEP Centres in the field of heavy metal pollution assessment performed in accordance with the 2016-2017 Workplan for implementation of the Convention [*ECE/EB.AIR/133/Add.1*]. It covers a number of topics including assessment of heavy metal pollution in the EMEP region, MSC-E activities focussed on further development and improvement of the modeling approaches, co-operation with national experts in the framework of case studies of heavy metal pollution in selected EMEP countries, collaboration with subsidiary bodies to the Convention, and other international organisations and programmes.

In addition, particular attention of the report is paid to the problem of mercury pollution both in the EMEP region and on a global scale. For this purpose, a digest of current MSC-E activities in this area is given in a separate chapter to support the forthcoming discussion at the Second Joint Session of the Steering Body to EMEP and the Working Group on Effects (Geneva, September 2016).

Assessment of heavy metal pollution of the EMEP region includes various aspects relating to emissions reporting, monitoring and modeling activities. Gridded data on heavy metal emissions is the primary information for the model assessment. However, uncertainties of the emission inventory remain significant. The key emission parameters affecting quality of model estimates have been analysed and ranked in terms of their priority. Monitoring of heavy metals at the EMEP network is important both for characterizing pollution levels and evaluation of modeling results. Measurements of lead, cadmium and mercury concentration in air and precipitation are reported along with brief analysis of data quality. The model assessment includes evaluation of current pollution levels of lead, cadmium and mercury in the region along with analysis of historical trends and forecasting future contamination. The analysis of long-term trends of heavy metal pollution served as a base for MSC-E contribution to the CLRTAP Assessment Report [*Maas and Grennfelt*, 2016], the TFMM Assessment

Report [*Colette et al*., 2016] and the WGE Assessment Report [*de Wit et al*., 2016] released by the Convention this year.

Further development of the Global EMEP Multi-media Modeling System (GLEMOS) is aimed at reduction of the model assessment uncertainties in the EMEP region. Following decisions of the Executive Body for CLRTAP [*ECE/EB.AIR/113/Add.1]* MSC-E continued the work on transition of the EMEP operational modeling to the new EMEP grid. Pilot simulations on the new grid have been performed for mercury and compared with measurements and the model results on the old grid. Besides, a multi-model analysis of mercury processes in the atmosphere has been performed to evaluate the key oxidation mechanisms in the atmosphere and identify future directions of model development. And finally, the model scheme of wind re-suspension of lead has been improved in terms of refinement of heavy metal re-emission fluxes from urban and industrial soils.

Co-operation with Parties to the Convention in the framework of case studies of heavy metal pollution is among priority tasks of EMEP. The country-specific research is continued for selected countries including Belarus, Poland and the United Kingdom. The pollution assessment for Belarus has been completed this year and main conclusions and recommendations have been formulated. New country scale studies have been initiated for Poland and the United Kingdom. Variety of input data has been collected in co-operation with national experts and preliminary simulations have been performed. In addition, MSC-E co-operated with national experts from Germany supporting their assessment of heavy metal emissions into surface water bodies.

Another important aspect of MSC-E activities is collaboration with subsidiary bodies to the Convention and other international organisations. All results of the research and development were presented and discussed at the EMEP Task Force on Measurements and Modeling (TFMM) and the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). Besides, information on ecosystem-specific deposition of heavy metals was provided to the Working Group on Effects (WGE) for evaluation of critical load exceedances. Moreover, the Centre worked in close co-operation with other international organizations and programmes (the United Nations Environmental Programme, the Arctic Monitoring and Assessment Programme, Helsinki Commission etc.) to broaden dissemination and facilitate exchange of the scientific and policy oriented information.

Finally, the main challenges of heavy metal pollution assessment, which need particular attention and further research in 2017 are summarized in the report. More detailed scientific information is presented in Annexes and in the MSC-E Technical Report 1/2016 [*Shatalov et al.*, 2016]. A variety of information on heavy metal pollution levels in the EMEP region, individual countries, marginal seas etc. is also distributed via the Internet at the MSC-E website [www.msceast.org].

1. PROBLEM OF MERCURY POLLUTION: REGIONAL AND GLOBAL ASPECTS

A problem of mercury pollution was recognized as one of the priority topics of the pollution assessment within EMEP at the recent Joint meeting of the Extended Bureaux of the EMEP Steering Body and the Working Group on Effects (March 2016, Geneva, Switzerland). MSC-E for a long period has performed various research activities focused on model assessment of mercury pollution both within the EMEP region and over the globe. This chapter presents a brief introduction to the mercury pollution problem and a digest of current MSC-E activities in this area described in the report below in more detail.

Mercury is widely known as a toxic pollutant capable of long-range transport, bioaccumulation in ecosystems and biota as well as adverse effects on human health and the environment. In spite of being a natural element its concentrations in the environment have been considerably enriched by human activities since the pre-industrial times [*Fitzgerald et al*., 1998; *Mason and Sheu*, 2002; *Krabbenhoft and Sunderland*, 2013]. Mercury differs from other atmospheric pollutants in that its major exposure routes are not related directly to the ambient air concentrations. While Mercury is mainly distributed through the atmosphere, its primary environmental and health impacts are through deposition to aquatic ecosystems, bioaccumulation in aquatic organisms and consumption of these by humans and wildlife [*Mahaffey et al*., 2004; *Sunderland et al*., 2010; *Mason et al*., 2012]. Therefore, atmospheric transport and deposition to the surface are essential parts of the mercury route from an emission source to final exposure.

Despite significant efforts to mitigate mercury pollution in the EMEP region, human health and ecosystems continue to be at risk in many countries. As it is stated in the recent **CLRTAP Assessment Report [***Maas and Grennfelt*, 2016] anthropogenic emissions of mercury were reduced by up to 60% in EMEP countries over the last two decades. However, both observations at the EMEP measurement sites and modeling results show less significant decrease (below 25%) of mercury concentration and deposition levels in the region over the same period. Similar or even lower rates of mercury pollution reduction are demonstrated by measurements of mercury in mosses presented in the **WGE Assessment Report** [*de Wit et al*., 2016]. It should be noticed that the decline of pollution levels considerably differs in different parts of the EMEP region from 35% in the European Union (EU28) to 20% in the EECCA countries [**TFMM Assessment Report**, *Colette et al*., 2016]. Besides, even slight increase of mercury wet deposition since 1995 was observed in the north of Scandinavia. It correlates with the observed evidence of growth in mercury concentrations in freshwater fish in Sweden and Norway [*de Wit et al*., 2016]. In addition, available measurements in soil show that mercury concentrations increased both in the forest floor and in deeper soil layers demonstrating continued soil accumulation and potentially hazardous effects on human and biota. In particular, estimates of critical load exceedances show that excessive deposition of mercury remains an issue of concern nowadays in many EMEP countries [*de Wit et al*., 2016]. Contribution of MSC-E to the mentioned above Assessment Reports is discussed below in Section 2.3.

Nowadays the highest levels of mercury deposition are characteristics of Southern and Central Europe (the northern part of Italy, southern Poland and the Balkan region) (Section 2.4). It is generally confirmed by measurements of mercury in precipitation at EMEP monitoring sites (Section

2.2). Relatively high deposition levels also take place in the Arctic due to the effect of intensive mercury oxidation and removal during springtime. In most of the EMEP countries the main contribution to mercury deposition is made by sources located outside the EMEP domain. The contribution of EMEP anthropogenic sources varies from 2% to almost 64% in different countries. Transboundary transport plays important role in mercury pollution in the EMEP countries. In one third of the countries mercury deposition from foreign emissions exceed deposition from national sources. On the other hand, up to 90% of mercury emitted by domestic sources is transported outside country's territory getting involved into the transboundary transport.

Due to its long residence time in the atmosphere mercury can be easily transported between continents [*Travnikov*, 2012; *AMAP/UNEP*, 2013]. Atmospheric transport from distant sources can significantly contribute to mercury deposition, particularly, in regions with low domestic emissions. Therefore evaluation of present-day mercury pollution levels as well as forecasts of future changes should take into account the global character of mercury atmospheric transport and dynamics of mercury emission changes in different continents. In particular, mercury deposition to EMEP countries consists of almost equal contributions of contemporary anthropogenic emissions and emissions from natural and legacy sources [*Ilyin et al*., 2014; *Maas and Grennfelt*, 2016]. Half of the anthropogenic part is contributed by domestic EMEP emission sources and the other half is by transport from sources located in other regions. The largest external contributors include East Asia (11%), Africa (4%), Southeast Asia (2%) and South America (2%).

Assessment of future levels of Mercury pollution and evaluation of different abatement scenarios are the key information for development of future mitigation measures. Model evaluation of future emission scenarios for 2035 (see Section 2.7) predicts general decrease of mercury deposition in Europe by 2035 from moderate reduction according to the Current Policy scenario to strong reduction for the Maximum Feasible Reduction scenario. Changes of mercury deposition levels as well as source apportionment differ in different parts of the EMEP region. The most significant decrease of mercury deposition is expected in Western and Central Europe. At the same time some of the scenarios predict deposition increase in limited number of 'hot spots' located in Eastern Europe and Central Asia. Decrease of mercury deposition in the EMEP region by 2035 is largely determined by reduction of domestic emissions and, to less extent, by emission change in other regions (first of all, in East Asia).

Current knowledge on mercury behaviour in the atmosphere and its potential to cycling between different environmental media is still incomplete. There are significant gaps in the understanding of chemical processes affecting mercury atmospheric transport and deposition, characteristics of the air-surface exchange and processes responsible for re-emission of mercury to the atmosphere. In order to reduce uncertainties of mercury pollution assessment MSC-E coordinated a multi-model study of mercury atmospheric processes performed in a form of model experiments complemented by extensive measurement data (Section 3.2). The study supports refinement of model parameterizations and, ultimately, quality of the model assessment in the EMEP domain. Further improvement of modeling approaches also includes the work on transition of the mercury operational modeling to the new EMEP grid (Section 3.1). Pilot simulations on the new grid show better model performance in comparison with measurements. Besides, simulations with finer

resolution provide more accurate pollution assessment of smaller EMEP countries as well as countries with complex relief.

Cooperation with other international organisations plays an important role in information exchange and outreach of the knowledge gathered within the Convention. The global aspect of mercury pollution resulted in development of the Minamata Convention on Mercury, a legally-binding treaty that was recently adopted by more than 140 nations. Since the very beginning of the development process EMEP participated in series of Global Mercury Assessments coordinated by the United Nations Environment Programme (UNEP). In particular, MSC-E took part in preparation of the UNEP Global Mercury Assessment 2013 used for negotiations of the Minamata Convention. A new Global Mercury Assessment 2018 is now in progress and MSC-E has been invited to take a lead of the assessment part focused on modeling of mercury pollution on global and regional scales (Section 5.2.1). In addition, evaluation of mercury pollution was considered as one of directions for possible future cooperation between the Convention and Arctic Monitoring and Assessment Programme (AMAP) (Section 5.2.3).

These and other aspects of mercury pollution in the EMEP region will be discussed at the Second Joint Session of the Steering Body to EMEP and the Working Group on Effects (Geneva, September 2016). The following topics are to be discussed at the session:

- Assessment of mercury transboundary pollution within EMEP
- Measurements of mercury levels in the EMEP monitoring network
- Mercury emissions in the EMEP countries and over the globe
- Biomonitoring of mercury pollution and evaluation of adverse effects on human and biota
- Co-operation with national experts and international organisations and programs

All the EMEP research Centres and representatives of the Parties to the Convention are invited to take part in the discussion.

2. HEAVY METAL POLLUTION OF THE EMEP REGION

2.1. Emission data for model assessment

Model assessment of heavy metal pollution levels in the EMEP domain for 2014 was carried out using gridded emission data on lead, cadmium and mercury provided by CEIP. Detailed information on heavy metal emissions in each country, as well as the gap-filling methods that have been used for the 2014 GNFR inventory (as reported in 2016) can be found in the CEIP Technical Report 01/2016 [*Tista et al.*, 2016].

The total values of anthropogenic heavy metal emissions in the EMEP countries for 2014 are higher than the corresponding values for 2013 in previous submission and amount to 3467 tonnes for lead, 204 tonnes for cadmium and 167 tonnes for mercury. The value of lead emission is about 318 tonnes higher in comparison to the lead emission used for model calculation for 2013. The total emissions of cadmium and mercury have increased in comparison to the previous year by 34 tonnes and 8 tonnes, respectively.

The changes of lead, cadmium and mercury emissions in each country are expressed as $100*(E_{2014} - E_{2013})/E_{2013}$ (%), where E_{2013} and E_{2014} are previous and current emissions in 2015 Submission and 2016 Submission, respectively. Changes are illustrated in Fig. 2.1. Negative values indicate a decrease in emissions, while positive values illustrate an increase in emissions from 2013 to 2014.

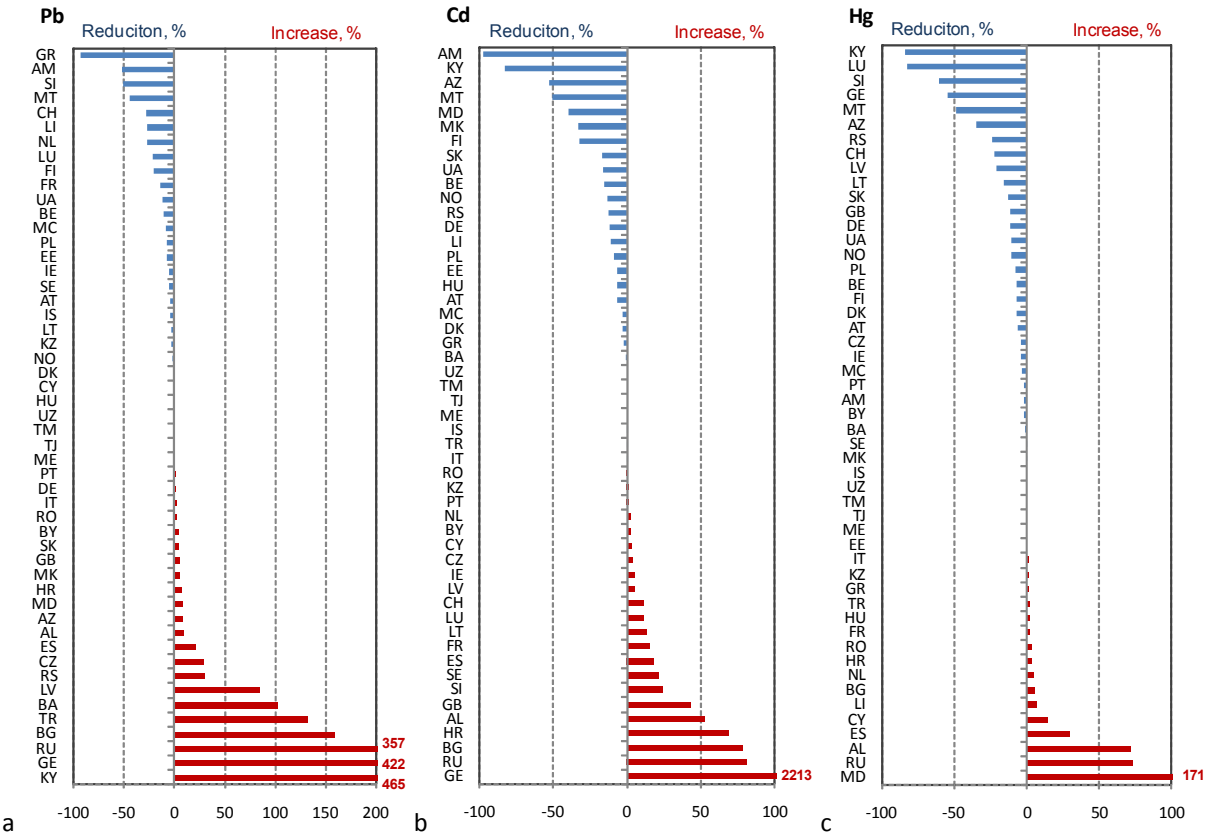

Fig. 2.1. *Relative changes of annual total emissions of lead (a), cadmium (b) and mercury (c) between 2013 and 2014, %*

Emissions in Armenia, Austria, Belgium, Denmark, Finland, Malta, Monaco, Norway, Poland and Ukraine decreased compared to emission values of all three metals for 2013. The increase of the heavy metal emission values was noted for Albania, Bulgaria, Croatia, Italy, Romania, the Russian Federation, Spain and Turkey.

In Georgia, emissions of lead and cadmium have increased over 5 and 23 times, respectively (Fig. 2.1a and b). In the Republic of Moldova, mercury emissions have increased almost 2.7 times (Fig. 2.1c). It should be noted that heavy metal emissions in the Russian Federation increased significantly compared to the previous year, example for lead - over 4.5 times (Fig. 2.1a). The increase is explained by update of methodology applied by CEIP for preparation of emission data for modeling. In particular, the emission value of GNFR sector 'A_PublicPower' was extrapolated from the previous period (2002-2006) [*Tista et al*., 2016]. Spatial distributions of lead, cadmium and mercury emissions used in the modeling are shown in Fig. 2.2.

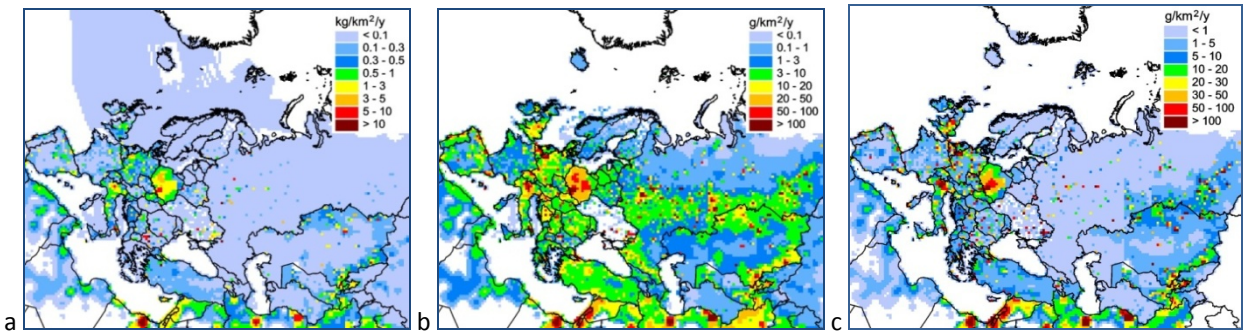

Fig. 2.2. *Spatial distribution of lead (a), cadmium (b)and mercury (c) anthropogenic emissions over the EMEP domain in 2014*

Mercury emissions on a global scale

Mercury is a pollutant capable of intercontinental transport in the atmosphere. Therefore, assessment of mercury pollution in the EMEP countries requires data on mercury anthropogenic emissions on a global scale.

The global inventory of mercury anthropogenic emissions for 2010 was used for mercury simulations on a global scale and evaluation of boundary conditions for regional modeling. A new updated emissions inventory has been developed as a part of the UNEP Global Mercury Assessment 2013 [*AMAP/UNEP*, 2013] that allows better characterization of differences between countries in terms of fuels and raw materials used as well as technologies and practices applied. The global mercury emission in 2010 is estimated at 1960 tonnes and comprises emissions from artisanal and small-scale gold mining (ASGM), combustion of fossil fuels (mainly coal) in power plants, industrial and residential boilers, metal production (ferrous and non-ferrous), cement production, product use, and cremation. More detailed information about the global inventory of mercury emissions can be found in the EMEP Status report 2/2014 [*Ilyin et al.*, 2014].

It should be noted that expert estimates of mercury anthropogenic emissions included to the UNEP global emissions inventory can differ significantly from national data reported by some EMEP countries because of different methodologies and statistical data applied [*AMAP/UNEP*, 2013].

Key emission parameters affecting quality of model estimates

Information on pollutant's emission to the atmosphere and other environmental media is one of the key parameters required for model assessment of pollution levels and transboundary fluxes. Completeness and uncertainties of emission data can significantly affect quality of the model estimates. Executive Body of CLRTAP initiated activity aimed at improvement of heavy metal emission inventories in the EMEP region [*ECE/EB.AIR/133/Add.1*].

Emission data for heavy metals have been characterized from the viewpoint of their use for the operational modeling within EMEP. Various emission parameters which affect quality of the model assessment have been discussed along with characterizing their uncertainties. Importance of different emission parameters and their influence on quality of the assessment results varies considerably for simulation of different pollutants. This fact should be taken into account when planning improvement of the emission data. Table 2.1 characterizes the key emission parameters for heavy metals in terms of their priority for the further improvement. It should be noted that lower priority level does not mean that no improvements are needed for this emission parameter but rather determines an order of their implementation.

Table 2.1. *Key emission parameters affecting quality of model estimates*

Emission parameter	Pb and Cd	Hg
Quality of gridded anthropogenic emissions	1	2
Chemical composition	-	1
Temporal variation	2	6
Vertical distribution	3	7
Global emissions inventory	4	3
Historical emissions	5	4
Emissions to other media	6	5

- First priority; - Second priority; - Third priority

As seen from Table 2.1, quality of gridded emission data (including completeness of reported data and quality of expert estimates used for gap filling) is among the first priority parameters. Other parameters with the highest priority include chemical composition of emissions (for mercury). Lower priority parameter, which is still important for mercury, is global emissions inventory. On the other hand, predominantly airborne substances (lead, cadmium) can be more affected by temporal variation of emissions and vertical distribution of emission sources. More detailed information on MSC-E contribution to the uncertainty analysis of heavy metal emissions is available on the MSC-E website: www.msceast.org.

Completeness of gridded emission data and chemical speciation of mercury emissions are the key parameters affecting quality of the model assessment. Other important parameters include data on temporal variability and vertical distribution of emissions for lead and cadmium, and global scale inventories as well as historical emissions for mercury.

2.2. EMEP monitoring network of heavy metals

Measurement network

In 2014, there were 31 sites measuring heavy metals (cadmium or lead) in both air and precipitation, and altogether there were 62 measurement sites. 29 sites were measuring mercury in either air and precipitation, 11 of these with concurrent measurements in air and precipitation. In total, 22 Parties to the Convention report heavy metal data to EMEP, but only 6 of these fulfil their monitoring obligations as defined in the EMEP monitoring strategy [*ECE/EB.AIR/GE.1/2009/15*] with at least one level 2 site with both air and precipitation measurements of heavy meals and mercury in air and precipitation. There are however some additional sites and Parties measuring heavy metals, though their reporting has been delayed, thus not possible to be included in this annual report. It is very important to improve their timeliness of reporting of data, which is set to 31 July. EMEP/CCC will have a workshop in October 2016 on data submission, and Parties with challenges with the new and extended reporting guidelines are encouraged to participate.

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 OSPAR. Detailed information about the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [*Aas and Nizzetto*, 2016]. All the data presented here are available at the EMEP database [http://ebas.nilu.no].

Observed concentration level of lead and cadmium in 2014

Annual averages of lead and cadmium concentrations in precipitation and in air in 2014 are presented in Fig. 2.3-2.6. Note that Cyprus data in aerosols are misplaced to fit inside the map. The lowest concentrations for all elements are generally found in northern Scandinavia. An increasing gradient can be seen from north to southeast, but the concentration levels are not evenly distributed, there are some "hotpots" for some elements, i.e. in Hungary and the BeNeLux countries. At IE01 the lead concentration in precipitation is relatively high, almost 4 µg/L. It is the highest observed annual mean in Europe and almost ten times higher than what was seen in 2013. Elevated level is also seen for other elements at IE01, but not to the same extent. The reason for this change is not clear. In IT01 high concentrations of cadmium in precipitation are probably influenced by the industrial region in the Roma area. There are several sites (in PT, FR, HU) with high detection limits and these only give an indicative measure for the upper limit. A more detailed discussion of temporal and spatial resolution of heavy metals in Europe is found in *Tørseth et al.* [2012] and in the EMEP/CCC data report for 2014 [*Aas and Nizzetto*, 2016]

The relatively high concentrations indicated at the few sites in Eastern Europe show the importance of getting more sites with continuous measurements in this region to get better knowledge of the pollution level there.

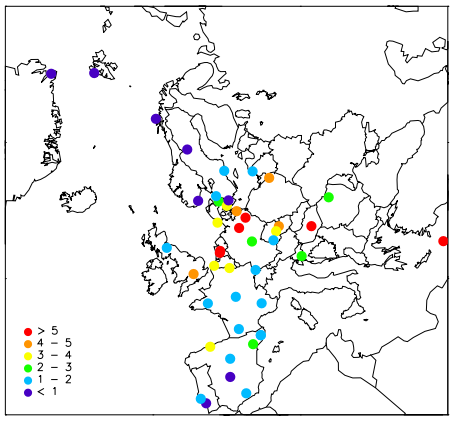

Fig. 2.3. *Lead in aerosol, ng/m^3*

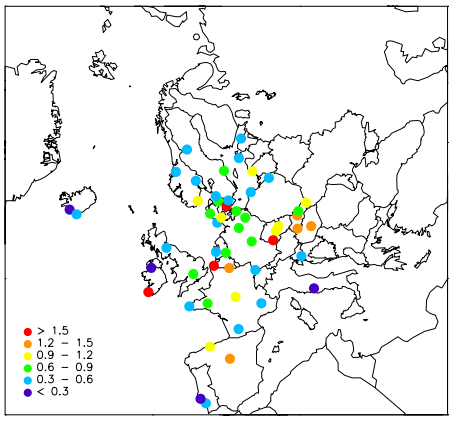

Fig. 2.4. *Lead in precipitation, µg/L*

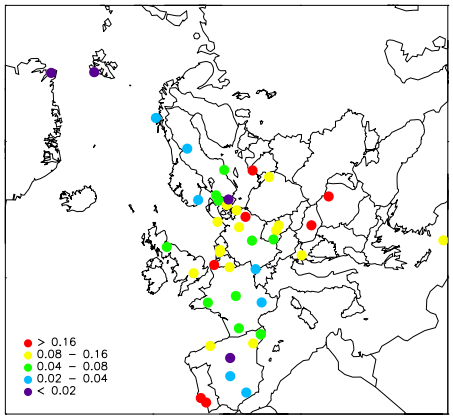

Fig. 2.5*. Cadmium in aerosol, ng/m^3*

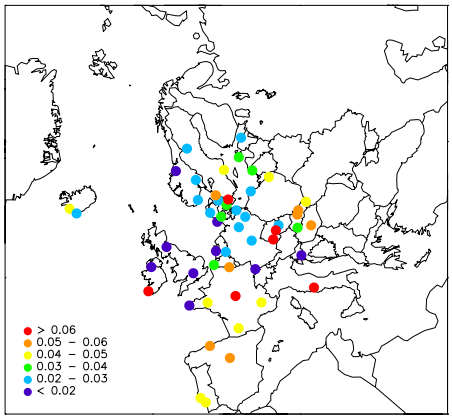

Fig. 2.6. *Cadmium in precipitation, µg/L*

Observed concentration level of mercury in 2014

Annual averages of mercury concentrations in precipitation and in air in 2014 are presented in Fig. 2.7-2.8. The spatial distribution of elemental mercury in air is scattered. A recent manuscript summarizing results from the GMOS project, present the mean background concentration of elemental mercury in European air to be 1.48 ng/m^3 in 2014 [*Sprovieri et al.*, 2016]. There is indication of elevated level in central Europe as expected due to influence from anthropogenic sources like coal combustion. An interesting observation is that the coastal Arctic sites in Norway are slightly higher than what is observed at Greenland and more inland in Finland and Sweden, which might be due to the summertime evaporation from the ocean or due to the fact that Svalbard i.e. experience several direct transport episodes from the continent, especially in winter and spring. PL05 and SI08 show unexpected low concentration, 1.2 ng/m^3 and 0.8 ng/m^3 respectively. The latter concentration level is even lower than what is seen in Antarctica [*Pfaffhuber et al.*, 2012]. Given the locations of these stations and the proximity to emission sources, it seems like a bias in the concentration level for these two sites. This bias is larger at ES08, which has an annual mean of 0.3 ng/m^3, which obviously is wrong. Results from a field intercomparison study of mercury measurement within EMEP performed in 2005 showed that the majority of the participating labs performed well and within the +-30% uncertainty being the EMEP data quality objective

[*Umweltbundesamt*, 2006]. However, the biased concentration results reported above, highlights the importance to follow QA/QC procedures. These three laboratories need to evaluate their methodology as it seems evident that there is an issue with either calibration or gold trap poisoning or a combination of both.

In precipitation, the highest levels are seen in Eastern Europe (SI, PL and CZ), which is reasonable since the anthropogenic emission sources are highest in this region, and in general the concentration decreases with distance to emission sources. Taking into account that precipitation measurements of mercury are more complex than air measurements, and that the expected measurement uncertainty is 42% [*Umweltbundesamt,* 2006], the observed concentrations and spatial pattern seems reasonable, for Poland most of the data is below detection limit so it is difficult to fully assess the spatial concentration pattern. Also Ireland and Portugal report most of the data below detection limit. Given that their applied measurement principle is capable to achieve detection limits an order of magnitude lower than the reported, they should evaluate their methodology for mercury measurements in precipitation, in particular with respect to sample treatment and handling, laboratory water quality and purity of chemicals.

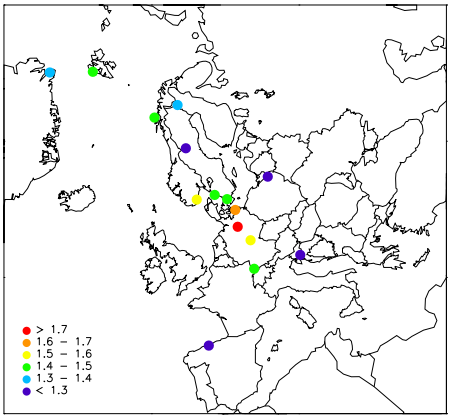

Fig. 2.7. *Mercury (g) in air, ng/m^3*

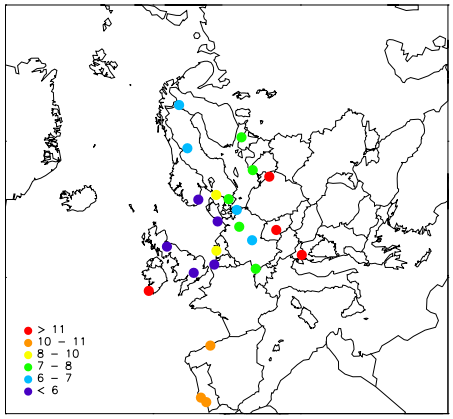

Fig. 2.8. *Mercury in precipitation, ng/L*

Nowadays heavy metal monitoring data in the eastern and southern parts of Europe and Central Asia is scarce. Therefore, more sites with continuous measurements in these regions are needed. Existence of low quality data at some monitoring sites supports the need of further evaluation of measurement techniques at these sites.

2.3. Long-term trends in the EMEP region (1990-2014)

Information on long-term trends of heavy metal (lead, cadmium, mercury) pollution in the EMEP region is important for the evaluation of the effectiveness of policy measures aimed at reduction of negative effects of the pollution on human health and biota. This year MSC-E has finalized assessment of the trends for the period from 1990 to 2012. Results of the assessment were contributed to the three reports: CLRTAP scientific assessment report [*Maas and Grennfelt*, 2016], TFMM assessment report [*Colette et al*., 2016], and WGE assessment report [*de Wit et al.,* 2015]. This section comprises main results contributed to the reports.

Trends of air concentrations and deposition of lead, cadmium and mercury over the last two decades were analyzed for the EMEP region as a whole, for particular countries and for grid cells where monitoring stations with long period of measurements were located. Because of scarce spatial coverage by these stations the main outcomes of the trend analysis were based on modeling results. Analysis of the trends is performed following the methodology described in [*Shatalov et al*., 2015].

Lead deposition demonstrated the most marked reduction (78%) of deposition over 1990-2012 period in the EMEP region, followed by cadmium (53%) and mercury (23%) (Fig. 2.9). The reduction of lead and mercury pollution levels was more significant in the first decade, while in the second it became smaller. Decline of cadmium deposition was almost uniform over the considered period. The main factors responsible for the decline of lead and cadmium were reduction of anthropogenic emissions and relatively slow decrease of deposition caused by secondary emissions (wind re-suspension). Mercury levels strongly depended on contribution of emission sources (anthropogenic, natural and historic) located outside the EMEP region.

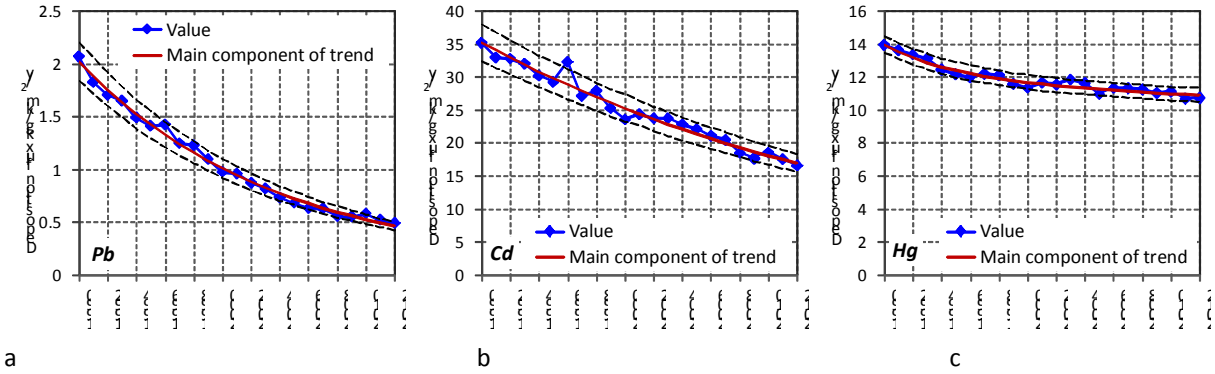

Fig. 2.9. *Long-term changes of modeled total deposition flux and of the main component of its trend for lead (a), cadmium (b) and mercury (c). Dashed lines indicate 95% confidence interval*

The reduction of heavy metal deposition levels varied among the EMEP countries. Characteristics of trends for each EMEP country are available in the internet at the MSC-E website [www.msceast.org].The highest total reduction of modeled metal deposition was noted for the EU28 countries. On average, this decrease occurred to be more than 80% for lead, 60% for cadmium and 35% for mercury (Fig. 2.10). In the EECCA countries the decline of heavy metal pollution levels was smaller: total reduction for 1990-2012 amounted to 76% (lead), 49% (cadmium) and 19% (mercury).

Information about reduction rate (% per year) was available for each of the EMEP 50-km grid cell. Where measurements with sufficiently long period of measurements were available, modeled reduction rates were compared with the observed ones. For example, both measured and observed air concentrations and wet deposition of lead in the central and western parts of Europe exhibited similar rates of reduction (Fig. 2.11).

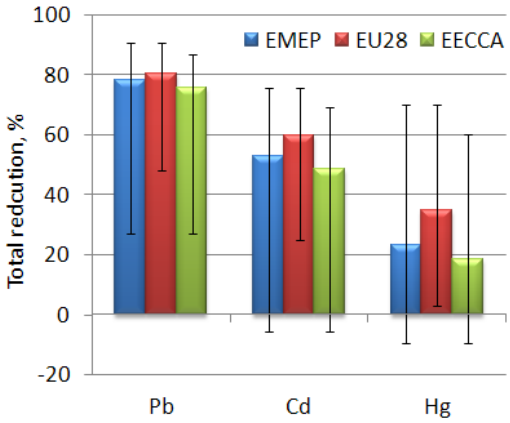

Fig. 2.10. *Total reduction of modeled lead, cadmium and mercury deposition in the EMEP region as a whole, in EU28 and in EECCA countries. Whiskers indicate range of the reductions among the countries. Negative values stand for an increase*

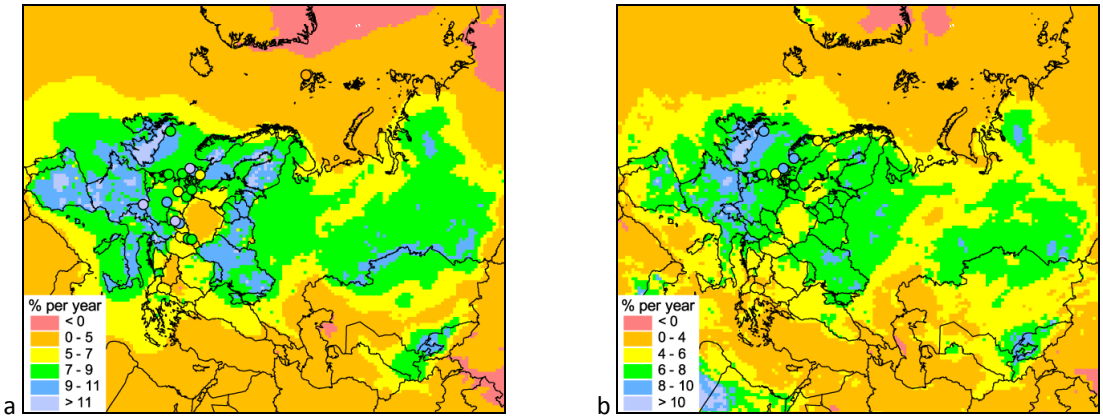

Fig. 2.11. *Average reduction rate of modeled (field) and observed (circles) lead concentrations in air (a) and wet deposition fluxes (b) for the period from 1990 to 2012*

Information on long-term changes of lead, cadmium and mercury deposition, calculated by MSC-E, were used by WGE for evaluation of trends of critical load exceedances. It was demonstrated that areas where heavy metal deposition exceeded critical loads, were still large. For example, relative area of ecosystems with exceedances of lead decreased from 68% in 1990 to about 20% in 2010. For mercury the corresponding values were 69% and 56%. Therefore, at present critical loads for lead are exceeded over about 1/5, and mercury - over more than 1/2 of ecosystem area of the EMEP region. These estimates justify that the problem of heavy metal pollution in the EMEP region is still highly relevant nowadays.

Calculated deposition of heavy metals for 2013 and 2014 has been added to the existing time series. It should be noted that interpretation of the obtained trends from 1990 to 2014 has to be done with caution because these time series are not homogeneous, since deposition for the last two years are based on updated anthropogenic emissions and different set of meteorological data (ECMWF analyses instead of ERA-Interim).

Cadmium levels demonstrated some increase since 2012 (Fig. 2.12). This increase can partly be explained by higher reported emission values in several countries, e.g. in Russia, Bulgaria, Croatia, Georgia, and partly by meteorological reasons. Lead values of EMEP-mean deposition flux continue the tendency of low decline taken place since 2000. In the last five years the range of lead deposition from 2010 to 2014 is within ±12%. Unlike cadmium, the lead levels do not exhibit the increase between 2013 and 2014 due to introduction of lower enrichment of lead concentrations in soils, resulted to some decline of wind re-suspension, as described in Section 3.3. Mercury levels demonstrate relatively low year-to-year variability because of predominant contribution of slowly changing non-EMEP emission sources to mercury concentrations and deposition in the EMEP region.

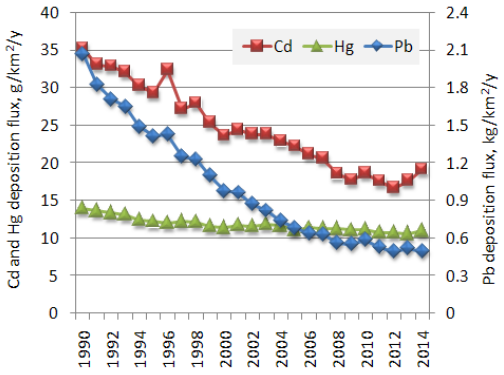

Fig. 2.12. *Long-term trends of deposition of lead, cadmium and mercury to the EMEP countries in 1990-2014*

Lead, cadmium and mercury deposition in the EMEP region declined by 78, 53 and 23%, respectively, from 1990 to 2014). Lead and mercury levels exhibit relatively low changes from 2012 to 2014. Cadmium levels demonstrated some increase since 2012 caused partly by higher emission values used in modeling, and partly by year-to-year variability of meteorological conditions.

2.4. Pollution levels and transboundary fluxes in 2014

Air concentrations, deposition and transboundary fluxes of lead, cadmium and mercury were calculated for 2014. This section provides description of the modeling results. Evaluation of modeling results against measurements is discussed in Annex A. Special attention is paid to comparison and analysis of the results for 2014 and 2013.

Heavy metal pollution levels in 2014

Modeling information about pollution levels of lead, cadmium and mercury in the EMEP region and in the individual countries in 2014 has been prepared and allocated in the Internet [www.msceast.org]. The main features of pollution levels, atmospheric transboundary transport of heavy metals and verification of the model results against measurements are overviewed in this section.

There are three groups of sources responsible for levels of heavy metal pollution in the EMEP region. They are anthropogenic sources of the EMEP countries, secondary emissions within the EMEP region (wind re-suspension of dust particles containing natural and previously deposited anthropogenic heavy metals and gas-phase natural emission and re-emission of mercury), and sources (anthropogenic and secondary) located outside the EMEP region (i.e., non-EMEP sources). With

regard to a particular country the anthropogenic sources, in its turn, can be presented as a sum of national sources (emissions within a country) and foreign sources.

Given very long residence time of mercury in the atmosphere it can be transported globally. To take into account contribution of intercontinental transport to mercury pollution of EMEP countries global scale modeling was applied for evaluation of the boundary conditions for the EMEP domain. Figure 2.13 shows distribution of Hg^0 air concentration in 2014 simulated with the global version of the GLEMOS modeling system.

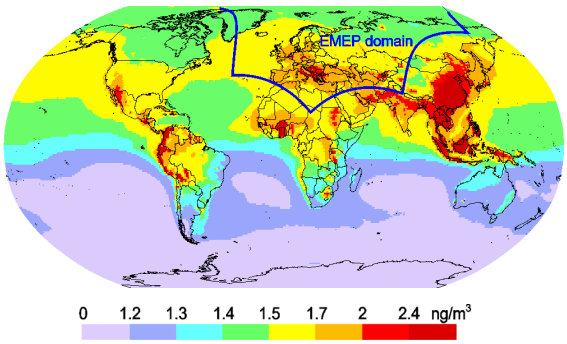

Fig. 2.13. *Global distribution of Hg^0 concentration in 2014. Blue line depicts boundary of the EMEP domain*

In 2014 the highest pollution levels of lead and cadmium in the EMEP region are noted for the northern part of Germany, the Benelux region, the eastern part of Ukraine and the northern part of Italy. Mean annual concentrations of lead in these regions vary from 8 to 30 ng/m^3, and of cadmium - from 0.3 to 0.5 ng/m^3 (Fig. 2.14a). The corresponding deposition levels of lead range from 1.5 to 5 kg/km^2/y, and of cadmium – from 40 to 150 g/km^2/y (Fig. 2.14b). Besides, relatively high cadmium concentrations and deposition are noted for the central part of Russia. Comparatively high values of concentration and deposition in the mentioned parts of the EMEP region are caused by combined influence of anthropogenic and secondary emission sources. The lowest heavy metal levels take place in Scandinavia and the northern part of Russia, which is explained by the remoteness of these regions from major emission sources. Besides, low deposition of lead and cadmium in the Central Asian region is caused by low precipitation.

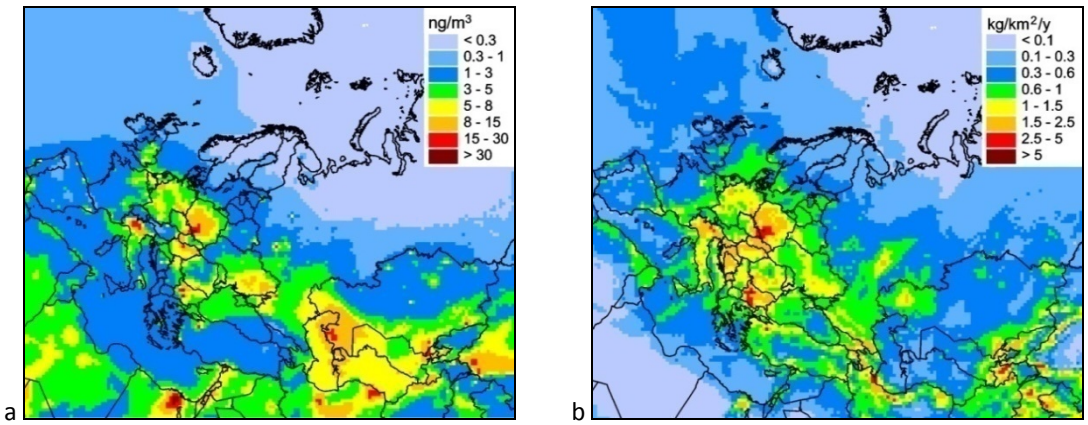

Fig. 2.14. *Mean annual air concentrations (a) and total deposition (b) of lead in 2014*

Spatial distribution of mercury net deposition fluxes in the EMEP region is similar to those of lead and cadmium. The highest levels (30-70 g/km^2/y) are noted for the northern part of Italy, the southern part of Poland, the Balkan region. Relatively high net deposition fluxes also take place in the Arctic region due to peculiarities of mercury atmospheric chemistry involving interactions with atmospheric bromine compounds (see also Section 5.2.3). Annual mean air concentrations of mercury are much

smoother compared to levels of other heavy metals like lead and cadmium. In the least polluted areas (north of Russia, Scandinavia, north-western Kazakhstan) of the EMEP region the levels of mercury concentrations range from 1.4 to 1.5 ng/m^3, and most polluted ones - from 1.7 to almost 2 ng/m^3. This low spatial variability is explained by long (0.5 – 1 year) residence time of atmospheric mercury. This long residence time results in significant mixing of mercury in the atmosphere and its global-scale dispersion.

Country-averaged deposition fluxes of lead, cadmium and mercury from anthropogenic, secondary and non-EMEP sources in 2014 were calculated for each EMEP country. Since 1990 the contributions of these three components have changed due to continuous reduction of anthropogenic emissions. In 2014 main contribution of lead and cadmium deposition in the EMEP countries is made by secondary sources. Their contribution to total country-averaged deposition exceeds 50% in 45 countries (lead) and 41 countries (cadmium). The highest deposition fluxes of cadmium (45 – 60 g/km^2/y) are noted for countries of eastern and south-eastern Europe (Bulgaria, Poland, Slovakia etc.) and in the Netherlands (Fig. 2.15). For lead the highest country-averaged deposition is calculated for Montenegro, followed by Monaco and Poland (1.7 - 1.8 kg/km^2/y). The lowest levels take place in countries without significant national emission sources, and located far from major countries-emitters, e.g., Ireland, Iceland, Finland, Norway.

The contributions of the three components to deposition of mercury in the EMEP countries differ from those of lead and cadmium. In most of countries the main contribution is made by non-EMEP sources. In 48 countries the contribution exceeds 50%, and in 28 countries – exceeds 75%. This high influence of non-EMEP sources on mercury levels in the EMEP countries is caused by global nature of mercury pollution. The contribution of anthropogenic sources varies from about 2% in Iceland to almost 64% in the FYR of Macedonia. The contribution of secondary emissions within the EMEP region is more or less uniform ranging from 2 to 5%.

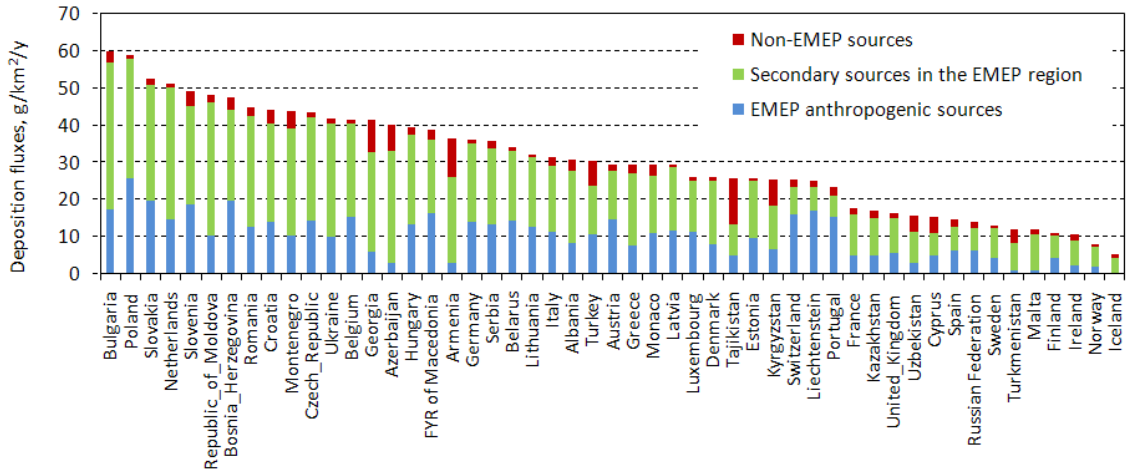

Fig. 2.15. *Country-averaged deposition fluxes of cadmium from the European and Central Asian anthropogenic, secondary and non-EMEP emission sources in 2014*

Transboundary transport in 2014

Anthropogenic deposition fluxes to countries are split in two parts: national, which comes from emission sources located within a country, and foreign sources located abroad and which influence is reflected through transboundary atmospheric transport. The fraction of deposition from foreign sources to anthropogenic deposition depends on numerous factors such as size of a country, magnitude and spatial distribution of national emission sources, proximity of major foreign emission sources to the state borders of a country, predominating wind patterns etc. Contribution of transboundary deposition of lead exceeds that of national sources in 24 countries (Fig. 2.16). The corresponding value for cadmium is 38 countries, and for mercury – 17 countries. As a rule, the highest contribution of transboundary transport to deposition is characteristic of countries with relatively small territory or with low density of national emissions, for example, Monaco, Lichtenstein, Kyrgyzstan, Latvia, Georgia etc., where this contribution exceeds 80%. In countries known for comparatively large emissions (e.g., Poland, Italy, Spain etc.) transboundary transport contributes less than 25% to deposition of lead, cadmium and mercury.

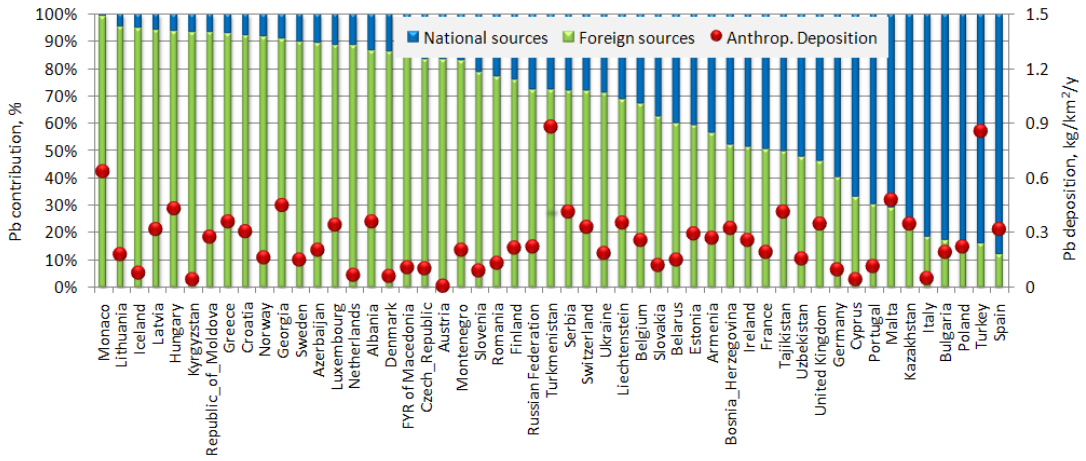

Fig. 2.16. *Relative contribution of the transboundary transport and national sources to anthropogenic lead deposition in the European and the Central Asian countries and deposition values from anthropogenic emission sources in 2014*

It is important to note that over country's territory the contribution of foreign sources to deposition differs significantly. Obviously, the highest contribution occurs along the state borders, while the lowest – nearby powerful national emission sources. Information about spatial distribution of contributions of foreign sources to each country is available in the internet [http://www.msceast.org/index.php/pollution-assessment/emep-countries-menu].

Heavy metals, emitted from sources of a country, partly fall out within the country's territory and partly get involved into transboundary transport, depositing to territories of other EMEP countries or elsewhere. Amount of the metals entering transboundary transport could be expressed in absolute and relative (fraction of emission) terms (Fig. 2.17). For example, emission of cadmium in Russia, used in modeling for 2014, is about 80 tonnes per year. About 60 tonnes of cadmium, emitted by Russian sources, are deposited within the country's territory, around 10 tonnes – to territories of

other EMEP countries and remaining mass (about 10 tonnes) is transported outside the EMEP countries. Therefore, about 20 tonnes (about 25% of national emission) of emitted cadmium enter transboundary transport.

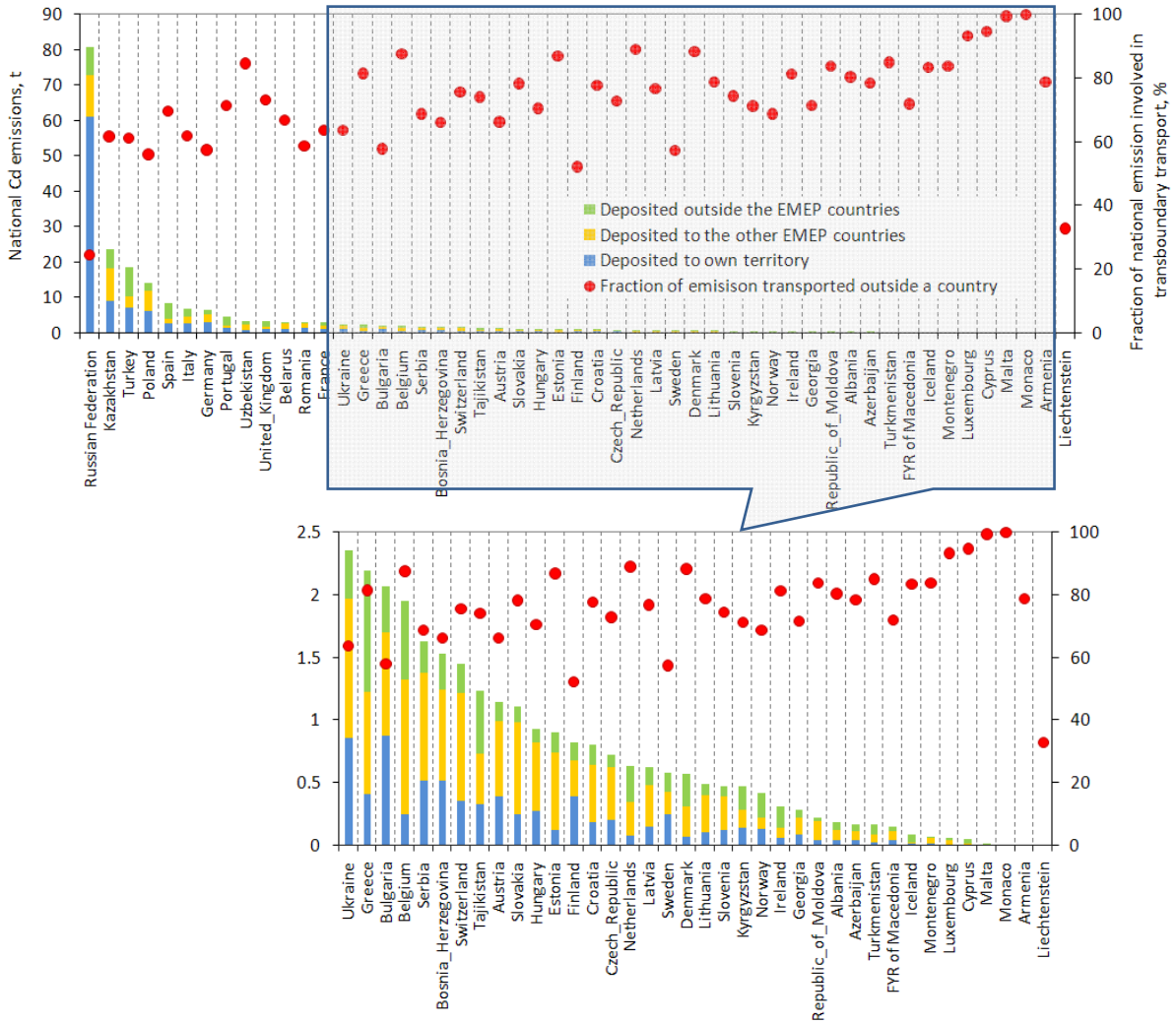

Fig. 2.17*. Distribution of cadmium emitted in the EMEP countries between deposition to own territory, deposition to the other EMEP countries and deposition outside the EMEP countries in 2014. Red dots indicate relative fraction of national emissions involved into the transboundary pollution*

In most of the EMEP countries from 60% to 90% of emitted lead and cadmium are transported outside territories of the countries. Russia is the exception: because of vast territory most of emitted lead and cadmium are deposited within the country, and relatively small fraction (25% for lead and cadmium) is transported outside.

Distribution of emitted mercury between deposition within a country, outside a country and outside the EMP region differs from that for lead and cadmium. While for lead and cadmium from 10 to 60% mass, emitted in countries (with few exceptions) is transported outside the EMEP region, for mercury this fraction varies from 60 to almost 97% (Fig. 2.18). As a rule, most of lead, cadmium and mercury, emitted by country's sources, is transported outside country's territory. However, most of lead and cadmium is deposited within the EMEP region, while most of mercury becomes involved in intercontinental transport.

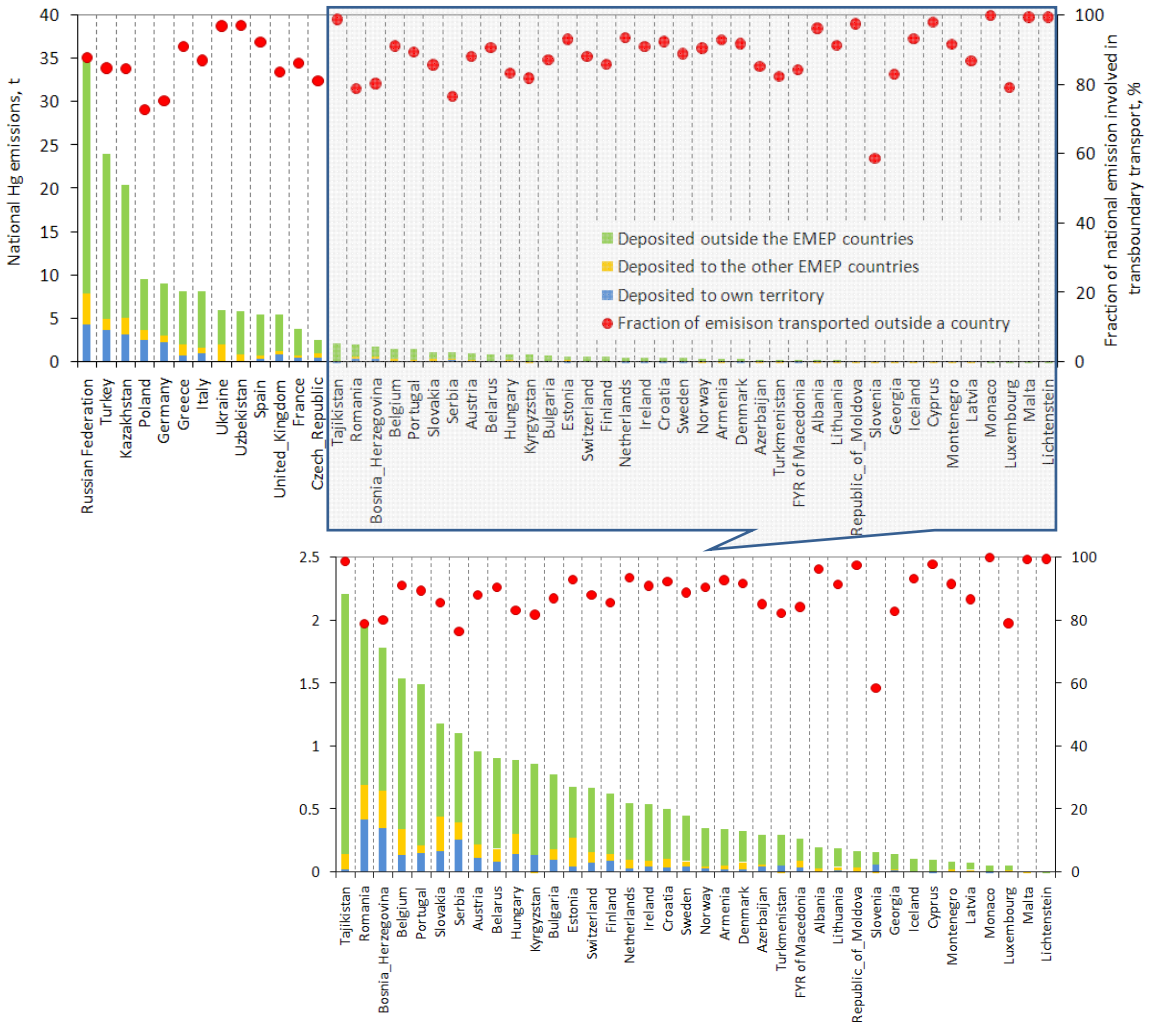

***Fig. 2.18**. Distribution of mercury emitted in the EMEP countries between deposition to own territory, deposition to the other EMEP countries and deposition outside the EMEP countries in 2014. Red dots indicate relative fraction of national emissions involved into the transboundary pollution*

Changes of pollution levels between 2013 and 2014

Changes of calculated pollution levels can be caused by various reasons. First of all, it is increase or decrease of reported anthropogenic emissions within the considered modeling domain. Another factor is variability of meteorological conditions, which affect changes of pollution levels directly – through changes of precipitation amounts, transport patterns or temperature, and indirectly – through modifying wind re-suspension of dust containing heavy metals. In this section changes of pollution levels between previous (2013) and current (2014) years are considered and exemplified by cadmium.

Changes of cadmium deposition between 2013 and 2014, expressed in the absolute terms, are the highest in the southern and southeastern part of Europe (Russia, Ukraine, Balkan countries), in the central part of the European territory of Russia and over south of Scandinavia (Fig. 2.19a). Relative changes were calculated as difference of deposition between 2014 and 2013 divided by deposition in

2013. The highest increase of deposition (> 60%) takes place also over the southeastern part of Europe, over the northern part of Russia and over Scandinavia (Fig 2.19b). Significant (30-60%) decline of deposition takes place in the western part of Europe (France, Germany) and in the western and eastern parts of the Mediterranean region.

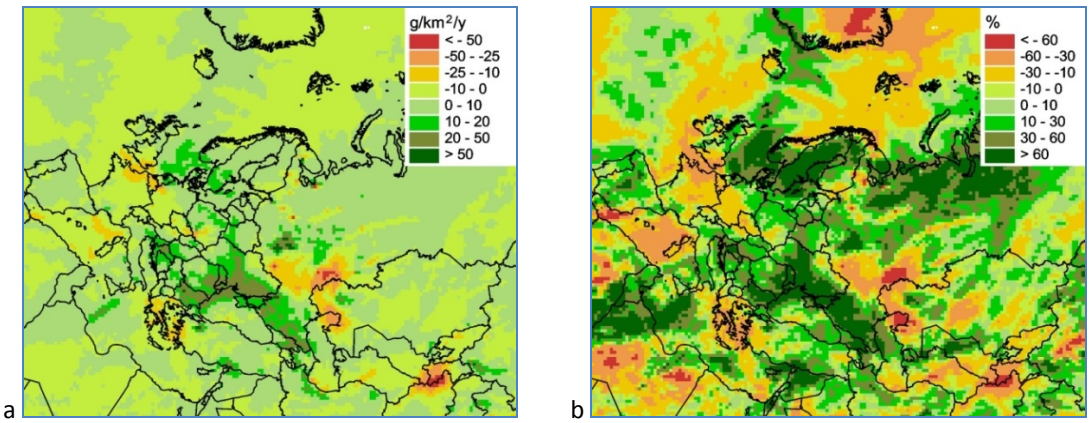

Fig 2.19. *Absolute (a) and relative (b) changes of cadmium total deposition between 2013 and 2014. Positive change means increase, and negative – decrease of deposition in 2014 compared to 2013*

Country-averaged relative changes of deposition are demonstrated in Fig. 2.20. The highest (35-75%) increase of deposition occurred in the Scandinavian countries (Sweden, Denmark) and in south/south-eastern 'belt' of Europe: Georgia, Ukraine, Moldova, Cyprus, Croatia, Bulgaria. Somewhat smaller (25-30%) increase is also noted for other 'south-eastern' countries such as Romania, Slovenia, Serbia, Hungary. The highest decrease takes place in Tajikistan (40%), followed by the western European countries (Belgium, Luxembourg, France) (20-30%). Besides, deposition in Greece declined by 23%.

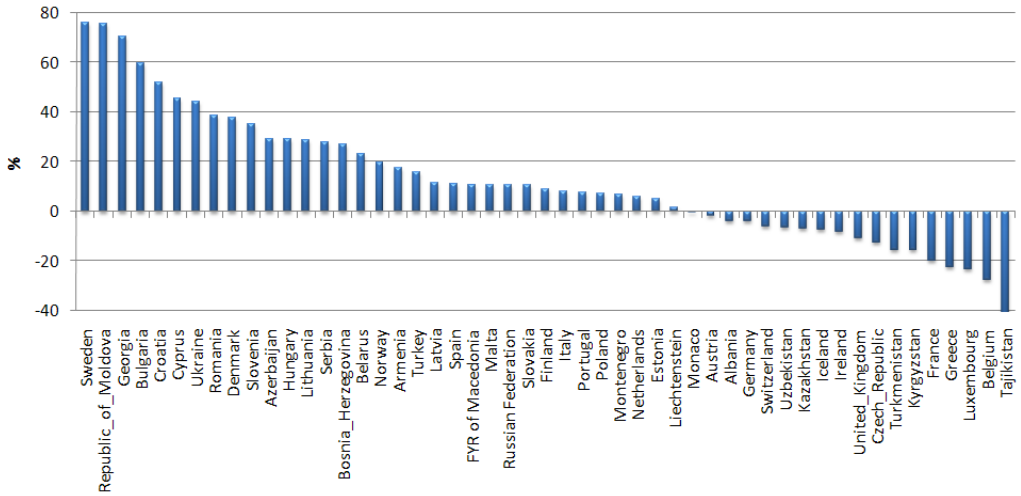

Fig. 2.20. *Relative changes of country-averaged cadmium total deposition between 2013 and 2014*

In order to distinguish three possible reasons of changes between 2013 and 2014 additional modeling tests are performed. In each test one of factors (emission, re-suspension, meteorological conditions) is changed to situation of 2013, while two others are kept at values for 2014. Comparing

deposition computed in these tests with the 'base-case' simulations (operational modeling for 2014) the effect of each of the factor is estimated (Fig. 2.21).

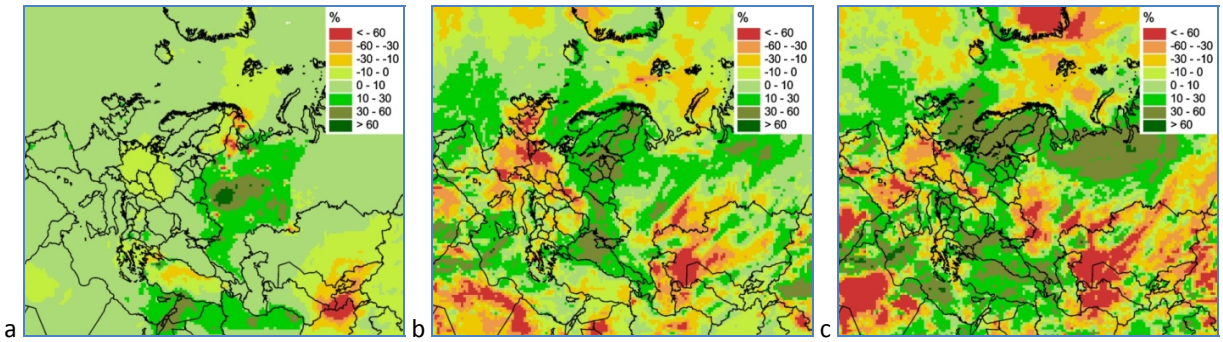

Fig. 2.21. *Effects of cadmium deposition changes (% relative to 2014) between 2013 and 2014 caused by changes of reported anthropogenic emissions (a), wind re-suspension (b) and meteorological conditions (c). Positive values mean increase of deposition from 2013 to 2014, and vice versa*

The effect of changes of emission data between calculations for 2013 and 2014 is the strongest for the central part of European territory of Russia (Fig. 2.21a). The increase of cadmium emissions in Russia from almost 45 to around 80 tonnes [*Tista et al*., 2016] leads to obvious growth of deposition in this country. In spite of 70-80% increase of reported cadmium emissions in Bulgaria and Croatia (Fig. 2.1b) the corresponding growth of deposition in these countries is much lower, mainly due to transboundary influence of emissions in neighbouring countries.

The effect of wind re-suspension change favours declining of deposition from 2013 to 2014 in the western part of Europe (the United Kingdom, France, Germany) and in the north-western part of Kazakhstan (Fig. 2.21b). At the same time this factor explains significant rise of deposition in Ukraine and the southern regions of Russia. This increase is associated with higher wind-blown dust emission, caused by drier weather conditions in certain months in 2014 compared to 2013 (January, March, July, September, August).

Another region where the factor of re-suspension is responsible for increase of pollution levels is Scandinavia and the Baltic region. Comparison of re-suspension fluxes in 2013 and 2014 in these regions demonstrates negative change (Fig 2.22). Therefore, the elevated contribution of re-suspension to deposition in Scandinavia and surrounding regions has to be explained by effect of atmospheric transport.

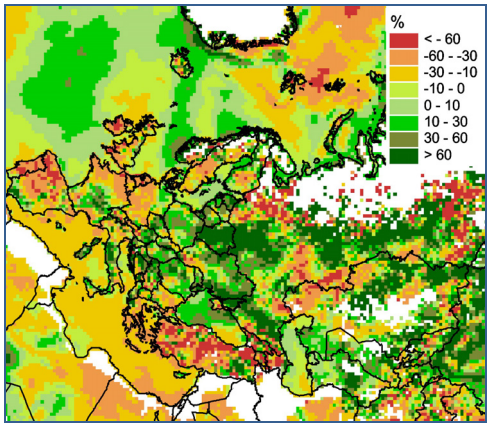

Fig. 2.22. *Relative changes of cadmium wind re-suspension between 2013 and 2014. Positive change means increase and negative – decrease in 2014 compared to 2013*

The factor of changes of meteorological conditions between 2013 and 2014 is the most pronounced in western and central Europe, western Kazakhstan, in the northern part of Russia, in Turkey, in the Balkan region and in Scandinavia (Fig. 2.21c). Negative effect in western and central Europe (France,

Germany, the Czech Republic) and in western Kazakhstan can be attributed to smaller annual precipitation in 2014 compared to 2013 (Fig. B.3b, Annex B). Increase of precipitation amounts over northern part of Russia, Turkey and especially over the Balkan region favours increasing of deposition fluxes in these regions.

Another situation takes place for Scandinavia. Change in meteorological conditions is the main factor responsible for deposition increase in Scandinavia (Fig. 2.21c). The change of precipitation in this region is relatively low: 10 - 30% increase in northern Sweden and Finland and 10 – 30% decline in southern Sweden. It is much lower than the overall change caused by meteorological factor (30-60%) (Fig. 2.21c). Therefore, the changes should be linked to variability of atmospheric transport.

Deposition to Sweden is used as an example for more detailed analysis of the changes between 2013 and 2014. Total deposition to the country is split in three components: deposition from anthropogenic, secondary and non-EMEP sources (Fig. 2.23a). As seen, the contribution of anthropogenic sources in 2014 is 1.7-fold higher, and of secondary sources two-fold higher compared to corresponding contributions in 2013. The contribution of non-EMEP sources is quite low and remains almost the same. On the base of source-receptor calculations contributions of particular countries to anthropogenic deposition are established (Fig. 2.23b). Deposition from national sources have risen by 30%, which is in line with 20% increase of national emissions and around 30% growth of precipitation in the southern, most populated and industrialized part of the country. Besides, almost two-fold increase of contribution of Russian sources is explained by higher emission values used in calculations for 2014 compared to 2013. Poland is the main source country of cadmium atmospheric pollution in Sweden in 2013 and 2014. Contribution of Polish sources increased by 2.1 times from 2013 to 2014, in spite of 9% decline of Polish cadmium emissions between these two years (Fig. 2.1b).

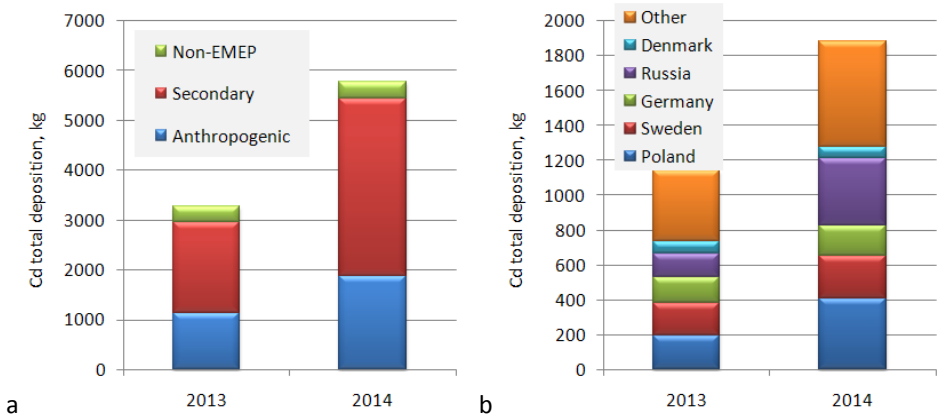

Fig. 2.23. *Contribution of anthropogenic, secondary and non-EMEP sources to cadmium deposition (a) and contributions of national and main foreign sources to deposition (b) in Sweden in 2013 and 2014*

In order to identify the effect of changes in peculiarities of atmospheric transport between 2013 and 2014 contribution to deposition in Sweden from each EMEP country in 2014 and 2013 was divided by corresponding country's emission. This ratio can be considered as simplified form of influence function and characterizes potential influence of a region (country) to Sweden. These ratios

calculated for foreign sources for 2014 and 2013 are depicted in Fig. 2.24. As seen, potential influence increased considerably in countries located towards the south or south-east of Sweden, such as the Baltic region (Poland, Latvia, Estonia, Lithuania), Balkans (Serbia, Croatia, Slovenia etc.), and central and south-eastern Europe (Slovakia, Romania, the Czech Republic etc.). It means that the role of meridianal atmospheric transport in 2014 has much stronger impact compared to 2013.

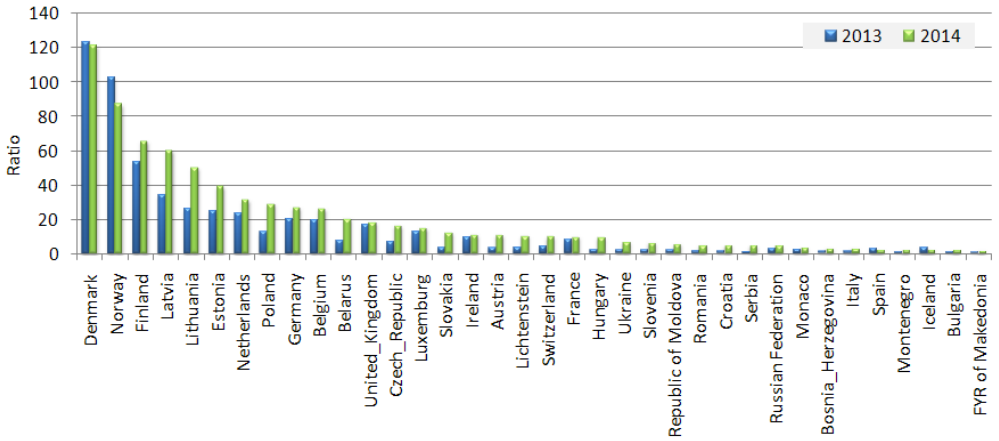

Fig. 2.24. *Potential influence of foreign countries on cadmium deposition in Sweden in 2013 and 2014, expressed as a ratio of deposition from country's emission sources to country's emissions*

Compared to 2013, in 2014 high deposition takes place in winter (January, February), in spring (March, May) and in autumn (November) (Fig. 2.25). It is worth mentioning that monthly deposition does not always correspond with monthly sums of precipitation: for example, high precipitation in July and August in 2014 are associated with relatively low deposition in these months.

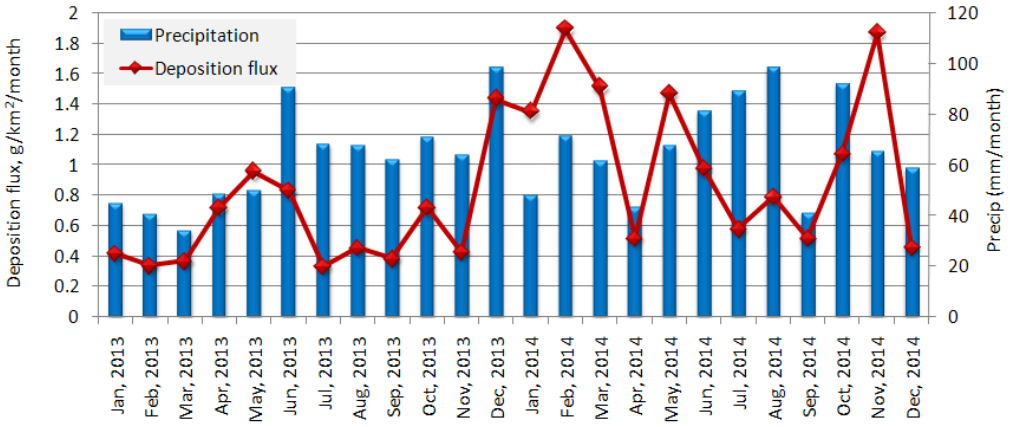

Fig. 2.25. *Monthly sum deposition of cadmium to Sweden and precipitation sums in 2013 and 2014*

Back trajectories are analyzed for one of monitoring stations in Sweden (SE14, Råö) in two months of 2014: November, when deposition is high and July, when low deposition of cadmium takes place. In November transport from the eastern part of Europe (south of Russia, Ukraine) and from central and western Europe (Poland, Germany, the Netherlands, Belgium, north of France) is distinct (Fig. 2.26a). In July, when deposition are relatively low, most of trajectories are passing over north of the United Kingdom or north-west of Russia (Fig. 2.26b).

Brief analysis of pollution level changes between 2013 and 2014 demonstrates that variability of meteorological conditions can markedly affect pollution levels in the EMEP region. In some countries changes of annual deposition of heavy metals exceed 60%. Contribution of particular countries to foreign deposition due to peculiarities of atmospheric circulation may change to similar extent. *Therefore, the effect of year-to-year meteorological variability on source-receptor relationships in particular countries can be as strong as or even stronger than inter-annual changes in country's emission data. It means that simulation of source-receptor matrices should be done regularly in order to take into account all peculiarities (emissions changes, meteorological variability) of a particular year, which affect pollution levels in the EMEP countries.*

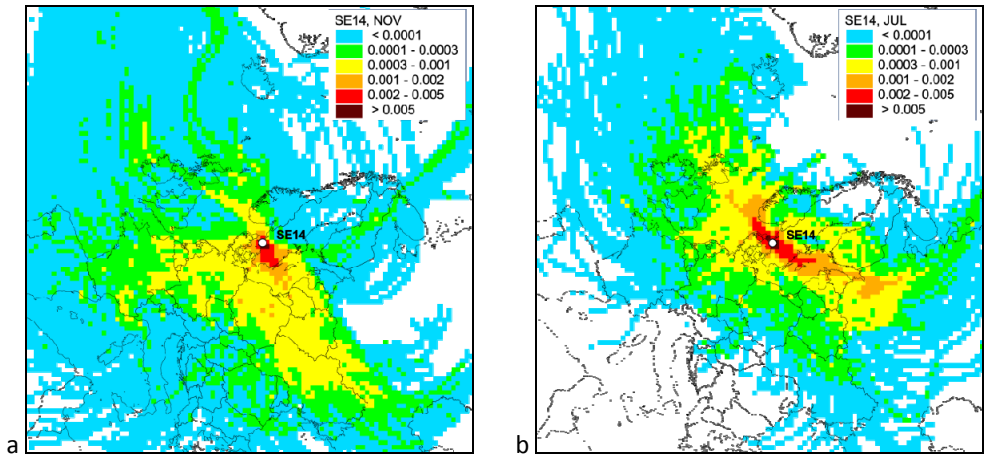

Fig. 2.26. *Density of back trajectories for station SE14 (Råö, Sweden) in November (a) and July (b), 2014*

2.5. Ecosystem-dependent deposition

Heavy metals are known for their harmful effects of human health and biota. They can accumulate in the environmental media and biomagnify in tissues of living organisms. At a certain deposition level (so-called critical load) steady-state concentrations in media (soil, waters, tissues etc) exceed their threshold values above which possible harmful effects may occur [*Hettelingh et al.*, 2015]. Exceedance of deposition over critical load is proposed to be used within Convention as a measure of negative effect to the environment.

Both deposition and critical loads differ deepening on particular ecosystem. In order to take it into account, deposition fluxes of heavy metals are calculated to different land-cover categories. For each heavy metal priority of considered ecosystems may differ depending on a pathway by which the metal makes an effect on human health or ecosystem functioning. For example, vegetable food is one of the main sources of lead and cadmium in humans [*WHO*, 2007]. Therefore, analysis of lead and cadmium deposition to agricultural lands is of priority attention. Besides, negative effects are indicated for soil species (microorganisms, fungi, nematodes, earthworms etc.) [*de Vries et al.*, 2015 and references therein]. For the last two decades deposition of lead to natural surfaces (grasslands,

semi-natural surfaces, wetlands, tundra and inland waters) declined by about 71%, and to forests – by almost 80% [*de Wit et al*., 2016]. The corresponding values for cadmium are about 57% and 53%. Measurements of lead and cadmium in upper (organic) forest soil layer also demonstrated decline in the upper soil layers between 1994 and 2011. However, these metals are being transferred to deeper soil layers and continue to accumulate there [*de Wit et al*., 2016].

Mercury is most toxic in its methylated form, which is produced in aquatic environments (lakes, rivers, wetlands etc). Deposition of mercury is declined by 23% for the EMEP region as a whole for the last two decades. In particular, the decline to forests almost corresponds to that averaged for EMEP and equals to 23%, and to the natural surfaces it amounts to 16% [*de Wit at al*., 2016]. Consequently, the area where mercury levels exceeded critical loads, also reduced from 69% in 1990 to 56% in 2010 [*de Wit et al*., 2016]. However, it is noted that concentrations of mercury levels in fish and in Swedish forest soils continued to increase [*de Wit et al*., 2016]. Fig. 2.27 demonstrates levels of mercury annual deposition to inland waters and forests. Spatial pattern of mercury deposition corresponds to spatial distribution of mercury emissions. Relatively high levels take place in regions of central and western Europe (Poland, the Balkan countries, northern Italy, Benelux), and in some regions of the EECCA countries (east of Ukraine, east of Russia, Kyrgyzstan). Deposition fluxes to inland waters are lower compared to deposition to forests. It is explained by higher deposition velocity values for forests compared to low-vegetation ecosystems and water surfaces.

Information about ecosystem-dependent deposition is prepared regularly for each heavy metal and for each type of land-cover. Country-specific information about ecosystem-dependent deposition is available at the MSC-E web-site for all the EMEP countries [www.msceast.org].

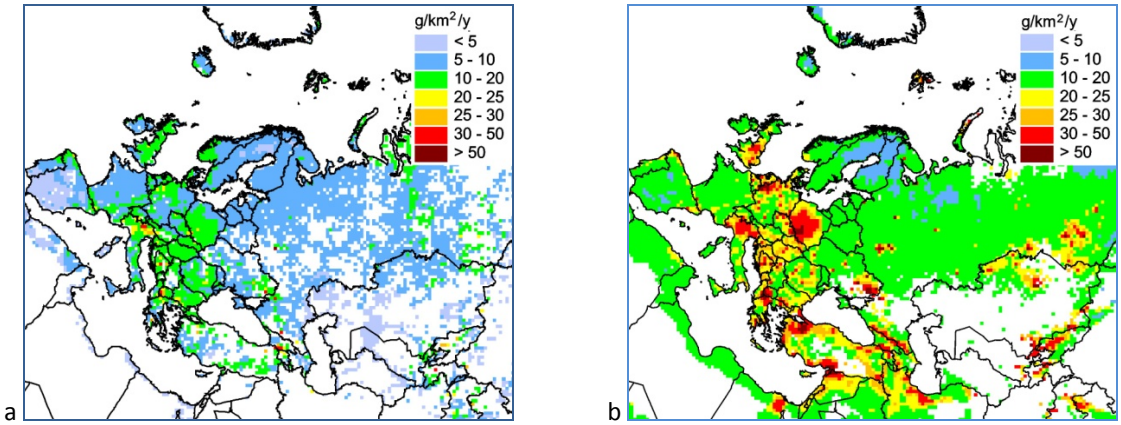

Fig. 2.27. *Annual mercury deposition flux to inland waters (a) and forests (b) in 2014*

Ecosystem-dependent deposition of lead, cadmium and mercury as well as critical load exccedances of these metals declined for the last two decades. Decrease of lead and cadmium concentrations in upper (organic) forest soils is also indicated. However, concentrations of all three heavy metals in deeper soil layers, and levels of mercury in fish continue to increase. Therefore, there is a need to continue activity on evaluation of negative effects of heavy metals in the environment.

2.6. Near-real time estimates of pollution levels (2015)

According to the current practice adopted in the Convention, submission of emissions and information about pollution levels becomes available for the EMEP countries with a two-year delay. In order to reduce the existing two-year gap, so-called 'near-real time' calculations of lead and cadmium levels were performed. The calculations were done on the base of meteorological data for 2015 and emission data for 2014.

Since the pollution levels for 2014 and 2015 have been calculated with the usage of the same emission data, the differences between results of these two calculations are explained by changes in meteorological conditions. These are both direct changes, for example, increase or decrease of deposition because of corresponding rise or decline of precipitation amounts, and indirect changes, caused by variability of wind re-suspension fluxes which are also affected by meteorological conditions.

Example of deposition changes, expressed in absolute and relative terms, is given for cadmium in Fig. 2.28. Absolute change is calculated as difference of the values related to 2014 and those related to 2015. Relative change was calculated as absolute difference divided by values for 2014 and expressed in per cents. As seen, in 2015 increase of deposition is expected in the central and southern regions of European territory of Russia, over northern part of France and Germany. Decline is expected over the central part of Europe, the Balkan region and Scandinavia.

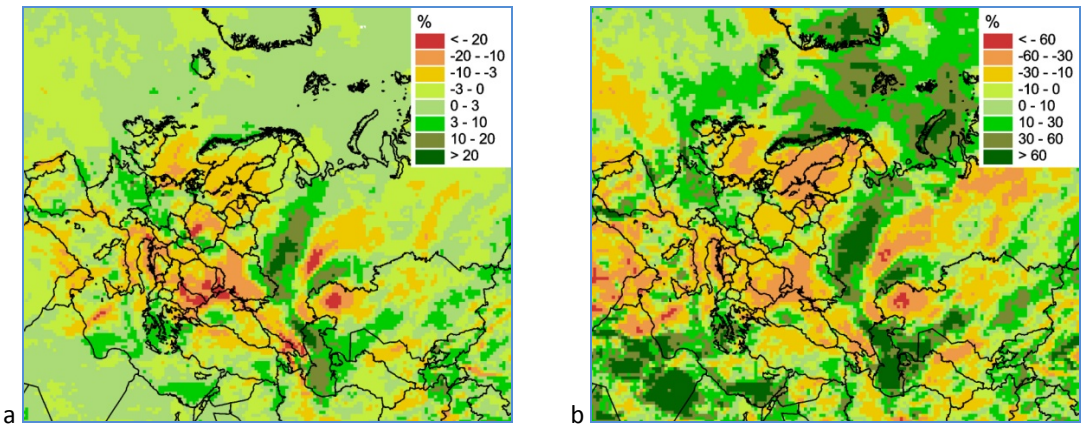

Fig. 2.28. *Absolute (a) and relative (b) changes of cadmium deposition between 2014 and 2015. Positive change mean increase, and negative - decrease of deposition between 2014 and 2015*

In most of countries the decline of country-averaged deposition is projected (Fig. 2.29). The strongest decrease is supposed to be in Ireland, Sweden and the United Kingdom (23-26%). The highest increase of country-averaged deposition of cadmium in 2015 is expected in Malta (almost 50%), followed by Kyrgyzstan and Kazakhstan (8-10%).

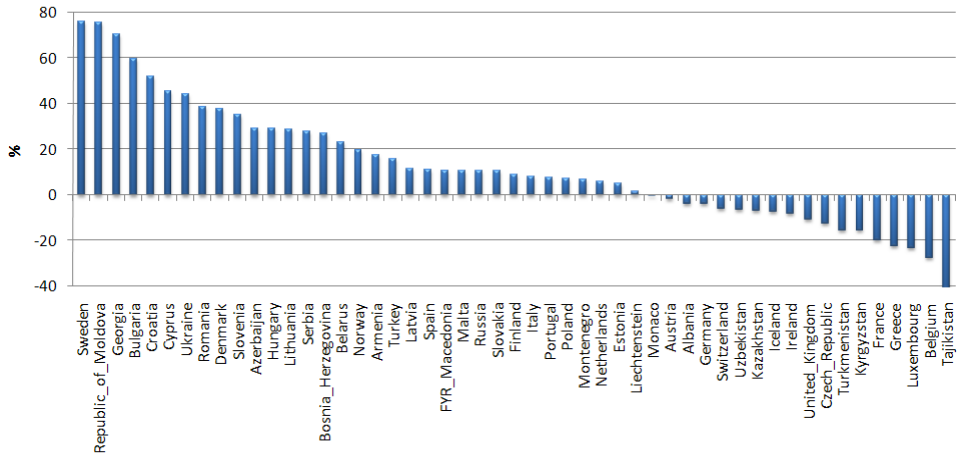

Fig. 2.29. *Relative change of country-averaged deposition of cadmium between 2014 and 2015. Positive change mean increase, and negative - decrease of deposition between 2014 and 2015*

Near-real time modeling provides the EMEP countries with the preliminary information on pollution levels related to the next reporting year. These data help to inform national experts about expected changes of the pollution caused by changing meteorological conditions.

2.7. Future changes of mercury pollution levels in Europe (2035)

Evaluation of future mercury pollution levels and different abatement scenarios are the key information for development of future mitigation measures. It is particularly topical in the context of the recently adopted Minamata Convention for Mercury – a global treaty to protect human health and the environment from the adverse effects of mercury [http://www.mercuryconvention.org/]. The current study is focused on model assessment of future changes of mercury atmospheric load in Europe as well as source apportionment of mercury deposition. For this purpose a number of model simulations have been performed for selected mercury emission scenarios for 2035. Description of the emission scenarios and evaluation of the future pollution levels on a global scale were presented in [*Ilyin et al.*, 2015]. Main results of the current study for Europe are discussed below.

Briefly, the scenarios used in the study include:

- The 'Current Policy' scenario (CP 2035) assuming that governmental policies and measures existing in 2010 have been adopted, including those that have not been fully implemented.
- The 'New Policy' scenario (NP 2035) assuming that policy commitments and plans announced by countries worldwide to reduce greenhouse gas emissions, as well as phase out fossil-energy subsidies, were fully implemented.
- The 'Maximum Feasible Reduction' scenario (MFR 2035) set out a target of all counties reaching the highest feasible reduction efficiency in each emission sector.

Figure 2.30 shows distributions of mercury deposition in the EMEP region simulated for the reference year 2013 and three scenarios of 2035. The largest deposition levels in Europe in 2013 are characteristics of Central Europe (Germany, Poland, the Czech Republic, and Slovakia) and the Balkan countries (The FYR of Macedonia, Albania, Bulgaria, Serbia, Greece, etc.) (Fig. 2.30a). The elevated

deposition fluxes (more than 20 g/km^2/y) are determined by two factors: significant emissions of oxidized mercury species from direct anthropogenic sources located in these areas and enhanced photo-oxidation of globally transported elemental mercury in the lower latitudes. The model also predicts increased deposition fluxes in the Arctic due to the effect of the Atmospheric Mercury Depletion Events, AMDEs [*Schroeder et al*., 1998; *Lindberg et al*., 2002; *Steffen et al*., 2008]. The lowest deposition fluxes are over remote aquatic and terrestrial areas of the Atlantic, Siberia, Central Asia and Northern Africa.

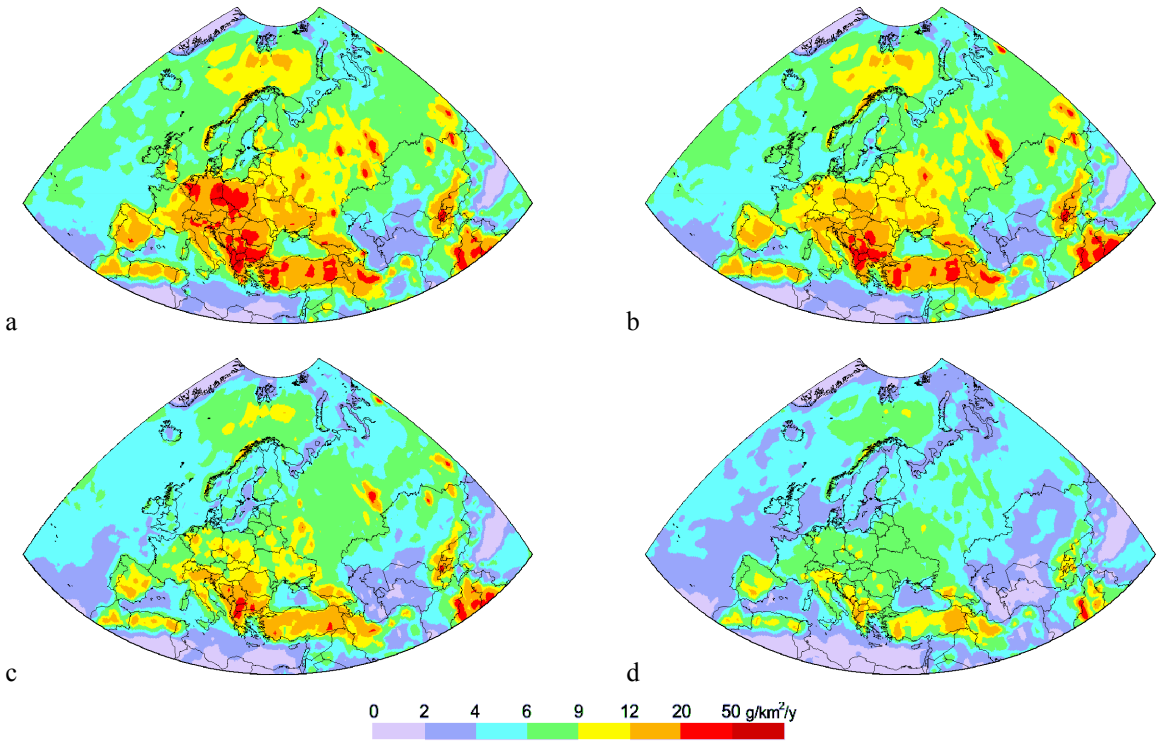

Fig. 2.30. *Spatial distribution of mercury deposition flux in Europe in 2013 (a) and in 2035 according to different emission scenarios: (b) – CP 2035; (c) – NP 2035; (d) – MFR 2035*

All three future scenarios predict general decrease of mercury deposition in Europe by 2035 from moderate reduction for CP 2035 to strong reduction for MFR 2035 (Figs 2.30b,d). Elevated levels of mercury deposition (above 12 g/km^2/y) will still remain in the Balkans and Southern Europe as well as in some 'hot spots' of Central and Eastern Europe as predicted by CP 2035 and NP 2035. According to the MFR 2035 scenario, mercury deposition will descend below 10 g/km^2/y almost over the whole European region.

Projections of relative changes of mercury deposition in the EMEP region between 2013 and 2035 according to the three selected scenarios are illustrated in Fig. 2.31. The 'Current Policy' scenario (CP 2035) supposes considerable decrease (20-50%) of mercury deposition in Western Europe and smaller reduction in other parts of the region (0-20%). In contrast, there are a number of 'hot spots' in Eastern Europe and Central Asia where significant growth (up to 50%) of mercury deposition is predicted. It results from the growth of local mercury emissions and impact of increasing emissions in South and East Asia. According to the 'New Policy' scenario (NP 2035) moderate decrease of mercury

deposition (more than 20%) is predicted over the whole European region, with stronger reduction in Central Europe (above 50%) and weaker reduction in contiguous regions (below 20%). Model predictions based on the 'Maximum Feasible Reduction' scenario (MFR2035) demonstrate significant reduction of mercury deposition over the whole domain with the largest decrease in Western and Central Europe and the Balkans (up to 70%).

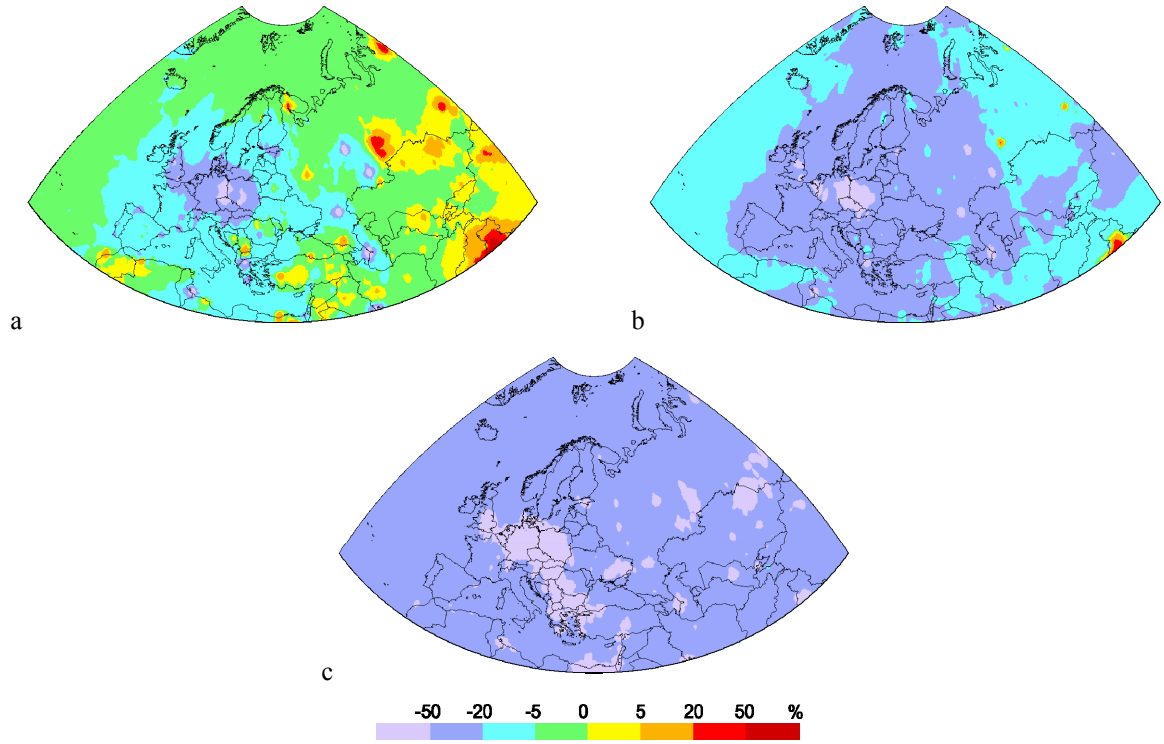

Fig. 2.31. *Relative change of mercury deposition flux in Europe between 2013 and 2035 according to different emission scenarios: (a) – CP 2035; (b) – NP 2035; (c) – MFR 2035*

Mercury is known as a global pollutant transported over long distances in the atmosphere. Therefore, changes of mercury pollution in the EMEP countries depend not only on emission dynamics in the region but also on emission changes in other regions of the globe. Figure 2.32 shows estimated contribution of various source regions to mercury deposition in the EMEP countries in 2013 and for the scenarios of 2035. Average mercury deposition in the EMEP countries in 2013 consists of contributions of domestic anthropogenic sources (30%), foreign anthropogenic sources (24%) and natural/legacy sources (46%). As it has been mentioned we assume that natural and legacy emissions will not changed significantly over the next 20 years. Thus, reduction of anthropogenic emissions in Europe and other regions leads to decrease of mercury deposition in the EMEP region and also to change of relative contribution of direct anthropogenic and natural/legacy sources. Decrease of contribution of the former leads to increase of relative contribution of the latter. Thus, total contribution of domestic and foreign anthropogenic sources decrease from 54% in 2013 to 15-50% in 2035. Decrease of mercury deposition in the EMEP region between 2013 and 2035 is largely defined by reduction of domestic emissions and, to less extent, by emission reduction in other regions (first of all, in East Asia).

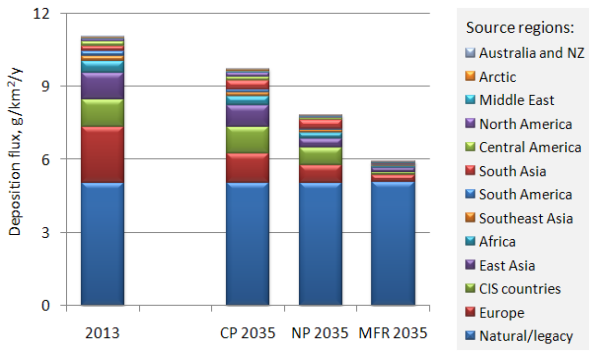

Fig. 2.32. *Source apportionment of average mercury deposition to the EMEP countries in 2013 and 2035 according to different emission scenarios*

Changes of mercury deposition levels as well as source apportionment differ in different parts of the EMEP region. Dynamics and composition of mercury deposition in two different parts of the region – European Union (EU28) and the EECCA countries – are shown in Fig. 2.33. For the sake of simplicity, the unchanged contribution of natural and legacy sources is not shown in the figure. The highest contemporary deposition and the largest reduction are characteristics of the EU countries (Fig. 2.33). All three scenarios expect significant emission reduction in Europe and mercury deposition in the EU mostly follows the changes of domestic emissions (Fig. 2.33a).

Mercury anthropogenic deposition in the EECCA countries depend on contributions of their domestic emission sources and, to less extent, on sources located in other parts of Europe and East Asia (Fig. 2.33b). The CP 2035 scenario does not predict significant deposition change in the EECCA countries by 2035 due to relatively small changes of deposition from domestic sources. Nevertheless, the predicted deposition decrease is noticeable for two other scenarios (NP 2035 and MFR 2035).

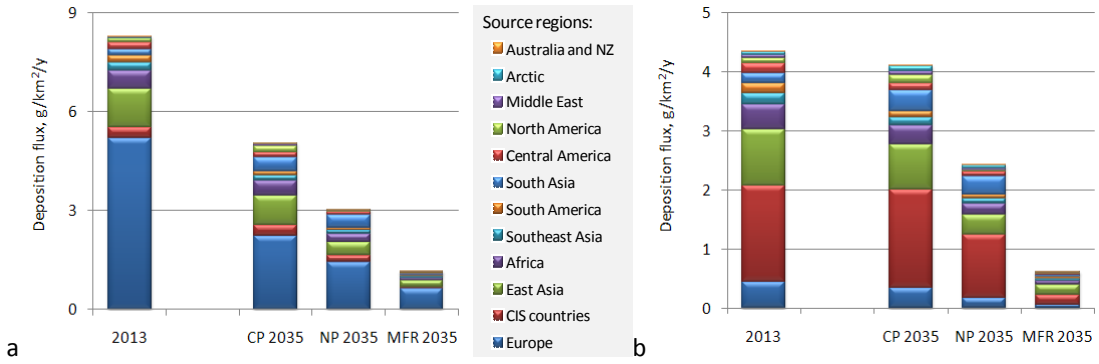

Fig.2.33. *Source apportionment of mercury deposition from direct anthropogenic sources in 2013 and 2035 in different parts of the EMEP region: (a) – European Union (EU28); (b) – EECCA countries*

Thus, the most significant decrease of mercury deposition during the next 20 years is expected in Western and Central Europe for all considered scenarios. At the same time some scenarios predict deposition increase in limited number of 'hot spots' located in Eastern Europe and Central Asia. Decrease of mercury deposition in the EMEP region between 2013 and 2035 is largely defined by reduction of domestic emissions and, to less extent, by emission reduction in other regions (first of all, in East Asia).

3. FURTHER DEVELOPMENT OF GLEMOS MODELING SYSTEM

Global EMEP Multi-media Modeling System (GLEMOS) is a new generation model developed by MSC-E for the operational modeling transboundary pollution of the EMEP countries by heavy metals, POPs and other contaminants. The progress of GLEMOS further update and development are discussed below.

3.1. Pilot calculations on the new EMEP grid

MSC-E continued the work on transition of the EMEP operational modeling of heavy metal and POP pollution to the new EMEP grid following decisions of the Executive Body for CLRTAP [*ECE/EB.AIR/113/Add.1]*. The grid changes include replacement of the polar-stereographic projection by the regular geographic (latitude-longitude) projection of the Earth surface, appropriate transformation of the EMEP domain and increase of the grid spatial resolution. The changes of the projection are motivated by requirement of consistent model studies on different scales (from national to global), better consistency with common databases of input data (emissions, meteorology, land cover etc.) and improved data exchange with other scientific and environment protection communities [*EMEP/SB*, 2012].

Preparatory work on transition to the new grid and first simulation results have been discussed in [*Ilyin et al*., 2014]. The pilot simulations on the new grid with the GLEMOS modeling system have been continued this year. For the sake of testing mercury atmospheric dispersion was calculated for 2014 both on the new grid with spatial resolution 0.2°×0.2° and on the old EMEP grid (50×50 km^2). In addition to the grid difference two regional versions of the model utilized different sets of meteorological and geophysical data as well as concentrations of chemical reactants. Simulation results of both model runs were compared and evaluated against observations. It should be noted that reported heavy metal emission data on the new grid with fine spatial resolution are not available yet. Therefore, to keep consistency between the regional simulations and global simulations used for generation of boundary conditions the most recent expert estimates of mercury anthropogenic emissions on a global scale [*AMAP/UNEP*, 2013] were used instead of the official emission data. The original emission fields with spatial resolution 0.5°×0.5° were interpolated into the old and new EMEP grids. Boundary conditions for regional modeling with 6-hour temporal resolution were retrieved from the GLEMOS simulations on a global scale.

Annual mean concentrations of elemental mercury (Hg^0) in the surface air simulated on the old and new grids are shown in Fig. 3.1. Both versions of the model produce similar spatial patterns with elevated concentrations over Southern Europe and the Mediterranean region and the south-to-north decreasing trend in accordance with available observations. Due to long residence time of Hg^0 in the atmosphere the atmospheric dispersion smoothes difference in the spatial resolution of the model runs. Generally, the new model version predicts somewhat higher concentration levels in the surface layer, probably, more correct reproduction of the vertical mixing and updated concentrations of chemical reactants. Nevertheless, the difference between two model runs is insignificant and commonly does not exceed 10-15%.

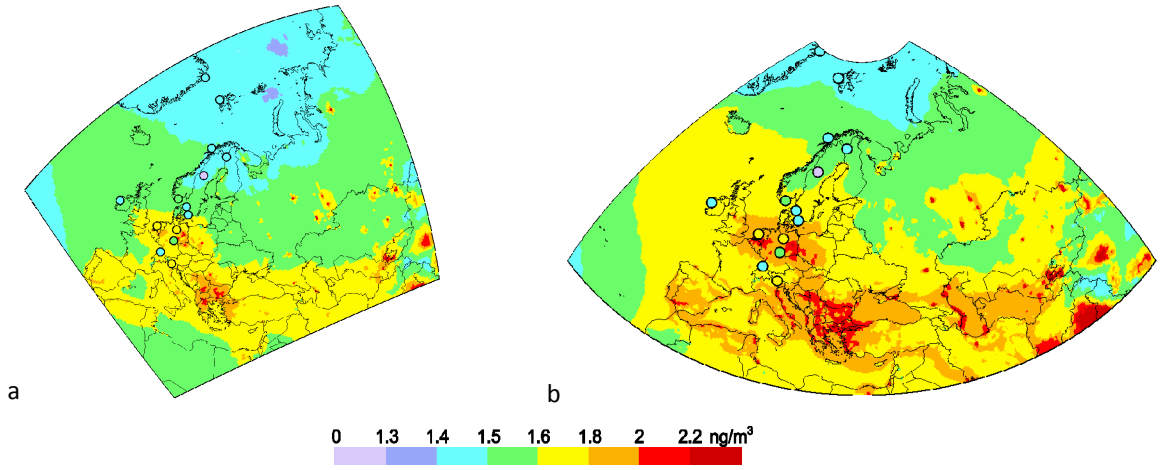

Fig. 3.1. *Simulated annual mean air concentration of Hg^0 in 2014 over the old (a) and new (b) EMEP domains. Coloured circles show mercury observations from the EMEP monitoring network*

Spatial distributions of simulated wet deposition flux for both grids are shown in Fig. 3.2. Both model runs predict similar patterns of wet deposition in Europe. Elevated deposition fluxes are characteristics of Central and Southern Europe as well as of the Caucasus region and Turkey. The lowest fluxes are in Northern Scandinavia and the southern Mediterranean region. Wet deposition of mercury in Europe is largely determined by anthropogenic emissions of oxidized mercury and local precipitation. Therefore, simulations on the new grid present more detailed deposition pattern due to higher spatial resolution of emissions and meteorological fields. On the other hand, there is considerable difference between the modeled deposition levels in remote regions of the Northern Atlantic and the Arctic, where mercury deposition highly depends by oxidation chemistry. Simulations on the new grid utilized updated information on atmospheric concentrations of major chemical reactants involved in reactions of mercury oxidation. It resulted in somewhat lower wet deposition fluxes in these regions.

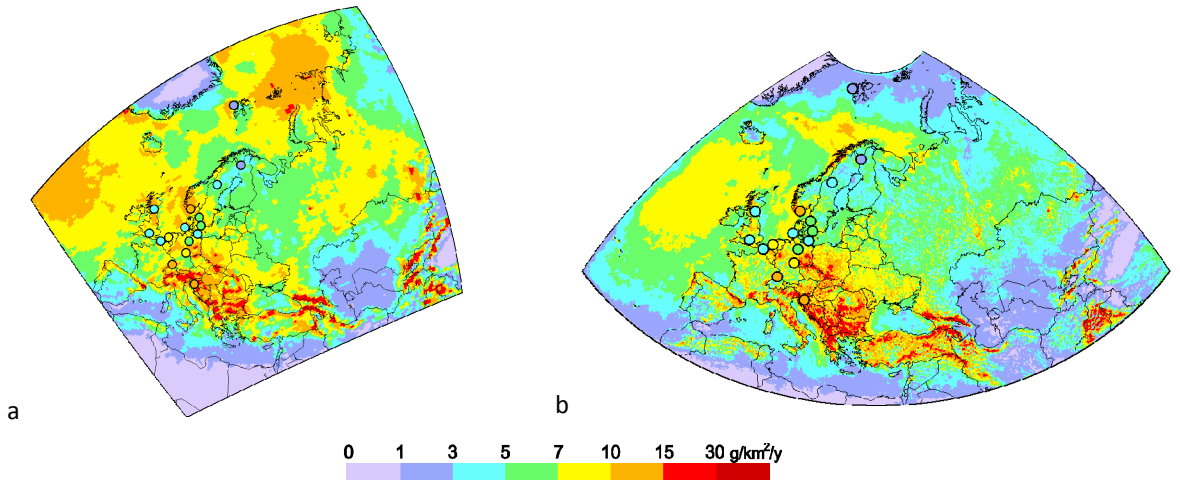

Fig. 3.2. *Simulated annual mercury wet deposition flux in 2014 over the old (a) and new (b) EMEP domains. Coloured circles show mercury observations from the EMEP monitoring network*

Evaluation of the modeling results against observations from the EMEP monitoring network is presented in Fig. 3.3. The major statistics summary is given in Table 3.1. In both runs the model well

reproduces measured air concentrations of Hg^0 (Fig. 3.3a). Levels of Hg^0 concentration in Europe are widely determined by the global background described in the regional modeling by the boundary conditions. That is why the difference between the runs on the old and new grids is negligible. Both model runs demonstrate some overestimation of observed wet deposition fluxes (Fig. 3.3b). However, the simulation on the new grid shows better correlation with measurements (0.92) due to higher spatial resolution and updated data on chemical reactants.

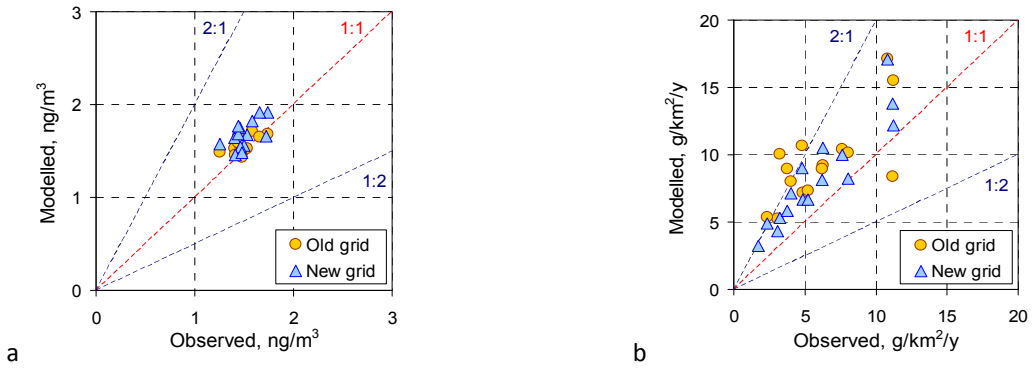

Fig. 3.3. *Evaluation of simulated annual Hg^0 air concentration (a) and mercury wet deposition (b) against EMEP measurements in 2014*

Table 3.1. *Statistics of the model-to-observation comparison for the old and new grids*

	Hg^0 concentration		Wet deposition	
	Old grid	New grid	Old grid	New grid
Correlation coefficient	0.63	0.60	0.72	0.92
Linear regression coefficient	1.06	1.11	1.4	1.35

Figure 3.4 shows simulated maps of total (wet and dry) mercury deposition over the old and new EMEP domains. This is the primary parameter of mercury pollution assessment since it characterizes the overall atmospheric load of mercury to the terrestrial and aquatic ecosystems and, ultimately, the impact on biota and human health. Taking into account that wet deposition prevails over dry deposition in most areas of temperate latitudes (except for arid regions) spatial pattern of total deposition generally follows the wet deposition pattern. On the other hand, total deposition is characterized by less pronounced the south-to-north decreasing trend. Besides, there are elevated deposition levels in Central Europe, Scandinavia and the northern part of the Russian Federation. The enhanced deposition fluxes in Central Europe are determined by dry deposition of oxidized mercury from direct anthropogenic emissions, whereas the elevated deposition in high latitudes is mostly affected by oxidation chemistry. As in the case of wet deposition, the model results on the new grid show significantly lower deposition levels in the high Arctic in comparison with the old grid due to updated data on chemical reactants.

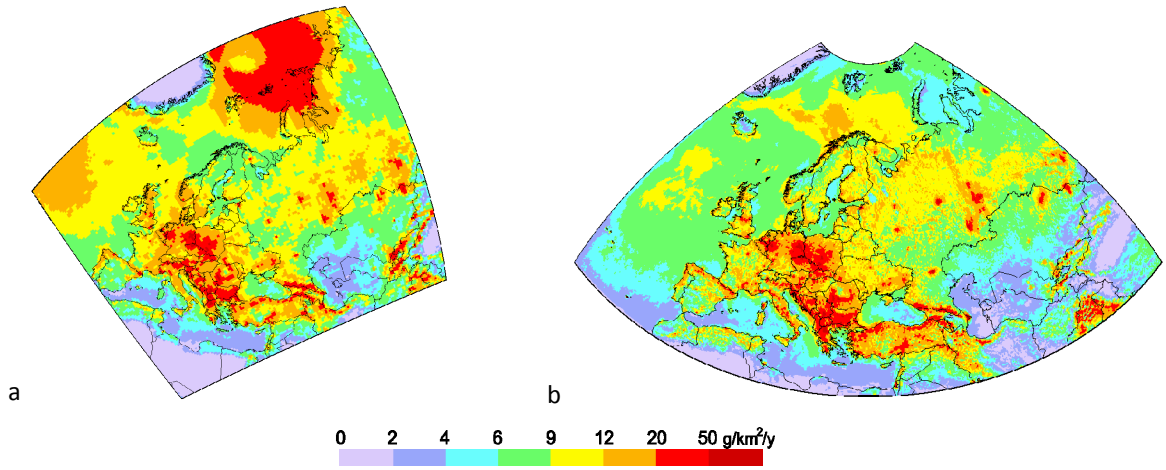

Fig. 3.4. *Simulated annual mercury total deposition flux in 2014 over the old (a) and new (b) EMEP domains*

Comparison of average mercury deposition to the EMEP countries simulated on the old and new grids is presented in Fig. 3.5. As seen average deposition levels are very similar for most of the countries. The difference does not exceed 20% in the three fourth of all countries. Considerable difference is obtained for two groups of countries. The first group consists of relatively small countries, whose territories are comparable or smaller than the 50 km gridcell (Monaco, Luxembourg, and Liechtenstein). Simulations with finer resolution provide more accurate pollution assessment of these countries. The other group includes countries located in the mountain regions (Switzerland, Armenia, Azerbaijan, Georgia, Tajikistan, Kyrgyzstan, etc.). Modeling on the finer grid allows better reproducing meteorological conditions over the complex relief of these countries.

Thus, the pilot simulations performed with GLEMOS on the new EMEP grid demonstrate reasonable model performance for the EMEP operational modeling. Increased spatial resolution of the model grid allows better reproduction of pollution dispersion on a country scale. Nevertheless, the model evaluation on the new grid will be continued to cover other heavy metals.

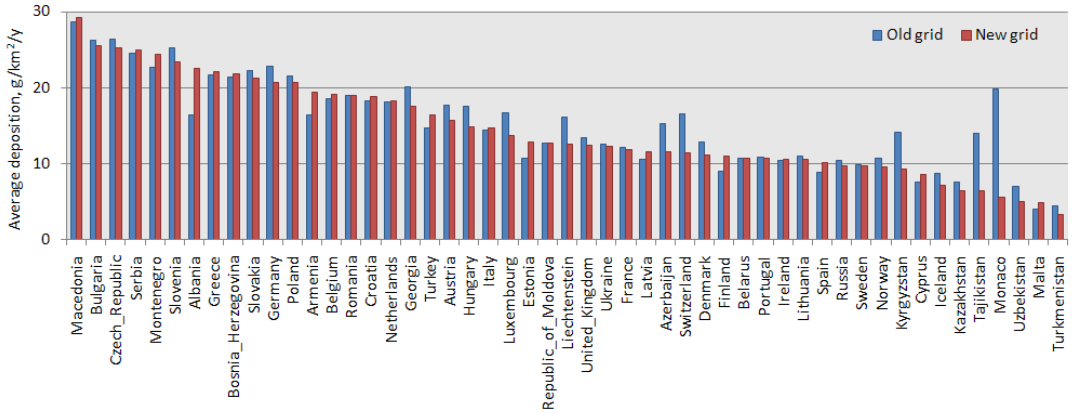

Fig. 3.5. *Comparison of simulated average mercury deposition in the EMEP countries in 2014*

Pilot simulations performed with GLEMOS on the new EMEP grid demonstrate better model performance in comparison with measurements. Simulations with finer resolution provide more accurate pollution assessment of smaller EMEP countries as well as countries with complex relief.

3.2. Multi-model study of mercury atmospheric processes

Oxidation chemistry determines mercury ability to long-term atmospheric transport and deposition to various terrestrial and aquatic regions. Contemporary understanding of mercury cycling in the atmosphere contains significant uncertainties. There are significant gaps in the knowledge of chemical processes affecting mercury atmospheric transport and deposition. Application of chemical transport models (CTM) complemented by extensive measurement data can facilitate a better understanding of the principal mechanisms governing mercury dispersion and cycling in the environment.

Multi-model study of mercury atmospheric processes has been performed as a part of Mercury Modeling Task Force organized within the framework of the EU funded GMOS (Global Mercury Observation System) project [www.gmos.eu]. Four global scale models took part in the study: GLEMOS (EMEP/MSC-E), GEOS-Chem (MIT, USA), GEM-MACH-Hg (Environment and Climate Change Canada), ECHMERIT (CNR-II, Italy). The study was organized in a form of multiple model experiments aimed at evaluation of particular processes and mechanisms of mercury atmospheric chemistry, anthropogenic and natural/secondary emissions, deposition etc. A summary of the major model experiments is given below:

- Simulation with the state-of-the-art model configuration (BASE)
- Model run with no anthropogenic emissions (NOANT)
- Model run with no natural and secondary emissions (NONAT)
- Simulations with atomic Br oxidation chemistry with two different sets of Br air concentrations (BRCHEM1 and BRCHEM2)
- Simulation with OH - initiated oxidation chemistry (OHCHEM)
- Simulation with O_3 - initiated oxidation chemistry (O3CHEM)

The simulation results were evaluated against observations from several monitoring networks. The measurements dataset is based on the EMEP regional network for Europe [*EMEP*, 2016], the NADP/MDN [*NADP/MDN*, 2016], AMNet [*AMNet*, 2016] and NAtChem [*NAtChem*, 2016] networks for North America as well as the global GMOS mercury network [*GMOS*, 2016]. More detailed introduction to the model experiments program, description of input information as well as discussion of initial results of the study have been presented in the MSC-E Status Report 2015 [*Ilyin et al*., 2015]. A summary of the major results of the study is given below.

As it was shown in [*Ilyin et al*., 2015] in the BASE simulation the models predict similar spatial patterns of mercury concentration with pronounced gradient between the Southern and the Northern Hemispheres and elevated concentrations in the major industrial regions – East and South Asia, Europe and North America. Detailed analysis of the meridional gradient of Hg^0 concentration in the surface air is presented in Fig. 3.6. As seen all four models successfully reproduce the observed inter-hemispheric increase of Hg^0 concentration. The lowers concentrations (about 1 ng/m^3) are typical for the temperate and high latitudes of the Southern Hemisphere. There is a weak maximum of zonal-mean Hg^0 concentration (1.4-1.6 ng/m^3) in the temperate latitudes of the Northern Hemisphere corresponding to location of the majority of anthropogenic emission sources. All the

models predict some decrease of concentration northward to the Arctic, which is not seen in the observations. It can be connected with overestimation of the role of Atmospheric Mercury Depletion Events (AMDE) in the Arctic or with underestimation of mercury re-emission from snow and seawater.

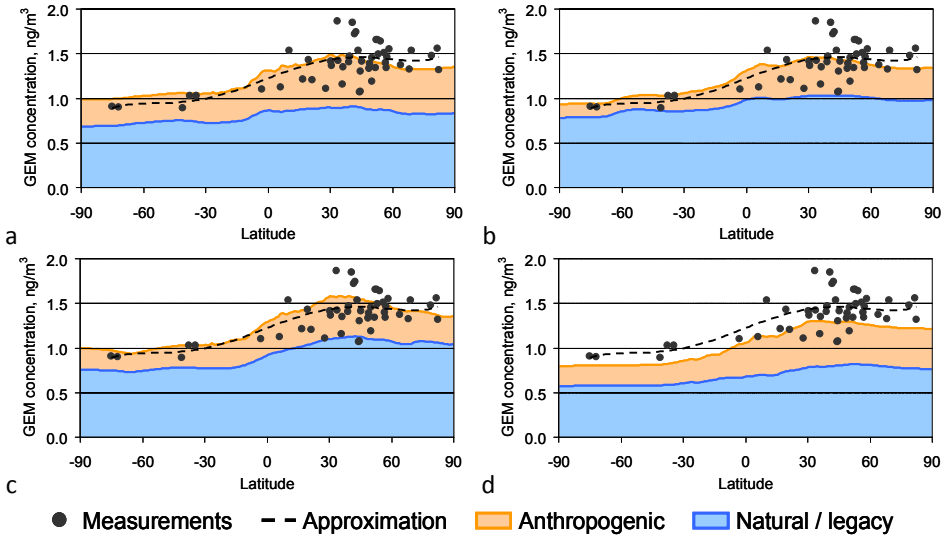

Fig. 3.6. *Global zonal-mean distribution of Hg^0 air concentration in 2013 simulated by four models: (a) – GLEMOS; (b) – GEOS-Chem; (c) – GEM-MACH-Hg; (d) – ECHMERIT. Black dots are observations and dotted line is a polynomial approximation*

Figure 3.6 also shows relative contribution of anthropogenic and natural / legacy emissions to Hg^0 concentration in the near-surface air. Generally, both absolute and relative contributions of anthropogenic emissions are higher in the Northern Hemisphere, where most of emission sources are located. Contribution of natural / legacy sources also increases northward but the gradient is considerably smaller. It should be noted that natural and legacy emissions are a part of mercury air-surface exchange and depend on mercury redox processes in other environmental media (soil, seawater, vegetation, etc.). Therefore, available estimates of natural / legacy sources contain significant uncertainties and largely differ between the models.

Statistical analysis of comparison of simulated and observed Hg^0 concentration in different model experiments are shown in Fig. 3.7 in terms of a symmetric relative bias (Fig. 3.7a) and a spatial correlation coefficient (Fig. 3.7b). In the BASE simulation all the models produce concentration distributions, which well agree with measurements (the bias is around zero and the correlation coefficient is about 0.7). Switching off anthropogenic emissions (NOANT) leads to decrease of Hg^0 levels in the atmosphere (the bias is ~40%) and some decrease of spatial correlation with measurements. It is worth to say that spatial distribution of mercury concentration in this model run is largely determined by natural and legacy sources and, therefore, the decrease of spatial correlation considerably differs between the models. Model results with only anthropogenic emissions (NONAT) are characterized by even larger negative bias (~100%) and again by some decrease of spatial correlation. Simulations with different chemical oxidation mechanisms (BRCHEM1, BRCHEM2, O3CHEM, OHCHEM) do not lead to strong changes of spatial distribution of

Hg^0 concentration in the surface air. All of these runs (except for BRCHEM1) result in some overestimation of Hg^0 levels indicating insufficient oxidation capacity of these mechanisms taken alone. The best spatial correlation is obtained for the oxidation reactions with Br (BRCHEM1) and OH radical (OHCHEM).

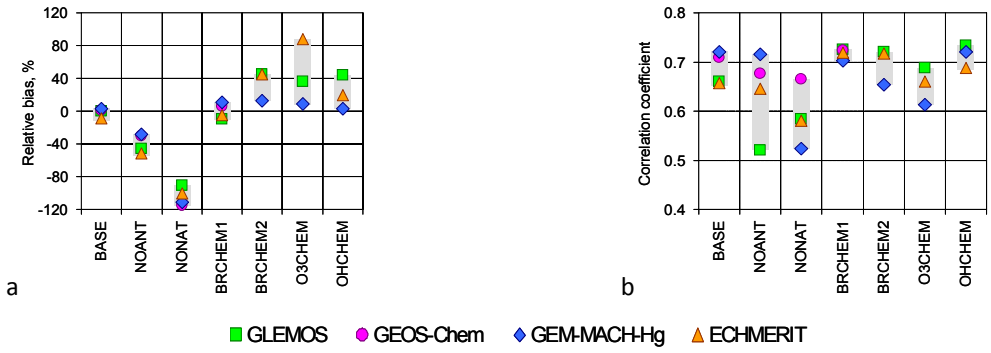

Fig. 3.7. *Relative bias (a) and spatial correlation coefficient (b) of simulated and observed annual mean Hg^0 air concentration for different model experiments*

In contrast to elemental mercury levels, air concentration of reactive mercury Hg(II) largely depends on mercury oxidation chemistry in the atmosphere. Ratio of Hg(II) concentration to Hg^0 concentration in the air can characterize oxidation capability of mercury atmospheric chemistry. Therefore, comparison of simulated and observed Hg(II)/Hg^0 ratios at measurement sites is used for evaluation of different oxidation mechanisms. Figure 3.8 shows evaluation of the ratio of annual mean concentrations of Hg(II) and Hg^0 simulated with application of three chemical mechanisms (reactions with Br, O_3 and OH). Br oxidation chemistry is applied with two different datasets of Br concentrations: generated by GEOS-Chem (BRCHEM1) and by p-TOMCAT (BRCHEM2) models. As seen the best agreement of the simulated and measured annual Hg(II)/Hg^0 ratios is for oxidation by Br with the first concentration dataset (BRCHEM1) and by ozone (O3CHEM). In the former case three of four models provide the simulated values that agree with measurements within a factor of three but the fourth model predicts strong overestimation the Hg(II)/Hg^0 ratios (Figs. 3.8a). It can be partly explained by underestimation of Hg(II) removal from the atmosphere via wet and dry deposition. In the latter case the models predict similar levels of the Hg(II)/Hg^0 ratio with some overestimation of observed values (Figs. 3.8c). It should be also mentioned that BRCHEM1 was the only mechanism that successfully reproduced observed seasonal cycle of Hg(II)/Hg^0 with maximum in spring and minimum in late summer (not shown here).

Mercury wet deposition is another parameter, which is strongly determined by atmospheric oxidation chemistry. Elemental mercury is a poorly soluble substance that is almost not scavenged by precipitation. Therefore, mercury concentration in precipitation and wet deposition flux mostly depend upon two factors – direct emissions of oxidized mercury forms from anthropogenic sources and mercury oxidation in the atmosphere. Besides, the influence of the first factor rapidly subsides with a distance from emission sources. Analysis of wet deposition can be also used for evaluation of chemical mechanisms of mercury oxidation in the atmosphere. Figure 3.9 shows comparison of modeled and measured seasonal variation of monthly mean wet deposition flux in Europe. The

observations demonstrate well pronounced seasonal cycle with the maximum in summer and the minimum during the cold season (winter and early spring). It is worth to note that measured precipitation amount does not reveal similar seasonality to explain intra-annual variation of wet deposition. As seen both model runs with Br oxidation chemistry (BRCHEM1 and BRCHEM2) predict maximum wet deposition during the spring months instead of summer (Figs. 3.9a and b). Simulations with the ozone oxidation mechanism (O3CHEM) do not provide a noticeable seasonality of deposition flux (Fig. 3.9c). The OH-initiated oxidation chemistry (OHCHEM) is the only mechanism that allows reproducing observed seasonal variation of wet deposition in Europe (Fig. 3.9d).

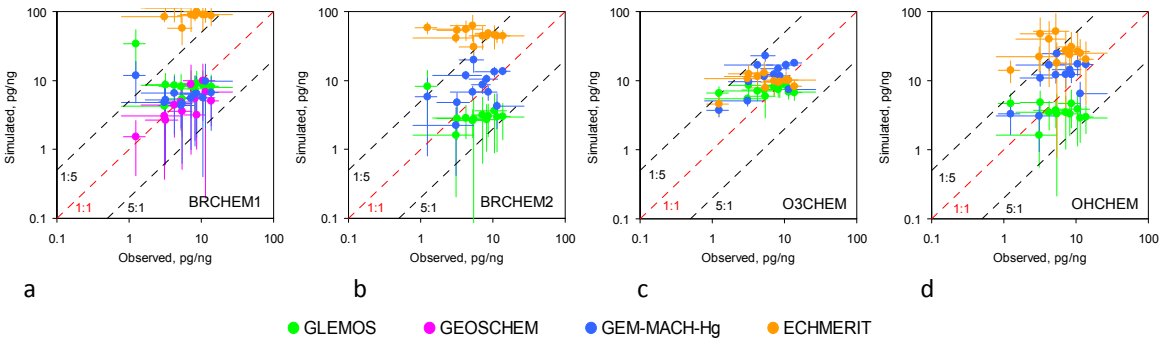

Fig. 3.8. *Scatter plots of simulated vs. observed ratios of annual mean Hg(II) concentration to Hg^0 concentration in 2013 for different model experiments: (a) – BRCHEM1; (b) – BRCHEM2; (c) – O3CHEM; (d) – OHCHEM. Whiskers show standard deviation of monthly mean simulated and observed values. Dotted red line depicts the 1:1 ratio; dashed black lines show deviation by a factor of 5*

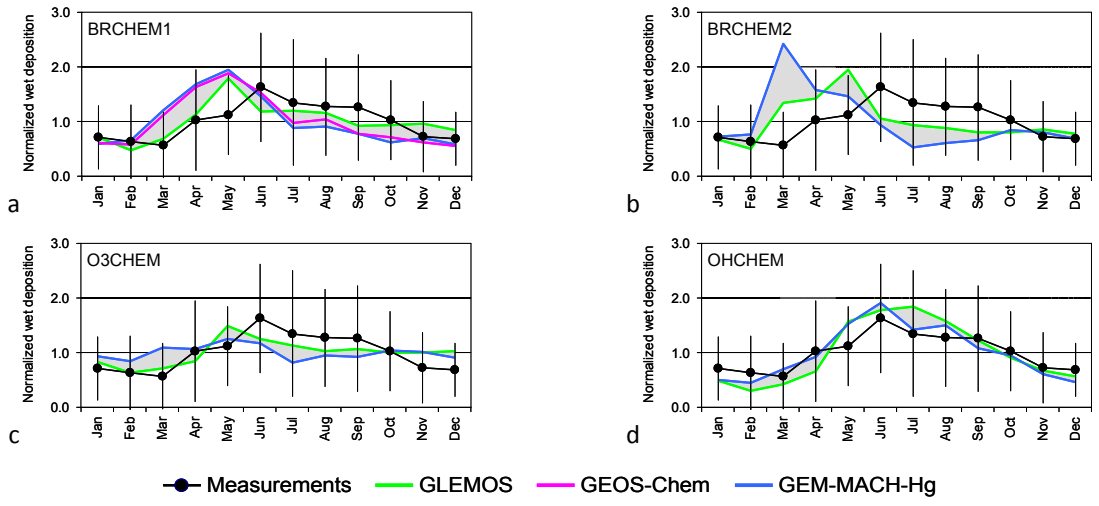

Fig. 3.9. *Normalized seasonal variation of monthly mean wet deposition flux in Europe. Black line with dots shows observations averaged over 14 sites in the region (whiskers are standard deviation). Colored lines present model simulations averaged over the same cites for different model experiments: (a) – BRCHEM1; (b) – BRCHEM2; (c) – O3CHEM; (d) – OHCHEM*

However, it should be noted that available emissions inventories do not include information on temporal variation of mercury anthropogenic emissions. Therefore, more detailed data on seasonal

variation and chemical speciation emissions could improve understanding of mercury processes and, ultimately, quality of the model assessment.

These and other results of the study have been presented at the workshop of the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) (Potsdam, Germany, February 2016). More information about co-operation with TF HTAP is available in Section 4.1.3.

Multi-model research activities performed within the GMOS project support further development of mercury modeling approaches and, ultimately, quality of the model assessment in the EMEP domain.

3.3. Update of re-suspension scheme of heavy metals

There are three main groups of sources responsible for heavy metal pollution levels in the EMEP region. First of all, these are primary (direct anthropogenic) emission sources such as industry, transport, energy production etc. Another type of sources is so-called secondary emissions. They include re-suspension of wind-blown dust containing previously deposited metals, and natural emission and re-emission of gaseous form of mercury. Finally, some contribution is made by sources (both anthropogenic and secondary) located outside the EMEP region.

Original version of parameterization of dust suspension as a secondary source of heavy metals included in modeling is described in [*Gusev et al,* 2006]. It is assumed that re-suspension takes place from agricultural lands in autumn and spring, urban territories and bare lands (mainly, deserts of the northern part of Africa, Central Asia and Middle East). Modeling of re-suspension of heavy metals is subject to large uncertainties because this phenomenon depends on numerous environmental factors such as meteorological conditions, soil texture, heavy metal concentration in soils etc.

Concentrations of heavy metals in soil are assumed to be enriched by heavy metals for the decades of atmospheric contamination. Amount of this enrichment is described by a coefficient called enrichment factor. Improvement of wind re-suspension scheme this year has been mainly focused on correction of the magnitude and spatial distribution of these factors in order to diminish deviation of modeled heavy metal pollution levels from the observed values. Similar activity has been already performed in the framework of country-specific assessment of lead pollution level in the Netherlands [*Ilyin et al*, 2014]. This year improvement of enrichment factors has been performed over the entire EMEP domain. Technical details of this work are available in [*Shatalov et al.*, 2016].

Model simulations for 2013, carried out with the usage of only anthropogenic emissions resulted to significant (by 60-70% on average) underestimation of the observed levels both for annual mean concentrations in air and wet deposition fluxes (Fig. 3.10). When re-suspension of heavy metals is included, the agreement between modeled and observed levels significantly improves. However, at some stations and in some periods of time it led to overestimation of the observed levels. For example, annual mean air concentrations of lead at the Dutch and Belgium stations were overestimated by a factor of 2-3 (Fig. 3.10a).

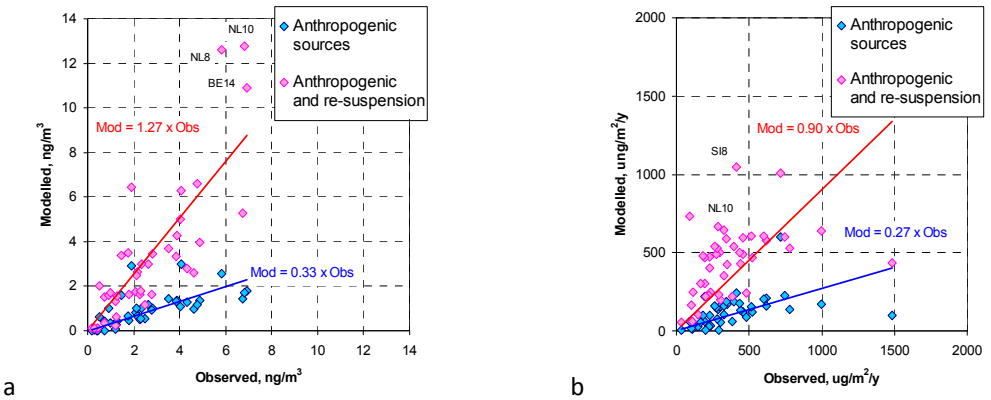

Fig. 3.10. *Modeled and observed annual mean lead concentrations in air (a) and annual wet deposition fluxes (b) in the EMEP region in 2013. Stations where significant overestimation is noted are indicated on the diagrams*

The major contribution to modeled concentrations and wet deposition fluxes at these stations is made by secondary sources, namely, re-suspension from urban type of the underlying surface. For example, at the station NL10 its contribution amounts to 84% for annual mean air concentrations and 62% for wet deposition (Fig. 3.11). Enrichment factor used for estimation of re-suspension flux contains significant uncertainty. Therefore, it is assumed that adjustment of this factor could facilitate diminishing deviation of modeled values from the observed ones. Update of the model parameterizations was done based on the adjustment of spatial distribution of the enrichment factor [*Shatalov et al.,* 2016].

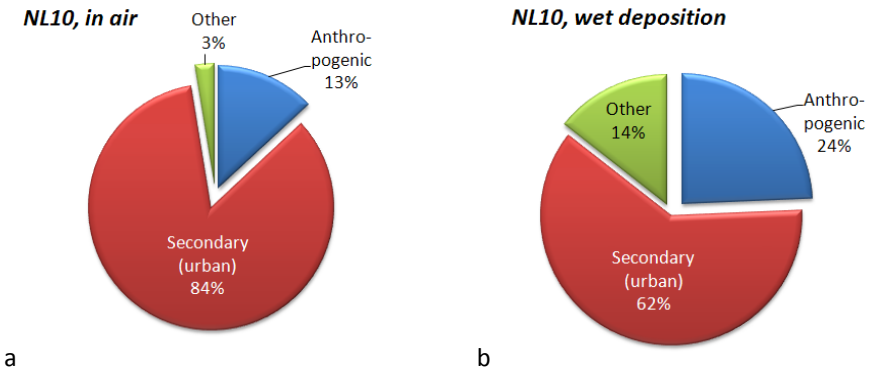

Fig. 3.11. *Contribution of anthropogenic, secondary (from urban surfaces) and remaining sources to annual mean concentrations of lead in air (a) and wet deposition fluxes (b) at station NL10 (Vredepeel, the Netherlands)*

Adjustment of enrichment factor (its magnitude and spatial distribution) led to decrease of discrepancies between modeled and observed concentrations in air and wet deposition fluxes of lead in 2013. Table 3.2 summarizes statistical indicators characterizing agreement between modeled and observed levels of air concentrations and wet deposition before and after the adjustment. Mean relative bias declined from 22 to –6% for concentrations in air and from 25% to -3% for wet deposition flux. Spatial correlation coefficients increased and Normalized Root Mean Square Error

(NRMSE) decreased which also meant the improvement of modeling results for the EMEP region as a whole.

Table 3.2. *Main statistical indicators of agreement between annual modeled and measured levels of air concentrations and wet deposition fluxes of lead in 2013*

	Original		Adjusted	
	C_{air}	Wet Dep	C_{air}	Wet Dep
Relative bias, %	22.2	24.6	-5.8	-3.1
Correlation coefficient	0.82	0.45	0.87	0.48
NRMSE	0.74	0.58	0.37	0.63
F2, %	73	79	75	87
F3, %	88	100	90	96

C_{air} – concentration in air

Wet Dep – wet deposition flux

NRMSE – Normalized Root Mean Square Error

F2 – fraction of values fitting to factor of 2 difference

F3 – fraction of values fitting to factor of 3 difference

Comparison of modeled (original and adjusted) and observed concentrations in air and wet deposition fluxes for particular stations is presented in Fig.3.12a,b. The positive effect for concentrations in air is noted for stations in the Netherlands and Belgium (Fig. 3.12a) as well as for French, British, Czech, and Hungarian stations. Improvement of agreement between modeled and observed wet deposition fluxes is also noted for stations in France, the United Kingdom, the Netherlands, Sweden and Slovenia (Fig. 3.12b).

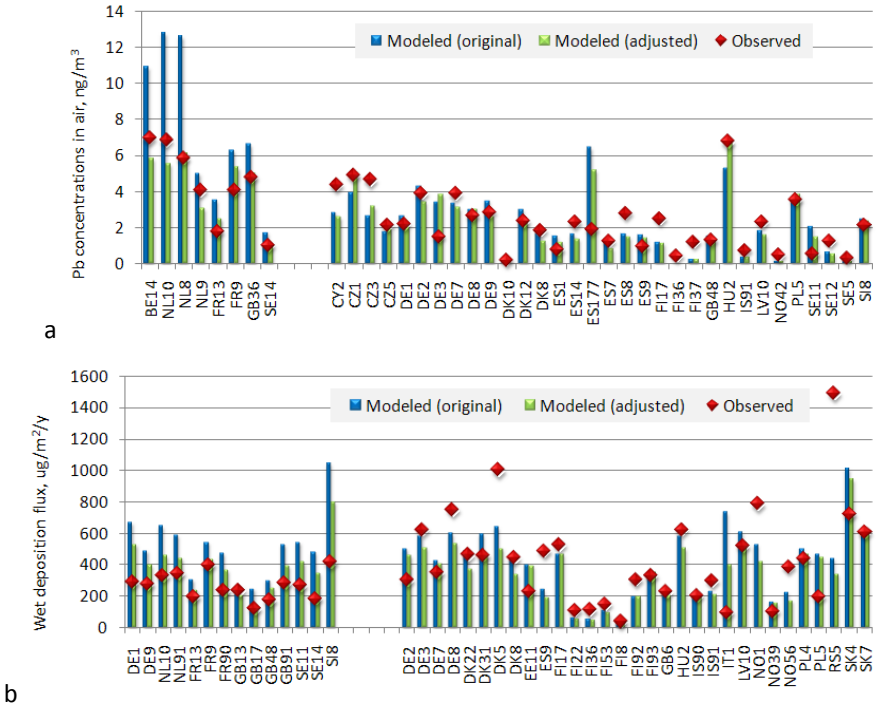

Fig. 3.12. *Modeled (original and adjusted) and observed concentrations in air (a) and wet deposition (b) of lead in 2013. Stations where modeled values decline due to adjustment of enrichment factor are grouped in the left part of the diagrams, and the rest- in the right part*

The adjustment of the enrichment factor favoured reducing deviation of modeled lead levels from the observations in the Netherlands, Belgium, France and the United Kingdom. However, there is a need of further investigation of issues related to wind re-suspension, and improvement of the model assessment in the EMEP region, including the following steps:

- Testing the developed algorithm for other years
- Detailed analysis of meteorological conditions favourable for initiation of dust suspension. The parameters mostly responsible for it are soil wetness and friction velocity.
- Collection of information on concentrations of heavy metals in soils of Asian and African desert regions as well as data on concentrations of heavy metals in street dust of cities in the EMEP region
- Investigation of possibility to extend wind suspension scheme through including other relevant processes such as traffic-induced dust suspension.

4. COOPERATION WITH PARTIES TO THE CONVENTION

4.1. Belarus

Long-term activity on country-specific case studies of heavy metal pollution assessment has been continued this year with the focus on Belarus. There were two main purposes of the study. First of all, it was to prepare the detailed information about pollution levels with fine (10x10 km^2) spatial resolution for the country. Another purpose was to investigate changes in model performance when transferring from coarse to fine spatial resolution. In this study lead pollution levels in 2012 were simulated and analyzed. This section provides the main results of the research, while full description of this work is available in the joint technical report of MSC-E and Institute of Natural Management of National Academy of Science of Belarus (INM) [*Ilyin et al.*, 2016].

Emission data involved in this study are based on two datasets. For the territory of Belarus lead emissions were provided by national experts. These data were presented as gridded values from sources of different source categories. Besides, emission values from particular large point sources (LPS) were extracted from these data. Total emission of lead from the country, according to the submitted national data for this study, amounted to 8.3 tonnes per year. For other countries the data officially provided by CEIP were used.

The only available background monitoring station in Belarus was '*Berezinskiy Reserve*'. Other measurement data were also involved, namely, EMEP Polish station Diabla Gora (PL5) and five urban background stations in Poland (PL0531A, PL0241A, PL0148A, PL0538A and PL0507A), extracted from the *AirBase* data base [*AirBase*, 2016]. In addition to this, urban measurements from 19 Belarusian cities were submitted by the experts from the country.

Information on lead pollution simulated for Belarus, include spatial distribution of pollution levels with fine resolution (10x10 km^2), source-receptor relationships, contribution of different emission sources categories to lead levels in Belarus, pollution levels caused by particular LPSs and modeled lead levels in main cities of the country. Figure 4.1 exemplifies spatial distribution of total deposition in Belarus and neighbouring countries with resolution 10x10 km^2.

Anthropogenic deposition was caused by combined influence of national sources and emission sources located outside the country's territory. The contribution of foreign sources to total anthropogenic deposition in Belarus was 94% (51 t/y), and from national sources – 6% (3.2 t/y). The main contributor to anthropogenic deposition to Belarus were Poland (52%), followed by Ukraine (11%) and Germany (3%) (Fig. 4.2a).

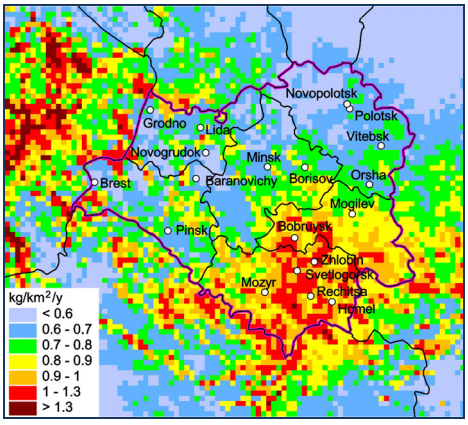

Fig. 4.1. *Annual deposition of lead with resolution 10x10 km^2 in 2012 in modeling domain*

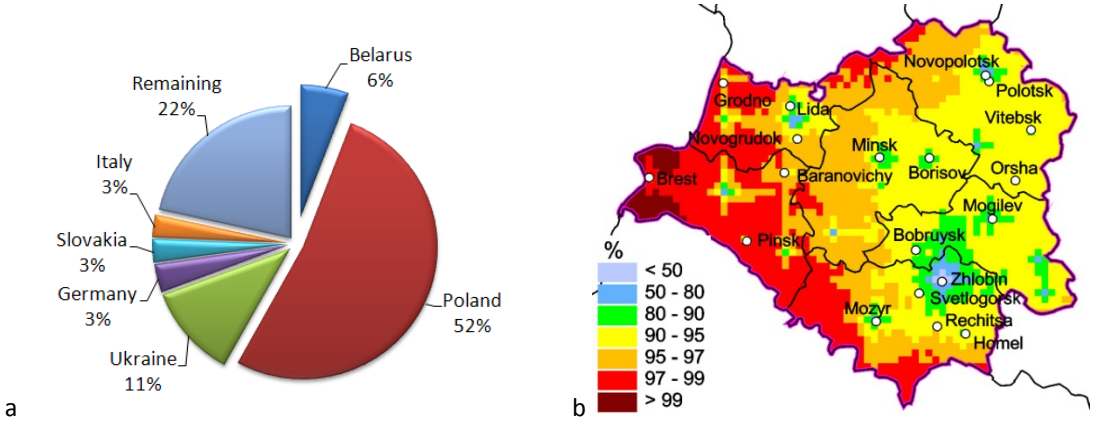

Fig. 4.2. *Contribution of national and foreign sources to anthropogenic deposition in Belarus (a) and spatial distribution of contribution of foreign sources to anthropogenic deposition in Belarus (b) in 2012*

Contribution of foreign emission sources to lead deposition over most part of the country's territory ranged from 90% in the east to almost 100% in the west of Belarus (Fig. 4.2b). Around main cities, where major national emission sources are located, the contribution was 80-90%. In Zhlobin city, where national emissions were the highest, the contribution of foreign sources was the lowest in the country and lay below 50%.

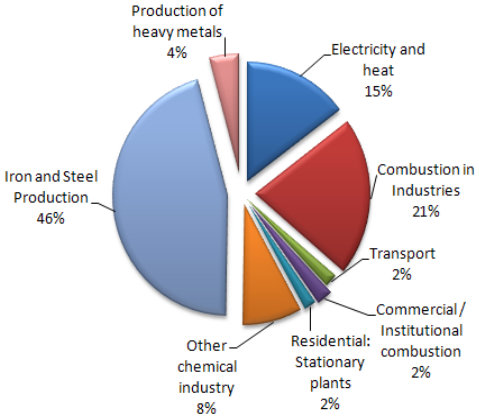

Fig. 4.3. *Contribution of groups of emission categories to deposition in Belarus form national sources*

Model calculations revealed that category '*Iron and Steel Production*' was the main national contributor to lead deposition in Belarus and provided 46% to deposition from national sources (Fig. 4.3). The other significant contributors were group '*Combustion in industries*' (21%), '*Electricity and Heat*' (15%) and '*Other Chemical Industry*' (8%). Contributions of different source categories differed among administrative regions of Belarus.

Modeling with fine spatial resolution was applied to estimate contribution of each of LPS to lead pollution in the country. For example, metallurgical plant in Zhlobin of the Homel region was the most powerful LPS of lead in Belarus emitting 2.7 tonnes of lead per year. In the vicinity of this plant deposition fluxes of lead exceeded 0.1 kg/km^2/y, or more than 10% of deposition from anthropogenic deposition (Fig. 4.4a). In neighbouring regions its contribution to anthropogenic deposition ranged from 0.5 to 10%, and in some regions of neighbouring countries (Russia) - 0.5 -3 %.

Contributions of various emission sources to lead pollution in capital cities (Minsk, Vitebsk, Mogilev, Brest, Grodno, Homel) of Belarusian administrative regions were calculated. Anthropogenic emissions in grid cells belonging to the cities were tagged and thus cities were considered as separate sources in source-receptor calculations. Therefore, anthropogenic sources were distinguished between 'city sources' and the 'external' (relative to a city) sources.

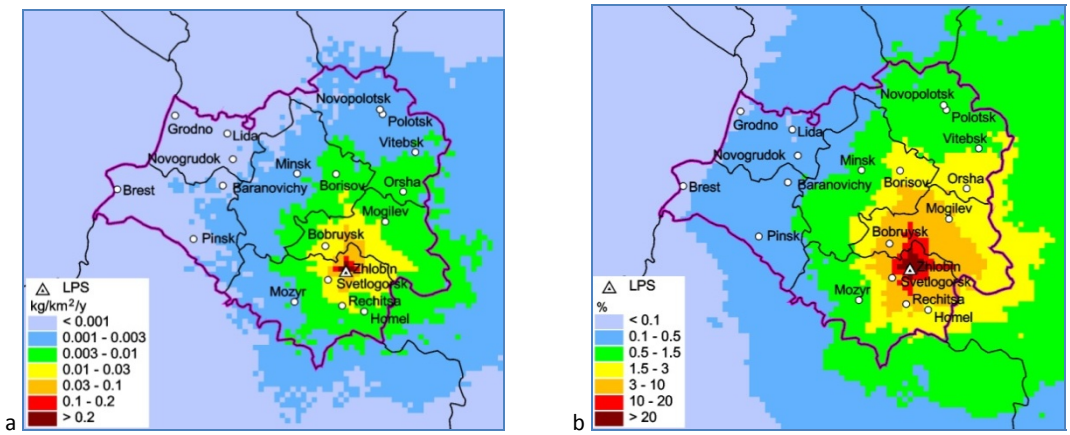

Fig. 4.4. *Deposition of lead (a) and its contribution to deposition from anthropogenic sources (b) from large point sources (LPS) "Belarusian metallurgical plant" in Zhlobin city in 2012*

External sources were split in two components: national (emitting lead from the territory of Belarus) and foreign (located outside Belarus). Among anthropogenic sources the foreign sources made the major (70-96%) contribution to deposition in the Belarusian cities (Fig. 4.5). City sources were responsible for 4-25% of deposition, and national external sources contributed from 0.5% to 5% of pollution.

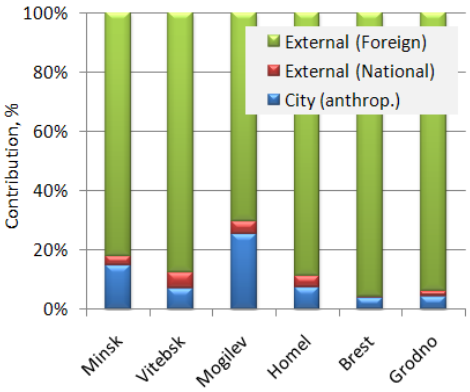

Fig. 4.5. *Relative contribution of city, foreign and national external sources to anthropogenic deposition in Belarusian cities*

Modeled results were compared with the available monitoring data. For the background Belarusian station 'Berezinskiy Reserve' the model underestimated the observed levels by almost 3 times. At the same time it was demonstrated that calculated annual mean concentrations in air for the Polish stations (both from EMEP and AirBase) agreed with observed levels within a factor of two. Possible reasons contributing to this discrepancy included uncertainties of the model, underestimation of the emissions data and uncertainties of measurement data. For the analysis of heavy metal emission data in the EECCA countries joint efforts of national emission experts of these countries together with CEIP and TFEIP are required. Besides, quality of monitoring data needs special attention. Finally, additional direction of the research could be focused on investigation of other pollutants in Belarus. It could help to understand if the considered situation is unique for lead or it is typical for other pollutants, for example, particulate matter or acidifying compounds.

In order to improve quality of assessment of heavy metal pollution levels in EECCA countries joint efforts of the EMEP Centres and national experts are needed in field of modeling, emission inventories and monitoring data.

4.2. Poland

This year detailed study of cadmium atmospheric pollution with fine spatial resolution has been started for Poland. At present the process of preparation of necessary input data is ongoing. In particular, information on cadmium emission data from point sources as well as gridded emissions with spatial resolution 0.1°x 0.1° in 2014 was submitted by national experts (Fig. 4.6). Emissions from four groups of point sources are considered: thermal power stations, production of coke, copper smelting and cement production. Gridded total emissions in Poland are split in source categories according to the GNFR classification (Nomenclature For Reporting of Gridded data and large point sources).

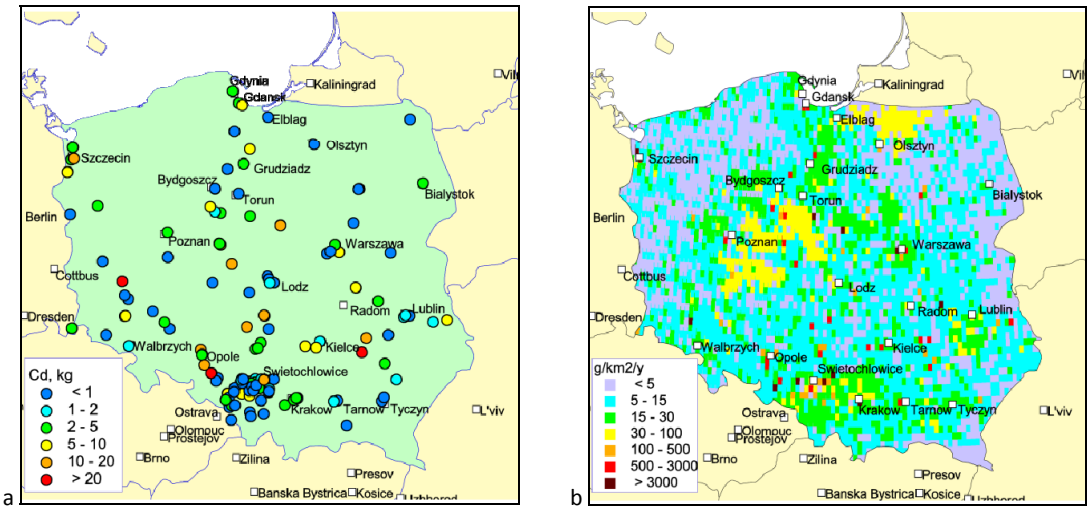

Fig. 4.6. *Emission of cadmium from point sources (a) and gridded emissions of cadmium with spatial resolution 0.1°x0.1° in 2014 (b) in Poland*

Further information needed for assessment of cadmium pollution levels in Poland comprises monitoring data, gridded meteorological fields, concentrations of cadmium in soils for calculation of wind re-suspension etc. The expected output information for the country includes spatial distribution of cadmium concentrations and deposition, source-receptor relationships for the country and its provinces (voevodships), contribution of particular source categories, point sources and cities to pollution in the country. The results of this work will be compared with measurement data and jointly analyzed by MSC-E and national experts from Poland.

4.3. The United Kingdom

Centre of Ecology and Hydrology (CEH) invited MSC-E to take part in country-specific study on assessment of heavy metal pollution over the United Kingdom. There were three purposes of the study. The first purpose is to provide the country with information on heavy metal pollution levels with high (10x10 km^2) spatial resolution. The second purpose was to carry out comparison of the results of MSC-E [*Travnikov and Ilyin*, 2005] and the national FRAME [*Dore et al.,* 2014] models. And finally, it is planned to pay special attention to investigation of role of wind-induced heavy metal re-suspension in pollution of the United Kingdom.

Currently experts from CEH provided MSC-E with information on emissions of lead in 2012, observations of heavy metal levels from national monitoring network and data on concentrations in soils. The national data contained splitting of emissions to different source categories and emission values from particular LPS. Spatial resolution of original emission data was 1x1 km^2. Further the emissions were re-gridded to 10x10 km^2 (Fig. 4.7a).

Measurement data on lead levels are available from 35 stations. Among them 27 stations measure concentrations in air, 8 stations – concentrations in precipitation, and 5 stations are co-located (Fig. 4.7b). Among stations, measuring concentrations in air, 12 are rural background, one – sub-urban background and the rest are urban stations. There are measurement data from 5 EMEP stations in 2012.

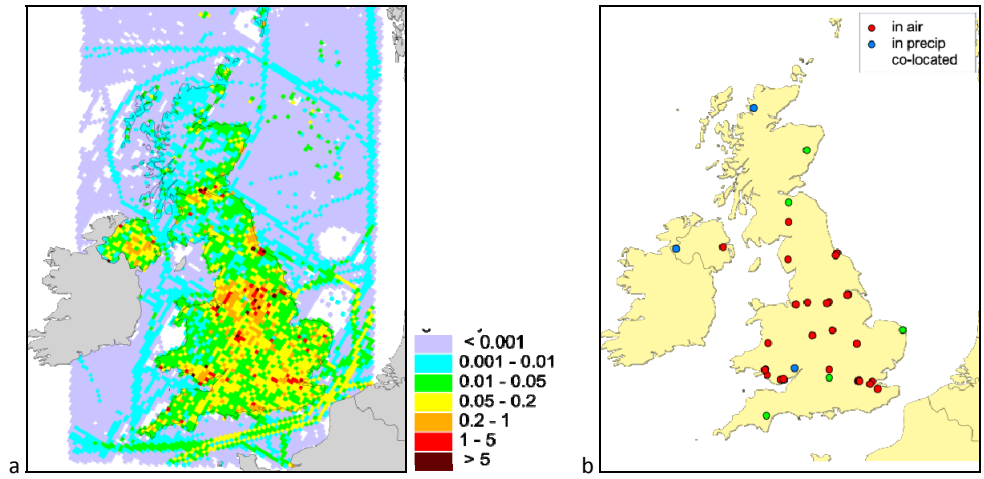

Fig. 4.7. *Total emission of lead in the United Kingdom (a) and location of monitoring stations (b)*

Preliminary results of modeling with fine (10x10 km^2) resolution based on MSC-E model were produced. Spatial distributions of annual mean air concentrations and total deposition are demonstrated in Fig. 4.8. As seen, the highest levels of lead in the United Kingdom are noted for the central and south-eastern parts of the country. The lowest levels occur in the northern part.

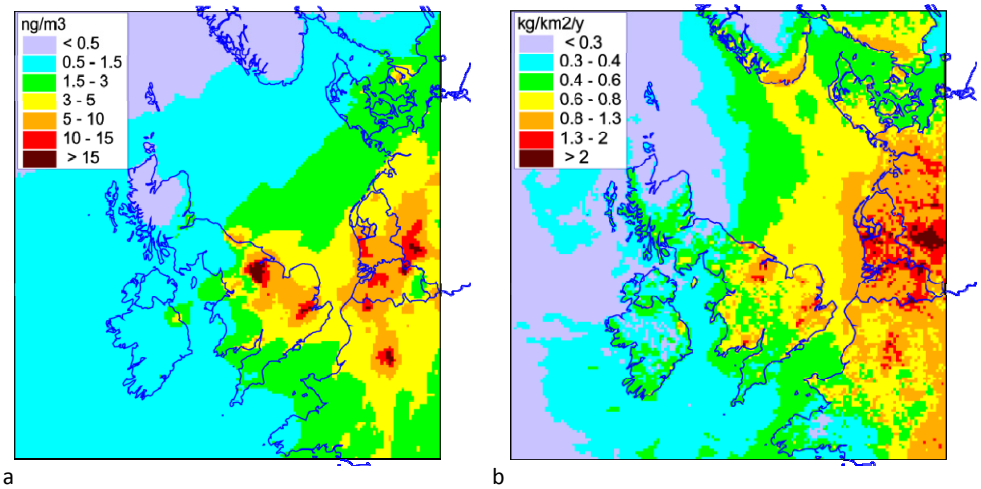

Fig. 4.8. *Annual mean concentrations (a) and total deposition (b) of lead in 2012*

Heavy metal pollution levels were also simulated by the FRAME model. Pilot modeling results of MSC-E and FRAME models, in particular, air concentrations at the rural background stations, were compared (Fig. 4.9). As seen from the Figure, both models adequately reproduce air concentrations of lead. The existing discrepancies between modeled and observed levels will be analysed further.

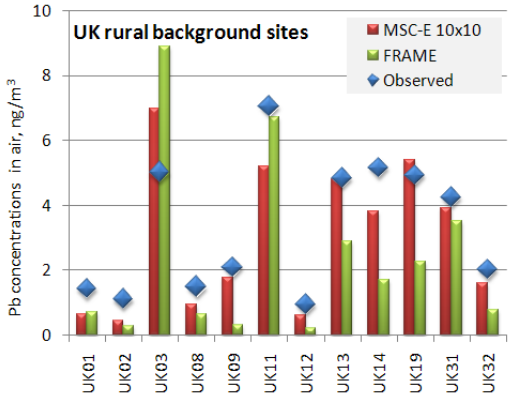

Fig. 4.9. *Modeled (MSC-E, FRAME) and observed annual mean air concentrations of lead at British rural background stations in 2012*

It is planned to further analyze the results of modeled heavy metal levels in the United Kingdom. In particular, special attention has to be paid to study of discrepancies between modeled and observed values. In addition to this, it is planned to investigate the role of wind re-suspension of heavy metals in pollution of the United Kingdom.

4.4. Germany

Modeling of Regionalized Emissions (MoRE)

This year MSC-E has cooperated with colleagues from Karlsruhe Institute of Technology (KIT) in Germany. Experts from KIT develop a model of emissions of heavy metals and nutrients to surface water bodies. MSC-E was requested to provide them with gridded lead, cadmium and mercury deposition fluxes in the EMEP region in 2012. This section, describing current status of the model development, is prepared by colleagues from KIT.

As the Directive 2008/105/EC [*EC*, 2008] requires an emission inventory for the PS, the model system MoRE (Modeling of Regionalized Emission) was developed on behalf of the Federal Environmental Agency of Germany. MoRE is an instrument for modeling emissions into surfaces water bodies and for identifying relevant emission pathways. MoRE is based on the model concept MONERIS [*Behrendt et al.*, 2000] and has been adapted for the modeling of pollutant emissions [*Fuchs et al.*, 2012].

The emission modeling is carried out with the Regionalized Pathway Analysis approach (RPA). The RPA comprises different pathways of substance emissions into water and distinguishes between point sources as pathways and pathways that are strongly influenced by diffuse sources.

Fig. shows relevant sources and pathways and the scope of RPA (Pathway Oriented). A detailed explanation can be found in EC (2012) In the MoRE system, the following pathways are implemented: “municipal wastewater treatment plants” (MWWTP), “industrial direct dischargers”, and “historic mining”, “sewer systems”, “surface runoff”, “erosion”, “groundwater”, “drainage”, “direct atmospheric deposition onto water surface”, and “inland navigation”.

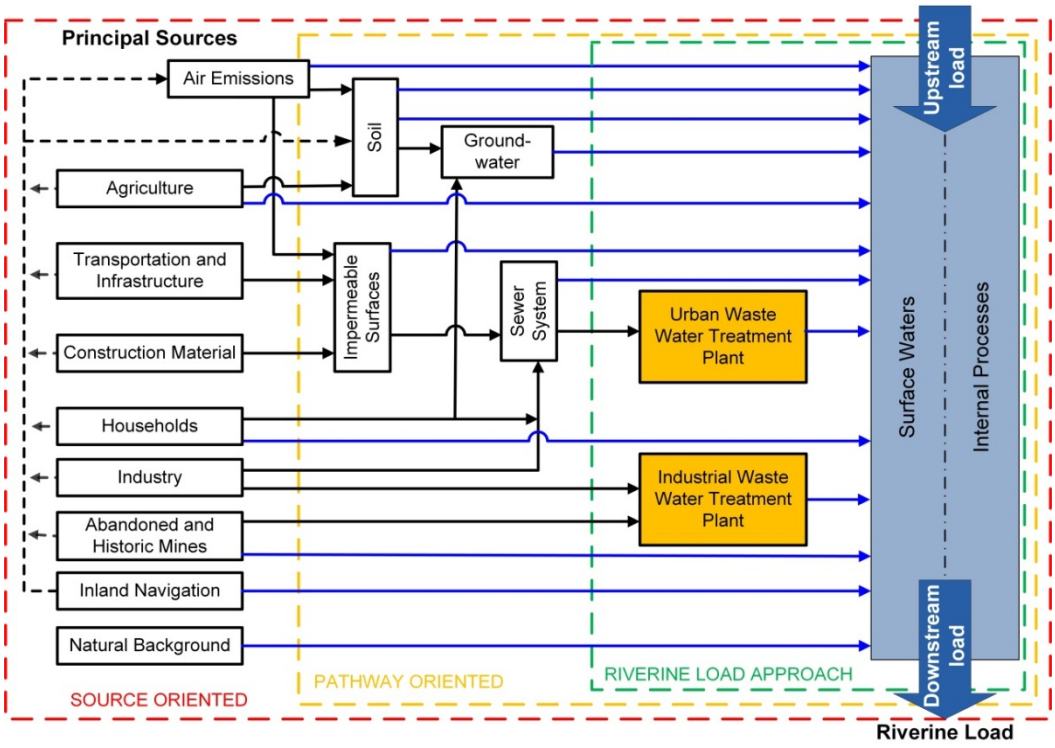

Fig.4.10. *General scheme and recommended approaches for pollutant inventories according to EC 2012*

At the current level of development MoRE can calculated the annual average emissions for nitrogen and phosphorus, the heavy metals cadmium, chromium, copper, mercury, nickel, lead, and zinc, polycyclic aromatic hydrocarbons ΣEPA[1] PAH_{16}, diethylhexylphthalate, nonylphenol, terbutryn, triclosan, diuron, isoproturon, as well as the pharmaceuticals diclofenac, ibuprofen, iomeprol, and sulfamethoxazol.

[1] 16 EPA-PAH are considered, i.e. the 16 individual substances selected from several hundred individual PAH compounds by the United States Environmental Protection Agency (US EPA) because of their particular relevance

The MoRE model has been implemented in an open source PostgreSQL/SQLite database comprising all input data, modeling approaches, results and a generic calculation engine. A user interface dynamically linked to the database allows both to edit input data and modeling approaches and carry out the modeling. All calculations are done on a single year basis. For more details on the software see https://isww.iwg.kit.edu/english/MoRE.php.

The methodologically complex RPA approach requires a large number of general and substance-specific input data, but allows a spatially differentiated representation of substance emissions into surface waters caused by the respected pathways. In particular, annual atmospheric gridded deposition of lead, cadmium and mercury, calculated by EMEP/MSC-E [*Ilyin et al.*, 2014], are involved in the modeling.

The quantification of emissions was carried out for analytical units with an average size of 130 km² representing hydrological subunit of the German water sheds [*Fuchs et al.*, 2010].

The basic modeling approach of MoRE is the mass balance approach, were any process which leads to a change in the mass flows during transport, is represented by empirical equation. Regional varying emissions are the result of different input data on land use, hydrology, geology, and statistical data like population density, distribution of combined and separate sewer system, design capacity of combined sewage treatment, etc. The RPA methodology uses also substances specific regionalized data (atmospheric deposition rate, substance concentrations and contents) for modeling emissions into water bodies.

Based on the general and substance-specific input data and approaches, the total and pathway specific emissions of nutrients, heavy metals and ΣEPA PAH_{16} were modeled using the regional pathway analysis for the period of 2006–2011. The results of the emission calculation were generated for Germany on the level of analytical units.

5. COOPERATION AND DISSEMINATION OF INFORMATION

5.1. Subsidiary bodies of the Convention

5.1.1. Task Force on Measurements and Modeling (TFMM)

In the framework of cooperation with TFMM MSC-E participated in the 17th meeting of the Task Force held in May 2016 in Utrecht (the Netherlands). Participants of the meeting were informed about the progress in the MSC-E activities related to the assessment of heavy metal pollution levels in the EMEP region.

In particular, information on current status of country-specific case studies, cooperation with WGE, evaluation of long-term trends of heavy metal levels, transition to the new EMEP grid and formulation of priorities to improve lead, cadmium and mercury emission inventories was presented. Besides, special attention was paid to mercury-related activities, such as multi-model assessment of global mercury pollution, study of mercury atmospheric chemistry (Section 2.2), contribution to the UNEP Global Mercury Assessment (Section 5.2.1).

Case study of lead pollution assessment with fine spatial resolution in Belarus was described in details. TFMM was informed about input data provided by national experts for this study, modeled output information with spatial resolution 10x10 km^2 produced for the country, and results of verification of modeling results. Major activity was focused on analysis of the uncertainties of the model, observations and emission data. On the base of the analysis main conclusions were drawn and recommendations for the further research in Belarus and the EECCA countries were formulated. Finally, progress and further plans regarding the case studies for the United Kingdom and Poland were demonstrated.

5.1.2. Working Group on Effects (WGE)

MSC-E took part in the annual Task Force meeting of ICP-Vegetation of WGE held in March 2016 in Dubna, Russia. The Task Force was informed about long-term trends of lead, cadmium and mercury in the EMEP region and about comparison of modeled values with observed concentrations in mosses.

Monitoring of concentrations in mosses is valuable source of supplementary information to characterize pollution levels. Compared to station-based monitoring, they are relatively cheap and are carried out over vast territories. Since mosses used in monitoring do not have roots they accept both nutrients and pollutants from the atmosphere. However, besides atmospheric deposition, concentration of pollutants in mosses depends on a number of other environmental factors, such as proximity to sea coast, surrounding land-cover, altitude etc. That is why monitoring data in mosses are more difficult to interpret compared to station-based measurements.

Regular moss surveys are carried out in the EMEP region once in five years. For the analysis of long-term trends the surveys held in 1990, 1995, 2000, 2005 and 2010 were used [*Harmens et al*., 2010, *Harmens et al*., 2013, *Steinnes at al*., 2011]. The data, collected and stored under the ICP-vegetation supervision, were provided to MSC-E for joint analysis of the results of atmospheric modeling and monitoring of heavy metals in mosses.

Information on measurements of lead and cadmium from all five moss survey was available from 10 countries (Finland, Sweden, Norway, Austria, the Czech Republic, Slovakia, Switzerland, Estonia, Iceland, Poland). For mercury the long-term trends were analyzed for the surveys for 1995, 2000, 2005 and 2010 because of lack of measurement data. Concentrations in mosses and total deposition cannot be compared directly. Therefore, for comparison of long-term trends these values were normalized. Reductions of lead concentrations in mosses and deposition for 1990-2010 were 82 and 79%, respectively (Fig. 5.1). The corresponding values for cadmium were 53% (concentrations in mosses) and 56% (deposition). Reduction of mercury from 1995 to 2010 was much lower: 16% for concentrations and 14% for deposition. Both model results and observations in mosses indicate some increase between 2005 and 2010.

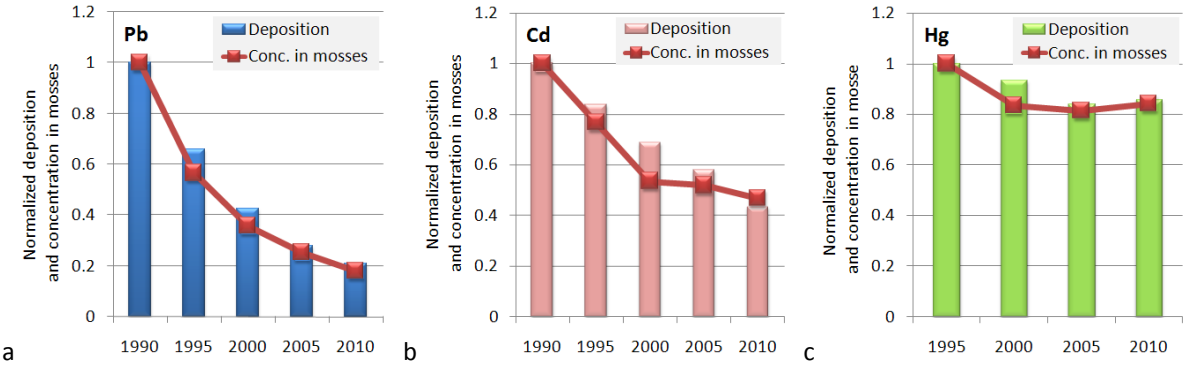

Fig.5.1. *Normalized modeled deposition and observed concentrations in mosses of lead (a), cadmium (b) and mercury (c) in the EMEP region in 1990-2010*

In particular countries magnitudes of reductions may differ from the mean values. For most countries reduction of lead and cadmium levels of modeled deposition and observed concentrations in mosses was similar (Table 5.1). However, there are some exceptions, for example, reduction of cadmium levels in France or Bulgaria. These situations require special investigation in cooperation with experts from the ICP-Vegetation.

Table 5.1. *Reduction (%) of calculated total deposition of heavy metals and their concentrations in mosses in the EMEP countries*

	Lead		Cadmium		Mercury	
	TD*	MC**	TD	MC	TD	MC
1990-2010						
Austria	77.6	83.8	39.2	57.6	-	-
Czech Republic	77.9	84.1	48.8	53.8	-	-
Estonia	78.4	82.6	62.6	54.5	-	-
Finland	82.2	80.0	66.2	55.0	-	-
Iceland	45.1	50.3	27.5	89.1	-	-
Norway	80.5	83.4	52.7	37.6	-	-
Slovakia	67.0	79.2	60.2	42.3	-	-
Sweden	81.3	83.3	56.8	44.6	-	-
Switzerland	83.1	86.5	49.2	59.3	-	-
1995-2010						
Bulgaria	55.1	38.8	51.7	0.6	-	-
France	74.2	62.4	55.3	17.6	-	-
1990-2005						
Germany	70.8	71.2	42.5	30.5	-	-
Latvia	73.4	62.1	44.1	2.1	-	-
Lithuania	73.1	55.5	48.1	63.2	-	-
United_Kingdom	74.3	60.1	46.8	77.3	-	-
Austria	-	-	-	-	10.4	38.0
Czech_Republic	-	-	-	-	29.8	40.3
Finland	-	-	-	-	5.7	16.0
Norway	-	-	-	-	11.5	4.2

* TD – total calculated deposition; ** MC – measured concentrations in mosses; '-' – no data

Special attention has been paid to investigation of spatial distribution of long-term pollution reductions in the EMEP domain. For example, in the Scandinavian region (Norway, Sweden, and Finland) country-averaged trends of modeled deposition and observed concentrations in mosses are very similar (Fig. 5.2). However, spatial distributions of long-term changes of these two parameters, expressed in terms of reduction between 1990 and 2010, do not fully coincide. Modeled and observed reductions demonstrate general south-to-north gradient (Fig. 5.3). However, the reduction of the observed concentrations in mosses is more mosaic compared to that of modeled deposition. Most likely, the differences in spatial distributions are caused by the fact that measurement in mosses may be influenced by local conditions, which are not fully resolved by the regional-scale model.

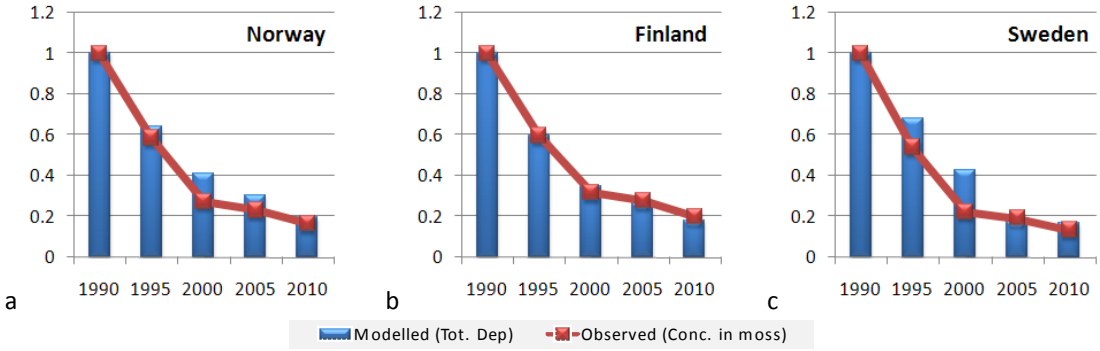

Fig. 5.2. *Normalized modeled deposition and observed concentrations in mosses of lead in Norway (a), Finland (b) and Sweden (c) in 1990-2010*

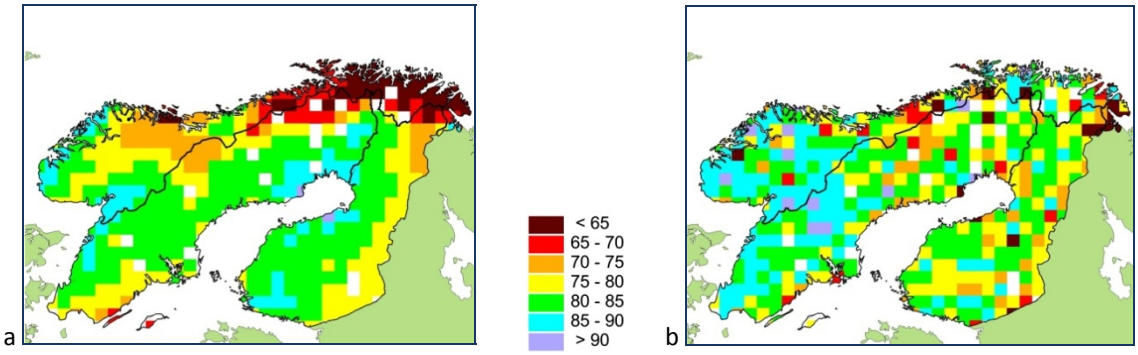

Fig. 5.3. *Spatial distribution of reductions of calculated total deposition (a) and concentrations in mosses (b) between 1990 and 2010 in Scandinavian region*

Measurements of heavy metals in mosses are characterized by wide spatial coverage and dense spatial resolution and could complement station-based EMEP monitoring data. Data on spatial distribution of lead, cadmium and mercury concentrations in mosses as well as on their long-term trends are used in verification of modeling results and assessment of heavy metal pollution in the EMEP region.

5.1.3. Task Force on Hemispheric Transport of Air Pollution (TF HTAP)

MSC-E continued co-operation with the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). In particular, it took part in the Task Force workshop held in Potsdam, Germany in February 2016. The Centre presented an overview of ongoing activities within EMEP and relevant research - projects focused on assessment of mercury pollution. It reported the final results of the multi-model research performed within the EU GMOS (Global Mercury Observation System) project and aimed at study of mercury atmospheric processes as well as assessment of mercury intercontinental transport and future scenarios.

It was demonstrated that oxidation chemistry of mercury in the atmosphere contained significant uncertainties. There is possibility of multiple oxidation mechanisms governing transformation and removal of mercury from the atmosphere. Contribution of current anthropogenic emissions to mercury deposition varies between 20% and 50% in different geographical regions. Besides,

contribution of intercontinental transport is comparable or prevails over domestic sources in most regions (except for South and East Asia) (Fig. 5.4). Speciation of mercury anthropogenic emissions strongly affects the range of mercury dispersion and deposition. On the other hand, mercury speciation in available emission inventories contains significant uncertainties. Model evaluation of future scenarios predicts decrease of mercury deposition in the next 20 years except for South and East Asia where the 'Current Policy' can lead to deposition growth.

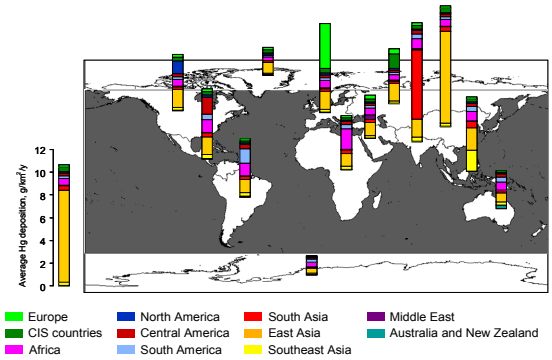

Fig. 5.4. *Source attribution of mercury deposition to different regions on a global scale in 2013*

Particular attention was paid to evaluation of mercury deposition to various aquatic regions, where mercury coming to freshwater and marine ecosystems from atmospheric deposition and other sources is biomagnifies in aquatic food webs. The presented results can contribute to the ongoing TF HTAP research activities as a part of the HTAP2 Modeling Experiments (Theme 2) and Model-Observation Evaluation and Process Diagnosis (Theme 3) [www.htap.org].

Importance of close cooperation with relevant on-going activities within international bodies including AMAP, UNEP and the Minamata Convention on Mercury was stressed. TF HTAP could play the role of an international platform for collection, discussion and dissemination of the research results on mercury and other pollutants.

5.1.4. Task Force on Emission Inventories and Projections (TFEIP)

One of the main goals of the Convention is an improvement of quality, transparency, consistency and completeness of emission inventories. Information on pollutant's emission to the atmosphere and other environmental media is one of the key parameters required for model assessment of pollution levels and transboundary fluxes. Uncertainties of emission data can significantly affect quality of the model estimates. At the recent session of the Executive Body [*ECE/EB.AIR/133/add.1*] it was stressed that scientific work to improve the quality and robustness of emission and projection data should be further maintained and reflected in the 2016-2017 workplan for the implementation of the Convention. This year it is planned to prepare a joint CEIP/MSC-E Technical report on current situation of heavy metal issues in the EMEP region, including methodologies used for gap filling, discrepancies between reported emissions and emission expert estimates, identification of sources of errors, proposals to upgrade current situation and practice, etc. The Report should serve as a basis for the EMEP strategy to alleviate existing problems and guide Parties.

MSC-E contribution concerning emission data for heavy metal modeling was presented at the EMEP Steering Body and WGE extended Bureaux (Geneva, March 2016), the TFMM meeting (Utrecht, May 2016) and sent to TFEIP and CEIP for consideration. Information on MSC-E activities in the field of heavy metal emissions for modeling is available on the MSC-E website: www.msceast.org.

5.2. International organizations

Cooperation with other international organisations and programs is one of the priority tasks of EMEP aimed at information exchange on environment pollution and outreach of the knowledge gathered within the Convention.

5.2.1. UNEP Minamata Convention

Mercury is a highly toxic pollutant capable of global dispersion in the environment [*Maas and Grennfelt*, 2016]. Global concern over mercury pollution resulted in development of the Minamata Convention on Mercury a legally-binding multilateral environmental agreement that was adopted by governments in 2013 [http://www.mercuryconvention.org/]. To support the negotiation process the United Nations Environment Programme (UNEP) coordinated preparation of a series of Global Mercury Assessments (GMA) [*UNEP*, 2002; *AMAP*/*UNEP*, 2008; AMAP/*UNEP*, 2013; AMAP/*UNEP*, 2015]. EMEP participated in all of the Assessments sharing information on mercury pollution and coordinating activities on global scale modeling.

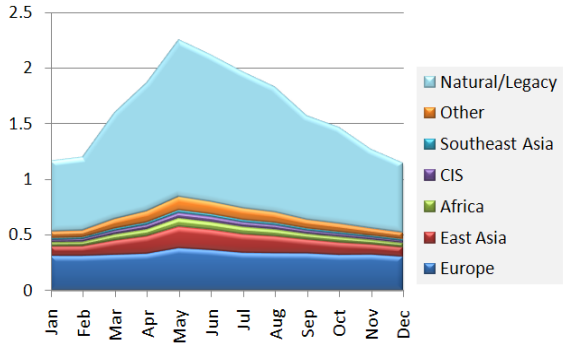

Fig. 5.5. *Seasonal variation of source attribution of average mercury deposition in Europe in 2013*

In particular, MSC-E took part in preparation of the UNEP Global Mercury Assessment 2013 (GMA 2013) used for negotiations of the Minamata Convention [*AMAP/UNEP*, 2013]. Variety of information was collected and made available for the assessment purpose including updated emissions inventory for mercury and observations on a global scale. These data are also useful in the EMEP operational activites on evaluation of mercury transboundary pollution of the EMEP countries.

MSC-E coordinated an update of GMA 2013 with new modeling results on evaluation of mercury intercontinental transport and source attribution of mercury deposition (Fig. 5.5) using the up-to-date global inventory of mercury anthropogenic emissions [AMAP/*UNEP*, 2015].

A new Global Mercury Assessment (GMA 2018) is now in progress in accordance with the request of the UNEP's Governing Council (Decision 27/12). MSC-E was invited to take a lead of the assessment part focused on modeling of mercury pollution on global and regional scales. Participation of the Centre in the assessment will be co-funded by UNEP. General structure of the assessment, participating experts and time schedule of the assessment were recently discussed at the first meeting of the GMA 2018 Project Coordination Group (Geneva, Switzerland, April 2016).

Co-operation with UNEP and the Minanata Convention on Mercury can support pollution assessment within EMEP by variety of data (including mercury emission inventories and observations on a global scale) as well as broaden dissemination of the scientific and policy oriented information generated within the Convention.

5.2.2. Helsinki Commission

Evaluation of airborne pollution load to marginal seas by heavy metals within the EMEP region is of interest by the marine conventions (e.g., HELCOM, OSPAR). In cooperation with other EMEP Centres, MSC-E performs regular model assessment of atmospheric pollution of the Baltic Sea by various pollutants including heavy metals. This work is carried out in accordance with the Memorandum of Understanding between the Baltic Marine Environment Protection Commission (HELCOM) and the United Nations Economic Commission for Europe (UN ECE) and is based on the long-term EMEP/HELCOM contract.

Current assessment of heavy metal pollution load to the Baltic Sea included estimation of long-term changes of lead, cadmium and mercury deposition during the period 1990-2013, source apportionment of heavy metal deposition in 2013 and verification of modeling results against measurements. Outcome of the assessment is available in a form of indicator fact sheets published on the HELCOM website [http://www.helcom.fi] and contribution to the EMEP Centres Joint report for HELCOM [*Semeena et al.*, 2015].

Anthropogenic emissions of the HELCOM countries dropped substantially from 1990 to 2013 (by 88% for lead, 65% for cadmium and mercury). In 2013 the biggest contributors to heavy metal emissions of the HELCOM area were Poland, Russia and Germany. The share of these countries in total emissions of heavy metals in the Baltic Sea region was about 90%.

Model evaluation of pollution levels indicates marked decrease of annual total atmospheric deposition of heavy metals to the Baltic Sea in the period of 1990-2013. The most significant drop of heavy metal deposition is obtained for lead (81%), followed by cadmium (68%), and mercury (29%) (Fig. 5.6a). Temporal changes of heavy metal pollution in different parts of the Sea are not homogeneous. Particularly, significant decrease of lead and cadmium deposition is noted in the Bothnian Bay sub-basin (about 90% and 80%) and of mercury in the Sound sub-basin (about 60%).

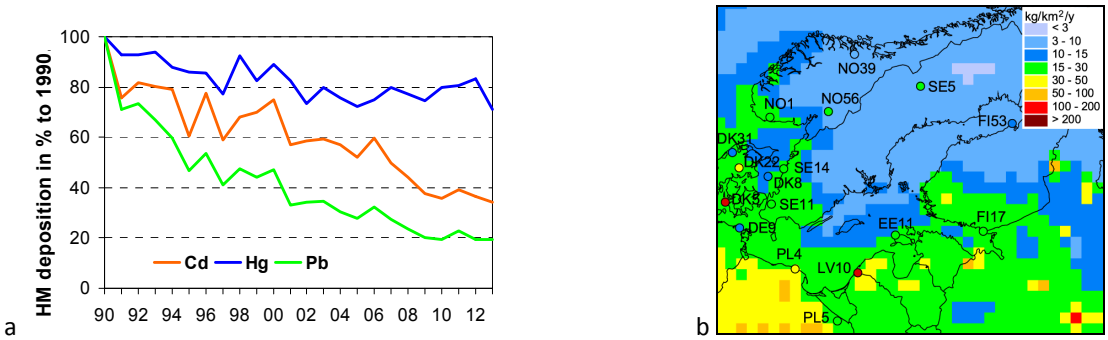

Fig. 5.6. *Relative changes of total annual atmospheric deposition of cadmium, mercury, and lead to the Baltic Sea for the period 1990-2013 (a) and spatial distribution of lead deposition over the Baltic Sea in 2013 along with annual mean observed lead deposition shown in coloured circles (b)*

Spatial variation of heavy metal deposition to the Baltic Sea in 2013 is illustrated in Fig. 5.6b by lead deposition fluxes. Elevated levels of pollution can be seen in the western part (the Kattegat and the Western Baltic sub-basins) and in the eastern part of the Baltic Sea (the Gulf of Finland). Along with

modeled values of heavy metal deposition the map provides observed deposition fluxes obtained at monitoring sites around the Baltic Sea. As seen, the model satisfactory describes spatial variations of measured lead deposition in the Baltic Sea region.

Anthropogenic emission sources of the HELCOM countries contributed to heavy metal deposition to the Baltic Sea more than 30% for cadmium, 20% for lead, and 10% for mercury. The largest contributions among the HELCOM countries to heavy metal deposition of the Baltic Sea are made by Poland, Germany, and Russia.

5.2.3. Arctic Monitoring and Assessment Program

EMEP has a long and successful experience of co-operation with the Arctic Monitoring and Assessment Programme (AMAP), which includes participation in joint assessments and projects. Specifically, MSC-E took part in a number of the AMAP Assessments of the Arctic pollution with mercury and other heavy metals [*AMAP*, 2005; 2011].

Recently, possible future cooperation between AMAP and CLRTAP was discussed at the joint meeting held in Potsdam, Germany in February 2016. Potential for collaboration was identified in a number of scientific and technical areas. *Assessment of mercury pollution of the Arctic and cooperation with UNEP on updating its Global Mercury Assessment were considered among priority directions of future collaboration.*

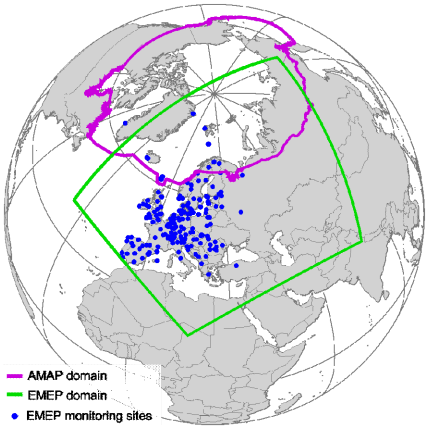

Fig. 5.7. *EMEP and AMAP domains*

The AMAP and EMEP domains have significant overlap over the European and Asian sectors of the Arctic (Fig. 5.7). Thus, the majority of information generated within EMEP as a part of its operational activity is also relevant to the Arctic pollution assessment. Moving towards the future collaboration between AMAP and CLRTAP in the field of heavy metal pollution MSC-E has prepared a brief summary of the pollution assessment for the overlap of the Arctic and EMEP domains.

Figure 5.8 shows spatial distribution of heavy metal depostion over the Arctic sector of the EMEP domain. Deposition of both lead (Fig. 5.8a) and cadmium (Fig. 5.8b) has a pronounced north-to-south gradient of deposition flux indicating transport of the pollutants northward from emission sources located in Europe and the Asian part of the Russian Federation. Elevated deposition fluxes are also characteristics of the Northern Atlantic as a result of wind suspension of heavy metals from the ocean with sea salt. Spatial distribution of mercury deposition differs significantly from those of lead and cadmium (Fig. 5.8c). The figure shows mercury net deposition flux defined as a difference between atmospheric deposition and prompt reemission from snow and ice surfaces. Along with the southward gradient increased net deposition is also predicted over the open water areas of the high Arctic. Relatively large deposition fluxes of mercury in the Arctic are determined by fast oxidation during the Atmospheric Mercury Depletion Events. However, significant amount of mercury

deposited to snow undergoes photoreduction and is reemitted back to the atmosphere. Therefore, net deposition over ice covered areas of the Arctic is not as high as over open waters.

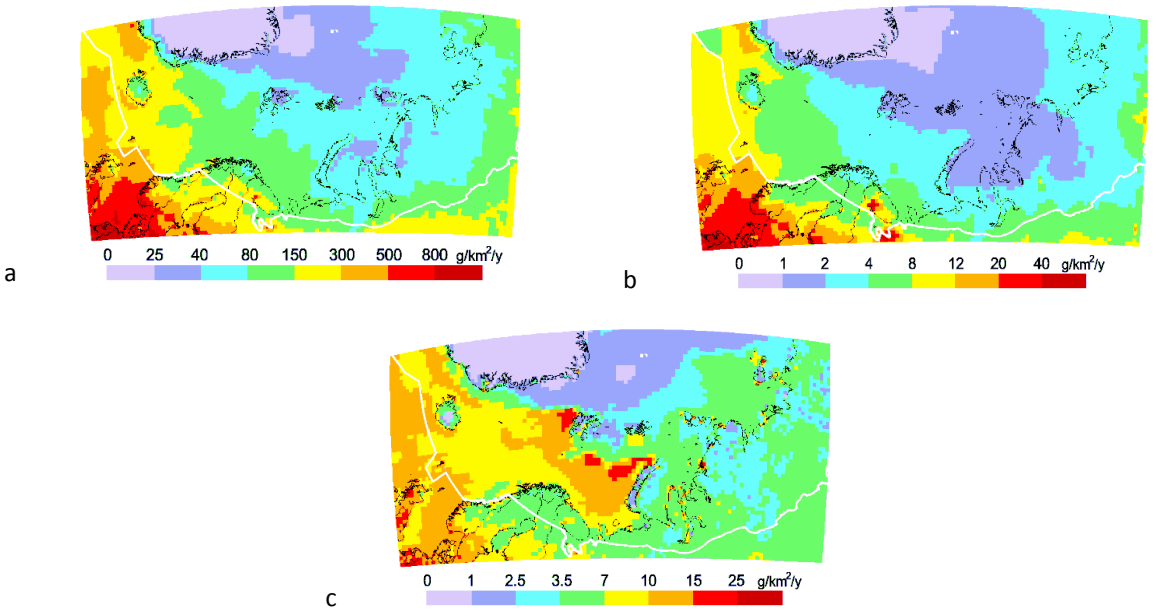

***Fig. 5.8**. Spatial distribution of annual total deposition of lead (a), cadmium (b) and net deposition flux of mercury (c) over the Arctic sector of the EMEP region in 2014. White line depicts the boundary of the AMAP domain*

Relative contribution of different source types to heavy metal deposition over the European and Asian terrestrial ecosystems of the Arctic is shown in Fig. 5.9. From 10% to 25% of lead and cadmium deposition to these remote areas are determined by EMEP anthropogenic sources (Figs. 5.9a and b). Besides, significant fraction (27-36%) of deposition is also caused by atmospheric trasnsport of lead and cadmium re-suspended from urban and bare lands (largely, from arid regions of Central Asia). The rest 50-55% of deposition are transported from non-EMEP anthropogenic and secondary sources. In contrast, mercury deposition to terrestrial areas of the Arctic is mostly defined by global sources. Long residense time of mercury in atmospheric air leads to global scale transport and mixing of this pollutant in the atmosphere. It should be noted that the fraction of "non-EMEP" sources in mercury deposition includes indirect contribution of EMEP emissions that flew out the region and mixed up with the global atmospheric mercury pool.

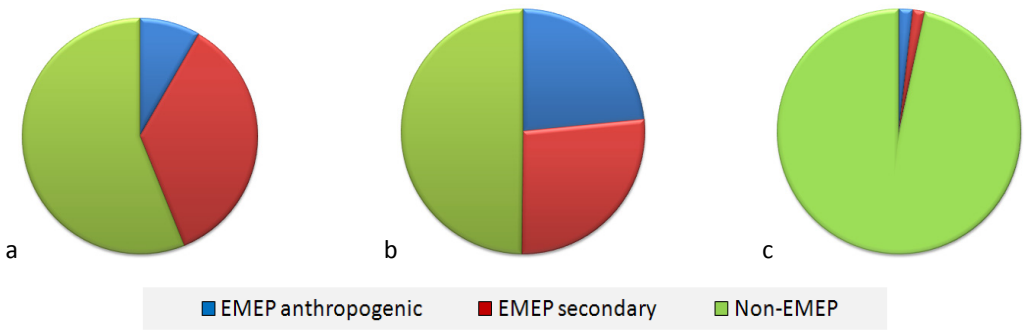

***Fig. 5.9.** Average contribution of different source types to annual deposition of lead (a), cadmium (b), and mercury (c) to the European and Asian terrestrial areas of the Arctic covered by the EMEP domain*

MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

Various aspects of heavy metal pollution assessment in the EMEP region in 2016 are addressed in this report. They include deposition, concentrations and transboundary transport of heavy metals in 2014, progress in model development, cooperation with the EMEP courtiers, CLRTAP subsidiary bodies and international organizations etc. Particular attention is paid to mercury pollution assessment. This section summarizes the main challenges revealed in the report and suggests directions to improve assessment of heavy metal pollution in the EMEP region.

- Background monitoring is one of the key sources of information used for description of current pollution levels and their temporal trends as well as for evaluation of modeling results. Nowadays heavy metal monitoring data are available mainly from the western, central and northern parts of Europe, whereas in the eastern and southern parts of Europe and Central Asia the monitoring network is scarce. Therefore, more sites with continuous measurements in these regions are needed. Partly this situation could be improved by use of biomonitoring data (mosses etc.) Another important issue is related to quality assurance and quality control of heavy metal monitoring in the EMEP region. Analysis of measurement data has shown existence of low quality data at some monitoring sites. Therefore, laboratories in these countries need to evaluate their methodologies.

- Uncertainties of the reported emission data are still high. The key parameters of heavy metal emission which affect quality of the model assessment are analysed and ranked in terms of their priority. It has been indicated that quality of gridded emission data (including completeness of reported data and quality of expert estimates used for the gapfilling) as well as chemical speciation of mercury emissions are at the top of the priority list. Other important parameters include data on temporal variability and vertical distribution of emissions for lead and cadmium, and global scale inventories as well as historical emissions for mercury. This ranking is a basis for further improvement of emission inventories under EMEP.

- Mercury pollution of the EMEP region attracts particular attention within the Convention. Therefore, improvement of the quality of mercury pollution assessment is among of the main tasks under EMEP. Performed multi-model analysis of mercury processes in the atmosphere has revealed a need of update of the current modeling approaches used for mercury pollution assessment within EMEP. Parameterizations of mercury atmospheric chemistry in the GLEMOS modeling system will be improved along with update of input data on air concentration of chemical reactants. Besides, evaluation of mercury pollution changes over long periods requires consideration of cycling and accumulation in different environmental media. The multi-media approach to mercury simulations should be employed in the GLEMOS modeling system.

- Further improvement of the modeling approaches also includes transition of the EMEP operation modeling to the new grid. Pilot simulations of mercury pollution on the new EMEP grid demonstrate promising results. However, additional development and testing are still required for simulation of other heavy metals. Another direction of the model development is connected with preparation and distribution of the GLEMOS modeling system for public use as an open code to support development of country-scale modeling approaches by national experts.

- EMEP national-scale assessments allow detailed investigation of country-specific pollution levels with fine spatial resolution in close cooperation with national experts. The study of lead pollution levels in Belarus reveals that anthropogenic emissions of lead in Belarus and some neighbouring countries are likely incomplete. For the analysis of heavy metal emission data in the EECCA countries joint efforts of national emission experts of these countries together with CEIP and TFEIP are required. Besides, in order to establish quality of measurements in the EECCA region, participation of national laboratories in the regular intercomparisons under the CCC supervision is appreciated. Further investigation of influence of emission data on a country-scale assessment will be continued in a case study for Poland, where possible uncertainties of temporal variation of emissions are identified. In addition, a case study for the United Kingdom has been also initiated. One of the important aims of this study is investigation of role of secondary sources in heavy metal pollution of the country.

- Cooperation between WGE and EMEP in the field of joint analysis and assessment of heavy metal pollution levels and their effects within the EMEP region is highly important. First of all, regular surveys of heavy metal concentrations in mosses, coordinated by the ICP-Vegetation of WGE, are valuable supplementary source of monitoring information for assessment of heavy metal pollution in the EMEP region and could complement the available station-based EMEP monitoring data. Further usage of these biomonitoring data in analysis of spatial distribution and long-term trends of heavy metal pollution and in verification of modeling results could be useful. Furthermore, in spite of long-term decline of lead, cadmium and mercury deposition and their critical loads exceedances, heavy metals continue accumulating in soils. Besides, levels of mercury in fish tend to increase. Further activity on evaluation of negative effects of heavy metals in the environment is significant.

WORK-PLAN FOR 2017

No	Activity description	Deliverable(s) and planned completion data	Input	Output
	Ongoing activities			
1.	Calculations of concentration and deposition fields on HMs for the EMEP domain for 2015. Evaluations against measurement data.	Annual status report on HMs	CCC, CEIP	EB, EMEP SB, WGSR, WGE, Parties
	Generation of data on ecosystem-dependent deposition of HMs. Analysis of deposition peculiarities.	Web-accessible data on ecosystem-dependent deposition		
	Source-receptor calculations	Web-accessible data on concentration and deposition fields and country-to-country deposition matrices (for 2015)		
2.	Estimates and analysis of transboundary pollution of marginal seas and the Arctic for 2015	Sections to annual status reports on HMs to EMEP SB Web-accessible data on transboundary pollution of marginal seas	CCC, CEIP	HELCOM, OSPAR, AMAP
3.	Detailed assessment of pollution levels of HMs in individual countries, based on model calculations and available national data	Individual country reports on transboundary air pollution for 2015 Web-accessible updated country specific data	CEIP CCC	Parties
4.	Assessment of transboundary pollution of HMs in the EECCA countries to facilitate ratification and implementation of the Protocol	Annually updated web-accessible information for 2015 in Russian		EECCA countries
6.	Preparation of meteorological data for regional and global modeling and for fine scale simulations on the new EMEP grid for 2015	Sections to annual status reports on HMs		
7	Cooperation with Convention subsidiary bodies	Sections to annual status reports on HMs		TFMM, TFHTAP, TFEIP, WGE
8	Exchange of information with national experts and international organizations	Sections to annual status reports on HMs		Parties, UNEP, Minamata Convention, AMAP, HELCOM

Research activity in 2017

9	Pilot simulations of long-range transport of HMs in the new EMEP grid and evaluation of modeling results: model performance and source-receptor relationships	EMEP status and MSC-E technical reports	CEIP, CCC	TFMM, SB
10	Ecosystem-dependent deposition fluxes of HMs to different land use types in the new EMEP grid	Model results (Section to status report)		TFMM, WGE
11	Assessment of HM pollution levels with fine spatial resolution generated in cooperation with national experts (EMEP case studies on HMs)	Technical reports jointly prepared with national experts. Poland and the United Kingdom foreseen.	Poland, the United Kingdom	SB, TFMM, Parties
12	Explore possible use of EMEP/WGE tools, data and infrastructure to support AMAP activities	Report to EMEP/WGE session		SB, WGE, AMAP
13	Continue collaboration with HELCOM related to atmospheric monitoring and modeling and data management	Report to the EMEP/WGE session		SB, HELCOM
14	Review and assess data, methodologies and competences available to deal with HMs issues in the ECE region and propose a strategy to improve emission inventories	Joint CEIP/MSC-E technical reports	CEIP	TFEIP, CEIP, SB
15	Contribute to air quality assessment in the EECCA countries	Report to the EMEP Steering Body	CEIP, CCC	SB EECCA countries
16	Improvement of model parameterization of pollutant exchange between different compartment (air, water, soil, vegetation) based on literature and national data on occurrence of HM (particularly Hg) in relevant compartments with global geographical coverage	Section to technical report		TFMM, National experts
17	Facilitate the use of the EMEP model by Parties	Make annual release of the GLEMOS open source code		TFMM, SB, Parties

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Annex A

EVALUATION OF MODELING RERSULTS VS. OBSERVATIONS

Uncertainties of the modeling results are combined of three components: uncertainties of emission data, of monitoring information and of the model as such. Uncertainties of anthropogenic emission data are estimated and reported by some EMEP countries [*CEIP*, 2016]. As seen, the uncertainties of annual emission data of lead, cadmium and mercury reaches almost 500%, 430% and 145%, respectively (Table A.1).

Table A.1. *Uncertainties (%) of the reported emission data in 2014*

2014	Pb	Cd	Hg
Belarus	182	175	107
Belgium	271	231	145
Croatia	136	278	76
Cyprus	95	81	13
Denmark	488	427	91
Estonia	139	130	138
Finland	±28	-30 +31	±21
France	148	28	20
Latvia	78	80	66
Poland	70	70	53
Sweden	18	35	56
United Kingdom	53	-30 to >50	-30 to 50

Monitoring uncertainties occur at different stages of obtaining measurements, such as sampling, transporting, storing, laboratory analysis. At present only uncertainty of laboratory analysis is regularly evaluated during annual laboratory intercomparison tests, undertaken under CCC supervision. Numerical characteristic of analytical uncertainty is relative deviation of concentration, analyzed in a laboratory, from theoretical value. In order to characterize quality of laboratory analysis, so-called Data quality objectives (DQO) was introduced by CCC. For heavy metals DQO are equal to ±15% of high concentrations (typical for background levels in the central and eastern parts of Europe) and ±25% for low concentrations (typical for Scandinavia). The most recent results of laboratory intercomparison tests for lead and cadmium demonstrated that for most of laboratories quality of the analysis fits DQO criteria [*CCC*, 2015].

Model uncertainties have been evaluated in [*Travnikov and Ilyin*, 2005]. It was demonstrated that intrinsic model uncertainty (i.e., uncertainty of the model as such, without effect of emission data) was amounted to ±30-40% for Europe as a whole (Fig. A.1).

Quality of the model assessment is evaluated via comparison with the available background monitoring data. Detailed information about the comparison is given in Technical report [*Shatalov et al.*, 2016], while the main results of the model evaluation are summarized in this section. Values of main statistical indicators used is the comparison for lead, cadmium and mercury are summarized in Table A.2. These indicators are Mean Relative Bias (MNB), Pearson's correlation coefficient and Normalized Root Mean Square Error (NRMSE). Besides, a share of model-measurement pairs of values differing from each other within 2-fold and 3-fold range (F2 and F3, respectively) is also presented.

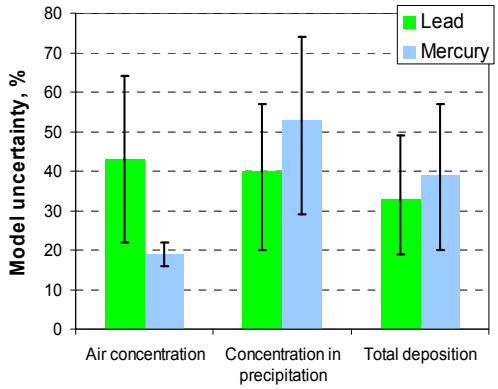

Fig. A.1. *Averaged for the EMEP region intrinsic (without effect of emission) model uncertainty for aerosol-born metals (lead) and for mercury. Whiskers indicate range of uncertainties over space.*

Table A.2. *Main statistical indicators of agreement between annual modeled and measured levels of air concentrations and wet deposition fluxes in 2014*

	Lead		Cadmium		Mercury	
	C_{air}	Wet Dep	C_{air}	Wet Dep	C_{air}	Wet Dep
Mean relative bias, %	4.3	-10.0	-10.8	-20.7	3.9	31.7
Correlation coefficient	0.82	0.50	0.79	0.46	0.17	0.22
NRMSE	0.39	0.59	0.44	0.53	0.07	0.54
F2, %	89	77	76	77	100	76
F3, %	97	94	88	87	100	100

C_{air} – concentration in air
Wet Dep – wet deposition flux
NRMSE – Normalized Root Mean Square Error
F2 – fraction of values fitting to factor of 2 difference
F3 – fraction of values fitting to factor of 3 difference

As seen, lead and cadmium air concentrations were reproduced with low relative biases (within ±10%) and with significant correlation coefficients (0.82 and 0.79, respectively). For wet deposition some underestimation is indicated. For lead it is around 10%, and for cadmium – about 20%. However, correlation for wet deposition of lead and cadmium in 2014 is relatively weak. Value of Pearson's correlation coefficient is very sensitive to outliers. Thus, the main reason of comparatively low correlation is large discrepancies between modeled and observed levels at few particular stations. Most of model-observation pairs of lead and cadmium concentration and deposition values agree within a factor of two.

Magnitude of mercury air concentrations reproduced reasonably well, since MRB is only around 4 % and all modeled and measured pairs of values agree within a factor of two. Correlation coefficient for

air concentrations is not high because of very low spatial variability of modeled and observed background mercury levels. This variability is comparable with uncertainties of the model, which results to low correlation. The reason for this is similar to that for lead and cadmium, i.e. discrepancies at some stations.

Maps with modeled pollution fields and measured values at stations allow to demonstrate where the model manages to reproduce observed levels, and where discrepancies between modeled and measure levels occur. Observed air concentrations were reproduced by the model almost in all regions where measurement station were located, namely in France, Spain, Germany, the United Kingdom, Denmark, the Netherlands, the Czech Republic (Fig. A.2a). At most of stations in these countries the agreement between annual mean modeled and observed air concentrations lay within ±50%. Higher discrepancies were noted for wet deposition fluxes. Satisfactory agreement between modeled and observed wet deposition fluxes took place for most of stations in Germany, Slovakia, Denmark (Fig. A.2b). Underestimation of lead levels is seen for Spanish, one Norwegian, one British and the Czech stations. For stations in Poland and some stations in Germany and Sweden overestimation of observed wet deposition fluxes is noted.

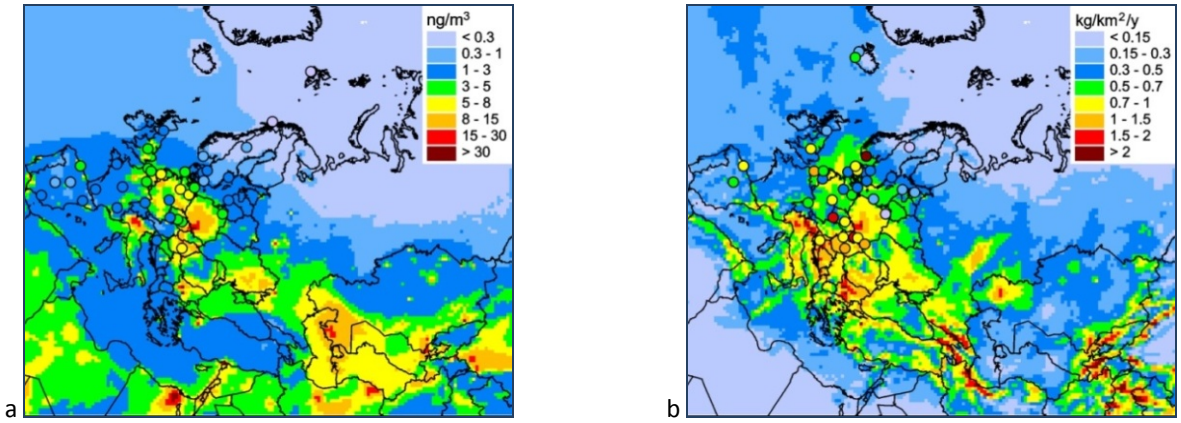

Fig. A.2. *Spatial distribution of modeled and observed lead concentrations in air (a) and wet deposition fluxes (b) in 2014*

Model performance for air concentrations of cadmium is similar to that for lead. The model reproduced cadmium levels at most of stations in various parts of the EMEP region. In particular, reasonable accuracy (within ±50%) was achieved for stations in the United Kingdom, Belgium, the Czech Republic, Hungary, most of German stations (Fig. 3a). Observed air concentrations were underestimated by the model at most of Scandinavian stations. Wet deposition fluxes, simulated by the model, match the observed fluxes within ±50% agreement at most of monitoring stations (Fig. A.3b). At some monitoring stations observed levels were much higher than the modeled ones. These stations are located in Spain, Norway, Hungary. Besides, the underestimation of observed fluxes is noted for one Swedish and one Slovak station. For one of German stations the model overestimates cadmium deposition levels by almost 70%. Discrepancies at these few stations resulted to relatively low spatial correlation coefficient (Table A.2).

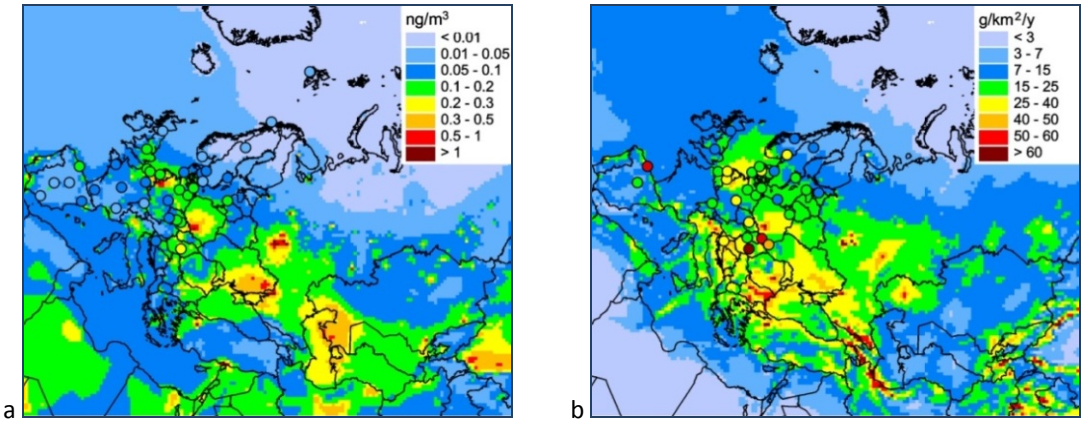

Fig. A3. *Spatial distribution of modeled and observed cadmium concentrations in air (a) and wet deposition fluxes (b) in 2014*

There are numerous reasons leading to the discrepancies between modeled and measured levels. To some extent the discrepancies can be explained by modeling uncertainties. Average value of the intrinsic model uncertainty is around ±40% for concentrations in air and ±30% for deposition, which is not enough to explain two-three-fold difference between modeled and observed levels at some stations. Another possible reason is uncertainties of anthropogenic or secondary emissions. As shown above, in some countries these uncertainties of their national emission can reach as much as 500%. Secondary emission is another source of the model uncertainties. Parameterization of this process is complicated because it requires various environmental data which are often uncertain, such as soil texture and water content and concentration of heavy metals in soils. Introduction of wind re-suspension of heavy metals allows to reach reasonable bias between measured and modeled concentrations and deposition. However, at some stations the contribution of re-suspension is likely too high and leads to the overestimation of observed levels. This year re-calculation of lead enrichment factor for topsoils has been undertaken, which favoured to reduce the overestimation. Nevertheless, further activity on improvement of parameterization of wind re-suspension should be continued. Finally, measurement data also contain uncertainty. Part of this uncertainty is identified by laboratory intercomparison tests. However, information on full measurement uncertainty is needed for interpretation of discrepancies between modeled and monitored heavy metal levels.

Discrepancy between modeled and observed concentrations of mercury lay within ±8% for all stations except SE5, which levels were significantly lower than those at other monitoring stations (Fig. A.4a). For most of stations modeled wet deposition fluxes matched the observed fluxes within ±50% range (Fig. A.4b). However, at some stations around two-fold overestimation of the observed fluxes took place. These are two stations in Germany, one in the United Kingdom, one in Finland and in the Netherland.

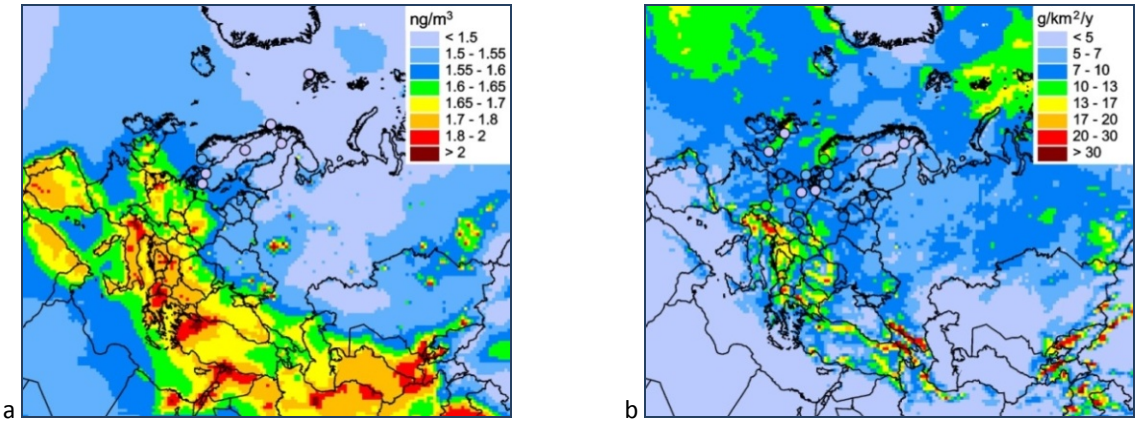

Fig. A.4. *Spatial distribution of modeled and observed elemental mercury concentrations in air (a) and wet deposition fluxes (b) in 2014*

Uncertainties of modeling, anthropogenic emissions and monitoring data, identified for lead and cadmium, are also relevant for mercury. However, unlike lead and cadmium, another important source of uncertainty of mercury modeling is parameterization of its atmospheric chemical transformations. Te role of chemical reactants as well as products of chemical reactions are still under discussion [*Lin et al*., 2006; *Gustin et al*., 2015]. Another peculiarity of mercury is speciation of anthropogenic emissions. The emission occurs in three forms: relatively inert and long-lived elemental mercury and short-lived particulate and gaseous oxidized mercury forms. Currently information about mercury emission speciation is not officially reported and obtained only in emission expert estimates.

Annex B

METEOROLOGICAL CONDITIONS OF 2014

Meteorological conditions markedly influence heavy metal pollution levels in the EMEP region. Peculiarities of atmospheric circulation determine transboundary transport patterns of pollutants. Wet scavenging, being the main mechanism of removal of heavy metals from the atmosphere, strongly depends on precipitation. Air temperature affects rates of chemical reactions and solubility of gaseous substances in cloud droplets and, thus, important for modeling of chemical transformations of mercury in the atmosphere. Besides, meteorological conditions influence wind suspension of dust particles containing previously deposited lead and cadmium, and, hence, affects secondary emission fluxes.

Since wind is caused by gradient of atmospheric pressure, anomalies (differences from climatic normal value) of pressure are often used to characterize circulation situations, affecting weather and atmospheric transport. Four main circulation modes (or 'patterns') for European region are singled out [*Polonskii and Kibal'chich,* 2015*; Barnston and Livezey,* 1987]. The most important mode is so-called *North Atlantic Oscillation* (NOA). This term is used to define variations of differences in pressure between Azores High and Icelandic Low. In its positive phase pressure gradient between the Azores and Icelandic Low is high, which results to strong westerly winds in central and northern Europe. In its negative phase the westerlies weaken. Another mode, also characterizing strength of zonal transport is *East Atlantic pattern* (EA). Unlike NOA, positive phase of EA means distinct westerly transport in subtropical of East Atlantic and Europe. *Scandinavian pattern* (SC) in its positive phase describes situation of antyciclonic blocking over Scandinavia and north-western Russia. The positive phase of the forth mode – *East Atlantic/West Russia pattern* (EA/WR) is associated with positive pressure anomalies over Central Europe and China and negative anomaly over Caspian region. Therefore, unlike NOA and EA, strength of SC and EA/WR most likely may characterize intensity of inter-latitude exchange of air masses.

Main typical weather features, linked with different modes of circulation are summarized in Table B.1 [*Polonskii and Kibal'chich,* 2015*, CPC*]. These features are described in terms of anomalies, i.e., deviations from climatic mean values. Hence, notations like 'positive', 'above-average' etc. mean values higher than climatic mean, and vice versa.

Numerical characteristic of these circulation modes is expressed via special indices. The simplified approach to produce the indices based on pressure gradient between centres of pressure anomalies. For example, NAO index can be estimated on the base of surface pressure gradient between Icelandic Low and Azores High. However, it should be noted that currently more sophisticated methods are used for circulation index calculation.

Table B.1. *Main circulation modes in Europe and typical associated weather patterns*

Mode	Typical circulation features within EMEP region	Typical weather patterns
NAO	Strength of zonal transport in mid-latitudes	In positive phases, above-averaged temperatures across northern Europe and below-average temperatures in southern Europe. Above-average precipitation over northern Europe and Scandinavia in winter, and below-average precipitation over southern and central Europe. In negative phase, opposite weather patterns take place.
EA	Strength of zonal transport in subtropical zone	In positive phase, surface air temperature above-normal over the most part of Europe. Precipitation deficit in Southern Europe. Opposite trends are observed in Northern Europe and Scandinavia.
SC	Anticyclones blocking over Scandinavia and associated meridianal transport	In positive phase, negative anomalies of surface temperature in Central Russia and Western Europe. Increase in the amount of precipitation in Central and Southern Europe. Precipitation deficit in Scandinavia and in the north east of Europe.
EA/WR	Positive pressure anomaly over Central Europe, negative anomaly over Caspian region and associated meridianal transport.	In positive phase, characterizes the conditions of anticyclonic blocking over the European part of Russia. Negative anomalies of surface air temperatures over the most part of the west of the European part of Russia and over the northeast of Africa. Decrease of precipitation over Central Europe and the Mediterranean region. The situation is opposite during the negative phase.

Table B.2 summarizes indices in European region for different seasons of 2014 [*WMO*, 2014]. As seen, in winter and autumn the westerly atmospheric transport was more distinct (positive NOA and high EA indices) compared to summer and spring. Comparatively high absolute values of EA/WR in winter are caused by spreading of ridge of Asian high over most part of Europe and linking to Azores high. Relatively high values of SC index in winter compared to summer denote more often blocking events in these seasons, suppressing westerly transport and favouring inter-latitude exchange of air masses.

Table B.2. *Indexes of main circulation modes in Europe in 2014*

Season	NAO	EA	EA/WR	SC
Winter 2013/14	0.56	1.6	-1.20	0.71
Spring 2014	-0.06	0.61	-0.11	0.68
Summer 2014	-0.91	0.10	-0.66	0.21
Autumn 2014	0.48	0.55	0.46	0.61

Mean annual air near-surface temperatures and annual sums of precipitation in 2014 are demonstrated in Fig. B.1. Annual temperatures in 2014 ranged from about -15°C in the Arctic to around 20-25° in the Mediterranean (Fig. B.1a). Obviously, seasonal and daily oscillations lead to much higher range. The areas of the highest precipitation amounts in 2014 take place over the North

Atlantic, western coast of Norway, the Balkan region and at up-wind ridge slopes of the Caucasus, Asia Minor and mountains of Central Asia (Fig. B.1b). The Arctic as well as desert regions of Central Asia and North Africa are characterized by the lowest precipitation sums.

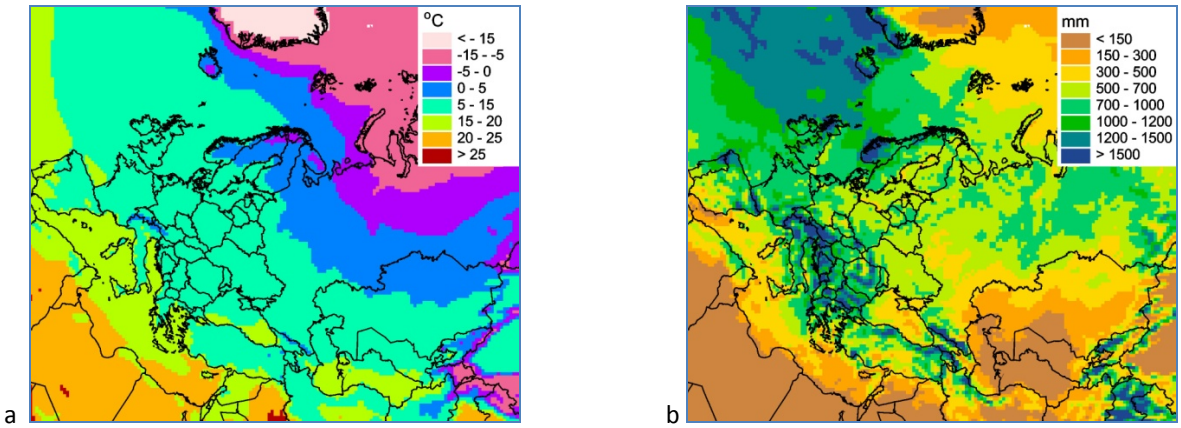

Fig. B.1. *Annual mean near-surface air temperature (a) and annual precipitation sums (b) in the EMEP region in 2014.*

Compared to climate, year 2014 is known for positive temperature anomalies over most part of Europe, Arctic and North Atlantic (Fig. B.2a) [*WMO*, 2014]. The highest anomalies (3-4°C) occur over Arctic. Over Scandinavia, the central part of Europe and the Balkan region positive anomalies of 2-3°C are noted. Anomalies of annual precipitation are insignificant over most part of the EMEP region (Fig. B.2b). However, large increase of precipitation occurred over Balkans, north of Italy and some areas of western and northern Europe. In the western part of Russia and in Scandinavia precipitation amounts in 2014 were lower than normal.

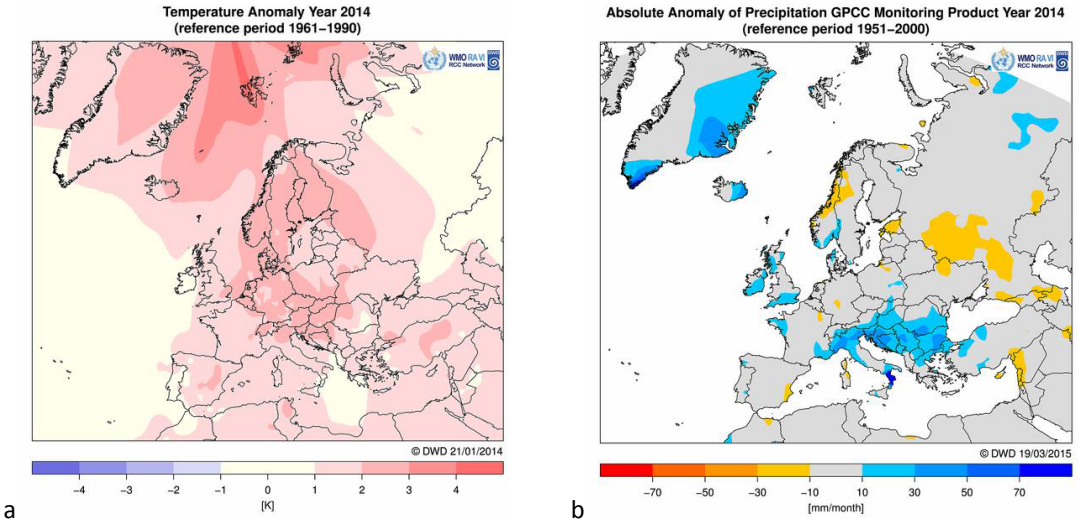

Fig. B.2. *Absolute anomalies of surface temperature (a) and precipitation (b) in 2014 [WMO, 2014]*

For analysis of pollution level differences between 2013 and 2014 the changes of annual air temperatures and precipitation were compared. In 2014 air temperatures were 0.5 – 1.5°C higher compared to 2013 over most part of Western, Central and North Europe (Fig. B.3a). Besides, similar difference occurs over North-East Atlantic and Arctic. Over most part of European Russia, Central Asia and North Atlantic lower (0.5 – 1.5°C) temperatures, compared to 2013, take place.

Spatial distribution of changes in annual precipitation sums is more mosaic compared to that of air temperatures. First of all, increase of precipitation in Balkans and Turkey by 30 – 60% and similar decrease in European Russia and Kazakhstan is noted (Fig. B.3b). Lower precipitation also occurs over Central and South Europe (Poland, Germany, France, Spain). Large relative differences are also noted for North Africa, but it should be noted that precipitation in this arid region are low and thus even high relative changes result to small effect in absolute terms.

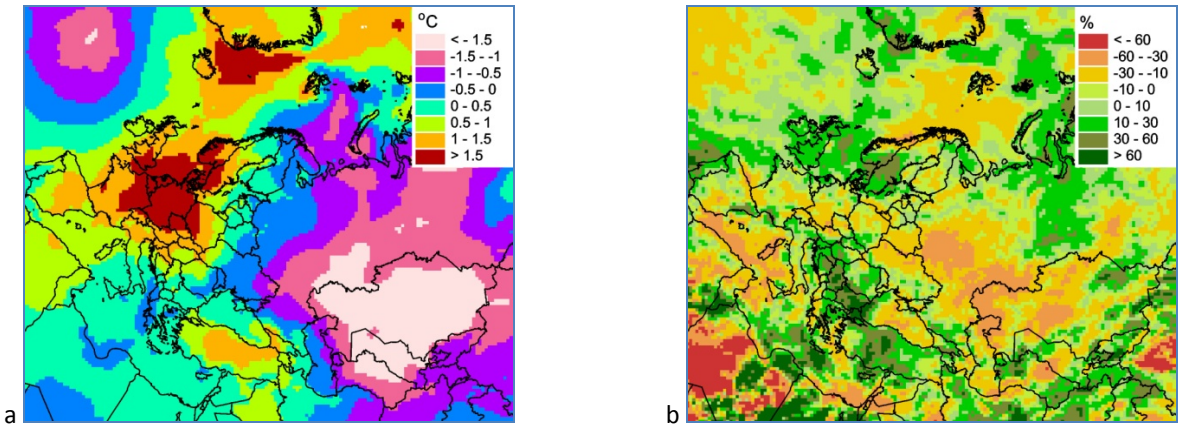

Fig. B.3. *Absolute difference of near-surface annual mean air temperatures (a) and relative difference of annual precipitation sums (b) between 2013 and 2014. Positive change mean increase, and negative – decrease of the parameters from 2013 to 2014*