

Heavy metals: Analysis of long-term trends, country-specific research and progress in mercury regional and global modelling

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Heavy metals: Analysis of long-term trends, country-specific research and progress in mercury regional and global modelling

METEOROLOGICAL SYNTHESIZING CENTRE - EAST

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

CHEMICAL CO-ORDINATING CENTRE

W.Aas



MCS-E

Meteorological Synthesizing Centre - East

2nd Roshchinsky proezd, 8/5
115419 Moscow
Russia
Phone.: +7 926 906 91 78
Fax: +7 495 956 19 44
E-mail: msce@msceast.org
Internet: www.msceast.org



CCC

Norwegian Institute for Air Research (NILU)

P.O. Box 100
NO-2027 Kjeller
Norway
Phone: +47 63 89 80 00
Fax: +47 63 89 80 50
E-mail: kjetil.torseth@nilu.no
Internet: www.nilu.no

EXECUTIVE SUMMARY

Heavy metals are known for their toxicity for human health and biota. Because of their ability to dispersion in the atmosphere over long distances (up to hundreds or thousands of kilometres) heavy metals are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP). Protocol on Heavy Metals to the Convention (Protocol on HMs), signed and ratified by most of the EMEP countries, the USA and Canada, is aimed at reduction of heavy metal emissions and decrease of the environmental pollution by these contaminants. Heavy metals targeted by the Protocol include lead, cadmium and mercury.

This report summarizes information on recent achievements of the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) in the field of heavy metal pollution assessment. It covers anthropogenic emissions and pollution levels of lead, cadmium and mercury in 2013 as well as transboundary atmospheric transport of these species based on both modelling results and measurement information. Particular attention is paid to the analysis of long-term changes of heavy metal pollution levels in the EMEP countries to support preparation of the CLRTAP Assessment Report. Furthermore, it describes further development of the Global EMEP Multi-media Modelling System (GLEMOS). Special focus is made on exchange of information between MSC-E and subsidiary bodies to the Convention, as well as dissemination of output information to national and international organizations.

Lead emissions in the EMEP region dropped by 90% between 1990 and 2012, whereas emissions of cadmium and mercury decreased approximately by 60%. However, the reduction rate differs significantly among the countries. The most noticeable emission reduction took place in countries of Western and Northern Europe (the United Kingdom, Luxembourg, Denmark, Norway, Sweden) and in some countries of Central Europe (Slovakia, Romania). Among the EECCA countries the highest emission decline was noted for Russia, Ukraine and the Republic of Moldova. There were also a number of countries, where some increase of anthropogenic emissions was indicated (e.g. Azerbaijan, Georgia and Liechtenstein). The data on heavy metal emissions for modelling within the EMEP domain for 2013 were provided by Centre on Emission Inventories and Projections (CEIP).

The EMEP monitoring network, as of 2013, includes 31 sites measuring heavy metals in both air and precipitation, and altogether there are 65 measurement sites. In total, 7 Parties to the Convention fulfil their monitoring obligations as defined in the EMEP monitoring strategy. Most of the EMEP monitoring stations are situated in the northern, western and central parts of Europe, while the background monitoring network is rather scarce in the eastern and south-eastern parts of Europe. The spatial coverage could be improved by more active involvement of the EECCA countries in the monitoring activity, and by taking into account data from international programmes and projects. Besides, alternative monitoring approaches such as biomonitoring in mosses and other passive sampling methods could be considered.

Pollution levels as well as long-term trends and transboundary transport among the EMEP countries are assessed by the environmental modelling. The areas with relatively high pollution levels of lead, cadmium and mercury in 2013 include the north-western part of Germany, the Benelux region, the southern part of Poland the northern part of Italy and the Balkans. Due to marked reduction of emissions in the EMEP countries for the last two decades deposition of heavy metals from anthropogenic sources strongly declined. Therefore, nowadays, relative contribution of secondary emission sources to pollution levels of cadmium and lead becomes higher than that in the nineties. Their contribution exceeds the contribution of direct anthropogenic sources in 14 countries for cadmium and in 6 countries for lead. For mercury, pollution levels are largely affected by non-EMEP emission sources (both anthropogenic and secondary), which dominate in 44 EMEP countries.

Long-term changes of heavy metal pollution in the EMEP region were analyzed for the period from 1990 to 2012. It was demonstrated that the overall pollution reduction in the EMEP region as a whole made up 78%, 53% and 23% for lead, cadmium and mercury, respectively. Commonly, the reduction rate in the EMEP countries was the highest at the beginning and the lowest at the end of the period. Besides, some countries (Armenia, Azerbaijan) were even characterized by increase of the pollution levels. The changes of pollution levels are mostly caused by emission reduction in the EMEP countries, by long-term changes of secondary emissions of lead and cadmium, and by dynamics of intercontinental transport of mercury. Following the reduction of heavy metal deposition, critical load exceedances for lead, cadmium and mercury, as well as the area over which the exceedances take place, have markedly declined in the EMEP region since 1990.

Transboundary transport plays an important role in pollution of the EMEP countries. Contribution of foreign sources to anthropogenic deposition of cadmium, lead and mercury exceeds prevail over contribution of domestic sources in 19, 24 and 17 countries, respectively. As a rule, contribution of foreign sources is higher near the state borders and is lower in the central parts of a country. From 60 to 90% of lead and cadmium, and from 75 to almost 100% of mercury emissions in the EMEP countries are transported outside country's territories and are deposited abroad. More detailed information about source-receptor relationships for the EMEP countries in 2013 is available on the MSC-E website (www.msceast.org).

Evaluation of future emission scenarios on a global scale has shown that the most significant changes in mercury pollution during the next 20 years will take place in the Northern Hemisphere and, in particular, in the largest industrial regions, where the majority of regulated emission sources are located. Considerable decrease of mercury deposition is expected in the EMEP region with slight change in relative contribution of various source regions.

MSC-E continues development of the modelling tool to improve quality and reliability of output information submitted to the EMEP countries. The Global EMEP Multi-media Modelling System, a perspective multi-scale modelling platform aimed at replacement of the previous MSC-E models for heavy metals, has been further evolved. The main directions of research and developments include improvement of the model parameterisation of atmospheric chemistry for mercury within the multi-model study of the key mercury processes, refinement of mercury interaction with atmospheric aerosol, update of the wind re-suspension scheme for heavy metals, and preparation of the model for operational calculations on the new longitude-latitude EMEP grid with fine spatial resolution.

Effects of the modelling grid refinement have been further examined in the framework of the EMEP country-specific case studies. Currently the study is focused on the assessment of lead pollution levels in Belarus. Special attention is paid to the analysis of initial data (emissions and measurements) and discrepancies between simulated and observed levels. Pollution assessment in the EECCA countries requires development of specific approaches, because of scarce background monitoring network and insufficient emission data in most of these countries. The research carried out for Belarus could serve as a test approach to evaluation of pollution levels in the other EECCA countries.

MSC-E continues cooperation with the subsidiary bodies to the Convention. In particular, in the framework of cooperation with the Task Force on Measurements and Modelling (TFMM) MSC-E elaborated a special methodology for long-term trend analysis applicable for wide range of pollutants including heavy metals (<https://wiki.met.no/emep/emep-experts/start>). Besides, information on ecosystem-specific deposition of heavy metals and their long-term changes have been provided to the Working Group on Effects (WGE) for evaluation of critical load exceedances as contribution to the WGE Assessment Report on Trends.

Information exchange with other international organizations and programmes broaden dissemination of the scientific and policy oriented information generated within EMEP and strengthen the status of the program on international level. In the framework of co-operation with the European Commission MSC-E takes part in the EU funded project GMOS (Global Mercury Observation System). MSC-E, as a leader of one of the project work packages, co-ordinates the modelling activities on a global scale as well as takes part in the regional scale model assessment. Research activities under GMOS will improve quality of model assessment of mercury pollution within the EMEP domain. Besides, the model studies under the project closely correlate with the current activities carried out within the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) and the UNEP efforts to support implementation of the Minamata Convention on Mercury. Results of the assessment were served as a basis for update of the modelling simulations in the UNEP Global Mercury Assessment 2013 and presented at the sixth session of the Intergovernmental Negotiating Committee on Mercury (INC6) (3-7 November 2014, Thailand). In addition, information about heavy metal pollution load to the Baltic Sea and its trends as well as source apportionment of heavy metal deposition in 2012 were submitted to the Baltic Marine Environment Protection Commission (HELCOM).

Future directions of MSC-E research will be focused on assessment of heavy metal pollution levels in the EMEP region and support of the EMEP countries with information required for the implementation of the Protocol on Heavy Metals. The main challenges of heavy metal pollution assessment, which need particular attention and further research in 2016-2017 have been formulated in accordance with the Key Messages of the CLRTAP Assessment Report. Evaluation of long-term trends for heavy metal pollution over the last two decades will be completed to support the CLRTAP Assessment Report. GLEMOS development will be focused on elaboration of the multi-media approach to improve knowledge on occurrence of heavy metals (particularly Hg) in relevant environmental compartments. The EMEP country-specific case studies are planned to be continued with particular attention to the EECCA countries.

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INTRODUCTION

Heavy metals are chemical elements naturally occurring in the environment. Their concentration and deposition levels have increased significantly since pre-industrial times due to anthropogenic activities causing a risk of adverse health effects in humans and wildlife. Because of their ability to dispersion in the atmosphere over long distances (up to hundreds or thousands of kilometres) heavy metals are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention). Protocol on Heavy Metals to the Convention (hereafter, Protocol on HMs or the Protocol), signed and ratified by most of the EMEP countries, the USA and Canada, is aimed at reduction of heavy metal emissions and decrease of the environmental pollution by these contaminants. Heavy metals targeted by the Protocol include lead, cadmium and mercury.

Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) provides the Executive Body for the Convention with information on effectiveness of emission control measures and on pollution changes of a range of contaminants including heavy metals. Various aspects of the assessment of heavy metal pollution in the EMEP region are covered by the EMEP Scientific Centres. Centre on Emission Inventories and Projections (CEIP) is responsible for collection, processing and quality control of data on emissions reported by countries, Chemical Co-ordinating Centre (CCC) provides methodological support of the EMEP monitoring network, Meteorological Synthesizing Centre – East (MSC-E) performs model assessment of pollution levels and transboundary transport. The work of the Centres is conducted in collaboration with the Task Force on Measurements and Modelling (TFMM), the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), Task Force on Emission Inventories and Projections (TFEIP) and the Working Group on Effects (WGE).

This report summarizes information on recent achievements of the EMEP programme in the field of heavy metal pollution assessment. It covers evaluation of pollution levels of lead, cadmium and mercury in 2013 as well as their transboundary atmospheric transport based on both modelling results and measurement information from the EMEP monitoring network. Particular attention is paid to the analysis of long-term changes of heavy metal pollution in the EMEP countries to support preparation of the CLRTAP Assessment Report. Furthermore, it describes further development of the Global EMEP Multi-media Modelling System (GLEMOS). Much attention is paid to exchange of information between MSC-E and subsidiary bodies to the Convention, as well as dissemination of output information to national and international organizations.

Anthropogenic emissions of heavy metals have been considerably reduced in many EMEP countries since the Protocol on HMs came into force. It has been followed by decline of pollution levels. Analysis of long-term changes of heavy metal pollution levels allows evaluation of effectiveness of the environmental policies implemented in the EMEP countries and, in particular, of the Protocol on HMs. A comprehensive analysis of heavy metal pollution trends in the EMEP region has been performed for the period 1990-2012 utilizing the specially developed methodology taking into account the non-linear character of heavy metal pollution reduction in Europe. Major factors affecting the long-term changes of pollution levels in the EMEP countries have been analysed.

MSC-E continues development of the modelling tool to improve quality and reliability of output information submitted to the EMEP countries. The Global EMEP Multi-media Modelling System, a perspective multi-scale modelling platform aimed at replacement of the previous MSC-E models for heavy metals, has been further developed. The main directions of research and developments include improvement of the model parameterisation of atmospheric chemistry for mercury within the multi-model study of the key mercury processes, refinement of mercury interaction with atmospheric

aerosol, update of the wind re-suspension scheme for heavy metals, and preparation of the model for operational calculations on the new longitude-latitude EMEP grid with fine spatial resolution.

Effects of the modelling grid refinement were further examined in the framework of the EMEP country-specific case studies. The studies allow taking into account particular features of a country's territory, relief, meteorological conditions, location of emission sources etc and involving national experts in more close cooperation within EMEP. Currently the study is focused on the assessment of lead pollution in Belarus. Pollution assessment in the EECCA countries requires development of specific approaches, because of scarce background monitoring network and insufficient emission data in most of these countries. The research carried out for Belarus could serve as a test approach to evaluation of pollution levels in the other EECCA countries.

Much attention was paid to cooperation with subsidiary bodies to the Convention. All results of the research and development are presented and discussed at the EMEP Task Force on Measurements and Modelling (TFMM). Besides, information on ecosystem-specific deposition of heavy metals and their long-term changes are provided to the Working Group on Effects (WGE) for evaluation of critical load exceedances. Moreover, contribution to the WGE Assessment report on trends was prepared. The information exchange with other international organizations and programmes (European Commission, the UNEP Minamata Convention on Mercury, the Arctic Monitoring and Assessment Programme, Helsinki Commission etc.) broaden dissemination of the scientific and policy oriented information generated within EMEP and strengthen the status of the program on international level. In addition to the annual Status reports a variety of information on heavy metal pollution levels in the EMEP region, individual countries, marginal seas etc. is distributed via the Internet at the MSC-E website (www.msceast.org).

Finally, the main challenges of heavy metal pollution assessment, which need particular attention and further research in 2016-2017 are summarized in the report in accordance with the Key Messages of the CLRTAP Assessment Report. More detailed scientific information is presented in Annexes and in the MSC-E Technical Report 1/2015 [*Shatalov et al.*, 2015].

1. POLLUTION OF THE EMEP REGION

1.1. Emission data for model assessment

Emission data for assessment of trends

This year the assessment of long-term trends of heavy metal pollution has been performed for lead, cadmium and mercury for the period 1990-2012 to characterize contemporary levels of pollution and their long-term changes in the EMEP countries in comparison to the base year.

The data on heavy metal emissions for modelling within the EMEP domain for the period 1990-2012 were based on emission data submitted in 2014 and generated by CEIP and MSC-E. CEIP provided datasets of gridded emissions for 2012. MSC-E compiled gridded emission data for the other years using official emissions reported by countries, emission expert estimates made by TNO [Berdowski *et al.* 1997; Denier van der Gon *et al.*, 2005] and expert estimates of emissions for the EECCA countries prepared by MSC-E.

Emissions of heavy metals have considerably decreased in the EMEP region over the last two decades. Lead emissions have dropped by 90% since 1990, whereas emissions of cadmium and mercury have decreased approximately by 60% (Fig. 1.1).

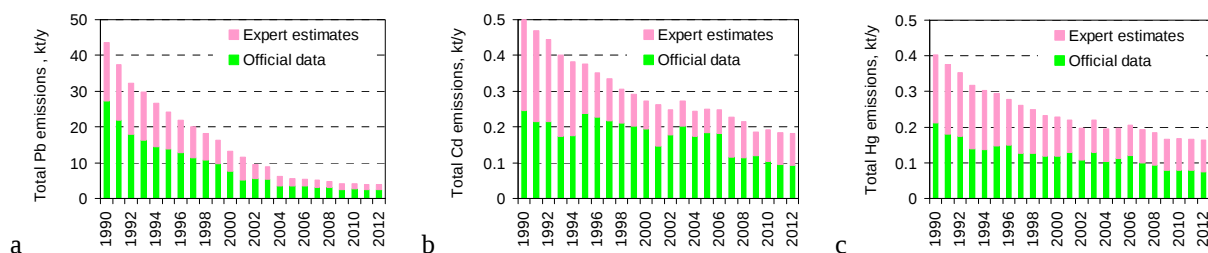


Fig. 1.1. Temporal changes of lead (a), cadmium (b), and mercury (c) anthropogenic emissions in the EMEP region from 1990 to 2012

In 2012 total anthropogenic emission of lead from the EMEP domain made up around 3895 tonnes (Fig. 1.1a). Lead emissions decreased by 39735 tonnes over the period 1990-2012 that corresponded to around 90% of total emission in 1990. In general, the emission reductions between 1990 and 2012 were significant for all countries, except for Azerbaijan, Georgia, Armenia and Malta, where emissions increased (Fig. 1.2). The most noticeable emission decreases (more than 95%) were in the Russian Federation, Monaco, Montenegro, Republic of Moldova, the United Kingdom, Luxembourg, Norway, France, Iceland, Lithuania, Sweden, Croatia, Latvia, Ukraine and Liechtenstein.

Total emission of cadmium in the EMEP domain in 2012 was 182 tonnes, which is 320 tonnes (65%) lower than in 1990 (Fig. 1.1b). The highest emission reductions (around 90%) were reported by Republic of Moldova, Malta, Ukraine, Luxembourg, Monaco, the United Kingdom and Romania (Fig. 1.3). Increases in emissions indicated in Cyprus (62%), Belarus (36%), Latvia (35%), Georgia (32%), Azerbaijan (25%), Liechtenstein (16%) and Turkey (9%).

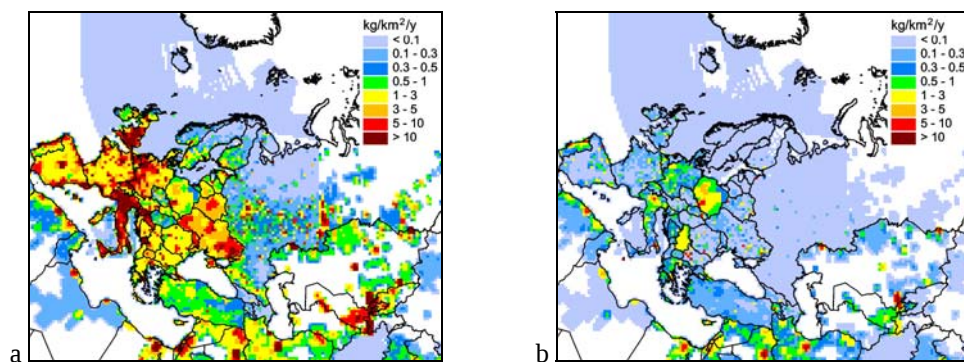


Fig. 1.2. Spatial distribution of lead anthropogenic emissions over the EMEP domain in 1990 (a) and 2012 (b).

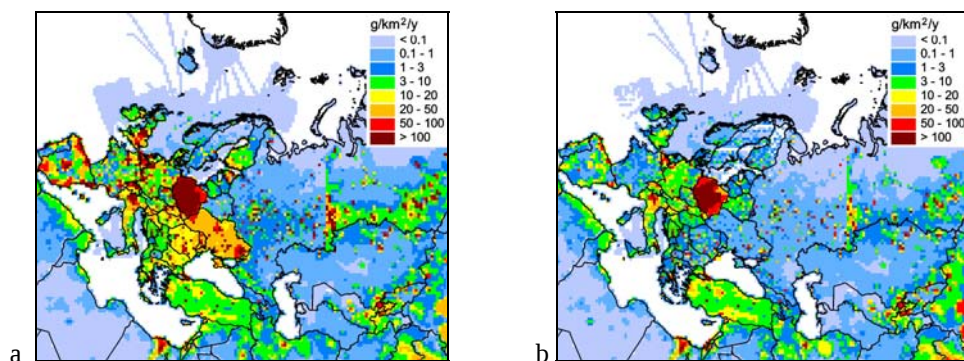


Fig. 1.3. Spatial distribution of cadmium anthropogenic emissions over the EMEP domain in 1990 (a) and 2012 (b).

Emissions of mercury in the EMEP domain decreased from 402 tonnes in 1990 to 164 tonnes in 2012, with reduction of 60% (Fig. 1.1c). Compared with 1990, mercury emissions declined in 2012 in 39 countries and increased in 6 countries (Fig. 1.4). The most substantial decreases of mercury emission were reported by Malta (99%), Republic of Moldova (97%), the Russian Federation (94%), Slovakia (90%) and Denmark (90%). Higher levels of emissions in 2012 (compared to 1990) were noticed in Iceland (119%), Liechtenstein (40%), Turkey (27%), Georgia (25%), Azerbaijan (23%) and Montenegro (19%).

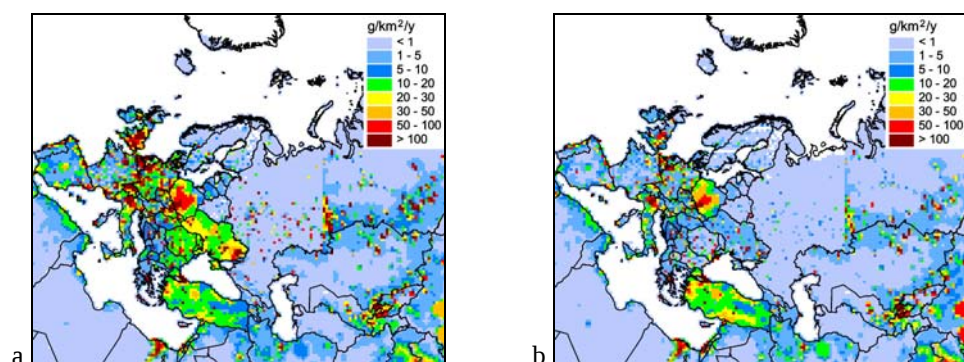


Fig. 1.4. Spatial distribution of mercury anthropogenic emissions over the EMEP domain in 1990 (a) and 2012 (b).

Lead emissions in the EMEP region have dropped by 90% between 1990 and 2012, whereas emissions of cadmium and mercury have decreased approximately by 60%. In particular countries changes in the emission data could differ significantly from the values of reduction for the EMEP region as a whole.

Emission data in 2013 for model assessment

In 2015, emissions of heavy metals, at least for one year of the period 1990-2013 were reported by 43 countries. Azerbaijan and Georgia submitted emission data for the first time. Data for the base year 1990 and the year 2013 were provided by 30 countries. 30 countries submitted gridded emissions of lead, cadmium and mercury with spatial resolution 50x50 km². It should be noted that the completeness and consistency of gridded data have improved, but still remain insufficient [Mareckova *et al.*, 2014]. The data on heavy metal emissions for modelling within the EMEP domain for 2013 were provided by CEIP.

The total anthropogenic heavy metal emissions in the EMEP countries in 2013 were lower than the corresponding emissions for 2012 in previous submission and amounted to 3427 tonnes for lead, 170 tonnes for cadmium and 159 tonnes for mercury.

Along with the reduction of emissions in most countries, changes of emission data in countries are associated with re-calculations of previously reported values. The aim of recalculations is to ensure consistency of the time series and thus improve the accuracy and completeness of the emission inventory.

30 countries recalculated their heavy metal emission data for the full time series in 2015. It was found that recalculations (differences between emissions reported in 2015 and in previous years) in half of them were larger than $\pm 10\%$. Significant differences were most frequently observed for Cd. Extreme differences were noted for Denmark (Cd 2010), Hungary (Cd 2012 and Hg 2011) and Republic of Moldova (Pb 2000, 2001, 2004, 2005, 2007 to 2011). An overview of large recalculations ($>\pm 10\%$) to the inventories of lead, cadmium and mercury for the period from 1990 to 2012 is summarized in Table A1 of Annex A. The explanatory information that describes main reasons of recalculations can be found in Informative Inventory Reports of countries (http://www.ceip.at/ms/ceip_home1/ceip_home/status_reporting/2015_submissions/).

Mercury emissions on a global scale

Mercury is well known as a global 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 inventories of mercury anthropogenic emissions to the atmosphere were produced for the years 1990, 1995, 2000 and 2005 [Pacyna and Pacyna, 2002; Pacyna *et al.*, 2006, 2009; AMAP/UNEP, 2008]. An inventory of the global anthropogenic Hg emissions for 2010 was prepared as a part of the UNEP Global Mercury Assessment 2013 [AMAP/UNEP, 2013].

Comparison of total anthropogenic mercury emissions in different regions and for different reference years is presented in Fig. 1.5. As seen the trend of total mercury emissions in East Asia was higher than that in the other regions. During the period 1990-2010 Hg emissions decreased in Europe, the EECCA countries and North America but significantly increased in Africa. Some increase also took place in South East Asia, Mexico & Central America and South America. However, it should be noted that the emissions inventory for 2010 differs from those for other years in terms of applied methodology. Therefore, the mercury emissions in 2010 are not directly comparable to the previous years [AMAP/UNEP, 2013].

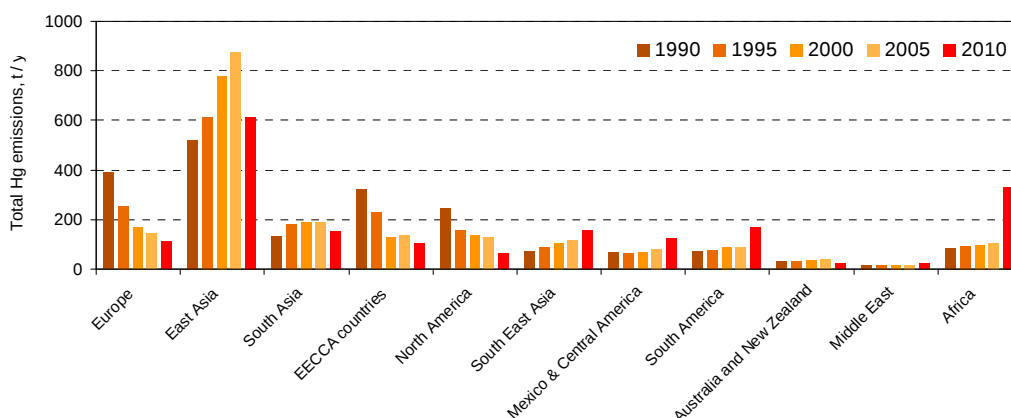


Fig. 1.5. Changes of global anthropogenic emissions of total mercury to the atmosphere from 1990-2010

1.2. EMEP monitoring network of heavy metals

Measurement network

In 2013, there were 31 sites measuring heavy metals in both air and precipitation, and altogether there were 65 measurement sites. 12 of these sites were measuring mercury in both air and precipitation. In total, 7 Parties to the Convention fulfil their monitoring obligations as defined in the EMEP monitoring strategy [UNECE, 2009] with at least one level 2 site with both air and precipitation measurements of heavy metals and mercury in air and precipitation. A number of countries have been reporting heavy metals within the EMEP area in connection with different national and international programmes such as HELCOM, AMAP and OSPARCOM.

Detailed information about the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [Aas and Bohlin-Nizzetto, 2015]. All the data presented here are available at the EMEP database (<http://ebas.nilu.no>).

Observed concentration level of Pb, Cd and Hg in 2013

Annual averages of Pb, Cd and Hg concentrations in precipitation and in air in 2013 are presented in Fig. 1.6-1.11. Note that Cyprus with measurements of heavy metals in air is outside the map domain so included as a dislocated point south of Turkey. The lowest concentrations for all elements 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 "hotspots" for some elements, i.e. in Hungary and the BeNeLux countries for lead and cadmium in air. 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 here. The spatial distribution of elemental mercury in air does not follow a general pattern; though somewhat elevated level in central Europe. In precipitation there are several sites (in PT, LV, IE) 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].

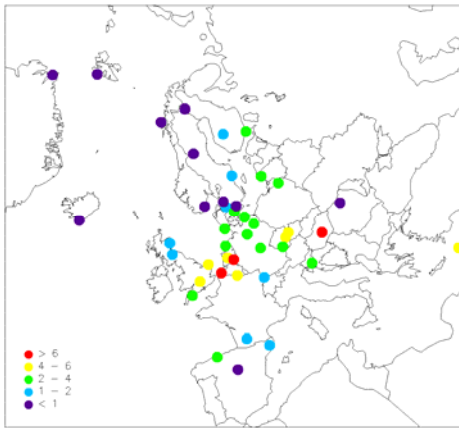


Fig. 1.6. Pb in aerosol, ng/m³

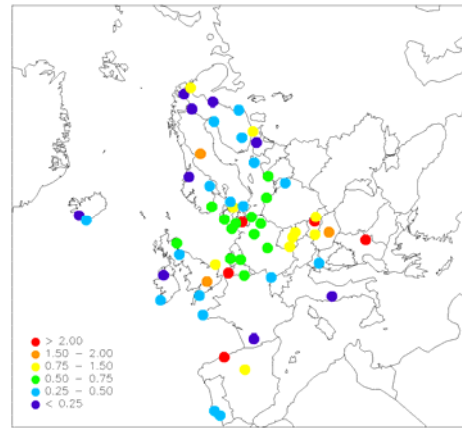


Fig. 1.7 Pb in precipitation, µg/L

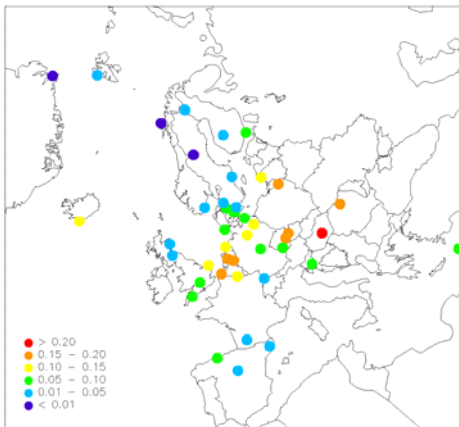


Fig. 1.8. Cd in aerosol, ng/m³

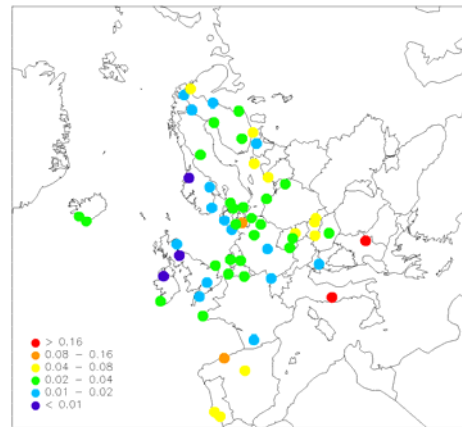


Fig. 1.9. Cd in precipitation, µg/L

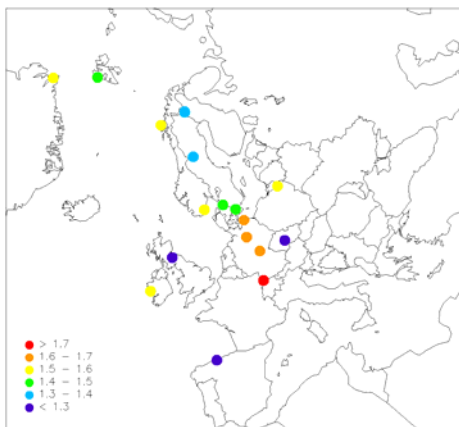


Fig. 1.10. Hg (g) in air, ng/m³

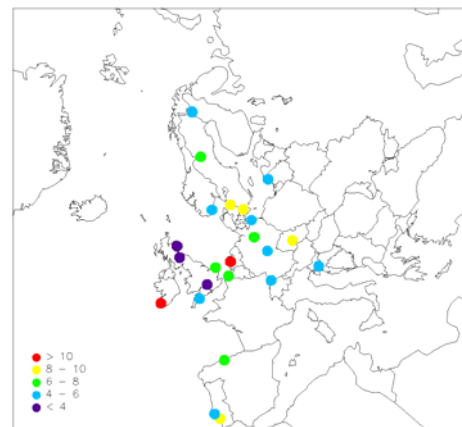


Fig 1.11. Hg in precipitation, ng/L

Trends in observed concentrations of Pb, Cd and Hg

Heavy metals were included in EMEP's monitoring program in 1999. However, earlier data are available and have been included in the EMEP database. Thus, the database includes measurements back to 1976 for a few sites, although most of the time series start around 1990. The Aarhus Treaty, signed in 1998 by nearly all European countries, targets Cd, Pb and Hg and committed the Parties to reduce their emissions for these three metals compared to 1990. An overview with trend analysis of

these element was done by *Tørseth et al.* [2012], and a trend assessment is being prepared by the EMEP TFMM, which is to be published during spring 2016. Here we only show an overview of the observed concentrations of Pb, Cd and Hg at selected sites with long term measurements in air and precipitation (Figs. 1.12-1.16). There are clear and significant reductions in lead and cadmium concentration in both compartments and most sites. For mercury there was a decrease in the beginning of the nineties, but in the last decade there is no clear change in the observed level.

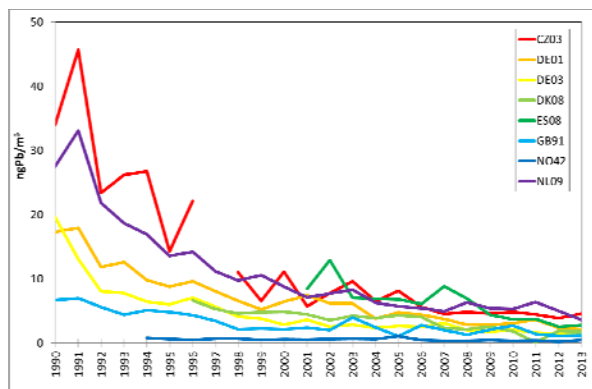


Fig. 1.12. Pb in aerosol, ng/m³

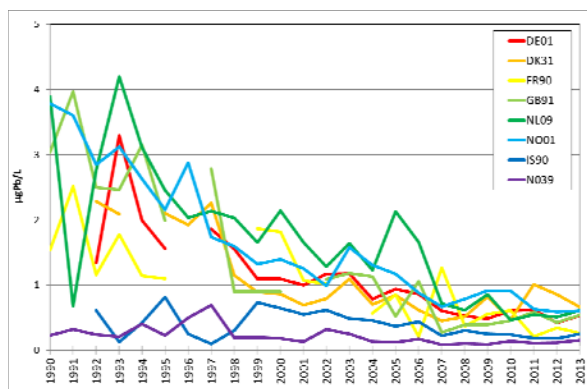


Fig. 1.13. Pb in precipitation, µg/L

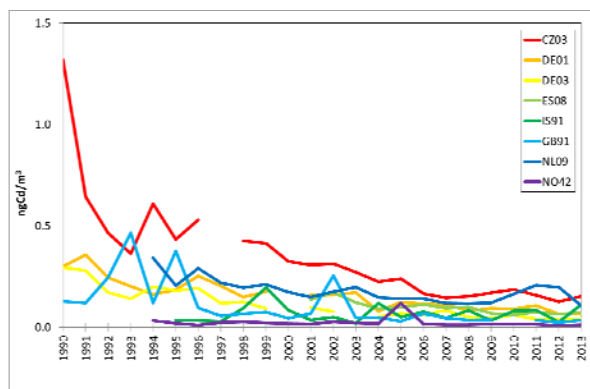


Fig 1.14. Cd in aerosol, ng/m³

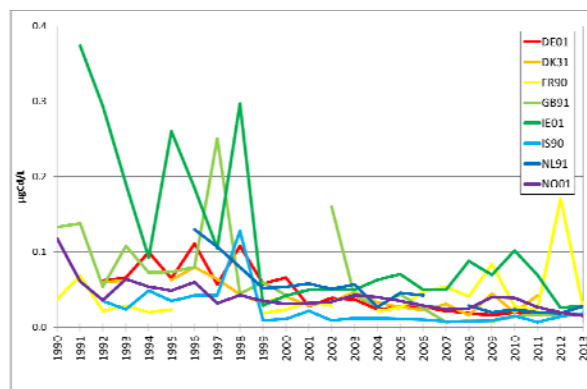


Fig. 1.15. Cd in precipitation, µg/L

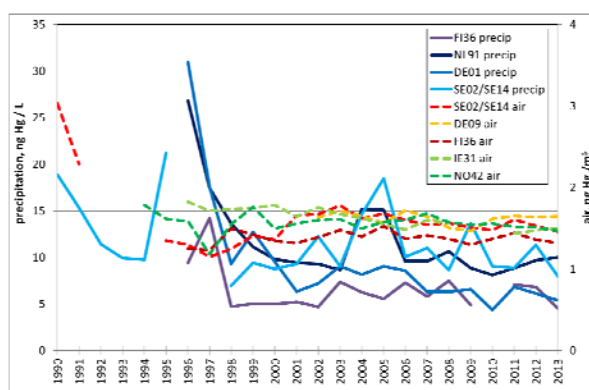


Fig. 1.16. Hg in air, ng/m³ and precipitation, ng/L

1.3. Pollution levels and transboundary fluxes in 2013

Heavy metal pollution levels in 2013

Modelling information about pollution levels of lead, cadmium and mercury in the EMEP region and in the individual countries in 2013 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.

Heavy metal pollution levels in the EMEP region were determined by three groups of emission sources (EMEP anthropogenic sources, secondary sources within the EMEP region and non-EMEP sources) and by peculiarities of meteorological conditions. The areas with relatively high air concentrations and deposition of lead and cadmium in 2013 were noted in the north-western part of Germany and the Benelux region, in the southern part of Poland, in the northern part of Italy and the Balkans (Fig. 1.17). Annual mean air concentrations in these regions ranged from 10 to 20 ng/m³ (lead) and from 0.2 to 0.6 ng/m³ (cadmium) or even exceeded these values. Total deposition fluxes in these regions exceeded 1.5 kg/km²/y for lead and 50 g/km²/y for cadmium. Comparatively high pollution levels were explained by location of major anthropogenic and secondary emission sources in the considered regions. Significant pollution levels of lead and cadmium in the southern part of Russia were mostly caused by influence of secondary sources, because national emissions in this part of the country were comparatively low (Fig. 1.2). Besides, relatively high concentrations of lead and cadmium in air were noted for the arid areas of Kazakhstan, Uzbekistan and Turkmenistan. These levels were caused by combined effect of anthropogenic and secondary sources and low precipitation amounts.

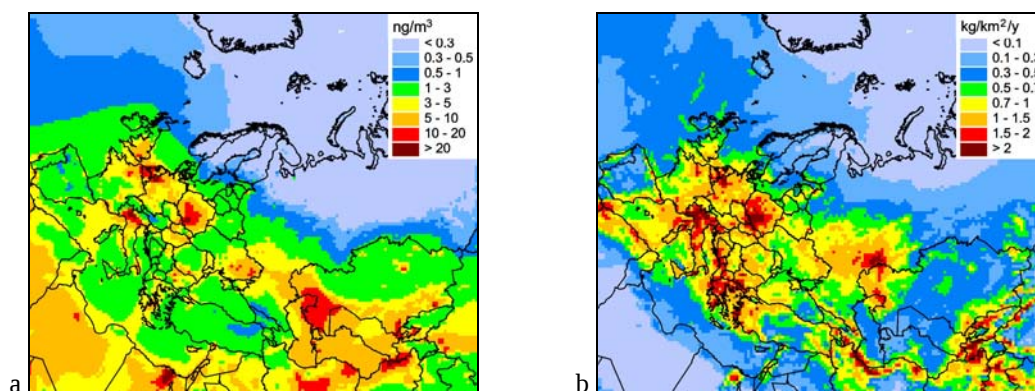


Fig. 1.17. Annual mean concentrations in air (a) and total deposition (b) of lead in 2013

Similar to lead and cadmium, areas of relatively high deposition of mercury were in Germany, the Benelux region, the southern part of Poland, the northern part of Italy and the Balkans. Mercury deposition fluxes in these regions ranged from 15 to 30 g/km²/y, which was explained mostly by location of anthropogenic emission sources. Unlike lead and cadmium, spatial distribution of mercury annual mean air concentrations was much more uniform. In most of polluted areas, associated with significant emission sources (north-western Germany, northern Italy, north-western Turkey, southern Poland), the levels were 1.8-2 ng/m³. Besides, somewhat higher levels, exceeding 2.2 ng/m³, took place in the southern part of Kazakhstan, Uzbekistan and Tajikistan. In Scandinavia, the northern part of Russia, parts of the Arctic concentrations were 1.4-1.5 ng/m³. Low spatial gradients of mercury air concentrations were explained by serious influence of intercontinental transport caused by long residence time of mercury in the atmosphere.

Country-averaged contributions of different emission sources (anthropogenic, secondary and non-EMEP) to deposition in the EMEP countries were determined. Due to marked reduction of emissions in the EMEP countries for the two last decades deposition from anthropogenic sources strongly declined. In 2013 significant contribution to the pollution levels of cadmium as well as of lead was made by secondary emission sources. Their contribution to total deposition of cadmium exceeded 50% in 45 countries (Fig. 1.18), and of lead – in 37 countries. Contribution of non-EMEP sources to levels of lead and cadmium was relatively low, lying below 10% in most of the countries. Significant contribution (20-70%) of non-EMEP sources was noted only for the EMEP countries situated in the vicinity of boundaries of the EMEP domain, such as Tajikistan, Uzbekistan, Armenia etc.

In case of mercury major contribution to deposition in the EMEP countries was done by non-EMEP emission sources (both anthropogenic and secondary). In 44 countries the contribution of global sources exceeded 50%, reaching 90% or more in Iceland, Norway, Russia and the other countries located close to the boundaries of the EMEP region. Deposition fluxes caused by secondary sources within EMEP were low because these sources were presented by long-lived elemental form of mercury which readily left the EMEP region and entered global pool of atmospheric mercury. Deposition from the EMEP anthropogenic sources of mercury was caused mostly by oxidized mercury forms easily removed from the atmosphere.

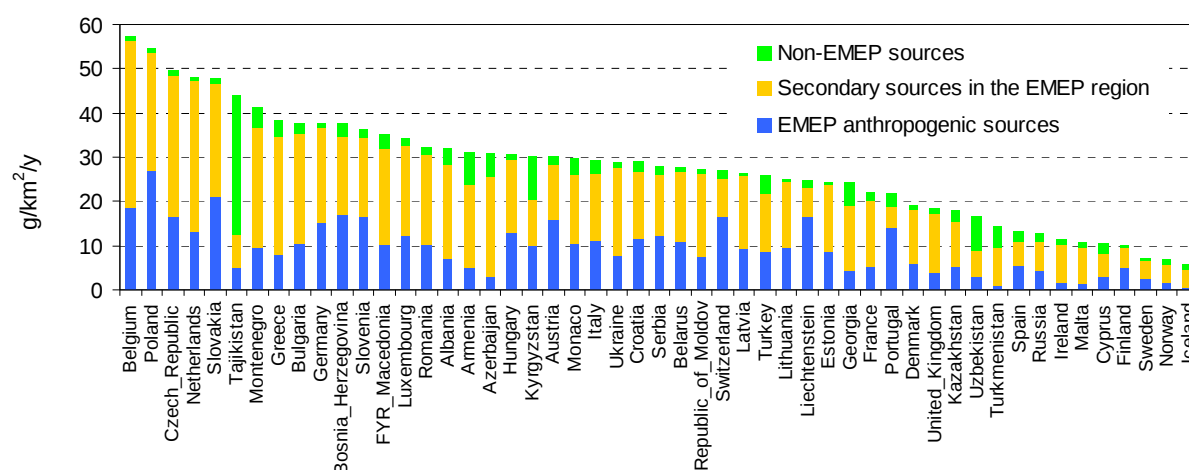


Fig. 1.18. Country-averaged deposition fluxes of cadmium from the European and Central Asian anthropogenic, secondary and non-EMEP emission sources in 2013

Changes of lead, cadmium and mercury pollution levels between 2012 and 2013 were overviewed. These changes were explained by differences in emission data used in calculations and by variability of meteorological conditions. In most of the countries the change between country-averaged deposition of lead in 2012 and 2013 was within $\pm 20\%$. Increase of lead deposition by 25-40% was noted for some Baltic counties (Latvia and Estonia), south-eastern Europe (Montenegro and the FYR of Macedonia), Ireland, the United Kingdom and the Czech Republic, mainly because of higher contribution of secondary sources compared to 2012. In Bulgaria, Cyprus, Malta, Serbia, Portugal and the Republic of Moldova deposition reduced by 20-45% mostly due to decrease of emission data used in calculations compared to 2012.

In general, similar tendencies were noted for cadmium. Besides, deposition of cadmium in Portugal increased because of rise of national emissions. Polish emissions demonstrated almost two-fold decline because of re-calculations of national emissions, which resulted to decrease of deposition by 30%.

Deposition changes of mercury between 2012 and 2013 were mostly within $\pm 25\%$. These changes could be attributed to variability of meteorological conditions and changes of emission values used in modelling. Marked changes were noted for the FYR Macedonia and Albania, where mercury deposition in 2013 was higher than those in 2012 by almost 100% and 25%, respectively. The main reason for this was changes of spatial distribution of emissions. Modelling for 2013 was based on spatial distribution developed by CEIP, while for 2012 the distribution of TNO [Denier van der Gon *et al.*, 2005] was applied.

Transboundary transport in 2013

Deposition from anthropogenic sources to each EMEP country was split in two parts: national (from national emission sources) and foreign (from the EMEP sources located outside a country's territory). Contribution of foreign sources to cadmium, lead and mercury anthropogenic deposition exceeded 50% in 19, 24 and 17 countries, respectively (Fig. 1.19). The highest contribution of foreign sources was found for countries which national emissions were low and territory was not large compared to other countries, for example, Lichtenstein, Montenegro, Albania, Georgia. The lowest contribution of foreign sources (and, hence, the highest contribution of national sources) was indicated for countries with large territory and considerable emissions (Russia, Kazakhstan, Germany, Poland). Besides, this statement is also true for countries located close to boundaries of the EMEP region, e.g., Portugal, Turkey, Spain. It should be noted that ratios between contributions from national and foreign sources in particular grid cells could markedly differ from the values for a country as a whole. As a rule, contribution of foreign sources near state borders is higher, and in the central parts of a country – lower than the country-averaged value. Information about spatial distribution of contribution of foreign sources to pollution levels for each EMEP country in 2013 is available in the Internet at MSC-E website (<http://www.msceast.org/index.php/emep-countries>).

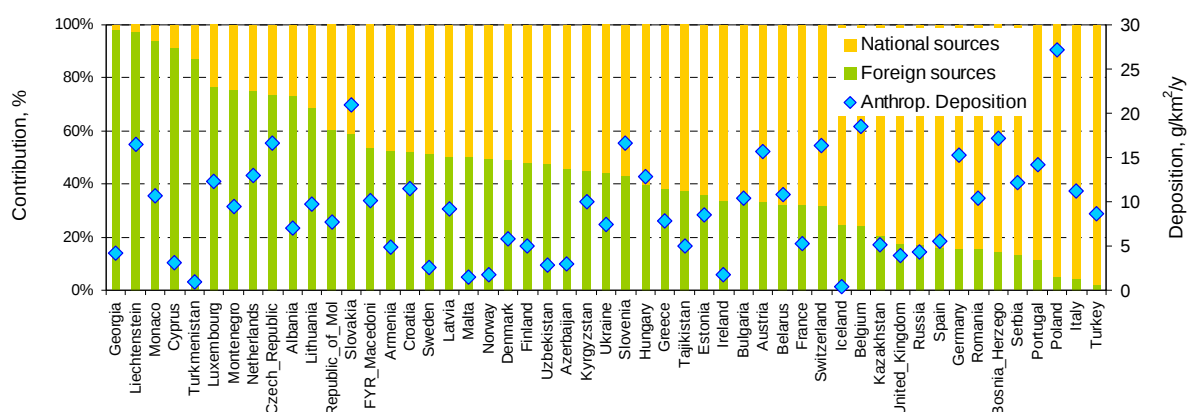


Fig. 1.19. Relative contribution of the transboundary transport and national sources to anthropogenic cadmium deposition in the European and the Central Asian countries and deposition values from anthropogenic emission sources in 2013

Heavy metals, emitted from sources of a country, partly deposited within the country's territory and partly got involved into transboundary transport, depositing to the other EMEP countries or elsewhere. Amount of the metals entering transboundary transport could be expressed in absolute and relative (fraction of emission) terms (Fig. 1.20). For example, about 710 tonnes of lead emitted in Kazakhstan in 2013 were distributed between deposition to own territory (270 tonnes), deposition to the other EMEP countries (around 275 tonnes) and deposited elsewhere (to other regions within the EMEP

domain or transported outside the domain) (about 165 tonnes). Thus, around 440 tonnes (62% of national emissions) were involved into transboundary transport.

As a rule, the major absolute contributors of heavy metals to transboundary transport were countries with significant national emissions. The highest contributors to transboundary transport of lead were Kazakhstan and Poland (440 and 320 tonnes, respectively). Kazakhstan and Turkey were the largest contributors of transboundary transport of cadmium (14 and 12 tonnes, respectively), and Turkey and Kazakhstan – of mercury (20 and 17 tonnes).

Heavy metal mass emitted by national sources and transported outside country's borders ranged from 60 to 90% for lead and cadmium, respectively, in most of the countries (Fig. 1.20). Russia was the exception: because of vast territory most of emitted lead and cadmium were deposited within the country, and relatively small fraction (25% for Pb and 21% for Cd) was transported outside. In case of mercury the fraction ranged from 75 to almost 100%.

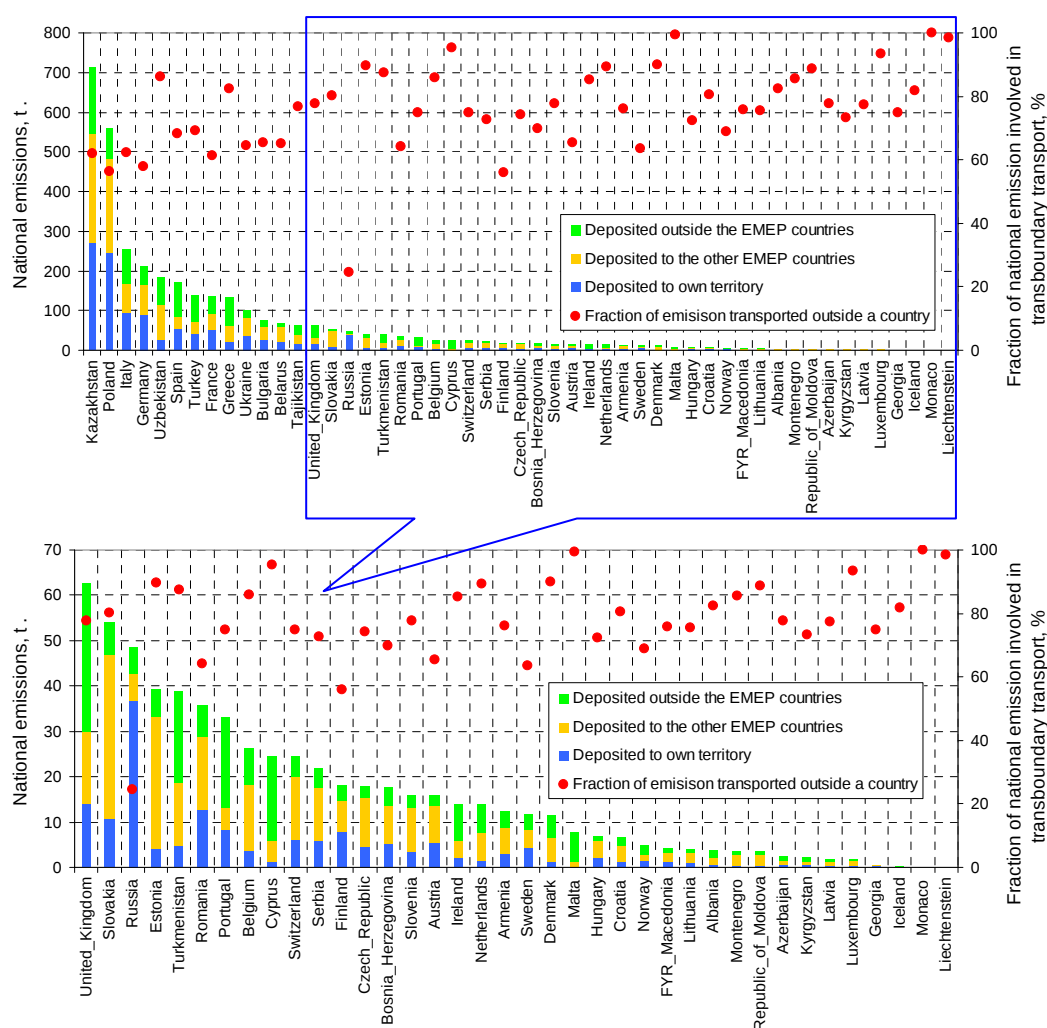


Fig. 1.20. Distribution of lead emitted in the EMEP countries between deposition to own territory, deposition to the other EMEP countries and deposition outside the EMEP countries in 2013. Red dots indicate relative fraction of national emissions involved into the transboundary pollution

Quality of the model assessment

Modelling is capable of producing information about pollution levels in each 50-km grid cell of the EMEP domain at any required temporal resolution. However, it is important to verify the modelling results via comparison of modelled values with the values observed at monitoring stations.

All components of the assessment of pollution levels, such as emissions, model parameterizations, and measurements are subject to various uncertainties. Therefore, modelled values of pollution levels can hardly fully coincide with the observed levels. However, if the difference between modelled and observed levels at a station is significant, e.g., exceeds factor of 2 or 3, analysis of these deviations is required. This section gives an overview of the results of the comparison of modelled air concentrations and wet deposition fluxes of lead, cadmium and mercury with the levels observed at the EMEP monitoring stations. More detailed information about comparison of modelled and measured values can be found in the MSC-E Technical Report [Shatalov *et al.*, 2015]. Agreement between modelled and measured values over long period could be characterized via statistical indices, such as mean relative bias (MRB), Pearson's correlation coefficient and normalized root mean square error (NRMSE). The indices are summarized in Table 1.1.

Table 1.1. Main statistical indicators of agreement between annual modelled and measured levels of air concentrations and wet deposition fluxes in 2013

	Lead		Cadmium		Mercury	
	C _{air}	Wet Dep	C _{air}	Wet Dep	C _{air}	Wet Dep
Relative bias, %	21.9	24.6	-4.7	-31.4	0.0	69.7
Correlation coefficient	0.86	0.45	0.57	0.65	0.63	0.92
NRMSE	0.81	0.58	1.00	0.51	0.05	0.74
F2, %	69	79	62	84	100	62
F3, %	86	100	76	100	100	92

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

Lead levels in 2013 were somewhat (by 20-25%) overestimated by the model. Spatial distribution of concentrations in air was reproduced relatively well (R_{cor} is equal to 0.86). For stations located in Germany, Poland, the Czech Republic, Denmark, Hungary, Slovenia and for most of Spanish sites the modelled concentrations agreed well with observed levels of air concentrations – the relative bias was within $\pm 25\%$ (Fig.1.21). Also relatively favourable agreement was noted for wet deposition fluxes at stations in Latvia, Denmark, Iceland, Slovakia and at the Polish station PL4. Overestimation for concentrations in air by 50-100% was noted for stations in the Netherlands, Belgium and France, and that for wet deposition fluxes – in Germany, France, Estonia and Slovenia. Most probable reason for it was overestimated contribution of secondary sources to air concentrations. For Finnish, one Norwegian (NO42) and Swedish (SE5) stations the model underestimated the observed concentrations in air.

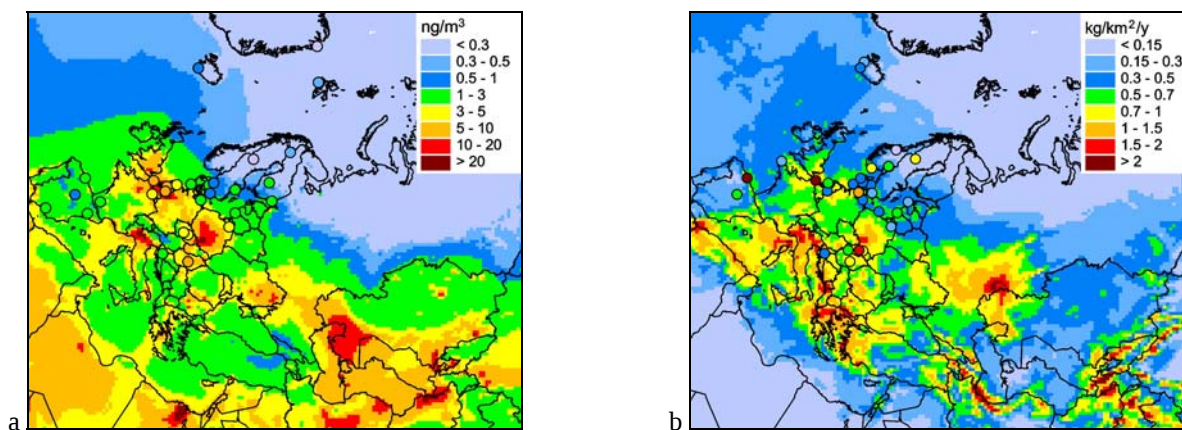


Fig. 1.21. Modelled and observed air concentrations of lead air concentrations (a) and wet deposition (b) in 2013

Modelled and observed cadmium concentrations in air agreed reasonably well with measurements on average: the mean relative bias is -5%. At stations located in Germany, Spain, Hungary and the Czech Republic (Fig. 1.22) the bias between modelled and observed values was within $\pm 50\%$. Modelled wet deposition fluxes matched the observed fluxes in Denmark, the Netherlands, Poland, Slovakia, Sweden and at most of Norwegian stations. Underestimation of 50% or more of the observed air concentrations was indicated at stations in Finland, Iceland and Latvia. Three-fold underestimation at the Polish station PL4 might be explained by uncertainties of measurement data, because concentrations observed at this station in 2013 were 1.5 – 3.5 times higher than those measured in several previous years. Significant overestimation of observed air concentrations (60-200%) in Belgium and the Netherlands and of wet deposition (60-80%) in Germany was explained likely by overestimated role of secondary sources in these countries. At Spanish stations the model underestimates observations by about 5 times. Most likely, it is connected with measurement uncertainties or influence of unaccounted local sources. These measurements were not included in the statistical indices in Table 1.1.

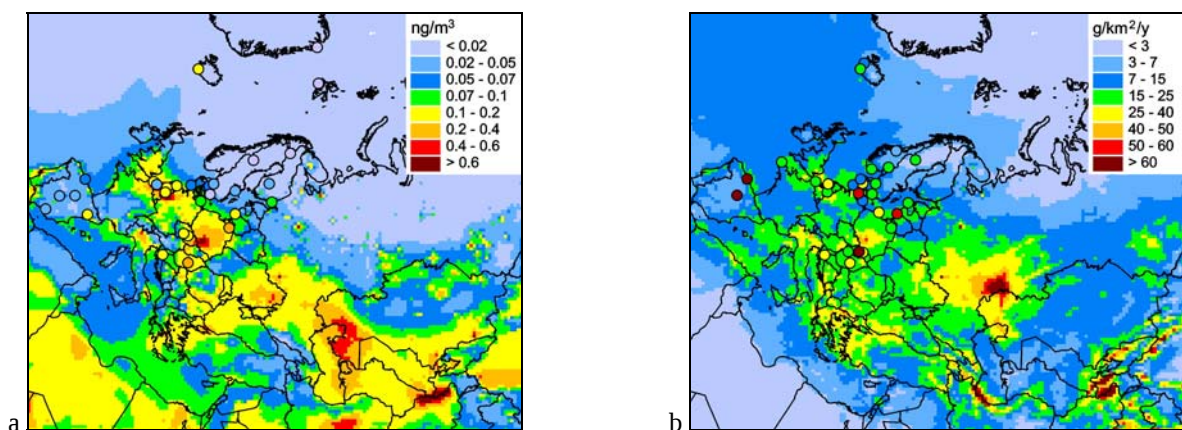


Fig. 1.22. Modelled and observed air concentrations of cadmium air concentrations (a) and wet deposition (b) in 2013

Both modelled and observed air concentrations of mercury were characterized by low spatial variability caused by long (about a year) atmospheric life time. The model adequately reproduced the levels of mercury air concentrations in the EMEP region: mean relative bias was zero, and for individual stations it was below 10% (Fig. 1.23). The exception was station GB48 (Auchencorth Moss,

the United Kingdom), where the model overestimated observed concentration by 65%. However, the observed value at this station seemed very low (0.9 ng/m^3) compared to levels typically measured at background stations of the Northern Hemisphere ($1.4 - 1.5 \text{ ng/m}^3$). Wet deposition of mercury was overestimated by the model by 70% on average (Table 1.1). At stations BE14, ES8, NL91 and SE5 the bias was around 40% (Fig. 1.23). For the other stations the overestimation was higher (60-150%). The main reason for this overestimation was uncertainty of atmospheric chemistry of mercury. Another factor of uncertainty was information on speciation of mercury in emissions, which was not available in official reporting. Current research of mercury processes in the atmosphere (Sections 3.1, 3.2) is aimed to improve the model performance.

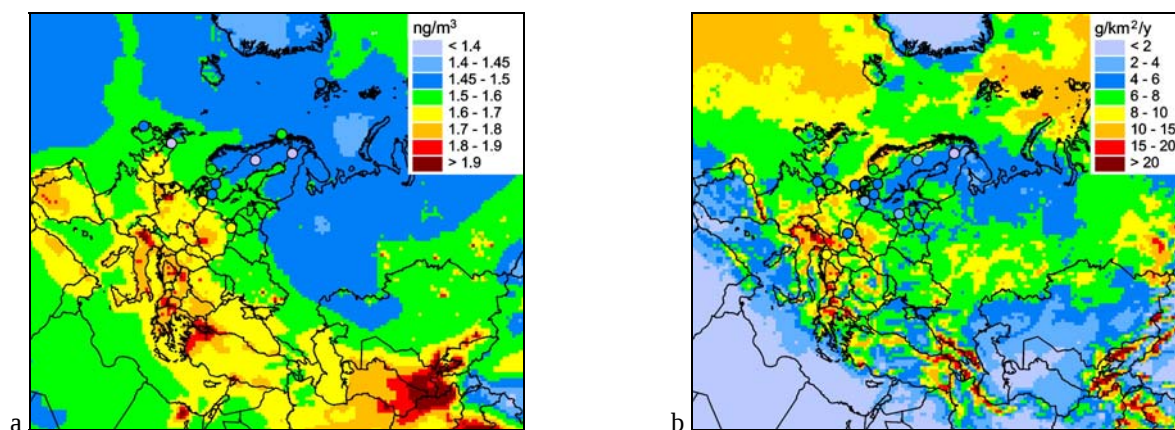


Fig. 1.23. Modelled and observed air concentrations of mercury air concentrations (a) and wet deposition (b) in 2013

Additional information on original modelling data on spatial distribution of concentrations and deposition of lead, cadmium and mercury in the EMEP region, country-specific information about pollution levels and source-receptor relationships is available in the Internet at the MSC-E website <http://www.msceast.org/>.

1.4. Ecosystem-dependent deposition

Heavy metals deposited from the atmosphere to the underlying surface can accumulate in soils and waters. When certain levels (so-called critical loads) of heavy metals are exceeded, harmful effects on human health and biota could be exerted. Evaluation of the effects caused by elevated levels of heavy metals is carried out by the Coordination Centre for Effects (CCE) of the Working Group on Effects (WGE). Necessary input information for this work is data on ecosystem-dependent deposition. MSC-E every year performs calculations of deposition of lead, cadmium and mercury to different types of the underlying surfaces. Information on ecosystem-dependent deposition in the EMEP region and in particular countries is available in the Internet (www.msceast.org).

Total deposition fluxes to various types of underlying surfaces are different depending on type of the surface. For example, deposition fluxes of lead to forested areas (Fig. 1.24a) are much higher than the fluxes to low-vegetation natural surfaces such as grasslands, wetlands etc (Fig. 1.24b). Similar is indicated for other heavy metals.

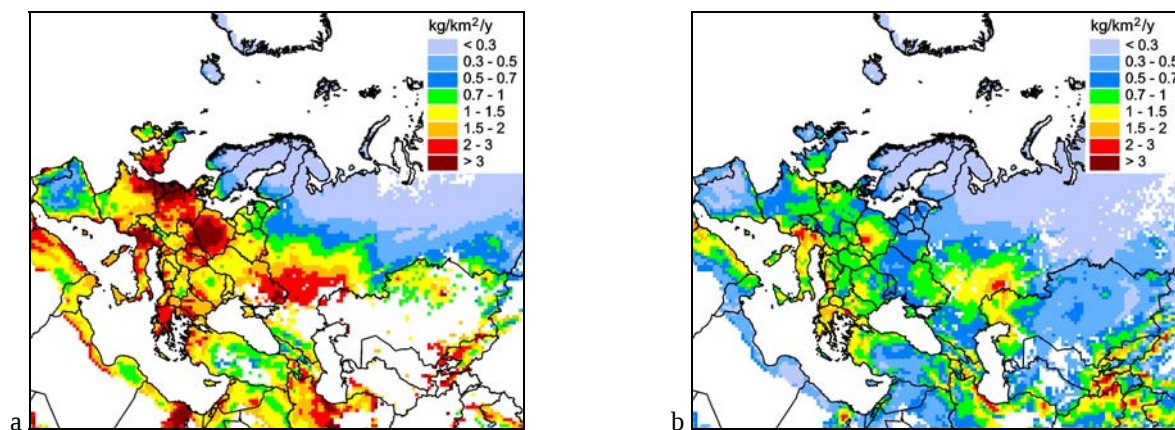


Fig. 1.24. Deposition of lead to forests (a) and non-forested natural surfaces (b) in 2013

Spatial distribution of ecosystem-dependent deposition fluxes was analyzed using frequency distribution histograms. In Figure 1.25 these histograms were depicted for lead, cadmium and mercury deposition to forests and to non-forested natural surfaces (grasslands, marshes, tundra etc.) in the EMEP region. Frequency distributions demonstrate that forests are characterized by relatively high deposition fluxes, compared to the other surfaces. For example, over 7% of grid cells deposition of lead to forests exceed 2.5 kg/km²/y (Fig. 1.25a), while for non-forested natural surfaces it takes place only over 0.2% of grid cells. Similar histograms could be produced for each type of ecosystem and for particular countries.

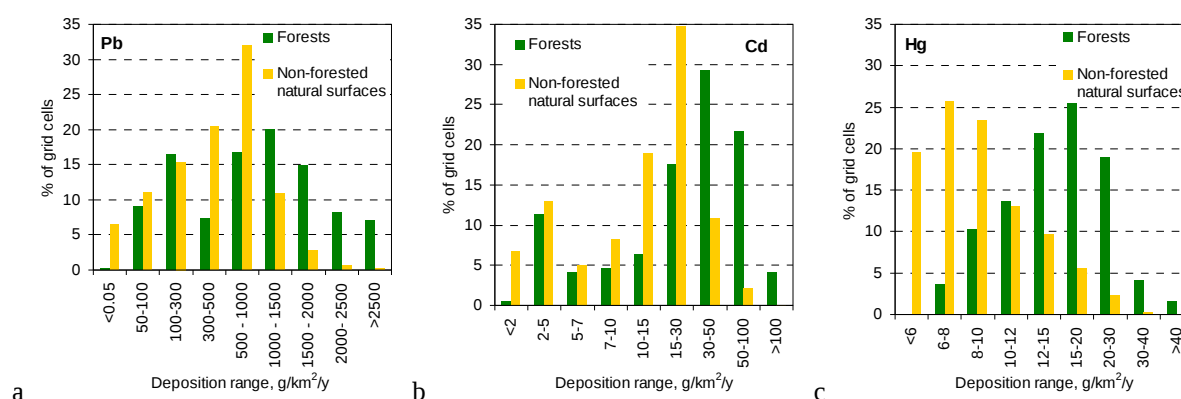


Fig. 1.25. Frequency distribution histograms of lead (a), cadmium (b) and mercury (c) deposition to forests and non-forested natural surfaces in 2013

On the base of the information on ecosystem-dependent deposition CCE calculated exceedances of heavy metal critical loads for different years of the period of 1990-2010. Both area where exceedances took place, and mean magnitude of exceedances of all three metals tended to decline (Fig. 1.26).

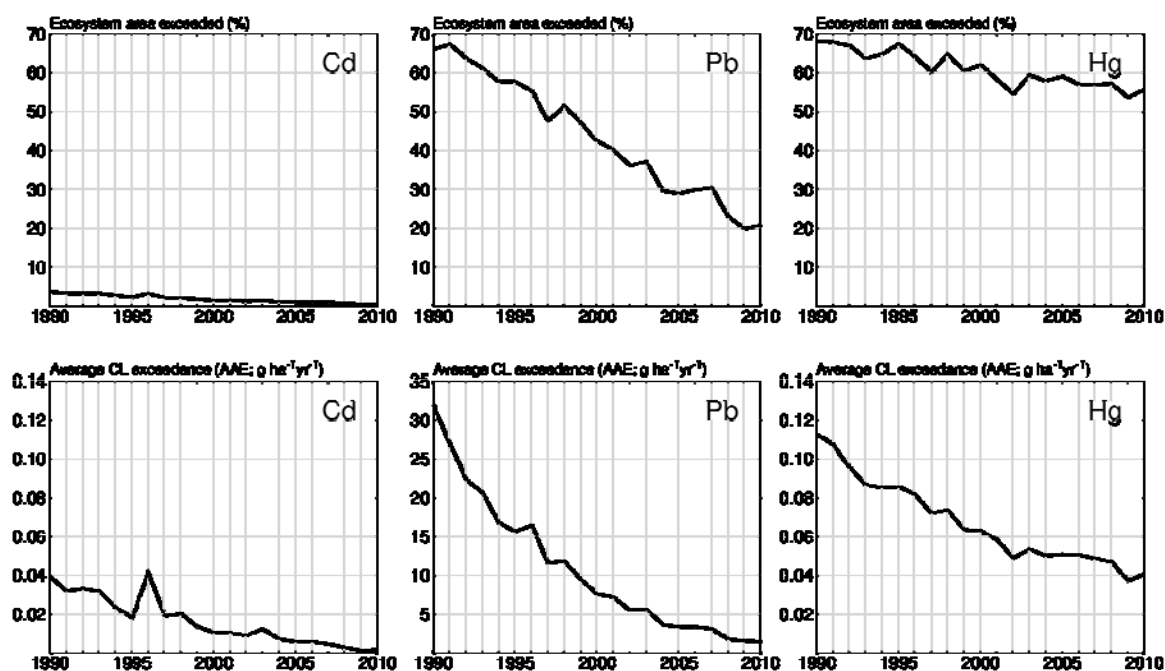


Fig. 1.26. Fraction of area where deposition exceeded critical loads (top) and average exceedance (lower) of cadmium (left), lead (middle) and mercury (right). The figure was kindly provided by CCE.

Following the reduction of heavy metal depositions, exceedances of lead, cadmium and mercury, as well as area over which exceedances took place, markedly declined in the EMEP region for the period 1990-2010.

1.5. Future changes of mercury pollution: A global perspective

Assessment of future levels of Hg pollution and evaluation of different abatement scenarios are the key information for development of future mitigation measures. Mercury differs from other heavy metals by its ability to long-range dispersion in the atmosphere. The bulk mercury atmospheric form – gaseous elemental mercury – can drift in the atmosphere for months moving over distances of thousands of kilometres. Therefore, changes of Hg pollution in the EMEP countries are largely affected by emission dynamics in other geographical regions and require assessment on a global scale. Evaluation of future Hg pollution levels in the EMEP and other regions has been performed as a part of collaborative MSC-E activities within the EU GMOS project (Section 5.2.1). Main results of the study are briefly discussed below.

The global inventory of mercury anthropogenic emissions for 2010 [AMAP/UNEP, 2013] was used as a reference point for evaluation of future changes. Spatial distribution of Hg anthropogenic emissions over the globe in 2010 is shown in Fig. 1.27a. As seen significant Hg emission fluxes were characteristics of industrial regions in East and South Asia, Central Europe and the eastern part of North America. Besides, high emissions are typical for some areas of Central and South America, Sub-Saharan Africa and Southeast Asia due to mercury releases from artisanal and small-scale gold mining. Definition of the geographical regions used in the study was based on the specifications adopted in the Phase II of the multi-model experiments under the CLRTAP Task Force on Hemispheric Transport of Air Pollution, TF HTAP (Fig. 1.27b).

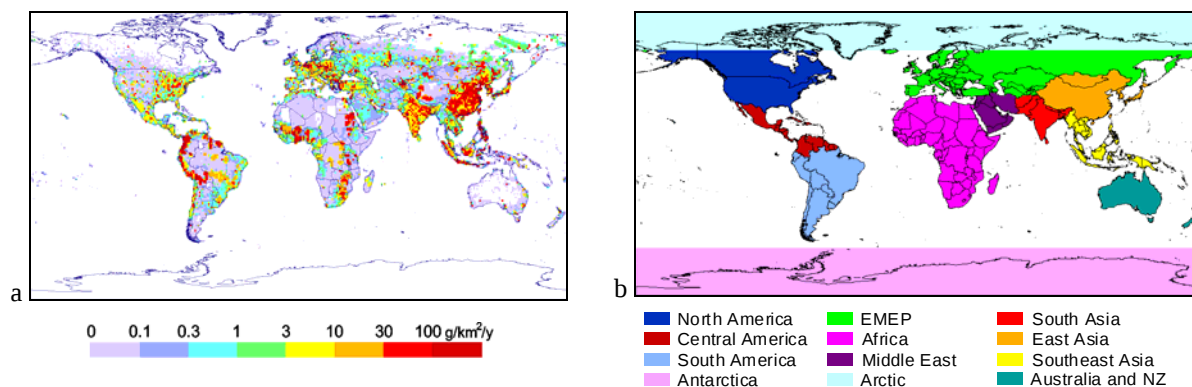


Fig. 1.27. Global distribution of mercury anthropogenic emissions in 2010 (a) and definition of source and receptor regions used in the analysis (b).

A number of future emission scenarios for 2035 has been developed within the EU GMOS project based on current and newly planned emission measures and energy consumption patterns. They consider the prospects for reducing Hg emissions from Hg specific measures or as a co-benefit from climate, pollutant and energy policy. 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. It gives a baseline vision on energy, industrial production as well as the use of air pollution control devices and waste management practices that are likely to change given no additional effort with regard to policy making.
- 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. The scenario illustrates the maximum possible Hg emission reductions that could be achieved if no other constraints have been taken into account, such as economy and increased demand.

Figure 1.28 shows total Hg anthropogenic emissions in various geographical regions according to the reference year 2010 and three selected emission scenarios for 2035. In the EMEP region as well as in the majority of other regions the CP 2035 scenario implies from weak to moderate reduction of Hg emissions between 2010 and 2035. Exceptions are East Asia and South Asia where the 'Current Policy' scenario expects emissions increase. The MFR 2035 scenario predicts essential Hg emissions decrease in all regions of the world. The NP 2035 presents an intermediate case with moderate reduction.

Long-term changes of Hg pollution in future have been evaluated with the GLEMOS model. Model simulation results for the three emissions scenarios of 2035 (CP2035, NP2035 and MFR2035) were compared with the reference case of 2010 to predict changes of Hg atmospheric dispersion and deposition on a global scale. Meteorological data for 2013 were used in all simulations to exclude the influence of inter-annual meteorological variability. Each model run consisted of the multi-year spin-up to reach the steady-state conditions and a one-year control simulation for the ultimate results. Geogenic and legacy sources were taken unchangeable over the considered period. Thus, the presented results reflect response of Hg atmospheric deposition to changes of direct anthropogenic emissions and do not take into account possible feedback of the ocean and terrestrial reservoirs to these changes.

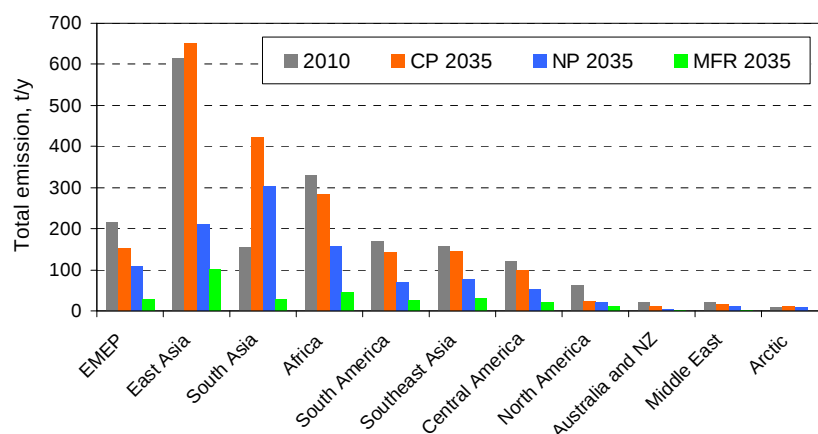


Fig. 1.28. Total Hg anthropogenic emissions in various geographical regions in 2010 and 2035 according to selected emission scenarios.

Figure 1.29a shows global distribution of Hg deposition flux simulated by GLEMOS for the reference year 2010. The largest deposition levels are characteristics of East and South Asia, which agrees with the spatial distribution of anthropogenic emissions (Fig. 1.27a). There are also significant deposition fluxes over the ocean, in the so called 'Inter Tropical Convergence Zone', which is characterized by enhanced precipitation. The model also predicts increased deposition fluxes in the polar regions due to the effect of the Atmospheric Mercury Depletion Events, AMDEs [Schroeder *et al.*, 1998; Lindberg *et al.*, 2002; Steffen *et al.*, 2008].

Projections of future changes in Hg deposition on a global scale simulated GLEMOS for three emissions scenarios of 2035 are illustrated in Figs. 1.29b-1.29d. The 'Current Policy' scenario (CP2035) supposes considerable decrease (20-30%) of Hg deposition in Europe and North America and strong (up to 50%) increase in South and Eastern Asia (Fig. 1.29b). In most of other parts of the Northern Hemisphere no significant changes ($\pm 5\%$) are expected, whereas slight decrease (5-15%) of deposition will take place in the Southern Hemisphere.

According to the 'New Policy' scenario (NP2035) moderate decrease of Hg deposition (20-30%) is predicted over the whole globe except for South Asia (India), where some deposition increase (10-15%) is expected due to the growth of regional anthropogenic emissions (Fig. 1.29c). Model predictions based on the 'Maximum Feasible Reduction' scenario (MFR2035) demonstrate consistent Hg deposition reduction on a global scale with somewhat larger decrease in the Northern Hemisphere (35-50%) and smaller decrease (30-35%) in the Southern Hemisphere (Figs. 1.29d). Thus, the most significant changes in Hg deposition (both increase and decrease) during the next 20 years for all considered scenarios are expected in the Northern Hemisphere and, in particular, in the largest industrial regions, where the majority of regulated emission sources are located.

Source apportionment of Hg deposition from direct anthropogenic sources in four geographical regions – EMEP, North America, East Asia and South Asia – is illustrated in Fig. 1.30. Contributions of natural and legacy emissions are not shown in the figure. Both CP 2035 and MFR 2035 scenarios suppose considerable reduction of Hg deposition in the EMEP region and in North America during the next 20 years (Figs. 1.30a and 1.30b). Relative contribution of various source regions to Hg deposition slightly changes for the both scenarios. The only exception is enhanced deposition from South Asia to these regions according to CP 2035. In contrast, in East and South Asia the CP 2035 scenario leads to substantial increase of Hg anthropogenic deposition due to increase of domestic emissions (Figs. 1.30c and 1.30d).

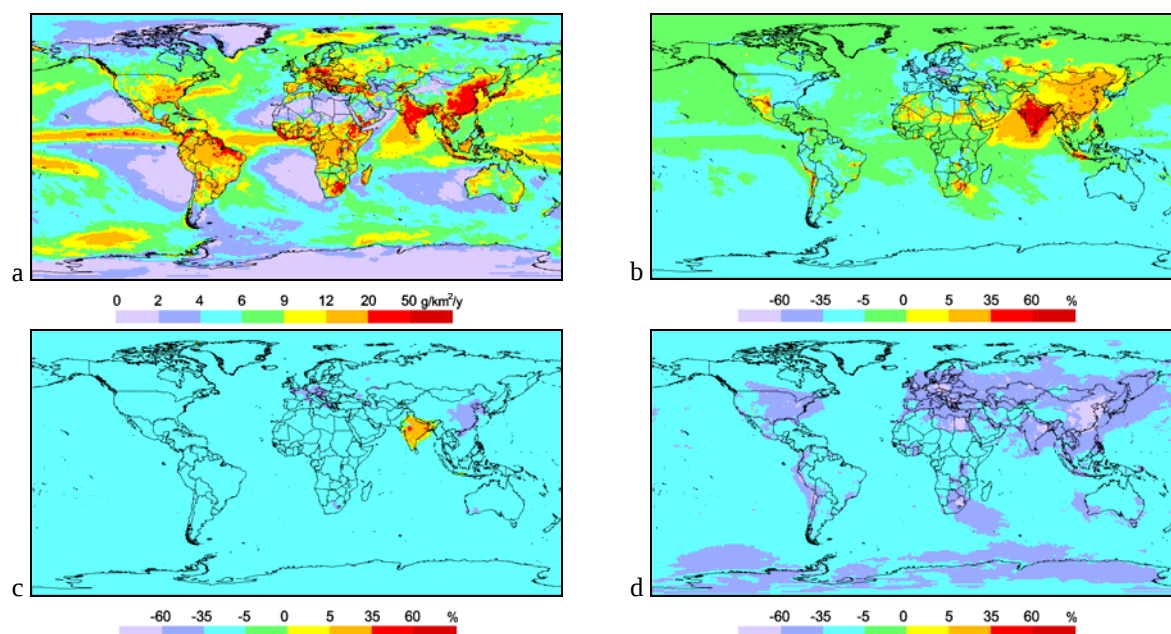


Fig. 1.29. Global distribution of Hg deposition in 2013 (a) and relative deposition changes by 2035 according to three emission scenarios: (b) – CP 2035; (c) – NP2035; (d) – MFR 2035.

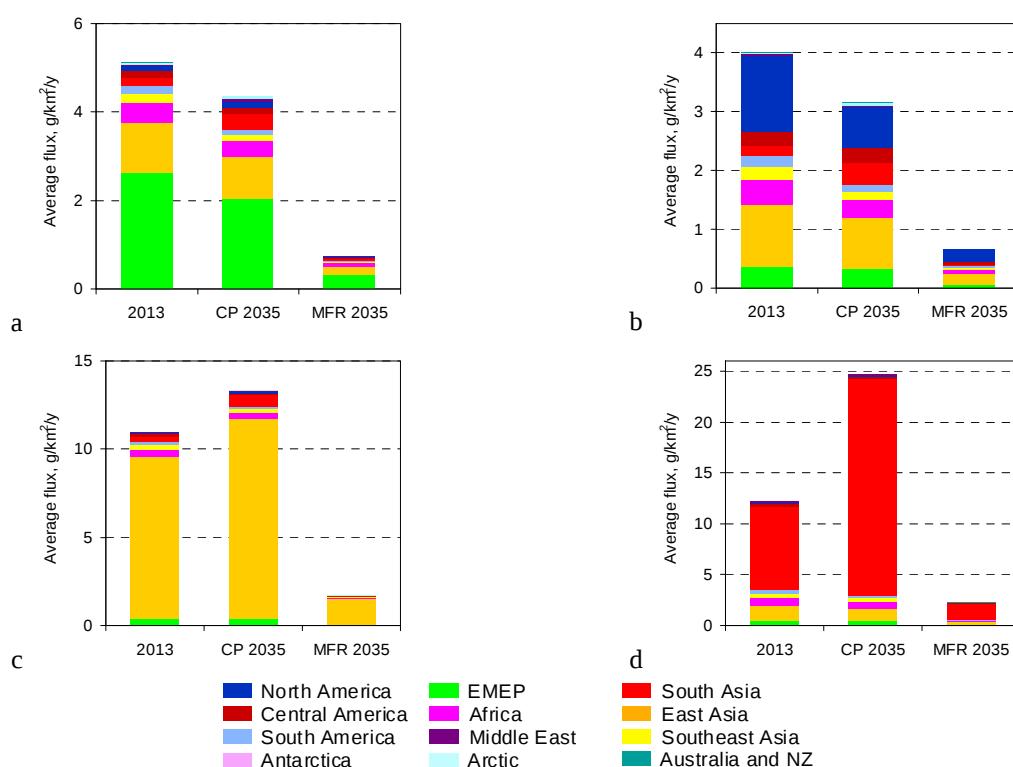


Fig. 1.30. Source apportionment of average Hg deposition from direct anthropogenic sources in 2013 and 2035 in various geographical regions: (a) – EMEP region; (b) – North America; (c) – East Asia; (d) – South Asia.

The most significant changes in Hg pollution during the next 20 years for all considered scenarios are predicted in the Northern Hemisphere and, in particular, in the largest industrial regions, where the majority of regulated emission sources are located. Considerable decrease of Hg deposition is expected in the EMEP region with slight change in relative contribution of various source regions.

2. LONG-TERM TRENDS OF HEAVY METAL POLLUTION FOR 1990-2012

Short overview of MSC-E approach to analysis of trends

Long-term trends of lead, cadmium and mercury pollution levels were analyzed for the period from 1990 to 2012. Methodology of the trend analysis was elaborated by MSC-E. Its description is available in Annex B and on the TFMM wiki page (<https://wiki.met.no/emep/emep-experts/start>). According to the methodology, initial time series (annual and/or monthly means of heavy metal deposition) were decomposed to the *regular component (trend)* consisting of *main component* and *seasonal component*, and *irregular component (residue)* (Fig. 2.1). The latter may be conditioned by short-term and/or random perturbations of the investigated series. The trend represents the considered time series free from these perturbations (in particular, year-to-year meteorological variability is thus excluded from the time series).

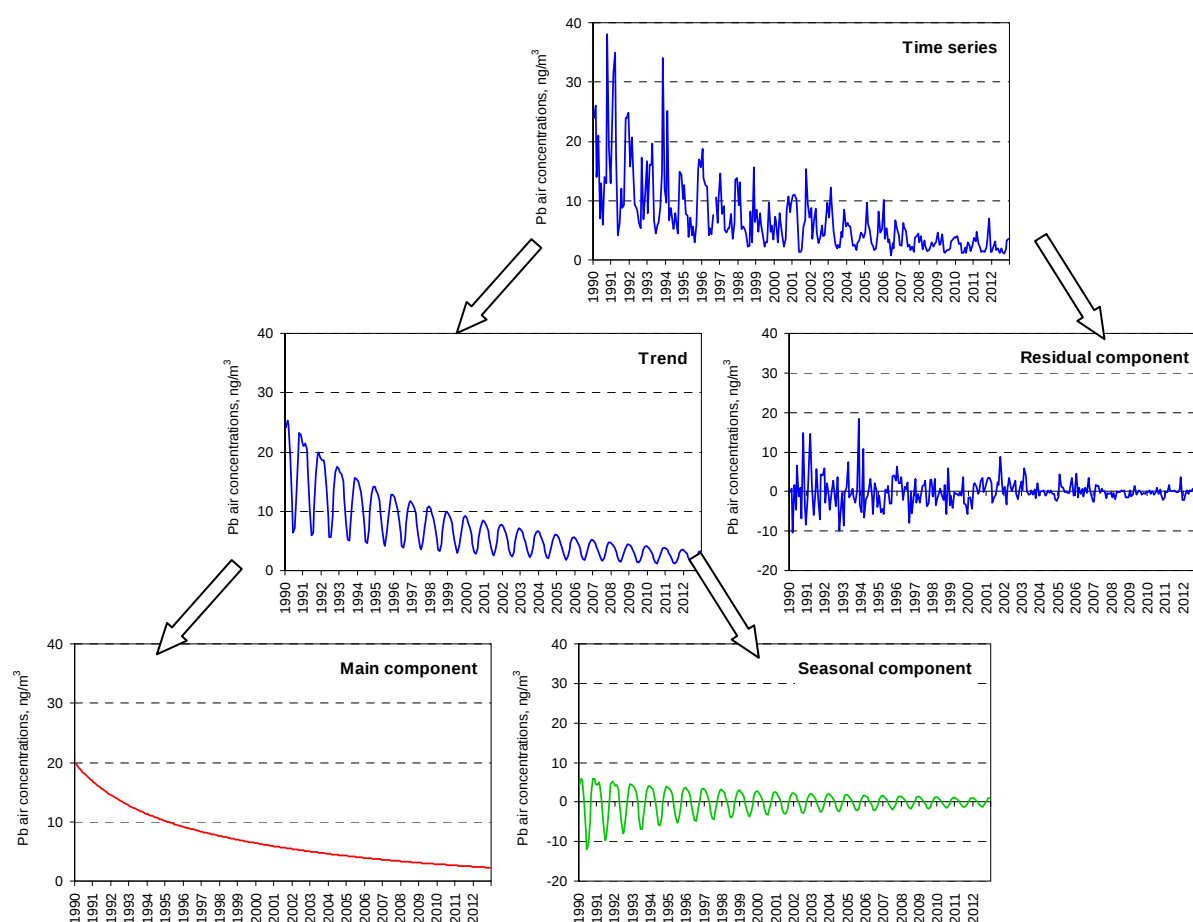


Fig. 2.1. Decomposition of time series to trend and residual component exemplified by measured concentrations of lead in air at EMEP station DE1

Reduction of pollution levels occurred at different rates for the period, therefore the trends were assumed to be bi-exponential. The decomposition allows evaluating the following trend parameters for heavy metals: *total reduction for the entire period, average, maximum and minimum annual reduction, and seasonality*. Maximum reduction rate takes place in the beginning, and minimum – in the end of the considered period. Seasonality is defined as average annual amplitude of seasonal component normalized by main component. This parameter expresses relative variations of main component due to seasonal variations.

HM deposition trends in the EMEP region and individual countries

Trends of pollution levels were analyzed for the EMEP region (Europe, Caucasus and Central Asia) and particular countries. Table 2.1 summarizes main characteristics of long-term trends in the EMEP region: total reduction, average, maximum and minimum reductions per year and seasonality. As seen, lead was characterized by the highest, and mercury – by the lowest magnitude of total reduction and average rate of reduction. Rates of deposition reduction of lead and mercury were higher in the beginning and lower in the end of the two-decade period, while the rate of cadmium annual decline remained constant. Deposition trends of all three metals were characterized by seasonality of around 30-40%. Since emission data did not include information on seasonal variations of heavy metal releases to the atmosphere, this seasonality could be attributed to the effect of meteorological variability.

Table 2.1. Main characteristics of long-term deposition trends of lead, cadmium and mercury for the period 1990-2012

Parameter	Pb	Cd	Hg
Total reduction, %	78.3	52.8	23.4
Average reduction, % per year	6.4	3.2	1.2
Maximum reduction (beginning of the period), % per year	6.8	3.2	3.5
Minimum reduction (end of the period), % per year	5.9	3.2	0.4
Seasonal variations, %	33.7	36.4	41.6

For simplicity time series and trends of total deposition over the EMEP region were given for annual mean values. As seen, the reduction of lead deposition is faster in the beginning of the considered period, and the lowest – in the end. The same is noted for mercury, but its reduction rate is smaller than that for lead (Fig.2.2).

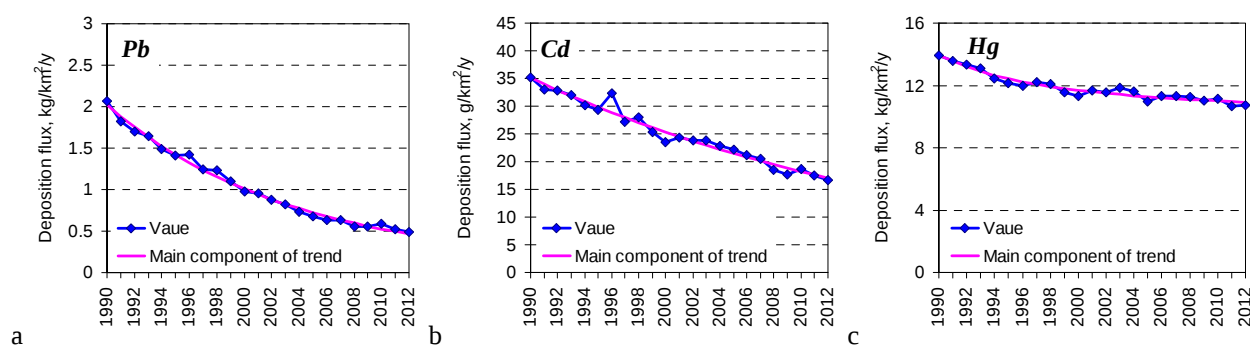


Fig. 2.2. Long-term changes of total deposition flux and of main component of its trend for lead (a), cadmium (b) and mercury (c)

The changes of pollution levels in particular countries could differ considerably from averaged values for the entire EMEP region. For example, lead deposition in Sweden were reducing with rate 19% per year in the beginning of the period, which was much faster than the average for EMEP value (Fig. 2.3a). Another situation is exemplified by trend of total deposition of mercury in Greece. From 1990 to 1996 the deposition were declining, with starting rate of 3.5% per year (Fig. 2.3b). However, the decline stopped in 1996 and was changed by growth, which rate reached 3% per year in 2012.

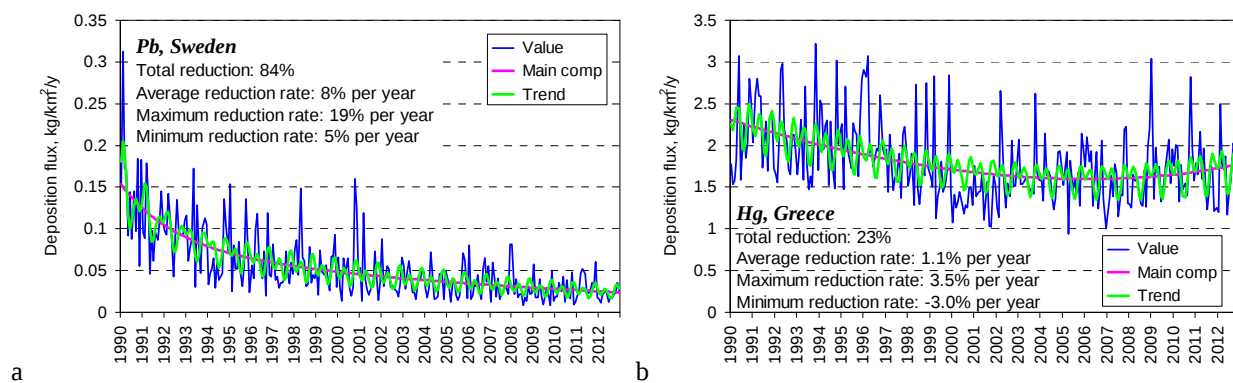


Fig. 2.3. Total deposition, trend and main component of lead in Sweden (a) and mercury in Greece (b)

For analysis of long-term pollution changes the EMEP countries were divided in three groups: EU28, EECCA, and the remaining countries related to the group 'other'. In most of countries of all three groups reduction rate of lead deposition was similar ranging from 4% to 8% per year (Fig. 2.4). The exceptions were Turkey and the Transcaucasian countries – Armenia, Georgia and Azerbaijan, where reduction rates were substantially lower – 1.4 – 2.7%. However, the highest rates of the reduction in the beginning of the considered period (15% – 18%) were noted for a number of the EU28 countries (Finland, Denmark, Germany, Spain etc...), for Monaco and for Norway from the group 'Other'. This fast decline followed by rapid reduction of anthropogenic emissions in these countries took place due to phasing out of leaded gasoline.

The main typical characteristic of reduction of cadmium deposition was its uniformity in time. Only in few countries (e.g., Estonia, Finland, Denmark, Russia etc.) annual reduction rate varied over the period. In the EU28 countries mean rate of annual reduction ranged from 2% to 6%. In the EECCA countries similar rate was noted only for countries in the European part of the region: Russia, Ukraine, Belarus and Moldova. In the Transcaucasian and the Central Asian countries the reduction was less than 1.3% per year, and in Armenia and Azerbaijan even slow growth (0.1%-0.2% per year) was indicated. Minor long-term changes of cadmium transboundary pollution in these countries was explained by relatively low rate of reduction or even growth of national emissions, and partly by contribution of non-EMEP sources, which long-term changes were neglected.

Mercury deposition rates were the lowest compared to those of lead and cadmium. In most of the countries they were lower than 3% per year. As a rule, annual reduction rates in the EU28 countries were higher than those in the EECCA countries. Although mean rates of deposition decline were low, they ranged substantially from year to year. In some countries (Belgium, Slovakia, the Netherlands etc.) the rates in the beginning of the considered period made up 10%-17% due to rapid reduction of anthropogenic emissions in these and neighbouring countries. In some countries (e.g., Belarus) even some long-term increase of deposition was noted followed by growth of national emissions.

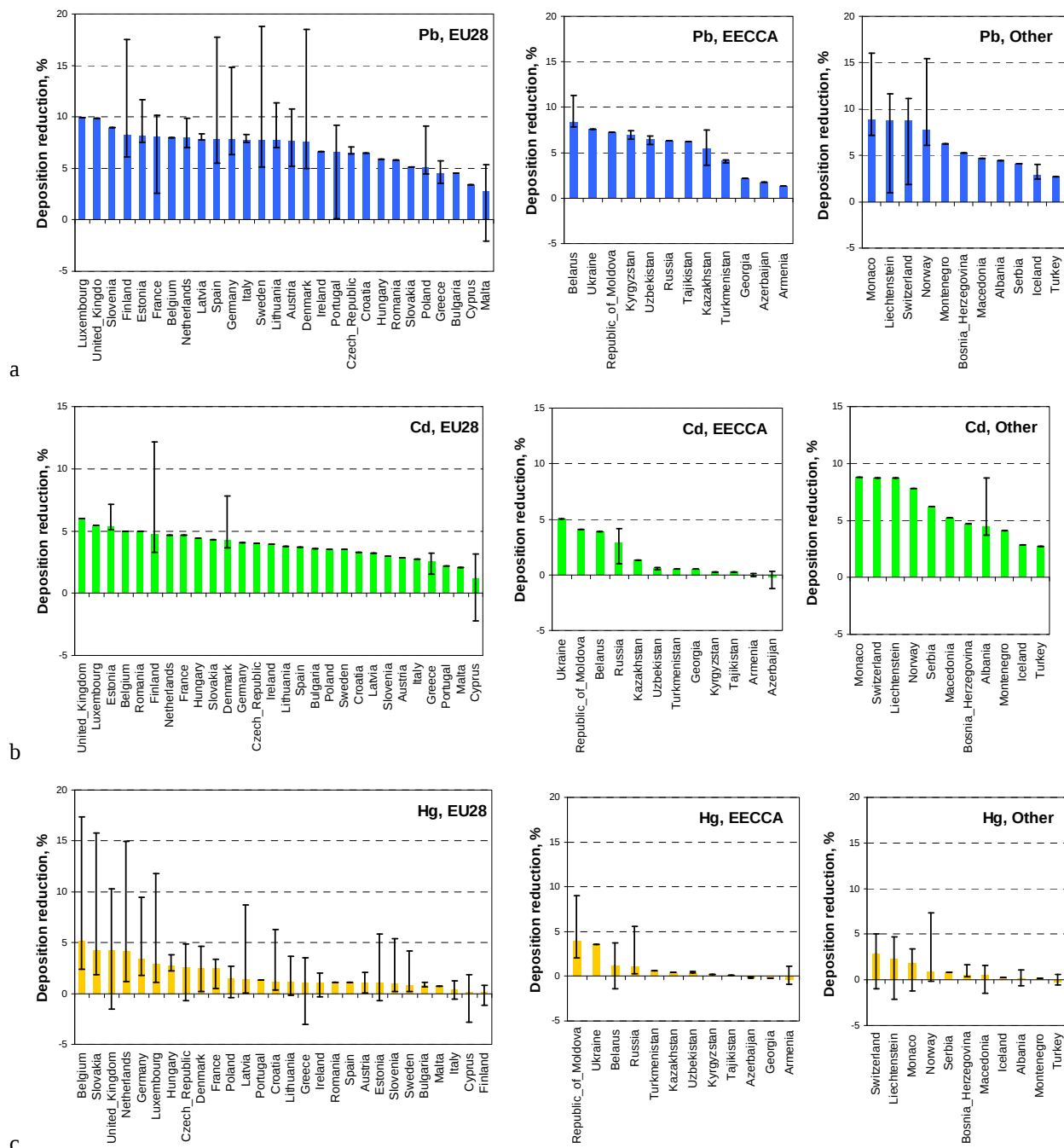


Fig 2.4. Country-averaged deposition reductions in countries of EU28 (left), EECCA (middle) and remaining regions (right) of lead (a), cadmium (b) and mercury (c). Negative values of reduction mean increase. Whiskers indicate range of annual reduction rates

Besides main component of trend describing gradual long-term changes of pollution levels, another important parameter is seasonality. Trends of cadmium deposition in Belgium and Kazakhstan were used to exemplify countries with low and high seasonalities (Fig. 2.5). Seasonality of cadmium deposition in Belgium was relatively low (24%) (Fig. 2.5a). In spite of marked month-to-month variability of deposition, the applied statistical methods did not deduce significant seasonality. The most probable reason for this is that seasonal variations were disturbed by strong episodic short-term variability of deposition. Kazakhstan is characterized by continental climate with marked seasonal variations of meteorological conditions. This meteorological variability resulted to strong (more than 60 %) seasonality of deposition levels (Fig. 2.5b).

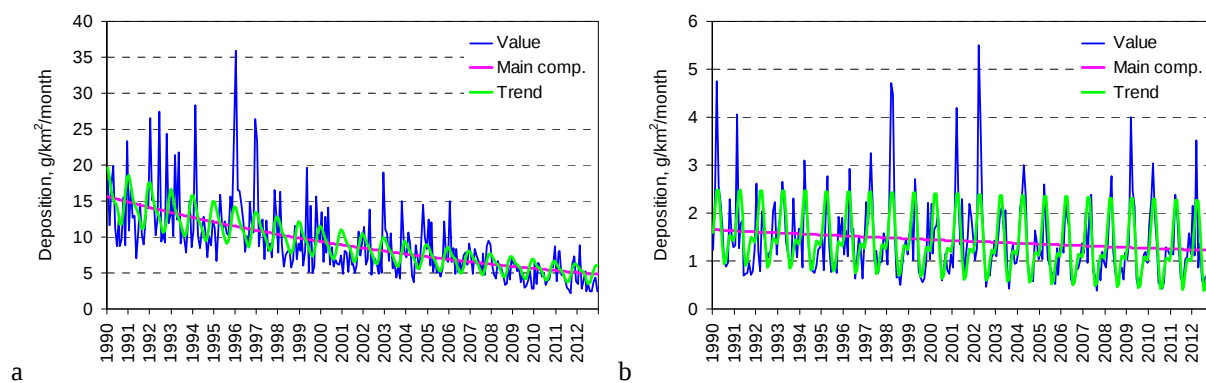


Fig. 2.5. Calculated monthly total deposition, trend and its main component of cadmium in Belgium (a) and Kazakhstan (b)

In particular countries seasonality ranged from 8% to 95% for the considered metals (Fig. 2.6). On average it amounted to 30-40%. Seasonality for mercury was somewhat lower than that for lead and cadmium, mostly due to long (about a year) atmospheric life time of mercury.

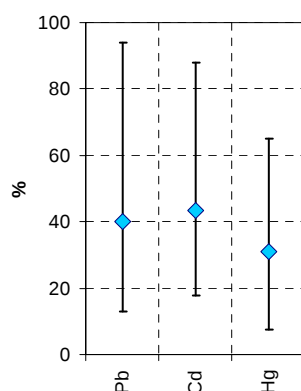


Fig. 2.6. Seasonality of lead, cadmium and mercury deposition averaged among the EMEP countries. Whiskers indicate range of seasonalities among particular countries

Factors affecting trends

Long-term trends of pollution levels depend on a number of factors including changes of anthropogenic and secondary emission data, long-term climatic trends, changes of land-use etc.

However, the main factors affecting the changes of heavy metal levels in the EMEP region from 1990 to 2012 seem to be anthropogenic and secondary emissions.

Reduction of anthropogenic emission of lead and cadmium for the period 1990- 2012 made up 90% and 60%, respectively (Fig. 2.7). However, total deposition reduction to the EMEP region was smaller (Table 2.1). The reason of this was significant contribution of secondary sources to pollution levels in the EMEP region. Reduction of sum of anthropogenic and secondary sources within the region made up 80% for lead and 57% for cadmium, which was very close to the reductions of deposition, because most of lead or cadmium emitted from the EMEP region was deposited within the region.

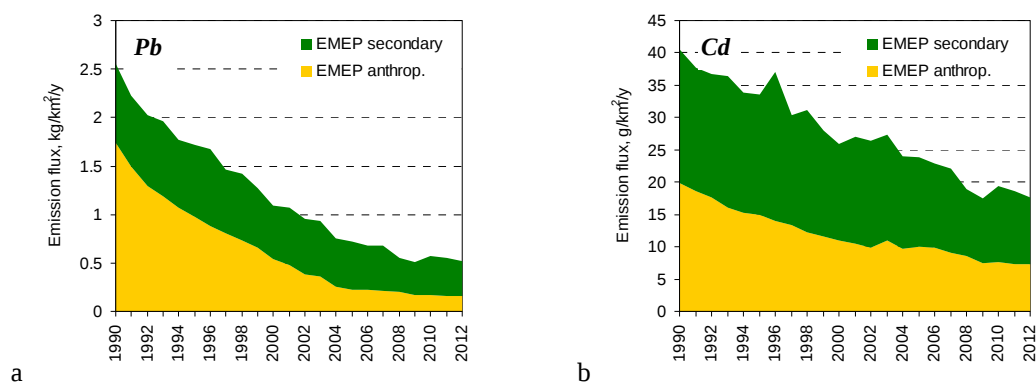


Fig. 2.7. Time series of anthropogenic and secondary emission fluxes of lead (a) and cadmium (b) within EMEP region

Situation with mercury differed from that for lead and cadmium. First of all, very large amount of mercury entered the region due to intercontinental transport from non-EMEP sources. Contribution of non-EMEP sources (both anthropogenic and secondary) ranged from 40% to almost 60% over the considered period (Fig. 2.8). However, in absolute terms their contribution remained almost the same for the whole period, because global emissions of mercury had not changed much (Section 1.1). And secondly, mercury speciation of emissions was very important factor affecting its transport and deposition. Since significant part of mercury was emitted as long-living elemental form (around 70% for anthropogenic sources and 100% for secondary sources), most of elemental mercury was transported outside the EMEP region. These two reasons resulted to relatively low (23%) reduction of mercury deposition in the region.

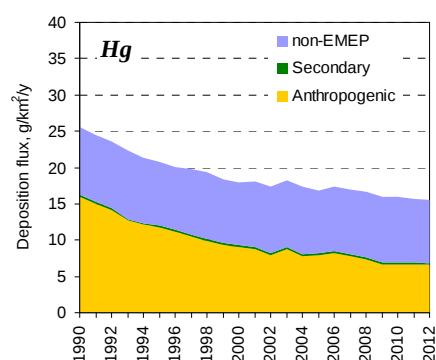


Fig. 2.8. Mercury deposition fluxes caused by anthropogenic and secondary sources within EMEP region and by non-EMEP sources

Similar to the EMEP domain, changes of pollution levels within each country could also be expressed through changes of anthropogenic and secondary sources. Anthropogenic sources were split into national and foreign ones. For example, deposition change of cadmium in the United Kingdom was around 75% (Fig. 2.9). This reduction was combined of deposition reductions due to national sources and EMEP secondary sources (around 35% each) and of deposition reduction from foreign emission sources (5%). There were countries, e.g., the Republic of Moldova, where the main factor responsible for cadmium deposition reduction was change of contribution of transboundary transport. In some countries, e.g., Latvia and Belarus, deposition from national emission sources even increased from 1990 to 2012. It was explained by increase of national emissions in these countries for the considered period. In some countries (e.g., Azerbaijan, Cyprus, Tajikistan) the increase of deposition was caused by growth of contribution of secondary sources. Although deposition from secondary sources tended to decline in the EMEP region, the increase in these particular countries was conditioned, most likely, by meteorological variability.

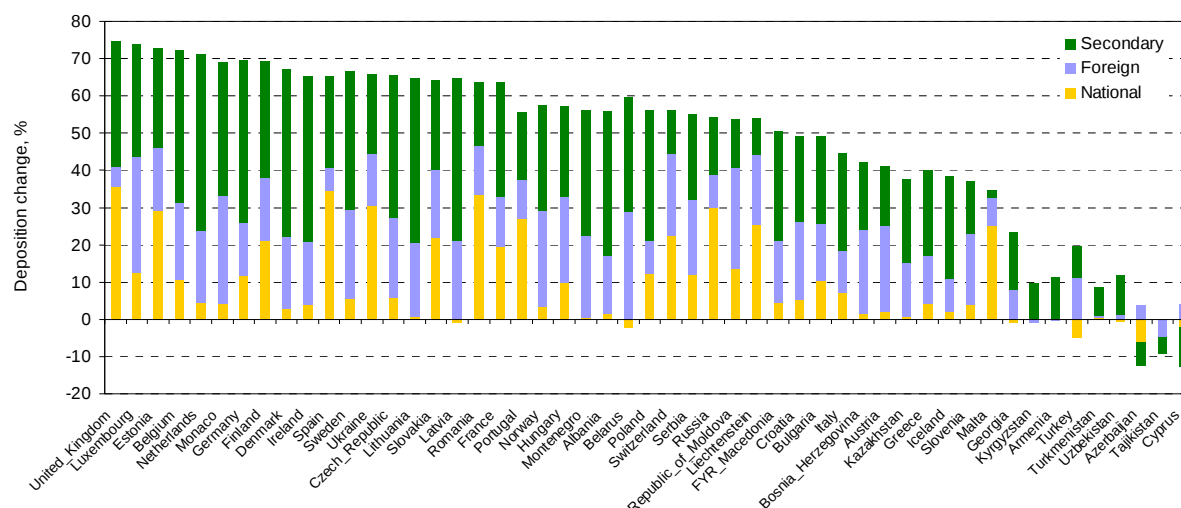


Fig. 2.9. Cadmium deposition change between 1990 and 2012 due to reduction of anthropogenic (national and foreign) and secondary emission sources within EMEP region

Trends of observed and modelled levels in particular gridcells

Data on long-term time series of heavy metal pollution levels were available for each gridcell of the EMEP domain. For those gridcells where monitoring stations were located, analysis of trends could be done jointly on the base of both modelled and measured values. There were 19 EMEP stations measuring lead and cadmium and 8 stations measuring mercury with long time series. Measurement data on lead and cadmium concentrations in air and wet deposition fluxes at these selected stations were available for the period started in 1992 or earlier and finished in 1998 or later. For mercury starting year for the selection of stations was 1995, because earlier measurements were either unavailable or unreliable. The selected stations were located mostly in the central and the northern parts of Europe (Fig. 2.10). Trends in the other parts of the EMEP region were characterized entirely by the modelling. Analysis of observed and modelled heavy metal levels at each selected station was performed on monthly basis in order to account for seasonal variability.

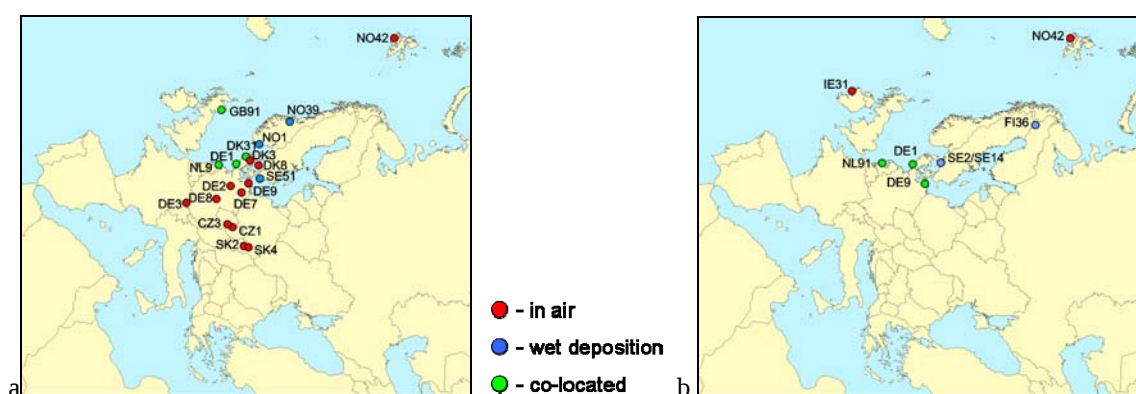


Fig. 2.10. Location of measurement stations with long time series selected for the analysis of long-term trends of lead and cadmium (a) and mercury (b).

At most of the EMEP stations both modelling results and monitoring data demonstrated significant reduction of pollution levels for the period. For example, mean reduction of air concentrations of lead at most of stations ranged from around 5% to more than 18% (Fig. 2.11a), and for wet deposition – from 5% to 9% (Fig. 2.11b). The exception was air concentrations at station NO42 where observed reduction was much higher than modelled one. This Arctic station was very remote from main emission sources and its levels were strongly affected by intercontinental transport of lead. For example, [Bazzano *et al.*, 2015] supposed that the main source of atmospheric lead in summer at this station were North American sources. Lack of long-term global lead emission data gave rise of uncertainties of modelling results in the remote regions.

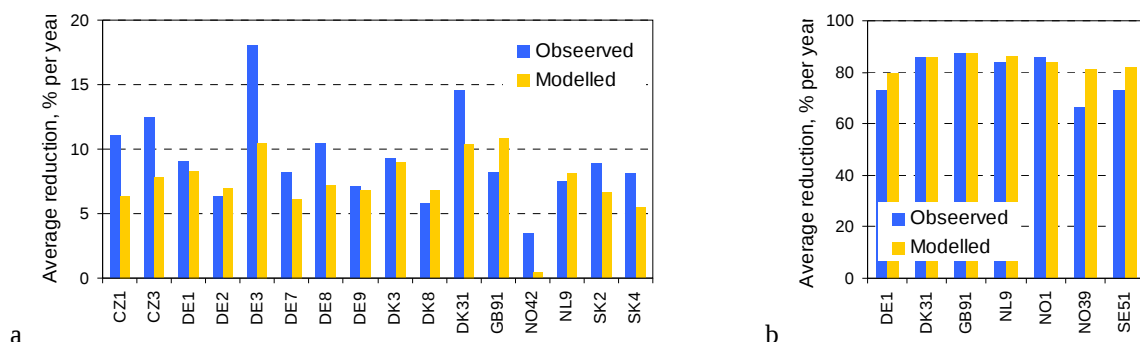


Fig. 2.11 Average reductions for modelled and measured trends of lead concentrations in air (a) and wet deposition fluxes (b) at monitoring stations, % per year.

On average, mean annual reduction of observed and modelled concentrations and wet deposition of lead for the period from 1990 to 2012 amounted to about 7-9% (Fig. 2.12a). For cadmium the reduction of modelled levels was around 4%, and that of observed levels – about 5-8% (Fig. 2.12b). Somewhat higher reduction of observed compared to modelled levels was caused by underestimation of high observed values in the beginning of the considered period. Time series of cadmium air concentrations at station DE7 were presented to exemplify this situation (Fig. 2.13). Reduction of modelled and measured mercury air concentrations was low (less than 0.5%) (Fig. 2.12c). Reduction of observed mercury wet deposition was higher (about 3% per year) than that of modelled levels. However, range of reductions of observed mercury deposition was quite high and fully overlapped the range for modelled values.

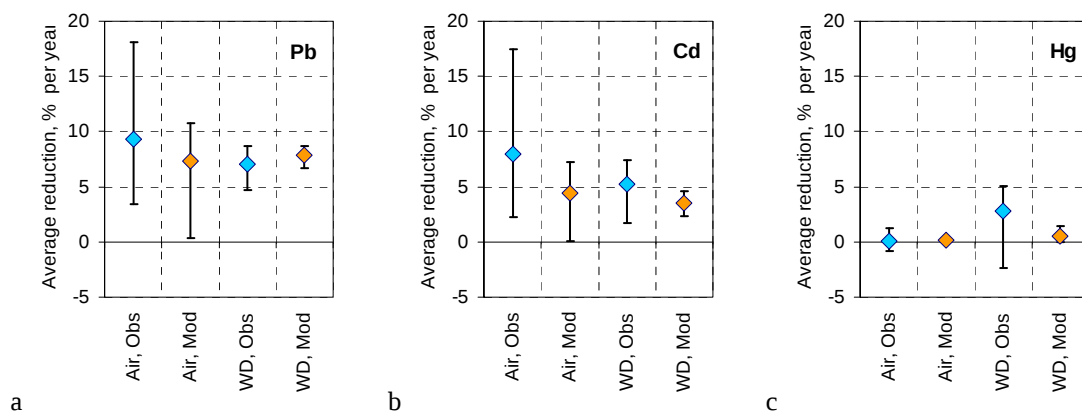


Fig. 2.12. Average annual reduction rates for modelled and observed air concentrations and wet deposition fluxes of lead (left), cadmium (middle) and mercury (right) over selected monitoring stations. Whiskers mean range of average reduction rates among stations

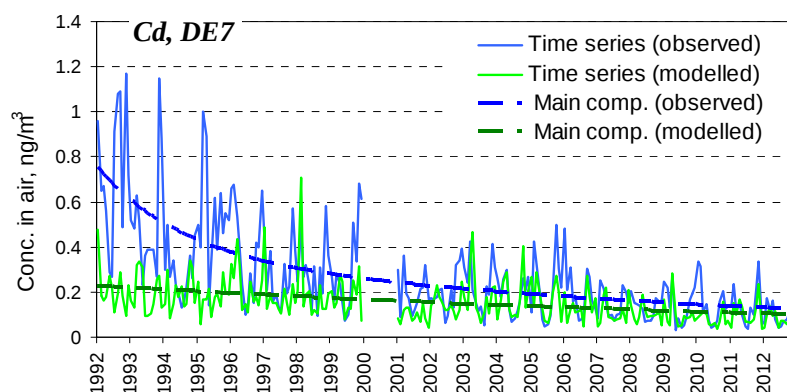


Fig. 2.13. Observed and modelled time series and main components of trend of cadmium concentrations in air at station DE7 (Neuglobsow, Germany)

Average seasonality of measured and observed lead and cadmium levels was about 40 - 50% (Fig. 2.14a and b). In case of mercury seasonality of air concentrations was low compared to the other heavy metals, amounting to about 10% (Fig. 2.14c). Observed seasonality of mercury wet deposition, ranging from 20 to 110%, was higher than modelled one, which could be linked with uncertainties of mercury chemical transformations as well as lack of data on seasonal variation of emissions.

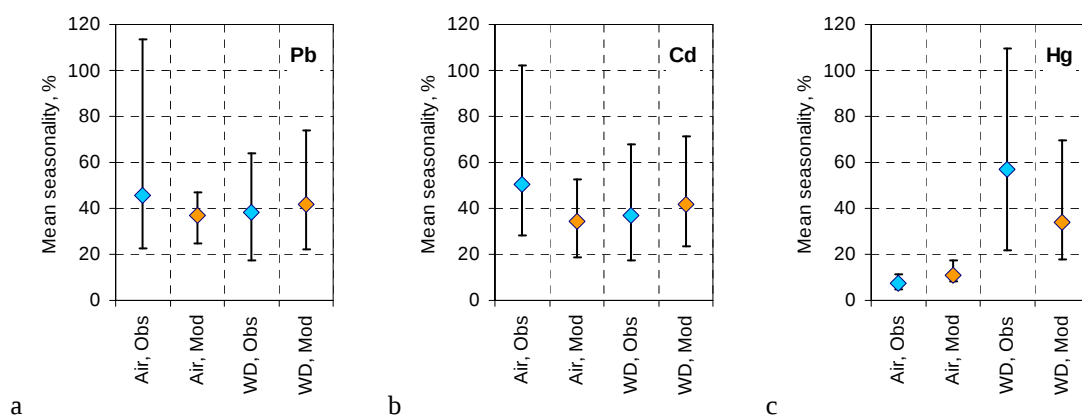


Fig. 2.14. Mean seasonality of modelled and observed air concentrations and wet deposition fluxes of lead (a), cadmium (b) and mercury (c). Whiskers mean range of seasonalities among stations

Verification of modelling results

Calculated concentrations in air and wet deposition fluxes of lead, cadmium and mercury were considered with the corresponding measurement values for 1990-2012. Verification was carried out for the EMEP stations with long time series of measurements. Statistical indices applied for the verification were the same as those used for the analysis of pollution levels in 2013: relative bias, correlation coefficient and normalized root mean square error (NRMSE). Values of these indices are summarized in Table 2.2.

As follows from the table, lead is characterized by the lowest relative bias: both for air concentrations and wet deposition fluxes it is below $\pm 10\%$. For most of model-observation pairs of values difference between modelled and observed levels lies within a factor of two. For cadmium the discrepancies between modelled and observed levels are somewhat higher than those for lead. The bias for air

concentrations is -26% and -35% for wet deposition. The reason for this is underestimation of the observed levels at some stations in the beginning of the period. Most likely, these high concentrations are caused by uncertainties of measurement approaches.

For mercury concentrations in air mean relative bias is the lowest (0%), and all modelled values agree with the observed ones within a factor of two. Relatively low correlation for mercury in air is explained by its small spatial and temporal variability. Bias for wet deposition of mercury is positive which means that the model tends to overestimate the observed levels. It could be connected with uncertainties in current understanding of mercury atmospheric chemistry.

Table 2.2. Main statistical indicators of agreement between annual modelled and measured levels of time series of air concentrations and wet deposition fluxes the period 1990-2012

	Lead		Cadmium		Mercury	
	C _{air}	Wet Dep	C _{air}	Wet Dep	C _{air}	Wet Dep
Relative bias, %	-9.9	6.2	-26.2	-35.6	0.0	24.5
Correlation coefficient	0.66	0.77	0.58	0.64	0.38	0.77
NRMSE	0.74	0.57	0.93	0.77	0.09	0.43
F2, %	82.7	85.4	79.9	72.3	100.0	82.2
F3, %	95.1	99.3	94.5	93.9	100.0	95.3

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

Analysis of long-term trends of lead, cadmium and mercury pollution levels in the EMEP region demonstrates that the levels have been declining since 1990. The highest reduction of pollution was noted for lead, followed by cadmium and then by mercury. The main reason of the decline was reduction of atmospheric emissions in the EMEP countries. However, other factors such as long-term changes of secondary emissions of lead and cadmium and contribution of intercontinental transport of mercury to pollution in the EMEP region were also important. Main characteristic of long-term changes (total reduction, annual reduction rate and seasonality) demonstrated considerable geographical variability, which was confirmed by analysis of modelled and observed levels at monitoring stations and of calculated deposition in particular EMEP countries.

Pollution levels of lead, cadmium and mercury in the EMEP region reduced by 78, 53 and 23%, respectively, mostly due to reduction atmospheric emissions in the EMEP countries, long-term changes of secondary emissions of lead and cadmium and contribution of intercontinental transport of mercury to pollution in the EMEP region. Main characteristics of trends (total reduction, annual reduction rate and seasonality) demonstrated considerable geographical variability.

3. DEVELOPMENT OF GLEMOS MODELLING SYSTEM

MSC-E continued development and improvement of its modelling tool used for assessment of heavy metal pollution in the EMEP region and on a global scale. In particular, this work is aimed at further elaboration of the Global EMEP Multi-media Modelling System (GLEMOS). The main activities in this direction include model-based study of processes governing mercury cycling in the atmosphere, evaluation of mercury interaction with atmospheric aerosol, identification of possible effect of the changing climate on mercury pollution based on literature survey and further improvement of the model description of heavy metal secondary emission due to wind re-suspension. A short summary of these activities are given below.

3.1. Multi-model assessment of mercury processes in the atmosphere

Mercury is a complex pollutant characterized by diverse properties in the environment. Chemical composition of mercury species and oxidation chemistry in the atmosphere determine its ability to long-term atmospheric transport and deposition to various terrestrial and aquatic regions. 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 (Fig. 3.1).

Application of self-consistent chemical transport models (CTM) in combination with extensive monitoring data could facilitate a better understanding of the principal mechanisms governing Hg dispersion and cycling in the environment and improvements in model parameterization. Application of CTM using different emission estimates and subsequent comparison of the modelling results with measurements can provide an additional evaluation of the emission inventories.

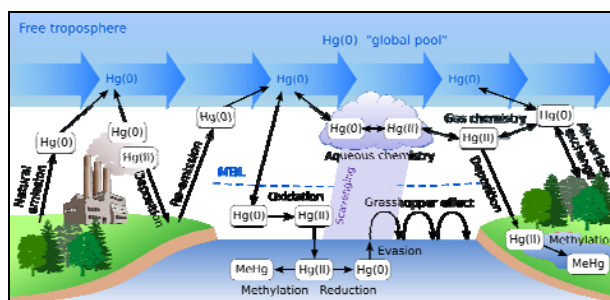


Fig. 3.1. General cycle of mercury in the environment
[Travnikov, 2011]

Multi-model study of processes governing mercury cycling in the atmosphere has been performed as a part of Mercury Modelling Task Force (MMTF) activities established within the framework of the EU funded GMOS (Global Mercury Observation System) project (see Section 5.2.1 for more details). The work of MMTF is organized in a form of model experiments aimed at answering particular scientific questions related to mercury atmospheric chemistry, anthropogenic and natural/secondary emissions, deposition etc. A summary of the MMTF model experiments is given in Table 3.1.

Table 3.1. Summary of model experiments under MMTF

Base case Simulation with the state-of-the-art model configuration (BASE)
Emission effects - Model run with no anthropogenic emissions (NOANT) - Modified speciation of Hg anthropogenic emissions (ANTSPEC) - Unified natural/secondary emissions (COMNAT) - Improved emissions base on inverse modelling (INVEMI)
Atmospheric chemistry - Model run with no atmospheric chemistry (NOCHEM) - Simulation with Br oxidation chemistry with two different sets of Br air concentration (BRCHEM1 and BRCHEM2) - Simulation with OH - initiated oxidation chemistry (OHCHEM) - Simulation with O₃ - initiated oxidation chemistry (O3CHEM)
Source region apportionment Evaluation of Hg dispersion from 14 major source regions (Europe, North America, East Asia, South Asia etc.)
Emission sectors apportionment Evaluation of Hg dispersion from 3 groups of emission sectors (Stationary combustion, Industrial sources, intentional use and product waste)

The model ensemble of the study includes six chemical transport models simulating mercury on both global and regional scales (Table 3.2). The models differ in their formulation, spatial resolution, and applied parameterizations of physical and chemical processes. All the models used the same inventory for anthropogenic emissions of mercury [AMAP/UNEP, 2013] but applied model specific definitions of natural and legacy sources. The target year of the simulations is 2013.

Table 3.2. Model ensemble of the study

Model	Scale	Institution	Model	Scale	Institution
GLEMOS	global/regional	EMEP/MSCE	GEOS-Chem	global	MIT, USA
ECHMERIT	global	CNR-IIA, Italy	CMAQ-Hg	regional	HZG, Germany
GEM-MACH-Hg	global	Environ. Canada	WRF-Chem	regional	CNR-IIA, Italy

The modelling results were evaluated against observations from several monitoring networks. The measurements dataset is based on the EMEP regional network for Europe [EMEP, 2015], the NADP/MDN network for North America [NADP/MDN, 2015] and the global GMOS mercury network [GMOS, 2015]. In total, the dataset includes 28 measurement sites measuring mercury concentration in the air (Fig. 3.2). Among them, 11 sites provide measurements of mercury species (Hg^0 , $\text{Hg(II)}_{\text{gas}}$, $\text{Hg(II)}_{\text{part}}$). Besides, there are 135 sites measuring

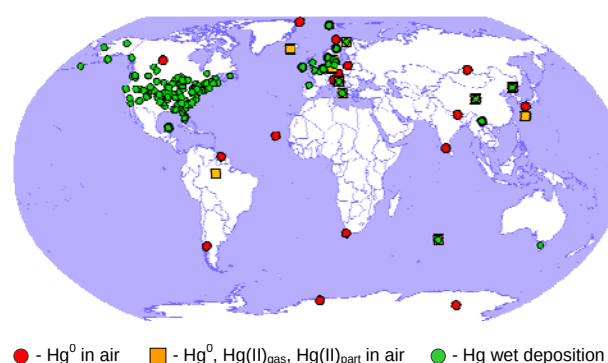


Fig. 3.2. Location of measurement sites

mercury wet deposition and concentration in precipitation. Air concentration measurements are relatively uniformly distributed over the globe with somewhat higher density in the Northern Hemisphere. In contrast, most observations of wet deposition are located in Europe and North America.

Global distributions of gaseous elemental mercury (Hg^0) concentration in the surface air simulated by four global models according to the Base case are shown in Fig. 3.3. The models simulate similar spatial patterns of mercury concentration with pronounced gradient between the Southern and the Northern Hemispheres and with elevated concentrations in the major industrial regions – East and South Asia, Europe and North America. Generally, the simulated results well agree with measured values shown by colored circles in Fig. 3.3. This means that the models reasonably well reproduce general circulation of mercury in the global atmosphere in spite of significant deviations in applied parameterizations of physical and chemical processes. It should be noted that the models also demonstrate acceptable performance simulating wet deposition flux (not shown here). However, model-to-model and mode-to-observation deviations are somewhat larger in this case due to stronger effect of uncertainties in atmospheric chemistry and some meteorological parameters (e.g. precipitation amount).

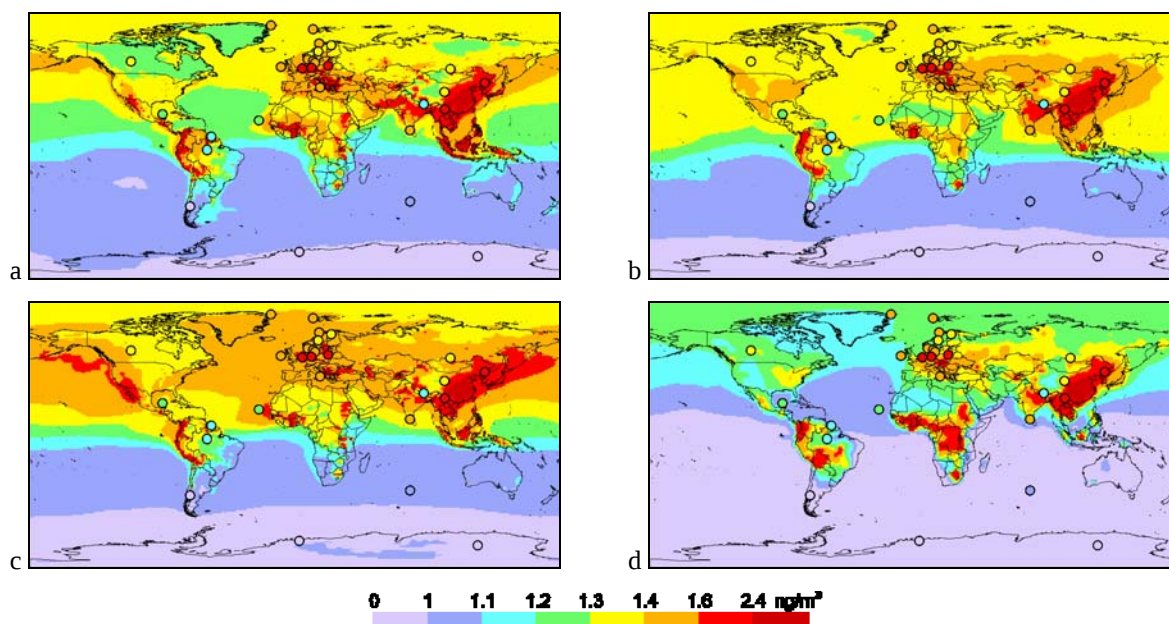


Fig. 3.3. Spatial distribution of Hg^0 air concentration in 2013 simulated according to the Base case by four global models: (a) - GLEMOS; (b) - GEOS-Chem; (c) – GEM-MACH-Hg; (d) – ECHMERIT. Circles show observed values in the same colour scale.

An example of the mercury process study involving both model experiments and measurement data is given in Fig. 3.4. In particular, it is shown that the Base case simulation of all models successfully reproduce the seasonal variation of mercury wet deposition in Europe with higher values in the warm season and lower values during winter. It is remarkable taking into account that the models apply three different chemical oxidation mechanisms of mercury in the atmosphere. The model experiment with no atmospheric chemistry provides completely different results with almost absent seasonal variability. On the other hand, the experiment with no anthropogenic emissions only slightly changes the seasonal variation in comparison with the Base case. It means that the seasonal variation of wet deposition of mercury in Europe is rather determined by atmospheric oxidation chemistry. 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.

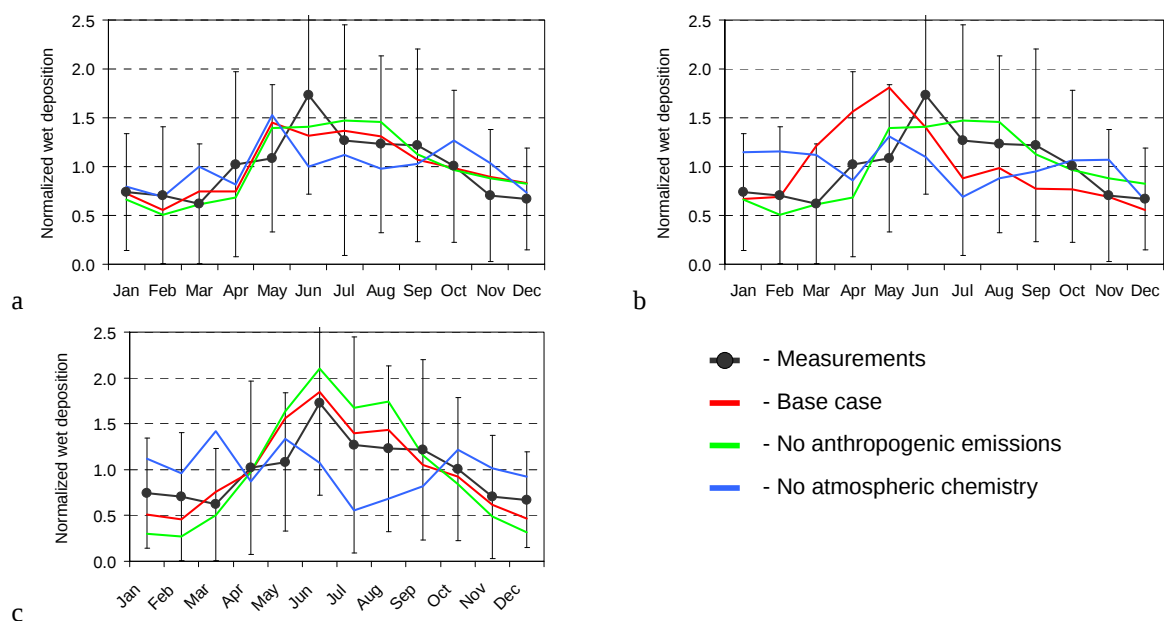


Fig. 3.4. Seasonal variation of wet deposition of mercury in Europe based on measurement data and model experiments by three global models: (a) – GLEMOS; (b) – GEOS-Chem; and (c) – GEM-MACH-Hg. Black line with dots show measured values averaged over all available sites. Whiskers show standard deviation ($\pm 1\sigma$) among the sites. Coloured lines present model simulations according to different model experiments averaged over the same sites. All monthly values are normalized by annual mean value.

The majority of the model experiments has been completed and analysis of the simulation results is underway. Preliminary results of the analysis include the following:

- The inter-hemispheric gradient of mercury concentration is largely defined by atmospheric chemistry and natural/legacy emissions;
- Both Br and OH-initiated oxidation chemistry allows reproducing the inter-hemispheric gradient. However, available data on Br air concentration are highly uncertain;
- Br and OH/O₃ mechanisms produce significantly different distributions of wet deposition in the inter-tropical convergence zone and in the Southern Hemisphere. More wet deposition measurements are needed in these regions for further evaluation.
- Seasonal variation of Hg wet deposition in Europe is mostly defined by oxidation chemistry. However, the role of temporal variation of anthropogenic emission is to be further identified.

Progress and preliminary results of the study have been presented at the International Conference on Mercury as a Global Pollutant 2015 (Jeju, Republic of Korea, June 2015).

Multi-model research activities performed under MMTF will improve understanding of mercury processes in the atmosphere and, ultimately, quality.

3.2. Gas-particle partitioning of mercury in the atmosphere

Mercury is a chemical element occurring in the atmosphere in various physical and chemical forms. It is emitted to the atmosphere from anthropogenic sources in a form of elemental vapour (Hg^0) as well as in divalent oxidized forms in various gaseous compounds ($\text{Hg(II)}_{\text{gas}}$) and in composition of aerosol particles ($\text{Hg(II)}_{\text{part}}$). Besides, transformations of mercury between different forms can take place in the atmosphere due to chemical oxidation or reduction reactions and sorption/desorption to particular matter. Various mercury species are characterized by diverse properties defining different aspects of mercury cycling in the atmosphere. Poorly soluble and relatively inert gaseous elemental mercury can drift in the air for months providing transport of mercury mass between different regions of the planet [Selin, 2009; Travníkov, 2011]. In contrast, oxidized mercury species are easily removed from the air by precipitation scavenging or the surface uptake. Besides, even gaseous and particulate phases of oxidized mercury differ significantly in their deposition rates. Therefore, correct treatment of mercury speciation in the model is critical for assessment of mercury pollution on both regional and global scales.

The GLEMOS model has been recently updated in terms of model parameterization of gas-particle partitioning of oxidized mercury in the atmosphere. For this purpose, the empirically derived parameterization of the gas-particle partitioning developed by Amos *et al.* [2012] has been implemented into the model. It has replaced the previously used approach when emissions and oxidation reaction products were treated either as particulate or gaseous species and were transported in the atmosphere independently as inert tracers. It often led to unrealistic levels of oxidized mercury species in comparison with available measurements. In the implemented scheme the gas-particle partitioning coefficient is a function of temperature and air concentration of aerosol particles ($\text{PM}_{2.5}$). The lower temperatures or higher $\text{PM}_{2.5}$ concentrations lead to increased fraction of oxidized mercury partitioned into the particulate phase [Rutter and Schauer, 2007; Amos *et al.*, 2012].

Global data on air concentration of particulate matter with high temporal resolution simulated by the MOZART model [Emmos *et al.*, 2010] were utilized to support the gas-particle partitioning scheme. Simulated monthly mean concentrations of $\text{PM}_{2.5}$ in January and July 2013 are shown in Fig. 3.5. As seen high concentrations of aerosol particles are characteristics of desert areas in Northern Africa, Middle East and surrounding areas due to wind suspension of mineral dust. The lowest PM concentrations are in the Arctic and Antarctica. The simulated $\text{PM}_{2.5}$ values reasonably well agree with observations from the EMEP monitoring network (shown by circles).

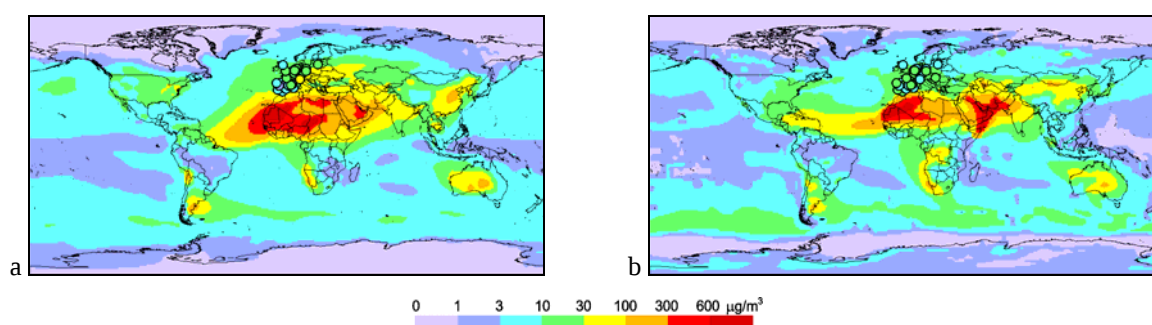


Fig. 3.5. Monthly mean air concentration of fine aerosol ($\text{PM}_{2.5}$) in January (a) and July (b) 2013 simulated by the MOZART model. Circles show $\text{PM}_{2.5}$ measurements at sites of the EMEP monitoring network

The implemented parameterization has been tested with the GLEMOS simulations on a global scale. Figure 3.6 illustrates predicted fraction of oxidized mercury partitioned into the particle phase in surface air in January and July 2013. The simulated spatial distributions of mercury partitioning differ considerably for different seasons. In January low temperatures and relatively high concentrations of

aerosol lead to elevated proportion of particulate mercury in the Northern Hemisphere (Fig. 3.6a). The fraction of $\text{Hg(II)}_{\text{part}}$ is particularly large over the continents (more than 70%). In the Arctic the proportions of gaseous and particulate phases are comparable despite of very low temperatures due to insignificant concentrations of particulate matter. In July the gas-particle partitioning of oxidized mercury shifts to the gaseous phase in the Northern Hemisphere except for areas affected by wind blown dust from desert and arid regions (Fig. 3.6b). In contrast, high fractions of particulate mercury move to the Southern Hemisphere due to the cold season and moderate concentrations of atmospheric aerosol. In general, these spatial patterns are confirmed by limited observations of speciated mercury from the GMOS monitoring network [GMOS, 2015].

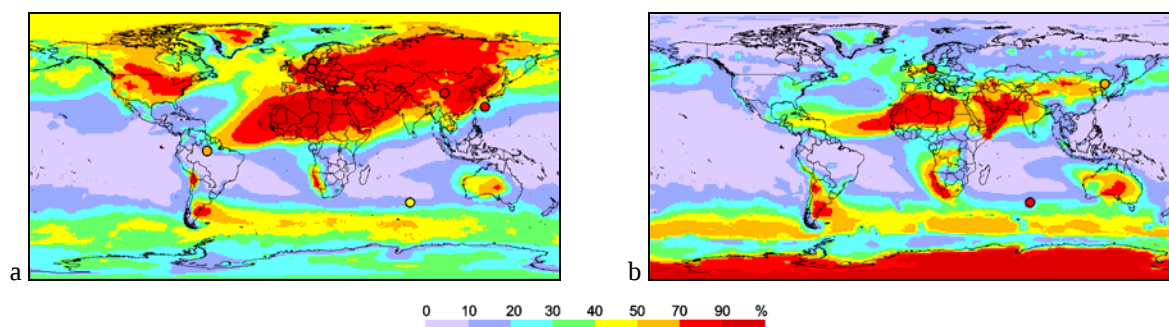


Fig. 3.6. Monthly mean fraction of oxidized mercury partitioned into the particle phase in surface air in January (a) and July (b) 2013. Circles show the measured particulate mercury fraction at sites of the GMOS network [GMOS, 2015]

Application of the improved gas-particle partitioning scheme resulted in more realistic levels of simulated particulate mercury when comparing with observations. Comparison of both simulated and measured seasonal variation of $\text{Hg(II)}_{\text{part}}$ concentration in surface air at two background sites of the EMEP monitoring network is shown in Fig. 3.7. As seen the model simulations with previous parameterization lead to significant overestimation of particulate mercury concentration during warm season. Application of the gas-particle partitioning results in much better agreement between the model and observations. However, it should be noted that observations of oxidized mercury species are available at very few monitoring sites and they are associated with large uncertainties [Gustin and Jaffe, 2010]. Therefore, extension of speciated mercury measurements along with improvement of measurement techniques are needed for further development and evaluation of the model parameterizations.

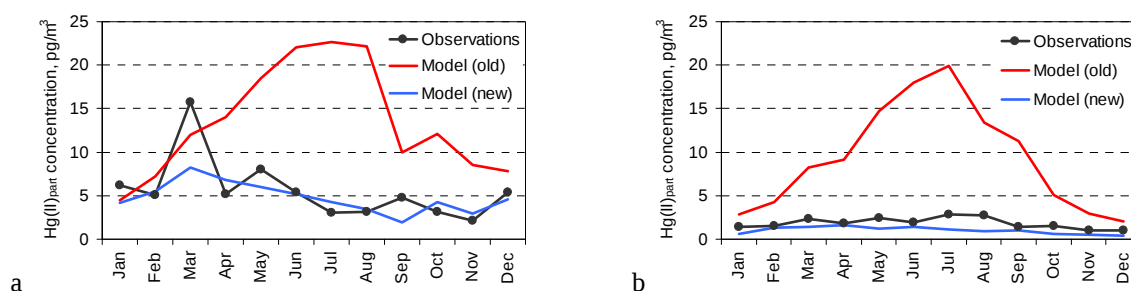


Fig. 3.7. Monthly mean air concentration of particle bound mercury ($\text{Hg(II)}_{\text{part}}$) at two EMEP sites in 2013: (a) – Råö (SE14) and (b) – Pallas (FI96). Black lines with dots show observations, coloured lines present GLEMOS simulations (red line – old parameterisation, blue line – simulation with gas-particle partitioning).

Update of the model parameterization of gas-particle partitioning of mercury in the atmosphere has allowed improving the GLEMOS model performance in mercury pollution modelling.

3.3. Model parameterization of wind re-suspension of heavy metals: further updates

According to the Work-plan of EMEP for 2014-2015, MSC-E continued to work on parameterization of heavy metal emissions due to wind re-suspension. First scheme describing re-suspension of lead and cadmium with wind-blown dust, containing heavy metals, was described in [Gusev *et al.*, 2006] and had undergone several modifications since then. The scheme calculates vertical flux of particulate matter (dust) re-suspended by wind from soils of different types of underlying surfaces. In order to obtain flux of heavy metals this dust flux is multiplied by concentration of heavy metals in soil. It is assumed that re-suspension of dust occurs from three types of underlying surfaces: bare land (mostly, deserts), arable lands and urban surfaces. According to currently used parameterization, re-suspension of lead and cadmium from urban surfaces is the main contributor to secondary emission of lead and cadmium in the EMEP domain.

In the current scheme, implemented in the model, it is assumed that re-suspension of wind-blown dust containing heavy metals is driven by wind erosion. However, in addition to this, traffic-induced turbulence can also be responsible for generation of dust flux from urban surfaces, mostly, from roads. One of schemes, describing this process, is already developed and implemented in the chemical transport model Lotus [Schaap *et al.*, 2009]. In order to improve quality of air pollution assessment, this scheme is planned to be applied in the modelling of heavy metal levels and transboundary transport for EMEP in 2016.

The scheme of wind re-suspension due to traffic links flux of dust with amount of vehicles moving along the roads (so-called vehicle-kilometer travelled, or VKT). Factor of proportionality between particle re-suspension flux and VKT is emission factor, which depends on type of road (highways, rural roads or urban roads) and on type of vehicle (heavy or light duty vehicles). Besides, meteorological conditions also affect the flux.

Input information for simulations of re-suspension of heavy metals due to traffic is spatially distributed data on VKT and concentrations of the metals in road dust. Currently this information is being collected. Pilot data on VKT, in a form of total values per European countries is derived from the Eurostat web site (<http://ec.europa.eu/eurostat/>). Example of VKT, averaged over 2005-2012 for some European countries, is shown in Fig. 3.8. Information on concentrations of heavy metals in road dust, observed in European cities was found in peer-reviewed literature [Amato *et al.*, 2013, Charlesworth *et al.*, 2003, Loreda *et al.*, 2004, Ordonez *et al.*, 2003, Robertson *et al.*, 2003, Christoforidis and Stamatis, 2009, De Miguel *et al.*, 1997] (Fig. 3.9).

In future it is planned to test the considered approach for calculation of wind re-suspension. However, further efforts are needed to collect necessary input data. Therefore, cooperation with national experts from the EMEP countries on this issue is appreciated.

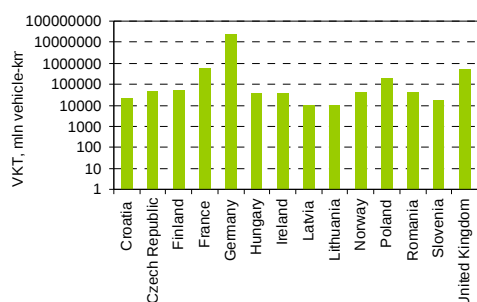


Fig. 3.8. Available VTK data for European countries averaged for 2005-2012

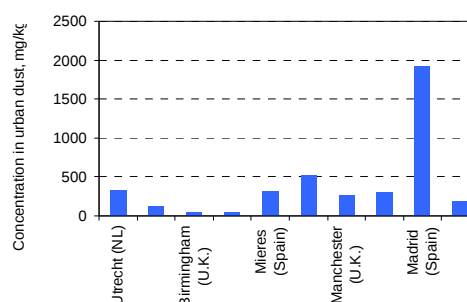


Fig. 3.9. Available information about concentrations of heavy metals in urban road dust in European cities

4. CASE STUDY OF HEAVY METAL LEVELS IN BELARUS

Country-specific case studies of heavy metal pollution levels with fine spatial resolution have been initiated by MSC-E and the Task Force on Measurements and Modelling (TFMM). The main purpose of the studies is to examine the effects of refinement of spatial resolution on the quality of pollution assessment in a country. The study allows taking into account specific features of country's relief, weather conditions, location of emission sources, national monitoring data etc. Therefore, additional objective of this activity is to provide countries with more detailed information on pollution levels, compared to that regularly produced under EMEP. Case studies for Croatia, the Czech Republic, and the Netherlands have already been completed. This year the research is focused on assessment of lead pollution levels in 2012 in Belarus.

4.1. Input data

Input data needed for simulation of pollution levels and their further analysis include emission and monitoring data, meteorological information, air concentrations at boundaries of modelling domain, geophysical information etc.

Emission data

Emission data for Belarus with resolution $10 \times 10 \text{ km}^2$ were provided by national experts, and for other countries – by CEIP with resolution $50 \times 50 \text{ km}^2$ (Fig. 4.1a). Total annual emission of lead in Belarus in 2012, submitted by national experts for this study was 8.3 tonnes. The highest emissions were concentrated in or nearby major cities of Belarus – Minsk, Vitebsk, Zhlobin etc., while outside the cities emission values were low (Fig. 4.1b). Emissions from large point sources ($> 100 \text{ kg/y}$) were concentrated in 19 grid cells, contributing about 90% to total emission in the country.

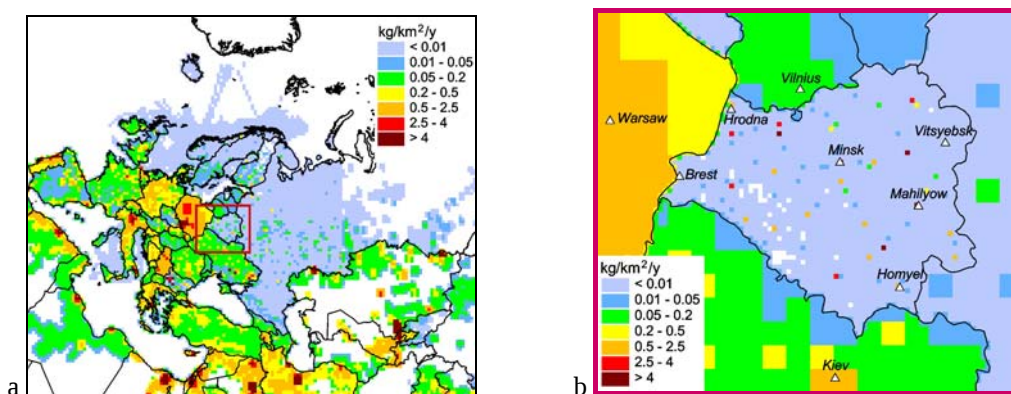


Fig. 4.1. Emission of lead ($\text{kg/km}^2/\text{y}$) in 2012 in the EMEP region (a) and in selected modelling domain including Belarus and surrounding areas (b)

Received emission data from Belarus were split into 15 emission source categories according to NFR classification. For model simulations these categories were combined in 8 groups of sources. The main group, responsible for 40% of total lead emission, was *iron and steel production*, followed by *combustion in industries* (21%) and *electricity and heat production* (19%) (Fig. 4.2). This information was used for formulation of vertical distribution of emissions in Belarus, following approach described in [Bieser et al., 2011].

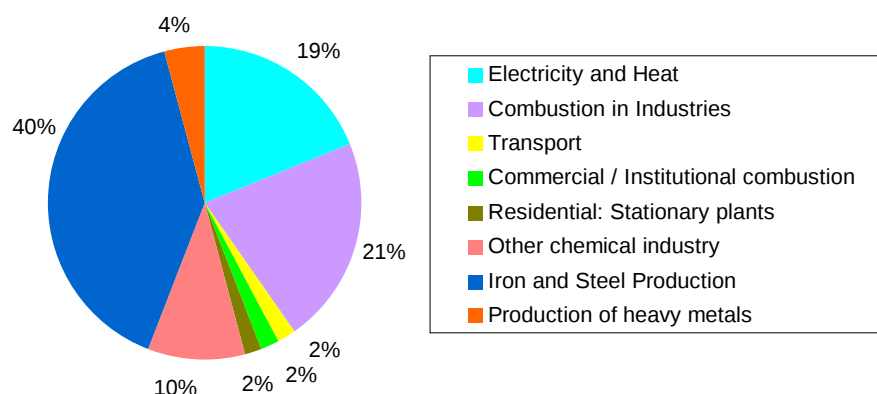


Fig. 4.2. Main groups of emission source categories of lead in Belarus

Monitoring data

Information on measured air concentrations was collected from various sources. First of all, daily-mean air concentrations of lead from Belarusian background monitoring station 'Berezinskiy reserve' were provided by national experts. Besides, information on monthly mean air concentrations measured at stations in 19 cities of Belarus, were also presented. EMEP station PL5 (Diabla Gora) in Poland is located within the selected modelling domain. Its measurements were used in the analysis of pollution levels. Finally, data from five Polish urban background stations, extracted from the EU monitoring database 'AirBase' were involved in the research. Location of stations measuring lead air concentrations in the agreed modelling domain is depicted in Fig. 4.3.

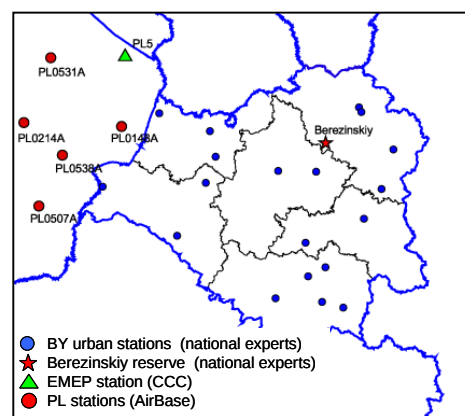


Fig. 4.3. Location of monitoring stations (Pb in air) in the selected domain.

Meteorological data

Meteorological data were prepared by MSC-E with 10x10 km² spatial resolution using nesting option of MM5 meteorological model [Grell *et al.*, 1995]. Comparison of meteorological variables simulated by MM5 with those calculated with coarser resolution (50 x 50 km²) and with observations demonstrated improvement of quality of meteorological information due to the increase of spatial resolution.

Meteorological parameters in the selected domain were exemplified by calculated and observed annual precipitation sums (Fig. 4.4a). As seen, spatial distribution of calculated precipitation generally agreed with observations, which was confirmed by scatter plot (Fig. 4.4b).

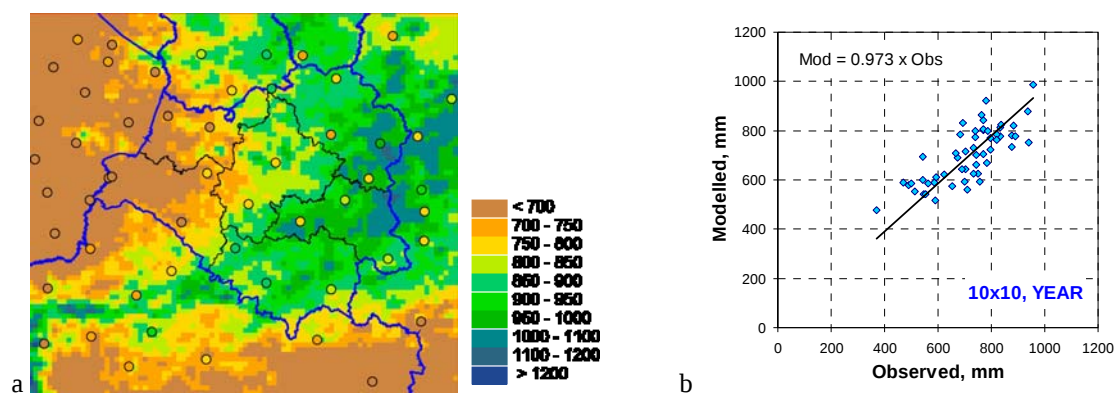


Fig. 4.4. Spatial distribution of modelled (field) and observed (circles) annual precipitation in the selected domain (a) and comparison of modelled precipitation against observed values (b)

Other data

Boundary concentrations at the borders of the selected domain were produced by modelling of air concentrations over the EMEP region with 50-km resolution. Land cover map with fine spatial resolution was generated on the base of MODIS satellite information (www.modis.bu.edu/landcover/). Data concentrations of lead in soils were applied for calculation of wind re-suspension. Map of concentrations of lead in soil used for these calculations, was constructed from two datasets. The first one included lead concentrations in soils in cities of Belarus, provided by national experts. The second dataset was derived from the European-wide project FOREGS (www.gtk.fi/publ/foregsatlas/).

4.2. Analysis of preliminary results

On the base of country-specific input information deposition and concentration levels with fine spatial resolution in Belarus have been simulated. Spatial distribution of lead deposition over the selected domain is non-uniform. Two zones of relatively high deposition are indicated. The first one is noted for Poland, caused mostly by significant emissions in this country (Fig. 4.5a). The second zone is located in the south-east part of Belarus, which caused by combination of two factors: emission sources in the Homel region of Belarus (Zhlobin, Rechitsa, Homel, Mozyr, Svetlogorsk cities) as well as peculiarities of spatial distribution of atmospheric precipitation.

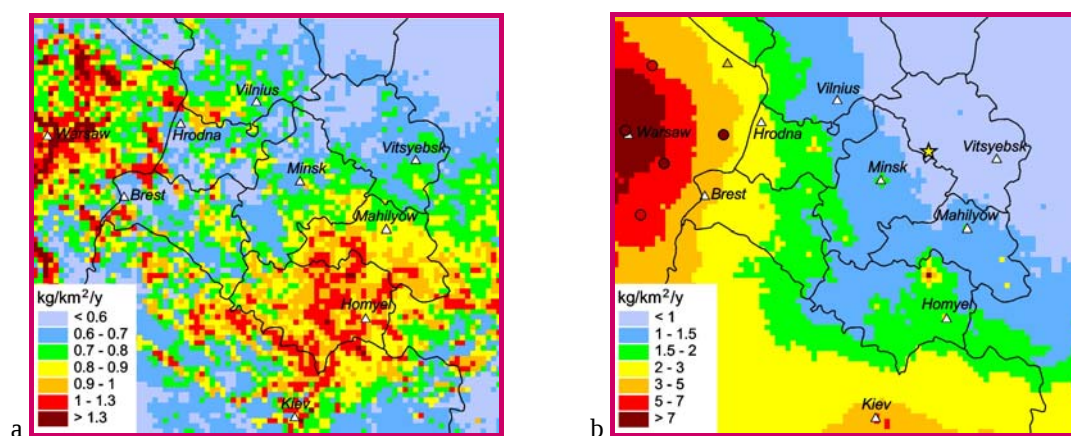


Fig. 4.5. Total annual deposition (a) and annual mean concentrations in air (b) of lead with resolution 10x10 km².

Spatial distribution of concentrations in air demonstrated gradient from the western and south-western parts of the country to the north-eastern part (Fig. 4.5b). This peculiarity was mostly caused by distribution of emissions in Belarus and in the neighbouring countries – Poland, Ukraine and Russia. The effect of fine resolution was clearly revealed over the Belarusian territory where local maximums of air concentrations were seen around cities.

Simulated concentrations of lead in air were compared with available measurement data. Concentrations of lead observed at the Belarusian background station 'Berezinskiy reserve' were underestimated by the model though the most part of the year (Fig. 4.6). Annual mean modelled concentration at this station was around 0.8 ng/m^3 , while the observed concentration was around 2.3 ng/m^3 . It should be noted that this station does not belong to the EMEP monitoring network. For better understanding of discrepancies between modelled and observed levels it would be good if national laboratory, which analyses heavy metal measurements from this station, could take part in regular intercomparison of analytical methods coordinated by CCC.

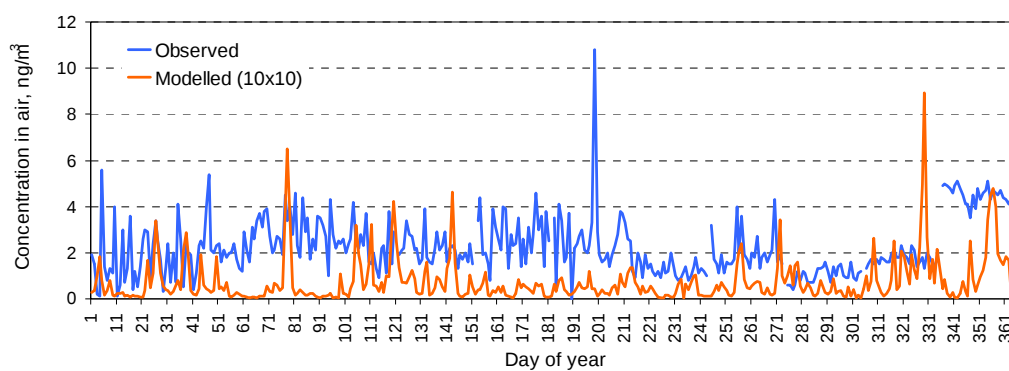


Fig. 4.6. Daily mean modelled and observed concentrations of lead at station 'Berezinskiy reserve' in Belarus in 2012.

Besides the Belarusian station, other monitoring information was involved in the analysis. At the EMEP station PL5 (Diabla Gora, Poland) the modelled concentrations well captured observed concentrations in warm period, while in cold period the model underestimated the observed levels. Similar situation took place for most of the other non-EMEP background urban Polish stations (Fig. 4.7). Relatively high levels of observed air concentrations at Polish stations could be caused by the effect of domestic heating in cold part of a year. One of possible reasons for this discrepancy is lack of information on seasonal variability of emissions in the reported emission data. Therefore, discussion about seasonal variability of emissions with experts from Poland would be useful for improvement of the model assessment in Belarus and surrounding regions.

In order to establish regions which emissions could contribute to observed levels not captured by the model, analysis of back trajectories has been started. In some episodes (e.g., 27-29 of April) air masses were transported through the southern part of Poland and the central regions of Europe, known for relatively high emissions (Fig. 4.8a). In this episode modelled levels were close to the observed ones. In other episodes (e.g., 04-08 of March), when the model significantly underestimated observed levels, air masses travelled from the north-western regions of the Russian Federation, where emissions were low (Fig. 4.8b). It is planned further to implement inverse modelling approach and to construct emission scenarios in cooperation with national experts from Belarus, in order to diminish discrepancies between observed and modelled values.

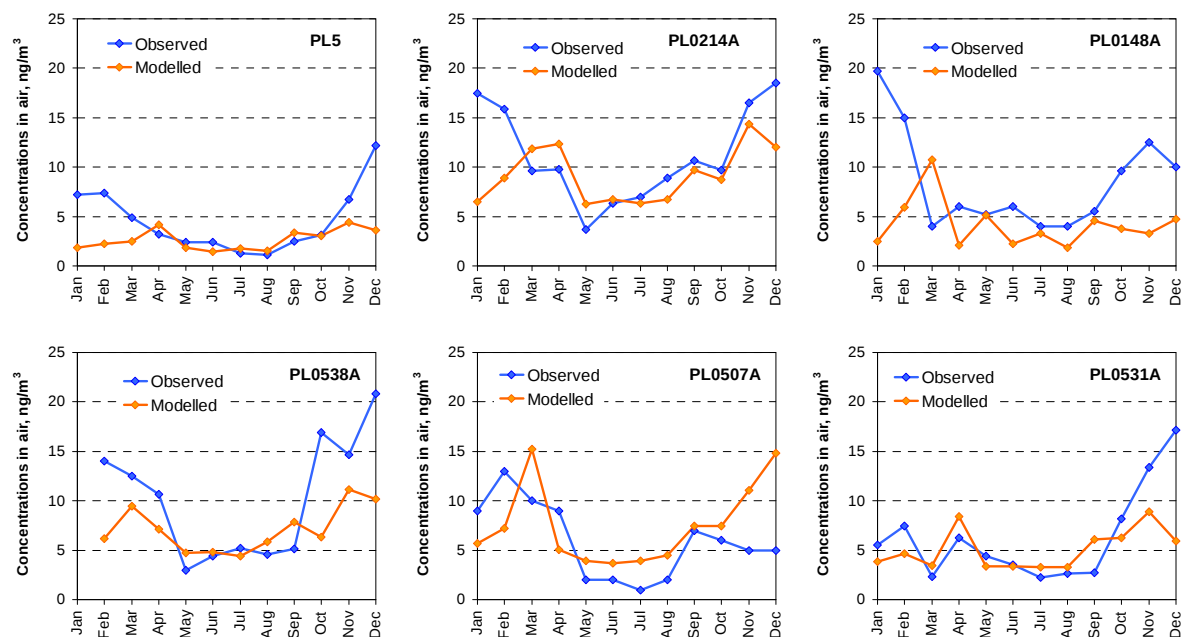


Fig. 4.7. Modelled and observed concentrations of lead for stations in Poland in 2012: EMEP station PL5 (top left diagram) and stations from EU AirBase monitoring data base (the rest diagrams).

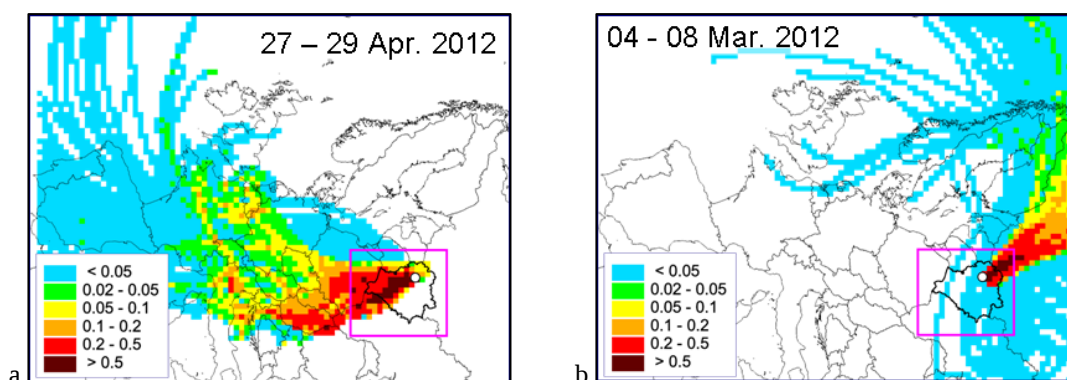


Fig. 4.8. Back trajectories (dimensionless units) for station 'Berezinskiy reserve' in the period from 27th to 29th of April (a) and from 4th to 8th of March (b).

Uncertainties of emission data could contribute to the discrepancies between modelled and measured levels. Available monitoring data were compared with emissions in Belarus and in the other EMEP countries. On one hand, observed concentrations at the background station 'Berezinskiy reserve' were similar to those observed at the EMEP stations in the other countries, namely in Germany, Spain, Slovenia, the United Kingdom (Fig. 4.9). The same was true for concentrations measured at urban stations in Belarus and other countries in different parts of Europe (Fig. 4.10a). On the other hand, comparison of emission fluxes indicated that country-averaged flux of lead

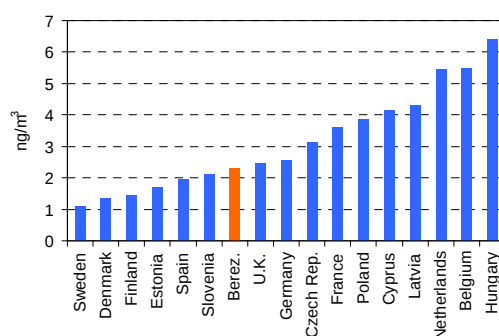


Fig. 4.9. Mean concentrations of lead observed at EMEP stations and in background station 'Berezinskiy reserve'

emission in Belarus was lower than those in other countries by an order of magnitude (Fig. 4.10b). However, when country-average emissions of other pollutants were considered, e.g., particular matter, the emission fluxes in Belarus and other countries were comparable (Fig. 4.10c). Cooperation between the EMEP centres (CEIP, CCC) and national experts from Belarus and neighbouring countries is needed to understand and explain the inconsistency between emission and monitoring data.

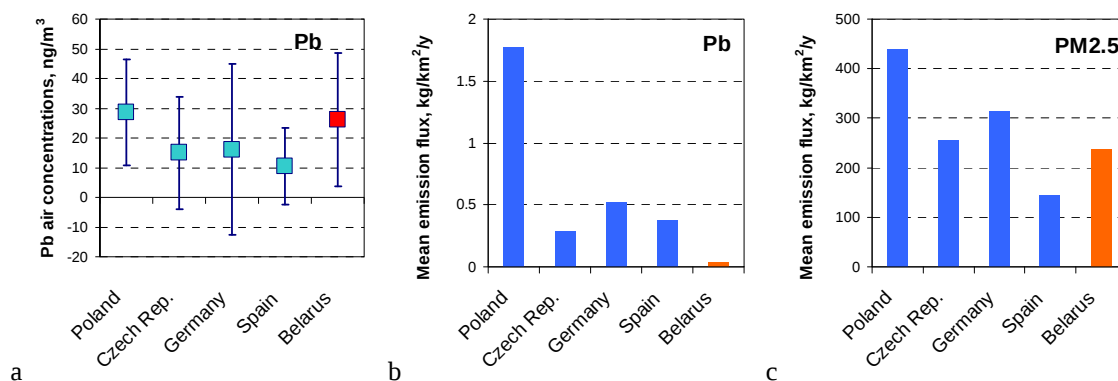


Fig. 4.10. Measured concentrations of lead at urban stations in selected countries and Belarus in 2012 (a) and spatially averaged emissions of lead (b) and PM2.5 (c) in the same countries. Boxes indicate mean value, and whiskers mean $\pm$ standard deviation.

4.3. Future steps of the country-specific study

Taking into account priorities of the Long-term strategy for the Convention and increasing interest to the EECCA countries [ECE/EB.AIR/106/Add.1], the work on assessment of heavy metal pollution levels in Belarus will be continued. The case study for Belarus could serve as a test approach to evaluation of pollution levels in the other EECCA countries.

The further possible steps in the field of assessment of heavy metal levels in Belarus are the following:

1. Application of inverse modelling approach and construction of emission scenarios in order to investigate uncertainties of emission data and to reduce discrepancies between modelled and measured values at monitoring stations
2. Further cooperation with the EMEP centres and national experts both from Belarus and from neighboring countries regarding emission and monitoring issues will be continued. Besides, taking into account considerable influence of the neighboring countries on pollution levels in Belarus of modelling domain is proposed to be extended in future.
3. A number of other pollutants, e.g., sulphur or nitrogen are characterized, as a rule, by lower uncertainties of their emission and monitoring data compared to heavy metals. Therefore, it is important to include these pollutants in the analysis.
4. Prepare detailed country-specific information for Belarus with fine spatial resolution, including:
 - a. Maps of concentrations and deposition
 - b. Contributions of foreign and secondary sources to deposition in Belarus and its regions

- c. Pollution levels in individual regions
- d. Contribution of emission source categories to pollution in the country
- e. Background pollution levels for Belarusian cities

Model assessment of lead pollution levels in Belarus with fine spatial resolution has been initiated. Special attention is paid to the analysis of initial data (emissions and measurements) and discrepancies between simulated and observed levels. Assessment of pollution levels in the EECCA countries requires development of specific approaches, because of scarce background monitoring network and insufficient emission data in most of these countries. The research carried out for Belarus could serve as a test approach to evaluation of pollution levels in the other EECCA countries.

5. COOPERATION AND DISSEMINATION OF INFORMATION

5.1. Subsidiary bodies to the Convention

5.1.1. Task Force on Measurements and Modelling (TFMM)

Analysis of trends

MSC-E took part in the Workshop on trend analysis held in Paris, France, 17-18 November 2014. The main aim of the workshop was to agree methodology for analysis of air pollution trends occurred in the last two decades. At the Workshop MSC-E presented methodology of analysis of long-term trends for heavy metals and POPs. The methodology was elaborated taking into account pronounced non-linearity of HM and POP trends during sufficiently large period (for example, from 1990 to 2012). It could be applied for the analysis of long-term tendencies in observed and calculated levels at monitoring stations as well as concentrations and deposition in the entire EMEP region, its sub-regions (including the EECCA region) and particular countries. A list of parameters characterizing trends was suggested. It included minimum, maximum, average and total pollution level reduction rates, non-linearity, seasonality and phase shift of trends. MSC-E was invited to give the description of the methodology and elaborate the tool for calculation of non-linear trends. At present the description of the methodology of trend analysis suggested by MSC-E and source code of the program for calculation of trends and its characteristics (together with operational manual and examples of application of the program to time series of air pollution) are allocated on the TFMM wiki-page.

The workshop participants agreed to consider two periods for analysis of trends: 1990-2012 and 1999-2012. Information on pollution levels should be analyzed with annual, monthly or daily temporal resolution. Besides, it was approved to study trends with regard to air mass transport sectors. Non-linear long-term trends were suggested to analyze using methodology proposed by MSC-E, and linear trends – using Sen's slope method. It was recommended to filter out values with unexpectedly high values, but to retain values below detection limits. It was decided to avoid meteorological correction or ignoring years with anomalous meteorological conditions.

The overview of the results of trend analysis for HM pollution for the period from 1990 to 2012 is presented in Section 1.3 of the present report. These results were reported at the annual TFMM meeting in Krakow, Poland (May 2015). A number of information related to long-term HM and POP pollution changes were proposed to be highlighted in the CLRTAP Assessment Report 2016. In particular, it includes trend characteristics for modelled and observed levels at the EMEP monitoring stations, reduction rates and total reductions of HM and POP levels in the EMEP countries, analysis of factors affecting trends (changes of anthropogenic and secondary emissions, meteorological variability, boundary conditions, transboundary transport). Besides, the effect of changes of source categories of emissions and large point sources could also be overviewed in the Report.

At the meeting application of MSC-E trend tool to calculation of trends was discussed by a number of national experts. It was advised to modify the MSC-E trend analysis tool for applications to different pollutants. At present the needed modifications are done and the source code of the modified program is allocated on the TFMM wiki-page.

Country-specific case studies

MSC-E continued country-specific activity on the EMEP case study on heavy metal pollution assessment with fine spatial resolution. Preparation of initial data and preliminary results on lead pollution levels in Belarus in 2012 and analysis of the results carried out jointly by MSC-E and national experts from Belarus were demonstrated at the TFMM meeting in Krakow. Future possible steps of this activity were overviewed. More detailed information about assessment of pollution levels in Belarus is presented in Chapter 4.

Model development

Progress in further development of the Global EMEP Multi-media Modelling System (GLEMOS) were reported and discussed at the TFMM meeting. In particular, the GLEMOS was involved in multi-model evaluation of Hg atmospheric processes in the framework of the EU GMOS project. Besides, the model calculated future mercury pollution levels over the globe and in the EMEP region based on various emission scenarios. Contribution of different emission source categories to mercury pollution over the globe was evaluated. All these activities were carried out in close cooperation with experts from other countries (USA, Germany, Italy and Canada) and international organizations, such as UNEP and AMAP. Recent results of the development of the GLEMOS are described in Chapter 3 of the Report.

5.1.2. Working Group on Effects (WGE)

MSC-E contributed to the WGE assessment report on trends providing the Group with information on long-term trends of heavy metal pollution levels. In particular, gridded data on ecosystem-dependent deposition of lead, cadmium and mercury for each year of the last two decades were submitted to CCE. For calculation of exceedances of critical loads of heavy metal deposition fluxes to different ecosystems were aggregated to deposition to forests (Temperate coniferous forests, Temperate deciduous forests, Mediterranean needleleaf forests, Mediterranean broadleaf forests) and deposition to non-forested natural surfaces (Grasslands, Semi-natural surfaces, Wetlands, Tundra and Inland waters).

Information about reduction rates of ecosystem-dependent and grid-averaged deposition to the EMEP region for 1990-2012 was submitted to WGE (Fig. 5.1). Trends of deposition were linked with long-term changes of anthropogenic and secondary sources and contribution of mercury intercontinental atmospheric transport. Besides, data on reduction rates of pollution levels in particular gridcells, where EMEP monitoring stations were located, were also delivered to the Working Group.

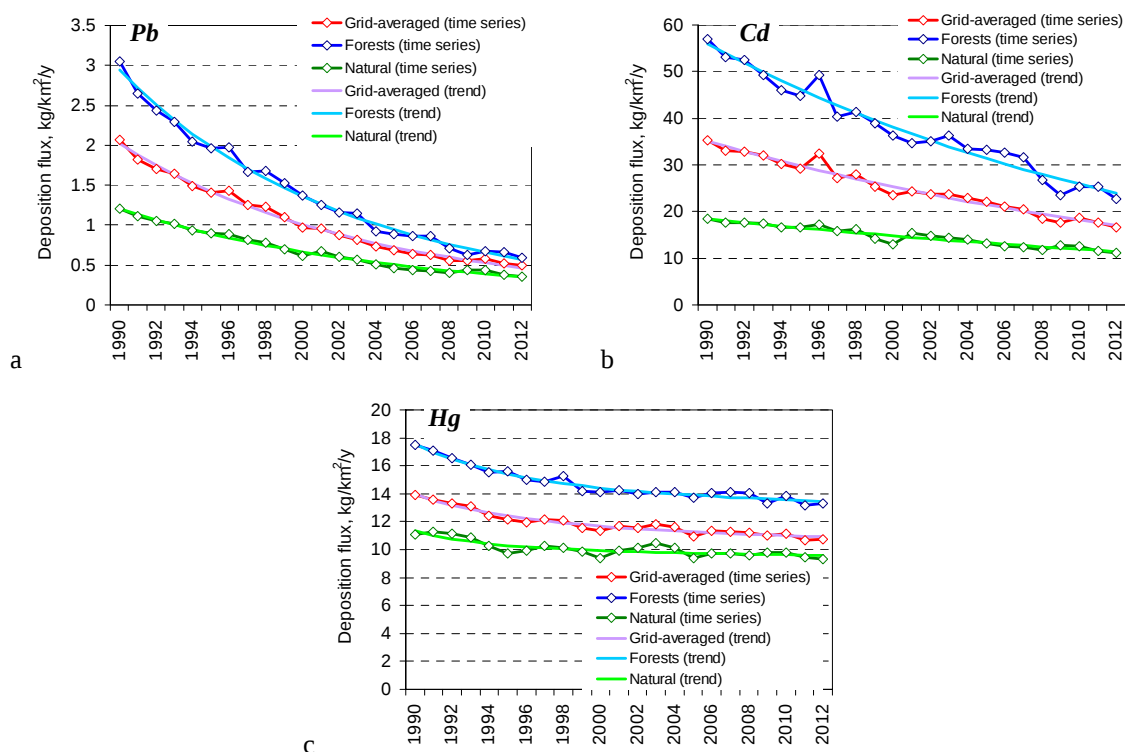


Fig. 5.1. Deposition fluxes of lead (a), cadmium (b) and mercury (c) and their main component of trends in the EMEP in 1990-2012

It was shown that deposition of lead to different ecosystems decreased by 70-80%, cadmium – by 40-60%, and mercury – by 15-23% for the considered period. Similar to deposition decline to the EMEP countries, reduction rate of deposition to ecosystems was non-uniform in time. It was the highest in the beginning and the lowest in the end of the period. It was demonstrated that the main factor responsible for decrease of ecosystem-dependent deposition of lead and cadmium was reduction of anthropogenic and secondary emissions, while in case of mercury large effect on the reduction was made by intercontinental transport.

5.2. International organizations

5.2.1. European Union and GMOS project

In the framework of co-operation with the European Commission MSC-E takes part in the EU funded project GMOS (Global Mercury Observation System). GMOS is a five-year project launched in 2010 and coming to the end this year. It is aimed at (see Fig. 5.2):

- Establishment of a global monitoring system for mercury including land-based, over-water and aircraft observations;
- Update of emission inventories for mercury and elaboration of future emission scenarios;
- Improvement and validation of regional and global scale atmospheric mercury models;
- Model application to evaluate source-receptor relationships, temporal trends and scenarios;
- Development of interoperable system for dissemination of the project output data.

MSC-E as a leader of one of the project work packages (WP7) co-ordinates the modelling activities on a global scale as well as takes part in the regional scale model assessment. The model consortium of the project consists of four global/hemispheric models and two regional scale models from different scientific groups of Europe and North America. The main achievements of model development and applications include:

- Update of modelling approaches and coupling global and regional-scale models;
- Model applications for the present-day conditions and reproduction of historical trends;
- Evaluation of mercury intercontinental transport patterns and contribution of global sources to mercury pollution in Europe;
- Forecasting mercury concentration and deposition patterns as well as source-receptor relationships for selected emission scenarios.

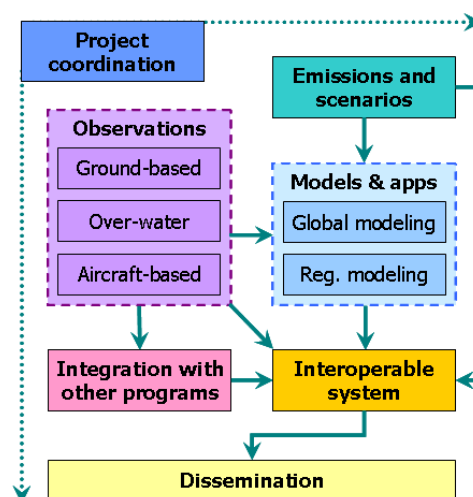


Fig. 5.2. General structure of the GMOS project

Some results of the model assessment focused on evaluation of mercury intercontinental transport and forecasting of future mercury pollution are described in Sections 1.5 and 5.2.2 of this report.

In addition, a new scientific cooperative initiative GMOS Mercury Modelling Task Force has been launched as a part of the GMOS project with support of the UNEP Mercury Air Transport and Fate Research Partnership (UNEP F&T) and the Group on Earth Observation (GEO, Task HE-02-C1). It is aimed at application of chemical transport models supplemented by comprehensive monitoring data for the improvement of current understanding of the key mercury processes in the atmosphere. MSC-E, as a GMOS partner and coordinator of global-scale modelling activities within the project performs general coordination of the Task Force and leads the multi-model assessment on a global scale.

The work of the Task Force is organized in the form of multi-model experiments aimed at answering particular scientific questions related to mercury atmospheric chemistry, anthropogenic and natural/secondary emissions, deposition etc. The simulation results are evaluated against variety of observations from the GMOS land-based network as well as measurement network of external partners, aircraft and ship-based measurements. Preliminary results of the multi-model assessment under the Task Force are discussed in Section 3.1.

MSC-E research and developments under the GMOS closely correlate with the activities carried out within the EMEP/TFHTAP and the UNEP efforts to support implementation of the Minamata Convention on Mercury.

5.2.2. UNEP Minamata Convention on Mercury

EMEP participates in various activities aimed at scientific support of international efforts to abate mercury pollution on global and regional scales. In particular, MSC-E took part in update of the UNEP Global Mercury Assessment 2013 (GMA 2013) [AMAP/UNEP, 2013]. The assessment was prepared in accordance with the request of the UNEP's Governing Council (Decision 25/5 III, paragraph 36) to support negotiations of the Minamata Convention on Mercury, a global treaty to reduce mercury pollution. GMA 2013 covered variety of aspects of mercury fate and transport in the environment including emissions to air and water, dispersion and chemical transformations in the atmosphere and aquatic environments, and exchange fluxes between different environmental media. Evaluation of mercury pollution levels on a global scale was based on the analysis of available observational data and modelling results.

The update of GMA 2013 was aimed at renewal of the assessment with new model simulation results focused on evaluation of mercury intercontinental transport and source attribution of mercury deposition using the up-to-date global inventory of mercury anthropogenic emissions. In the study MSC-E coordinated multi-model assessment of mercury global pollution. An ensemble of three chemical transport models – GLEMOS (MSC-E), GEOS-Chem (Massachusetts Institute of Technology), GMHG (Environment Canada) – was applied for evaluation of mercury atmospheric dispersion and deposition on a global scale in 2013. The models differ significantly in their formulation including different horizontal and vertical spatial resolution, description of mercury atmospheric chemistry and parameterization of specific processes. Thus, the multi-model ensemble reflects variety of contemporary approaches applied for simulations of mercury contamination and partly characterizes uncertainties associated with gaps in knowledge on mercury processes in the environment.

Simulated spatial distributions of GEM concentration in ambient air and total mercury deposition fluxes are shown in Fig. 5.3. As seen concentration of GEM has a pronounced south-to-north gradient. Mercury concentrations in the Southern Hemisphere are mostly below 1.2 ng/m³, whereas in the Northern Hemisphere they range between 1.3 and 1.4 ng/m³ over the ocean and commonly exceed 1.4 ng/m³ over land. Along with high Hg deposition fluxes over large industrial regions (East and South Asia, Europe, North America etc.) and regions with significant mercury emissions from ASGM (Southeast Asia, Central and South America, Sub-Saharan Africa) relatively large deposition is also predicted over some remote areas of the oceans and in the polar regions. The former include, for instance, elevated mercury deposition in the Intertropical Convergence Zone (ITCZ) due to high precipitation intensity. The latter are characterized by intensive oxidation of GEM in the lower troposphere during AMDEs leading to increased Hg deposition in springtime.

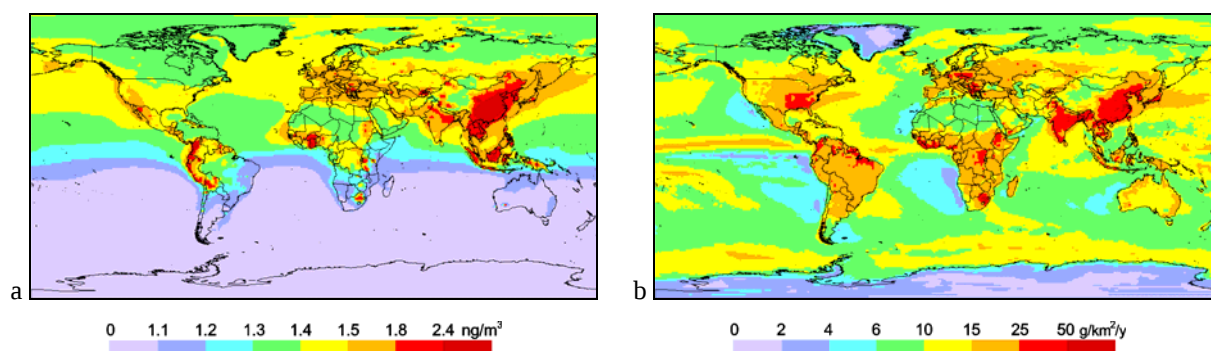


Fig. 5.3. Global distribution of ensemble mean annual GEM concentration in ambient air (a) and annual total mercury deposition (b) in 2013.

Atmospheric deposition of mercury includes significant contribution of natural and legacy sources. The shares of contemporary anthropogenic emission sources and natural/legacy emissions are shown in Fig. 5.4. The diagram also presents average Hg deposition fluxes in various geographical regions. As seen, relative contributions of these two types of emissions are comparable only in three source regions – South Asia, East Asia and Europe. In the other regions the share of current anthropogenic sources varies between 20% and 35%. Contribution of natural and legacy sources is generally higher in remote regions with lower atmospheric deposition. It should be noted that models relatively well agree in simulation of the current anthropogenic deposition. In contrast, estimates of mercury deposition from legacy and natural sources vary within a factor of 2 indicating higher uncertainty of the multi-model results for this deposition component.

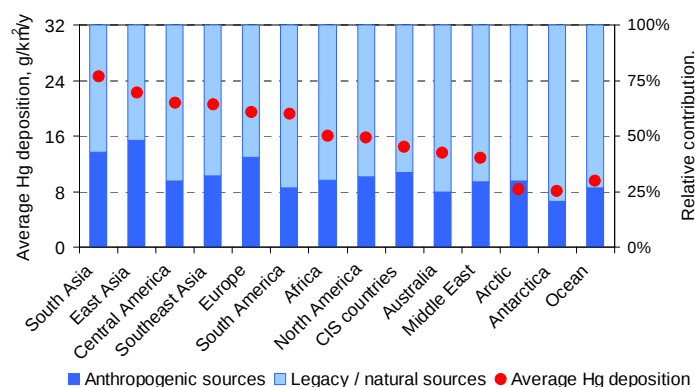


Fig. 5.4. Average mercury deposition flux over various geographical regions in 2013 and relative contribution of different source types to deposition as simulated by the model ensemble.

Comparison of relative contributions of domestic and foreign anthropogenic sources to total mercury deposition in various regions simulated by the model ensemble is illustrated in Fig. 5.5. The share of domestic sources varies from zero in Antarctica to 36% in East Asia. In most regions (except for East Asia) contribution of foreign sources is within the range 15-30%. In two regions (East and South Asia) contribution of domestic sources (25-36%) exceeds contribution of sources located outside the region (12-18%). It is determined by significant anthropogenic emissions in these regions and by dominating role of emissions from industrial and combustion sources, which include essential fraction of Hg in oxidised forms. In Europe both domestic and foreign emissions contribute almost equally (20%) to total mercury deposition. On the other hand, some other large contributors to global mercury emission – Africa, South America and Southeast Asia – are characterized by considerably lower contribution of domestic sources (6-11%). It can be explained by considerable portion of emissions from ASGM in these regions, which contain mercury in gaseous elemental form.

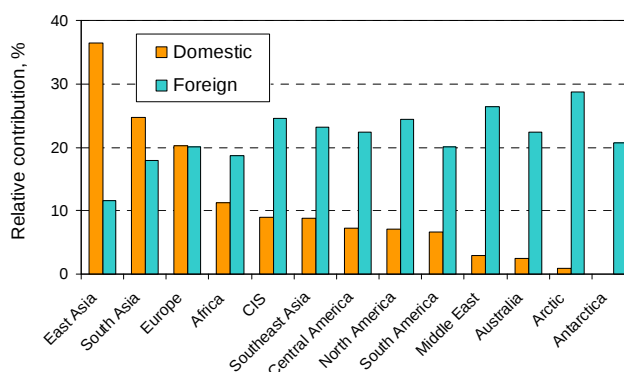


Fig. 5.5. Relative contribution of domestic and foreign anthropogenic sources to total Hg deposition over various regions

Figure 5.6 presents source apportionment of mercury deposition from contemporary anthropogenic sources to various geographical regions of the world. As it has been mentioned above mercury anthropogenic deposition to Europe consists of almost equal contributions of domestic and foreign anthropogenic emissions. The largest external contributors include East Asia (20%), Africa (8%), the CIS countries (5%), and South Asia (3%). Contribution of domestic sources to mercury deposition to North America (23%) is smaller than contribution of East Asia (32%). Africa (12%), the CIS countries (6%) and Central America (6%) are among other significant contributors of deposition from contemporary anthropogenic emissions. In contrast, anthropogenic deposition of mercury to East and South Asia is dominated by contributions of domestic sources (76% and 58%, respectively). Mercury deposition to remote regions such as the Arctic and Antarctica is determined by long-range atmospheric transport from major source regions. The major contributors for the both regions are East Asia and Africa. The Arctic is also affected by emission sources located in Europe and the CIS countries, whereas Antarctica is influenced by South and Central Americas.

The model ensemble was also applied for simulation of mercury deposition from different anthropogenic emissions sectors. An additional subset of the global mercury emissions inventory 2010 that provided sector specific emissions data was used for this purpose [AMAP, 2014]. In the subset the sectors of mercury anthropogenic emissions were aggregated into three general groups:

- Stationary combustions sources including power plants and distributed heating;
- Industrial sources including stationary combustion for industry;
- Intentional use and product waste associated sectors including artisanal and small-scale gold mining.

The sectoral composition of mercury deposition in different geographical regions is illustrated in Fig. 5.7. As seen anthropogenic mercury deposition in the major source regions is largely determined by stationary combustion and industrial sources. These emission sectors dominate in Asia, Europe, North America and the CIS countries. In contrast, regions with smaller emissions (Central and South America, Australia and New Zealand, the Arctic etc.) are significantly affected by mercury from intentional use and product waste associated sectors.

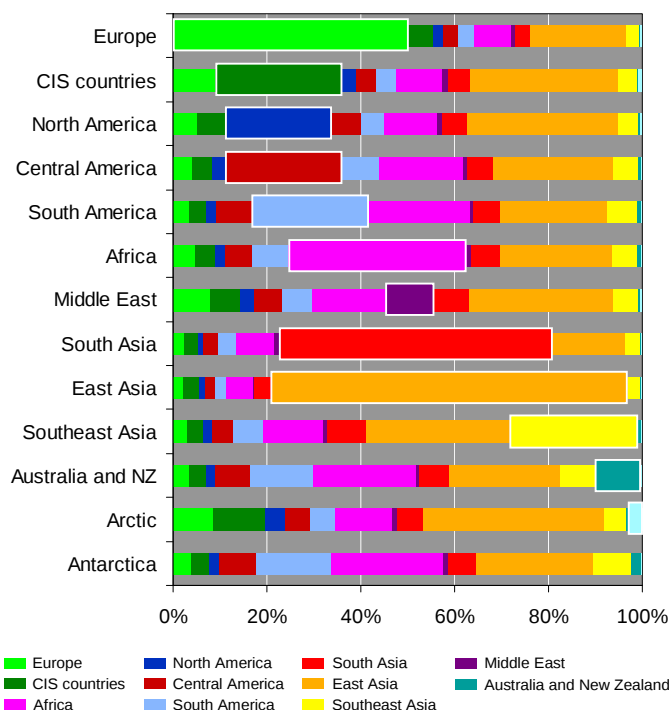


Fig. 5.6. Source apportionment of mercury deposition from contemporary anthropogenic emissions to various geographical regions in 2013. The wider bars indicate the mercury contribution from sources within the region itself.

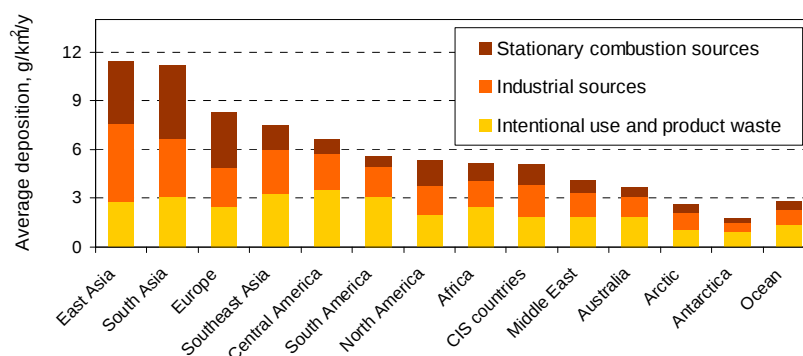


Fig. 5.7. Contribution of the groups of emission sectors to average mercury deposition from contemporary anthropogenic sources over various regions in 2013 simulated by the model ensemble.

Results of the assessment were published in a special AMAP/UNEP Technical Report [AMAP/UNEP, 2015] and presented at the sixth session of the intergovernmental negotiating committee on mercury (INC6) (3-7 November 2014, Thailand).

5.2.3. Helsinki Commission

Information on airborne pollution load to marginal seas by heavy metals within the EMEP region is of interest for 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 and is based on the long-term EMEP/HELCOM contract.

In 2014 assessment of HM pollution load to the Baltic Sea [Valiyaveetil *et al.*, 2014] included information on long-term changes of Pb, Cd, and Hg deposition during the period 1990-2012. In addition, the source apportionment of HM deposition was carried out for 2012. Short summary of information on the Baltic Sea contamination by HMs is available in a form of indicator fact sheets published in the Internet on the HELCOM website (<http://www.helcom.fi>). Anthropogenic emissions of the HELCOM countries dropped substantially since 1990 (by 89% for lead, 58% for cadmium, and 65% for mercury). Among the HELCOM countries Poland, Russia and Germany were the biggest contributors to HM emissions in 2012.

Model evaluation of pollution levels indicates marked decrease of annual total atmospheric deposition of heavy metals to the Baltic Sea in the period from 1990 to 2012. The most significant drop in HM deposition is obtained for lead (79%), followed by cadmium (53%), and mercury (23%) (Fig. 5.8a). Changes of heavy metal pollution in different parts of the Sea are not homogeneous. Particularly, significant decrease in HM deposition is noted for the western sub-basins (more than 80% for lead and about 60% for cadmium and mercury). Changes of HM pollution levels in the northern and the eastern sub-basins are lower.

Spatial variations of heavy metal deposition to the Baltic Sea in 2012 are illustrated in Fig. 5.8b by mercury deposition fluxes. Elevated levels of pollution can be seen in the western part of the Baltic Sea (the Kattegat and the Western Baltic sub-basins) and in the eastern part (the Gulf of Finland). The largest contributions among the HELCOM countries to heavy metal deposition of the Baltic Sea are made by Poland, Germany, and Russia.

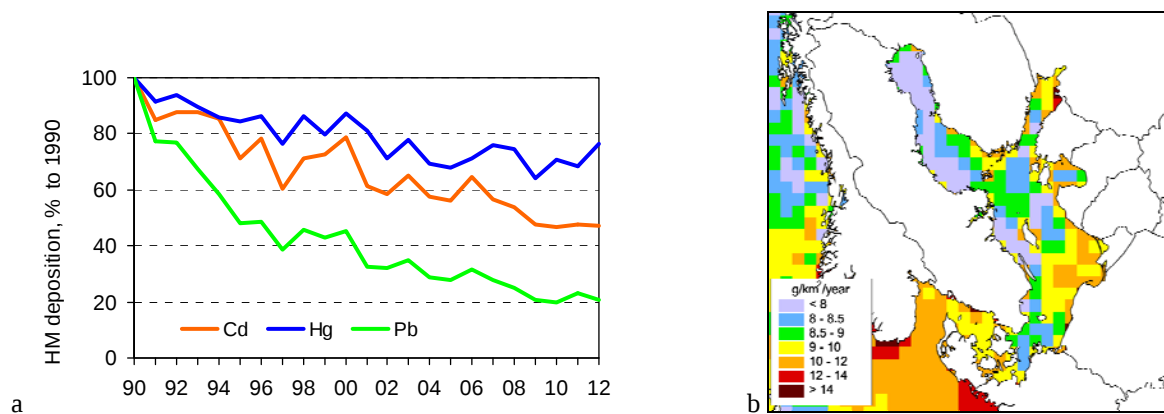


Fig. 5.8. Relative changes of total annual atmospheric deposition of cadmium, mercury, and lead to the Baltic Sea for the period 1990-2012 (a) and spatial distribution of mercury deposition over the Baltic Sea in 2012 (b)

6. MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

Main activities of the EMEP Centres MSC-E and CCC in 2016 will focus on assessment of heavy metal pollution levels in the EMEP region and support of the EMEP countries with information required for the implementation of the Protocol on Heavy Metals. Special efforts will be undertaken to characterize status of heavy metal pollution in the countries of Eastern Europe, Caucasus and Central Asia. The main challenges of heavy metal pollution assessment, which need particular attention and further research in 2016-2017 in accordance with the Key Messages of the CLRTAP Assessment Report, are summarized below. Proposals for the EMEP workplan for heavy metals in 2016-2017 are given in Annex C.

- Information on long-term changes of heavy metal pollution is important for understanding effectiveness of environmental policy in the EMEP countries and, in particular, for the implementation of the Protocol on Heavy Metals. On-going evaluation of long-term trends for heavy metal pollution over the last two decades will be completed to support the CLRTAP Assessment Report. The provided information will include characteristics of pollution trends both in the EMEP region and in particular EMEP countries as well as factors affecting the long-term changes of pollution levels.
- As a basis for model development, there is a need for better knowledge on occurrence of heavy metals (particularly Hg) in relevant environmental compartments and with global geographical coverage. To meet this purpose the Global EMEP Multi-media Modelling System will be further developed. The research and development will be devoted to elaboration of the multi-media approach for mercury modelling with focus on marine and terrestrial environments. It will allow improving model capability to simulate pollutant exchange between different compartments and will facilitate better understanding of mercury bioaccumulation/biomagnification in food chains.
- In order to investigate effects of transition to finer spatial resolution, the EMEP country-specific case studies focused on heavy metal pollution assessment on a country scale will be continued. Assessment of pollution levels in the EECCA countries requires development of specific approaches, because of scarce background monitoring network and insufficient emission data in most of these countries. The research carried out for Belarus could serve as a test approach to evaluation of pollution levels in the other EECCA countries.
- There is a need for better coverage of Eastern and Southern Europe and, especially, the EECCA countries with monitoring data on heavy metals. The data coverage could be improved through more active involvement of national data as well as data from international programmes and projects. In addition to this, alternative monitoring approaches could be used, e.g., biomonitoring, passive sampling methods etc.
- Uncertainties of the available emission data are still significant. Alternative methods for emission inventories for heavy metals could be applied based on a combination of monitoring and modelling at regional and local scales including inverse modelling techniques. In addition, historical emission data are needed to be able to take into account long-term accumulation and re-emissions of mercury.

REFERENCES

- Aas W., Bohlin-Nizzetto P. [2015]. Heavy metals and POP measurements 2013, Norwegian Institute for Air Research. EMEP/CCC-Report 3/2015.
- AMAP [2014] Global Anthropogenic Emissions of Mercury to the Atmosphere (<http://www.amap.no/mercury-emissions/datasets>).
- AMAP/UNEP [2008] Technical Background Report to the Global Atmospheric Mercury Assessment. Arctic Monitoring and Assessment Programme / UNEP Chemicals Branch. 159 pp. Online at: www.chem.unep.ch/mercury/Atmospheric_Emissions/Technical_background_report.pdf
- AMAP/UNEP [2013] Technical Background Report for the Global Mercury Assessment 2013. Arctic Monitoring and Assessment Programme, Oslo, Norway / UNEP Chemicals Branch, Geneva, Switzerland. vi + 263 pp. (<http://www.amap.no/documents/download/1265>).
- AMAP/UNEP [2015] AMAP/UNEP. Global Mercury Modelling: Update of Modelling Results in the Global Mercury Assessment 2013. Arctic Monitoring and Assessment Programme, Oslo, Norway/UNEP Chemicals Branch, Geneva, Switzerland. iv + 32 pp. Available at <http://www.amap.no/documents/doc/global-mercury-modelling-update-of-modelling-results-in-the-global-mercury-assessment-2013/1218>
- Amato F., Schaap M., Denier van der Gon H.A.C., Pandolfi M., Alastuey A., Keuken M., Querol X. [2013]. Short-term variability of mineral dust, metals and carbon emission from road dust resuspension. *Atmospheric Environment*, vol. 74, pp. 134 - 140.
- Amos H.M., Jacob D.J., Holmes C.D., Fisher J.A., Wang Q., Yantosca R.M., Corbitt E.S., Galarneau E., Rutter A.P., Gustin M.S., Steffen A., Schauer J.J., Graydon J.A., St. Louis V.L., Talbot R.W., Edgerton E.S., Zhang Y., Sunderland E. M. [2012] Gas-particle partitioning of atmospheric Hg(II) and its effect on global mercury deposition. *Atmos. Chem. Phys.*, 12, 591–603.
- Bazzano A., Ardini F., Becagli S., Traversi R., Udusti R., Cappelletti D. and Grotti M. [2015]. Source assessment of atmospheric lead measured at Ny-Ålesund, Svalbard. *Atmospheric Environment* 113, 20 – 26.
- Berdowski J.J.M., Baas J., Bloos J.P.J., Visschedijk A.J.H. and P.Y.J. Zandveld [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, UBA-FB report 104 02 672/03, Apeldoorn, 239 p.
- Bieser J., Aulinger A., Matthias V., Quante M., Denier van der Gon H.A.C. [2011]. Vertical emission profiles for Europe based on plume rise calculations. *Environmental Pollution*, Volume 159, Issue 10, October 2011, Pages 2935–2946.
- Charlesworth S. M., Everett M., McCarthy R., Ordonez A., and De Miguel E. [2003]. A comparative study of heavy metal concentration and distribution in deposited street dusts in a large and a small urban area: Birmingham and Coventry, West Midlands, UK. *Environment International*, vol. 29(5), pp. 563–573.
- Christoforidis A. and Stamatis N. [2009]. Heavy metal contamination in street dust and roadside soil along the major national road in Kavala's region, Greece. *Geoderma*, vol. 151, pp. 257–263
- De Miguel E., Llamas J. F., Chacon E., Berg T., Larssen S., Røyset O., and Vadset M. [1997] Origin and patterns of distribution of trace elements in street dust. Unleaded petrol and urban lead. *Atmospheric Environment*, vol. 31, pp. 2733–2740.
- Denier van der Gon D. H.A.C., van het Bolscher M., Visschedijk A.J.H. and Zandveld P.Y.J. [2005] Study to the effectiveness of the UNECE Heavy Metals Protocol and costs of possible additional measures. Phase I: Estimation of emission reduction resulting from the implementation of the HM Protocol. TNO-report B&O-A R 2005/193.
- EMEP [2015] EMEP monitoring network. <http://ebas.nilu.no/Pages/DataSetList.aspx?key=D6326693BE254F6F9F3DDBDB750037DA>. Accessed on June 25.
- Emmons L. K., Walters S., Hess P. G., Lamarque J.-F., Pfister G. G., Fillmore D., Granier C., Guenther A., Kinnison D., Laepple T., Orlando J., Tie X., Tyndall G., Wiedinmyer C., Baughcum S. L., and Kloster S. [2010] Description and evaluation of the model for ozone and related chemical tracers, version 4 (MOZART-4). *Geosci. Model Dev.* 3, 43–67.

- GMOS [2015] GMOS global monitoring network for mercury. <http://www.gmos.eu/sdi/>. Accessed on June 25.
- Grell G., Dudhia J. and Stauffer D. [1995] A Description of the Fifth Generation Penn State/NCAR Mesoscale Model (MM5). NCAR Tech. Note, TN-398+STR, p.117.
- Gusev A., I. Ilyin, L.Mantseva, O.Rozovskaya, V. Shatalov, O. Travnikov [2006] Progress in further development of MSCE-HM and MSCE-POP models (implementation of the model review recommendations) EMEP/MSC-E Technical Report 4/2006.
- Gustin M. and Jaffe D. [2010] Reducing the uncertainty in measurement and understanding of mercury in the atmosphere, *Environ. Sci. Technol.*, 44, 2222–2227, doi:10.1021/es902736k.
- Lindberg S. E., Brooks S., Lin C.-J., Scott K. J., Landis M. S., Stevens R. K., Goodsite M., and Richter A. [2002] Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise, *Environ. Sci. Technol.*, 36, 1245–1256.
- Loredo J., Ordonez A., Charlesworth S., and De Miguel E. [2004] Influence of industry on the geochemical urban environment of Mieres (Spain) and associated health risk. *Environmental Geochemistry and Health*, vol. 25(3), pp.307–323.
- Mareckova K., R. Wankmueller, L. Moosmann, M. Pinterits and M. Tista [2014] Inventory Review 2014, Technical Report 1/2014, CEIP, ISBN 978-3-99004-300-4.
- NADP/MDN [2015] National Atmospheric Deposition Program (NRSP-3). Mercury Deposition Network (MDN). NADP Program Office, Illinois State Water Survey, 2204 Griffith Dr., Champaign, IL 61820. <http://nadp.sws.uiuc.edu/MDN/>. Accessed on June 25, 2015
- Ordonez A., Loredo J., De Miguel E., and Charlesworth S. M. [2003]. Distribution of heavy metals in the street dusts and soils of an industrial city in northern Spain. *Archives of Environmental Contamination and Toxicology*, vol. 44, pp.160–170.
- Pacyna E.G. and J.M. Pacyna [2002] Global emission of mercury from anthropogenic sources in 1995. *Water, Air and Soil Pollution*, 137: 149-165.
- Pacyna E.G., J.M. Pacyna, F. Steenhuisen and S. Wilson [2006] Global anthropogenic mercury emission inventory for 2000. *Atmospheric Environment*, 40, 4048-4063.
- Pacyna E.G., J.M. Pacyna, K. Sundseth, J. Munthe, K. Kindbom, S. Wilson, F. Steenhuisen and P. Maxson [2009] Global emission of mercury to the atmosphere from anthropogenic sources in 2005 and projections to 2020. *Atmospheric Environment*, 44:2487-2499.
- Robertson L. [2004] Extended back-trajectories by means of adjoint equations. SMHI Report RMK No. 105, 37 p.
- Rutter A.P. and Schauer J.J. [2007] The effect of temperature on the gas-particle partitioning of reactive mercury in atmospheric aerosols, *Atmos. Environ.*, 41, 8647–8657, doi:10.1016/j.atmosenv.2007.07.024.
- Schaap M., Manders A.M.M., Hendriks E.C.J., Cnossen J.M., Segers A.J.S., Denier van der Gon H.A.C., Jozwicka M., Sauter F.J., Velders G.J.M., Matthijsen J., Builtjes P.J.H. [2009] Regional modelling of particulate matter for the Netherlands.
- Schroeder W.H., Anlauf K.G., Barrie L.A., Lu J.Y., Steffen A., Schneeberger D.R., and Berg T. [1998] Arctic springtime depletion of mercury. *Nature* 394, 331–332.
- Selin N.E. [2009] Global biogeochemical cycling of mercury: A review. *Annu. Rev. Environ. Resour.* 34, 43-63.
- Shatalov V., Ilyin I., Gusev A., Rozovskaya O., Travnikov O. [2015] Heavy Metals and Persistent Organic Pollutants: development of multi-scale modeling and trend analysis methodology. EMEP/MSC-E Technical report 1/2015.
- Steffen A., Douglas T., Amyot M., Ariya P., Aspmo K., Berg T., Bottenheim J., Brooks S., Cobbett F., Dastoor A., Dommergue A., Ebinghaus R., Ferrari C., Gardfeldt K., Goodsite M. E., Lean D., Poulain A. J., Scherz C., Skov H., Sommar J., and Temme C. (2008) A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow. *Atmos. Chem. Phys.* 8, 1445–1482.
- Smith G.K. [2002] Non-linear regression, in Encyclopedia of Environmetrics, Vol. 3, pp. 1405 – 1411.

- Tørseth K., W. Aas, K. Breivik, A. M. Fjæraa, M. Fiebig, A. G. Hjelbrekke, C. Lund Myhre, S. Solberg, and K. E. Yttri [2012]. Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972–2009. *Atmos. Chem. Phys. Discuss.*, 12, 1733-1820.
- Travnikov O. [2011] Atmospheric Transport of Mercury, in Environmental Chemistry and Toxicology of Mercury (eds G. Liu, Y. Cai and N. O'Driscoll), John Wiley & Sons, Inc., Hoboken, NJ, USA. doi: 10.1002/9781118146644.ch10.
- Valiyaveetil S., J. Bartnicki, A.Gusev, W.Aas [2014] Atmospheric supply of nitrogen, lead, cadmium, mercury and dioxins/furans to the Baltic Sea in 2012. Summary Report for HELCOM. MSC-W Technical Report 2/2014.
- UNECE [2009] EMEP monitoring strategy. ECE/EB.AIR/GE.1/2009/15.

The Table A.1 summarizes information about large ($>\pm 10\%$) differences between emissions reported by countries in 2015 and in the previous years (so-called re-calculations) covering period from 1990 to 2012.

Table A1. Large recalculations of lead, cadmium and mercury

Country	Pollutant	Range of re-calculations (larger than $\pm 10\%$)	Main reason
Bulgaria	Pb	-37% to -17%	Implementation of abatement technology in the primary copper production.
	Cd	-44% to -24%	
Croatia	Pb	-84% to -66%	Methodological improvements. Harmonization of AD (fuels amount) in stationary energy sectors. Update EF according to GB2013.
	Cd	-26% to 34%	
	Hg	-39% to -13%	
Cyprus	Cd	-21% to 67%	Methodological improvements. The implementation of the provisions of GB2013.
	Hg	-49% to -35%	
Denmark	Cd	10% to 212%	Update EF for residential wood combustion according to GB2013.
Estonia	Cd	11% to 48%	Update EF for the residential combustion sector according to GB2013.
Germany	Pb	10% to 21%	Update EF according to GB2013. Additionally calculated emission from the use of fireworks.
	Cd	15% to 45%	
Hungary	Cd	19% to 121%	Update EF according to GB2013. The use of new calculation methods.
	Hg	19% to 106%	
Ireland	Cd	-58% to -18%	The implementation of the provisions of GB2013.
	Hg	-24% to 47%	
Latvia	Hg	11% to 18%	Update EF according to GB2013.
Netherlands	Pb	-15% to -13%	Recalculations in the road transport sector.
Norway	Cd	10% to 28%	Update EF for the agricultural residue burning according to GB2013.
Moldova	Pb	-95% to 345%	Update AD. Changes in technology and calculation methods. The replacement manual data handling on automated calculations.
	Cd	-89% to 91%	
	Hg	-89% to 42%	
Poland	Cd	-62% to -45%	Change of methodology. Verification of EF for power plants, heating plants and households.
Portugal	Pb	-84% to -28%	The revision according to GB2013. The revision made by the INE (National Statistical Office) concerning the quantities of industrial waste incinerated.
Romania	Pb	-31% to 18%	Update methodology and EF according to GB2013.
	Cd	23% to 58%	
	Hg	-56% to -27%	
Serbia	Pb	-81% to -10%	The use of AD from National energy balances. The implementation of the provisions of GB2013.
	Cd	-17% to 21%	
	Hg	-34% to 12%	

Notes: AD – activity data; EF – emission factor; GB2013 - EMEP/EEA Emission Inventory Guidebook 2013

METHODOLOGY OF TREND ANALYSIS OF AIR QUALITY DATA

Here the methodology of the trend analysis for operational evaluation of trends of air quality data (air concentrations and deposition fluxes of airborne pollutants) in the framework of CLRTAP is proposed. This methodology is worked out following the decision of TFMM Workshop on trend analysis held in Paris (France) 17 – 18 November 2014.

The dynamics of air concentrations can be decomposed to trend and residual component. First of them (*trend*) describes regular variations of the initial time series, and the second (*residual component*) is conditioned by short-term and/or random perturbations of the investigated series. In turn, regular component is split into *main component* describing general decrease/increase of air concentrations during the considered period and *seasonal component* describing intra-annual variations of air concentrations.

For heavy metal time series main component normally occurs to be non-linear. Thus, for the description of the trends for these pollutants multi-exponential approach with harmonic components is applied. In fact, the approach is based on the combination of exponential trends with standard description of periodic time series (see, e.g. the monograph by T. W. Andersson, Statistical analysis of time series, Wiley-Interscience, 1994).

Analytically, the decomposition is described as follows.

For annual means (without seasonal variation component) the trend is searched in the multi-exponential form:

$$y_t = a_1 \cdot \exp(-t / \tau_1) + a_2 \cdot \exp(-t / \tau_2) + \dots + a_m \cdot \exp(-t / \tau_n) + \omega, \quad (\text{B.1})$$

where y_t are the values of the considered time series, $t = 1, N$, N being the length of the series (years), n is the number of exponentials taken into account, $\tau_1 \dots \tau_n$ are characteristic times of the considered exponentials, and $a_1 \dots a_n$ are constants. The values of τ_i and a_i , $i = 1, \dots, n$ are calculated with the help of least square method minimizing standard deviation of residue ω . The number of exponentials considered (n) should be chosen according to the form of time series. Generally, it is recommended to use two exponentials for the trend analysis (in some rare cases three exponentials are needed). The number of exponential components needed can be evaluated for the considered series using F-statistics (see [Smith, 2002]).

The following trend parameters are considered for annual resolution:

- relative reduction for the entire period R_{tot} ;
- average annual relative reduction R_{av} ;
- relative reduction for the first year (usually maximum) R_{max} ;
- relative reduction for the last year (usually minimum) R_{min} ;
- standard deviation of residual component normalized by trend component.

For monthly and daily means the trend is searched in the multi-exponential form with several harmonic compounds:

$$y_t = \exp(-t / \tau_1) \cdot (a_{10} + H_{11}(t) + \dots + H_{1k}(t)) + \exp(-t / \tau_2) \cdot (a_{20} + H_{21}(t) + \dots + H_{2k}(t)) + \dots + \exp(-t / \tau_n) \cdot (a_{n0} + H_{n1}(t) + \dots + H_{nk}(t)) + \omega, \quad (\text{B.2})$$

where k is the number of harmonic compounds considered, and $H_{ij}(t)$ is given by

$$H_{ij}(t) = (a_{ij1} \cdot \cos(2\pi/T_j \cdot t) + b_{ij1} \cdot \sin(2\pi/T_j \cdot t) + \dots + a_{ijm} \cdot \cos(m \cdot 2\pi/T_j \cdot t) + b_{ijm} \cdot \sin(m \cdot 2\pi/T_j \cdot t)), \quad (\text{B.3})$$

where T_j are periods of the considered harmonic compounds and m is the number of harmonics in each harmonic component. The numbers n , k and m should be chosen depending on the nature of time series, and all the rest coefficients are calculated by the least square method.

For monthly resolution it is recommended to use just one harmonic compound with period of 1 year (seasonal variations). In the case of daily resolution additional harmonic compounds with various periods can be used (for example weakly variations with the period 7/365.25 years). It was found that the number m of harmonics in each harmonic compound can be normally put to 2.

As described above, the trend can be decomposed to seasonal component (sum of all terms including H_{ij}) and main component (all the rest terms except for ω).

In this case the above listed parameters are calculated using main component. In addition, the following parameters are considered (normalization is performed by values of main component, including the last parameter in the above list):

- average value of normalized seasonal component (seasonality)
- average value of phase shift, that is the time from the beginning of the year and maximum of normalized seasonal component).

The above described form of trends based on the supposition that characteristic times are constant for each individual component within the entire period of consideration. However, for some types of dependence of initial time series this supposition can be violated. For such series the form of trends can be modified by the assumption that time dependence parameters $\lambda_i = 1 / \tau_i$ can slowly depend on time. For the description of such trends polynomial approximation of first or second order can be used.

The program for calculating trends of time series of air quality data for annual, monthly or daily resolution was elaborated by MSC-E following the decision of TFMM Workshop on trend analysis held in Paris (France) 17 – 18 November 2014. The program together with operational manual and Fortran codes can be downloaded from TFMM wiki page.

PROPOSALS FOR 2016-2017 WORK-PLAN

No Acti- vity	Activity description	Deliverable(s) and planned completion data	Input	Output (Cooperation with Parties and international bodies)
Ongoing activities				
1.	Calculations of concentration and deposition fields on HMs and POPs for the EMEP domain for 2013 and 2014. Evaluations against measurement data.	Annual status report on HMs (2016/2017). Annual status report on POPs (2016/2017).		EB EMEP SB WGSR WGE Countries
	Simulations of long-range transport of HMs and POPs in the new EMEP grid	Sections to annual EMEP status reports on HMs and POPs		
	Trend assessment. Analysis of temporal and spatial annual changes of pollution levels	Contribution to the CLRTAP Assessment Report		
	Generation of data on ecosystem-dependent deposition of HMs and POPs. 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 (2014/2015)		
2.	Estimates and analysis of transboundary pollution of marginal seas and the Arctic for 2014, 2015	Sections to annual status reports on HMs and POPs to EMEP SB (2016/2017) Web-accessible data on transboundary pollution of marginal seas		International bodies (HELCOM, OSPAR, AMAP)
3.	Detailed assessment of pollution levels of HMs and POPs in individual countries, based on model calculations and available national data.	Individual country reports on transboundary air pollution for 2014-2015 Web-accessible updated country specific data.	CEIP CCC	Countries
4.	Assessment of transboundary pollution of HMs and POPs in the EECCA countries to facilitate ratification and implementation of the Protocols of HMs and POPs.	Annually updated web-accessible information for 2014/2015 in Russian	EECCA countries	EECCA countries
5.	Assessment of HM and POP pollution levels with fine spatial resolution generated in cooperation with national experts. (EMEP case studies on HMs).	Technical reports jointly prepared with national experts "Assessment of pollution levels of HMs in selected countries" (2016/2017).	Belarus (cont.) Poland (2016) EECCA (2017) (to be confirmed)	SB, TFMM National experts
6.	Preparation of meteorological data for regional and global modelling and for fine scale simulations on new EMEP grid for 1990-2015.	Sections to annual status reports on HMs and POPs (2016/2017) Updated web-accessible input dataset for 2014/2015	CEIP	Countries CEIP, TFEIP

Research activity in 2016-2017 based on KMs of the CLRTAP Assessment Report

7.	Transition of operational calculations of HMs and POPs to the latitude-longitude projection with fine resolution. Analysis of changes in the pollution assessment.	Section on transition to the latitude-longitude projection to the technical report.	National experts, SB, EB, TFMM, TFHTAP
8.	Improvement of model parameterizations of atmospheric chemistry related to interaction of Hg and POPs with atmospheric aerosols	Section on atmospheric aerosols to the technical report.	TFMM National experts
9.	1.3. 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) and POPs in relevant compartments with global geographical coverage	Section to technical report	TFMM National experts UNEP/AMAP
10.	Develop alternative methods for emission inventories for HMs and POPs (including inverse modelling) based on a combination of monitoring and modelling at regional and global scales	Section to technical report on application of inverse modelling to the analysis of agreement between measurements and modelling results	TFEIP CEIP UNEP/AMAP
11.	Develop integrated approach to exploit synergies in research of emissions, long-range transport and exposure of HMs and POPs to allow a systematic identification of risks and for evaluation of options for emission control:		WGE
	11.1. Ecosystem-depended deposition fluxes of HMs and POPs to different land use types in the new EMEP grid.	Section to technical report Results and data published on web	National experts, TFMM, WGE
	11.2. Evaluation of background levels in selected cities of the EMEP countries.	Results and data published on web. Presentation of results at TFMM	National experts, TFMM
	11.3. Generate data on global transport and deposition of Hg and POPs to terrestrial and marine environment to better understand bioaccumulation/biomagnifications processes in food chains	Section to technical report on long-term accumulation of HMs and POPs in different environmental compartments	Minamata SC Conventions