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*Co-operative programme for monitoring
and evaluation of the long-range
transmission of air pollutants in Europe*

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Heavy Metals: Transboundary Pollution of the Environment

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Heavy Metal Transboundary Pollution of the Environment

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EXECUTIVE SUMMARY

Heavy metals are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) for a long time as toxic pollutants of the environment. International cooperation for pollution abatement within CLRTAP supported by continuous scientific monitoring and assessment led to considerable reduction of heavy metal pollution during the last two decades. However, significant pollution levels still remain in different parts of the UNECE region. It particularly relates to the countries of Eastern Europe, the Caucasus and Central Asia (EECCA), where information on heavy metal contamination is rather limited.

In accordance with the Protocol on Heavy Metals the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) provides the Convention with information on effectiveness of emission control measures and on pollution changes of heavy metals within the EMEP region. Different aspects of the assessment of heavy metal pollution are covered by the EMEP Scientific Centres: Centre on Emission Inventories and Projections (CEIP), Chemical Co-ordinating Centre (CCC) and Meteorological Synthesizing Centre – East (MSC-E) in collaboration with the Task Force on Measurements and Modelling (TFMM), the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), the Task Force of Emission Inventories and Projections (TFEIP), and the Working Group on Effects (WGE).

Following the recommendations of the Bureau of the EMEP Steering Body this report is focused on the major results of the EMEP Centres' activities in support of implementation of the Protocol on Heavy Metals. It presents a short summary of the current status and long-term changes of heavy metal pollution in the EMEP region. The information on heavy metal pollution of the EMEP countries includes brief description of emission data available for model assessment, overview and status of the monitoring network, and analysis of heavy metal pollution based on model estimates. A short overview of recent model developments is given in the annex to the report. Detailed description of the research and development activities is available in the Technical report [Shatalov *et al.*, 2013]. Progress in these activities was reported and discussed at the meetings of the EMEP Task Forces (TFMM, TF HTAP, TFEIP).

Emission reporting is an essential part of the pollution assessment. According to the Protocol on Heavy Metals each Party to the Convention shall report information on heavy metal emissions using methodologies specified by the EMEP Steering Body. Data on official emissions of heavy metals were provided by CEIP. As of 2013, national data on heavy metal emissions for at least one year in the period 1990-2011 were reported by 41 of the 51 Parties to the Convention. Information on the base and the last years of the period was provided by 33 countries and 27 countries submitted gridded emissions. According to the available data anthropogenic emissions were significantly reduced in the EMEP countries over the considered period. Lead emissions dropped by 90% since 1990, whereas emissions of cadmium and mercury decreased approximately by 60%.

Completeness, consistency and uncertainties of officially reported emission data on heavy metals have been recently discussed by the EMEP Task Forces (TFMM and TFEIP). Among the most critical parameters required for improvement of the assessment quality there were distinguished the completeness of the emission time-series and availability of emission spatial distribution for all EMEP countries, information on large-point sources and the key emission source categories as well as updated expert estimates used for gap-filling of emission data within the EMEP region and on a global scale.

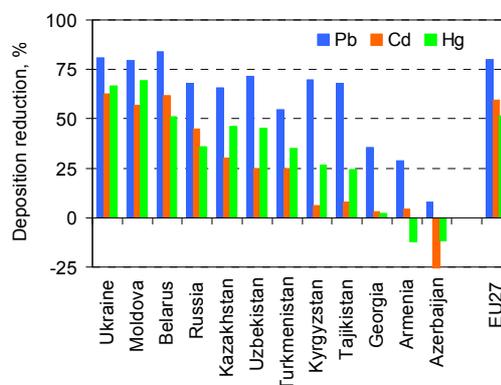
The EMEP monitoring network for heavy metals has been developing continuously since 1999. In 2011, there were 34 sites measuring heavy metals in both air and precipitation, and altogether there

were 67 measurement sites. Thirty four sites measured at least one mercury form but not all sites had complete dataset for the whole year. Among them, 15 sites measured mercury in both air and precipitation. There is a need for better coverage of heavy metal measurements and especially those of mercury in the southern and eastern parts of Europe and Central Asia. It could be partly achieved by involvement of national monitoring networks available in the countries.

According to the assessment results, deposition levels of cadmium, lead and mercury decreased in the EMEP countries between 1990 and 2011 on average by 51%, 75% and 37%, respectively. The major reduction took place in 1990-2000, whereas since 2000 the rate of pollution reduction declined. Cadmium deposition decreased by 40-60% over the most part of the EMEP domain. However, an increase of deposition is noted in the Mediterranean region as well as in the central part of Russia. Decline of lead deposition in the EMEP domain is much deeper than that of cadmium, and typically exceeds 60%. Besides, the increase is noted for the easternmost part of the Mediterranean region, while in the central part deposition has declined. As for mercury, deposition decreased in most of Europe and Central Asia by 20-60%, whereas increase of deposition occurred in the Mediterranean region and in the northern Scandinavia. It should be mentioned that meteorological variability is responsible for about 30% variation of heavy metal deposition levels in the most of the EMEP countries. It can lead to deposition increase in some countries, even when emissions continue declining.

Long-term changes of heavy metal deposition in the EMEP region differ between ecosystems and land cover types. The largest reduction takes place for urban areas located close to anthropogenic sources that are characterized by relatively rapid emission changes. Deposition to arable lands and natural surfaces (forests, grasslands, water bodies) decreases slowly due to stronger impact of slowly changed natural and secondary sources.

Pollution reduction in the Eastern Europe, Caucasus and Central Asia (EECCA) countries currently is of high priority within the LRTAP Convention. Changes of heavy metal pollution in the EECCA countries do not follow the common European trend. Decline of heavy metal deposition is generally lower, in particular, in the Central Asian and the Caucasus countries. The comparable level of pollution reduction is only a characteristic of the western part of the EECCA region, which is largely affected by emission changes in other European countries.



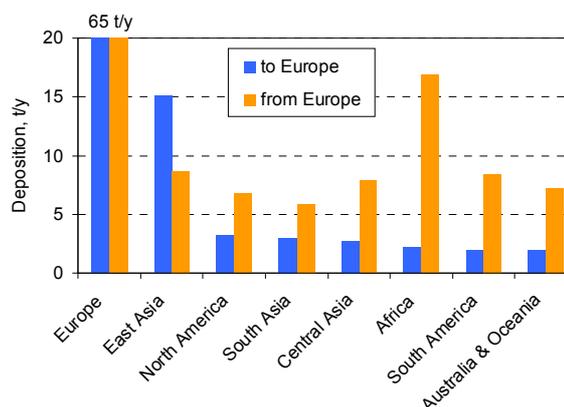
Deposition reduction of lead, cadmium and mercury in the EECCA countries and in EU27 between 1990 and 2011

However, given the large gaps of knowledge on national emissions and a lack of monitoring data, assessment of pollution levels in this region is rather uncertain. Official data on anthropogenic emissions are reported only by 5 of 12 EECCA countries. Among them, two countries report information on spatial distribution of emissions. Besides, no monitoring data on heavy metal concentration in air and precipitation are reported so far. Therefore, additional efforts are needed to facilitate development of national emission inventories and monitoring networks in these countries.

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

significant in the most of them. Long-term trend of heavy metal deposition in the EMEP countries is caused by both reduction of national emissions and changes in deposition from transboundary transport. Their relative contributions differ in different countries. Pollution changes in countries with large national emissions are mostly defined by dynamics of national sources. Nevertheless, in two thirds of the EMEP countries changes of transboundary pollution prevail over variation of domestic sources. Contribution of non-EMEP sources to lead and cadmium levels in the EMEP countries is relatively low. Therefore, contamination by these metals is mainly caused by sources located within the region. In contrast, intercontinental transport largely contributes to mercury pollution levels in the EMEP domain.

Mercury differs from other heavy metals by its ability to long-range dispersion in the atmosphere. Therefore, mercury pollution levels in Europe are largely affected by emission sources from other regions. The major external contributor to mercury anthropogenic deposition in Europe is East Asia. Transport from this region to Europe is almost double of the reverse transport from Europe to the region. As to other regions including North and South Americas, South and Central Asia, Africa, Australia and Oceania, Europe acts as a net exporter of atmospheric mercury transporting significantly more pollution to these regions than receiving from them. Assessment of mercury intercontinental transport is performed in co-operation with the Task Force on Hemispheric Transport of Air Pollution (TF HTAP).



Mutual atmospheric transport of mercury between Europe and other continents

Dissemination of the assessment results and other relevant information aimed at support of political decisions is of high importance. Annual reports containing current status of heavy metal pollution within the EMEP region is supplemented by presentation of the information on the web. It provides more flexible and targeted assistance to national experts and authorities with data required for the environment protection regulations. The country-specific information contains variety of data on emissions, measurements and model assessment for particular country collected in one place. Moreover, an additional work has been initiated to provide the EECCA countries with similar country-specific and other information in Russian.

Another important aspect is the information exchange with other international organizations and programmes. EMEP is the unique instrument within the LRTAP Convention providing regular pollution assessment and support Parties to the Convention with information on heavy metal pollution levels in Europe and other regions. Therefore, there is a wide interest outside the Convention to the data products and analysis performed by the EMEP Scientific Centres. In the context of the co-operation MSC-E regularly exchanges information with different international bodies (UNEP, AMAP, EU, HELCOM, OSPAR, etc.). These activities are performed in accordance with the EMEP workplan. However, it is important in future to formulate ways of working, outreach and information sharing to ensure more close long-term cooperation between the Convention and other relevant international bodies.

Future activities of the MSC-E research work will include further development and evaluation of the Global EMEP Multi-media Modelling System (GLEMOS). The updates will include transition to operational calculations of heavy metals in latitude-longitude projection with fine resolution, improvement of the process parameterizations related to mercury chemistry and interaction with atmospheric aerosols as well as development of multi-media approach to simulation of mercury cycling in the environment with focus on the aqueous ecosystems. Besides, future work will also include evaluation of secondary emissions of heavy metals and application of the inverse modelling approach for analysis of emissions data and modelling results. Finally, it is planned to pay a special attention to updating of the EMEP/MSC-E website and development of its Russian version to support the EECCA countries with relevant information.

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1. INTRODUCTION

Heavy metals are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) for a long time as pollutants toxic for human health and the environment. International cooperation for pollution abatement within CLRTAP supported by continuous scientific monitoring and assessment led to considerable reduction of heavy metal pollution during the last two decades. However, significant pollution levels still remain in different parts of the UNECE region. It particularly relates to the countries of Eastern Europe, the Caucasus and Central Asia (EECCA), where information on heavy metal contamination is rather limited.

EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) provides the Convention with information on effectiveness of emission control measures and on pollution changes of a range of contaminants including heavy metals. Different 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 (TFHTAP), the Task Force on Emission Inventories and Projections (TFEIP) and the Working Group on Effects (WGE).

This report presents a short summary of the current status and long-term changes of heavy metal pollution in the EMEP region. The main information on heavy metal pollution of the EMEP countries includes brief description of emission data available for modelling, overview and status of the monitoring network, and analysis of heavy metal pollution based on model estimates. The model assessment, in particular, contains characteristics of long-term changes of deposition fluxes of the priority heavy metals (lead, cadmium and mercury) in the EMEP region, transboundary transport between the EMEP countries and ecosystem-specific deposition fluxes. Additionally, atmospheric transport of mercury on a global scale is characterized in the context of the impact of other regions and continents on mercury pollution in Europe and vice versa.

Particular attention is paid to the assessment of pollution levels in the EECCA countries. Given the large gaps of knowledge on the state of emissions and a lack of monitoring data, assessment of pollution levels in this region is rather uncertain. Available estimates show that pollution changes in these countries do not follow the common European reduction, probably, because of application of outdated standards in heavy metal emitting industries and transport. Therefore, the involvement of the EECCA countries into the active co-operation is one of the priority tasks within the Convention.

Dissemination of scientific information aimed at support of political decisions is of high importance. Along with traditional annual reports significant efforts are undertaken to develop functional and effective presentation of the results on the web. The main focus of the web presentation is availability of country-specific information that could be used by national authorities for the environment protection regulations. Another important aspect is information exchange with other international organizations and programmes, which broaden the dissemination scale and strengthen the EMEP status on the international level.

Finally, significant efforts are undertaken to update and improve the modelling tools applied for the pollution assessment. Regular activities in this area are aimed at improvement of the assessment quality and include evaluation of the modelling results against measurements and development of the model parameterizations. A short overview of recent model developments is given in the annex to the report. Detailed description of the research and development activities is available in the Technical report [Shatalov *et al.*, 2013].

1. POLLUTION OF THE EMEP REGION

1.1. Emissions data for model assessment

Heavy metal emissions at least for one year in the period 1990-2011 were reported by 41 out of the 51 Parties to the Convention. Data for the base year 1990 and the year 2011 were provided by 33 countries and 27 countries submitted gridded emissions of the considered heavy metals.

The data on heavy metal emissions within the EMEP domain for 1990 and 2011 used for modelling were generated by CEIP and MSC-E. CEIP provided datasets of gridded emissions for 2011. MSC-E prepared gridded emission data for the base year 1990 using official emissions reported by countries, emission expert estimates made by TNO [Denier van der Gon *et al.*, 2005] and expert estimates of emissions for the EECCA countries made by MSC-E.

Information of emission distribution with height is important for modelling of atmospheric transport of heavy metals. Pollutants emitted at higher altitude tend to be transported further due to increase of wind velocity with height. Vertical distribution of the pollutant concentration in the vicinity of emission sources also depends on height of the emission source. In order to estimate distribution of emissions with height MSC-E utilized sector-split emission information provided by the EMEP countries. Height distributions for different emission sectors were averaged taking into account a sector contribution to the total emission. It was assumed that heavy metal emission was distributed between three lowest model layers (0-70 m; 70-150 m and 150-300 m).

Mercury is emitted to the atmosphere in elemental, gaseous oxidized and particulate forms. Atmospheric behaviour of mercury strongly depends on its form. Gaseous oxidized and particulate mercury is removed relatively quickly, while elemental mercury persists in the atmosphere over long time (around a year) and takes part in intercontinental transport. The speciation of mercury emissions is not included in the information reported by the Parties to the Convention. Therefore, expert estimates of the mercury emission speciation have been made by MSC-E. Detailed information on vertical distribution of the emission data and speciation of mercury emissions is presented in [Travnikov and Ilyin, 2005].

Information about uncertainties of emission data is important for understanding and interpretation of the model results. However, only few countries report uncertainties of their national emission data.

Changes of heavy metal emissions in EMEP countries in period 1990-2011

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

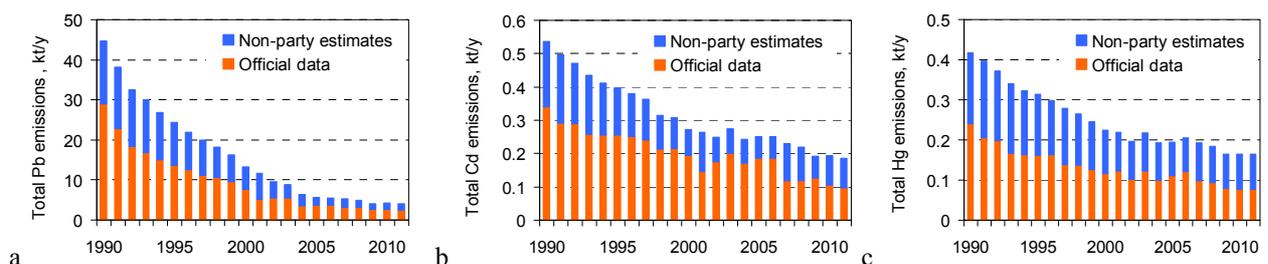


Fig. 1.1. Temporal changes of lead (a), cadmium (b), and mercury (c) emissions in the EMEP region from 1990 to 2011

In 2011 total emission of lead from the EMEP domain made up at around 4039 tonnes. The emission reductions between 1990 and 2011 took place in all EMEP countries, except for Azerbaijan, Georgia and Malta (Fig. 1.2). The most noticeable emission decreases (more than 95%) were in Republic of Moldova, the Russian Federation, Monaco, Hungary, Lithuania, the United Kingdom, Luxembourg, Norway, France, Iceland, Sweden, and the Ukraine.

In the period from 1990 to 2011 lead emissions in the EMEP region reduced by 90%, and cadmium and mercury emissions – by about 60%

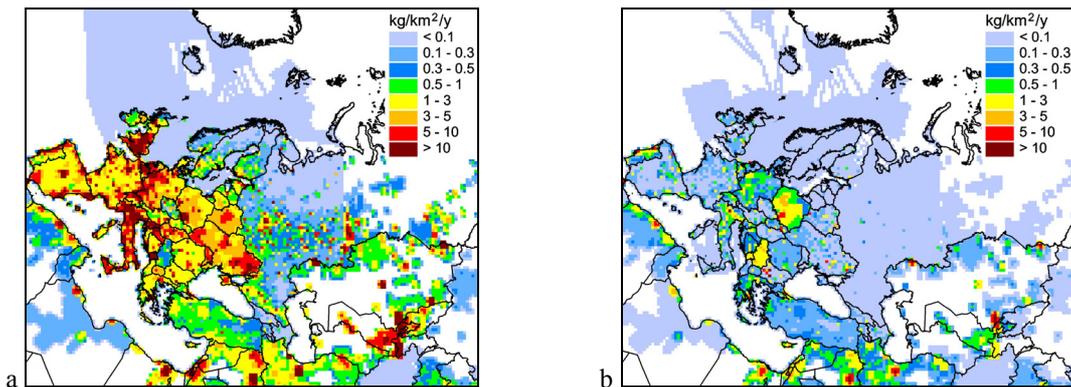


Fig. 1.2. Spatial distribution of lead emissions over the EMEP domain in 1990 (a) and 2011 (b).

Total emission of cadmium in the EMEP domain in 2011 was 185 tonnes. Compared with 1990, lower cadmium emissions for 2011 can be seen in 39 EMEP countries (Fig. 1.3). The highest emission decreases compared to 1990 took place in Lithuania (96%), Malta (96%), Republic of Moldova (95%), the Ukraine (95%), Luxembourg (92%), and Monaco (91%). Increases in emissions indicated in Cyprus (46%), Belarus (42%), Georgia (29%), Azerbaijan (23%), Liechtenstein (13%) and Turkey (8%).

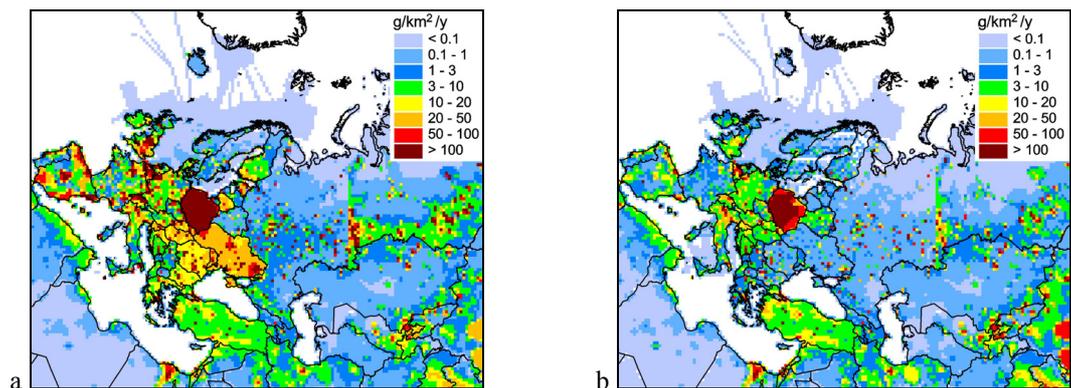


Fig. 1.3. Spatial distribution of cadmium emissions over the EMEP domain in 1990 (a) and 2011 (b).

Emission of mercury in the EMEP domain in 2011 amounted to 165 tonnes. During the period 1990 and 2011, mercury emissions decreased in 38 countries and increased in 7 countries (Fig. 1.4). The most substantial decreases of mercury emission values took place in Malta (99%), Lithuania (95%), Republic of Moldova (94%), the Russian Federation (94%), and Slovakia (91%). Higher levels of emissions (compared to 1990) in 2011 were noticed in Iceland (120%), the FYR of Macedonia (72%), Turkey (25%), Georgia (23%), Azerbaijan (21%), Montenegro (19%), and Liechtenstein (17%).

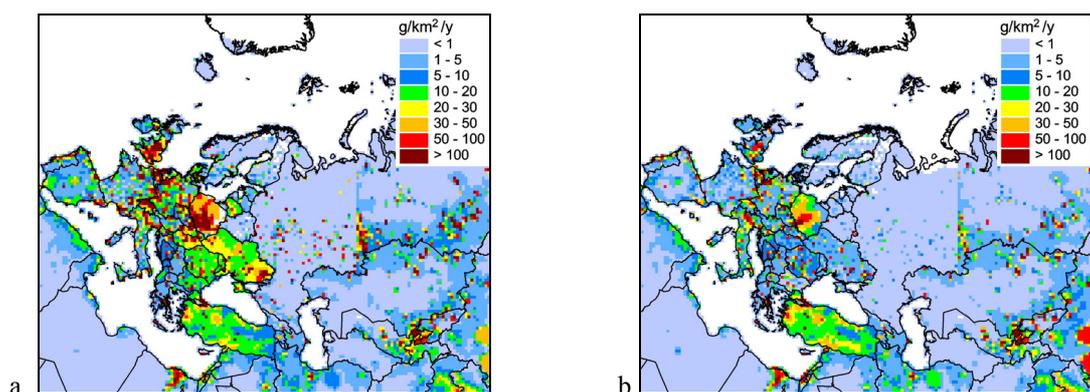


Fig. 1.4. Spatial distribution of mercury emissions over the EMEP domain in 1990 (a) and 2011 (b).

Further improvement of heavy metal emissions

Completeness, consistency and uncertainties of officially reported heavy metal emissions were the subject of discussion at the recent meetings of the EMEP Task Forces (TFMM in Zagreb and TFEIP in Istanbul in May 2013). Particularly, the meetings addressed the quality of currently available official emissions and considered the ways of its further improvement. Additionally, following the request of the EMEP Steering Body Bureau and TFEIP, EMEP modellers were invited to present their thoughts and wishes concerning reported emissions, and ways of their improvement to add certainty to modelling results.

In particular, it was emphasized that the completeness of emission time-series and availability of emission spatial distribution for all EMEP countries was one of the most important characteristics for model assessment of heavy metal pollution within the EMEP domain. Differentiation of reported emissions by sectors and coverage of all potentially contributing sectors of emissions is of importance for the evaluation of vertical distribution of emissions and their temporal disaggregating.

Considering the transition to finer resolution of gridded emissions the data on large point sources and their characteristics were also recognized as important information for modelling, especially for modelling with fine scale resolution (e.g. $0.1^\circ \times 0.1^\circ$). Additionally, it was indicated that most of the available emission expert estimates used currently for the purposes of gap-filling of emissions within the EMEP region and for the modelling at global scale became outdated and required updating.

Discussing the wishes of modellers with regard to emission data and their characteristics, not currently reported by countries, importance of the information on chemical speciation and historical trends of mercury emissions for model assessment was considered at the TFMM and TFEIP meetings.

Completeness of emission data, information on large-point sources, key emission source categories and contemporary emission expert estimates with fine spatial resolution are needed for further progress in assessment of heavy metal pollution in the EMEP countries.

1.2. EMEP monitoring network of heavy metals

Measurement network

Heavy metals were included in EMEP's monitoring program in 1999. However, earlier data has been available and collected, and the EMEP database (<http://ebas.nilu.no>) thus also includes older data, even back to 1987 for a few sites. A number of countries have been reporting heavy metals within the EMEP area in connection with different national and international 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 Breivik, 2013]. In 2011, there were 34 sites measuring heavy metals in both air and precipitation, and altogether there were 67 measurement sites, which is more than in 2010. There were 34 sites measuring at least one form of mercury which is three more than previous year, but not all sites do have complete dataset for the whole year. 15 sites were measuring mercury in both air and precipitation, though 12 of these measure gaseous mercury (in difference from particulate mercury) and fulfil the monitoring obligations. There is a need for better coverage of especially mercury in large part of Europe. That said however, the measurement obligations set by the EMEP monitoring strategy [UNECE, 2009] and the EUs air quality directives [EU, 2004, 2008] have clearly improved the site coverage the last years.

Observed concentration level of Pb, Cd and Hg in 2011

Annual averages of Pb, Cd and Hg concentrations in precipitation and in air in 2011 are presented in Fig. 1.5-1.10. Note that Cyprus with measurements of heavy metals in air is outside the map domain so included as a dislocated point south of Turkey. Further, the aerosol data from Slovakia have not been reported in time to be included in the report. The lowest concentrations for all elements in air as well as precipitation are generally found in northern Scandinavia. An increasing gradient can in general be seen southeast, but the concentration levels are not evenly distributed, there are some "hotspots" for some elements, i.e. in the BeNeLux countries for lead and cadmium in air. The 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 PL, LV, IE) with high detection limits and these only give an indicative measure for the upper limit. For sites with more accurate measurements the highest level is seen at single sites in Germany and Belgium. A more detailed discussion of temporal and spatial resolution of heavy metals in Europe was discussed in last year status report [Travnikov *et al.*, 2012] and by Tørseth *et al.* [2012].

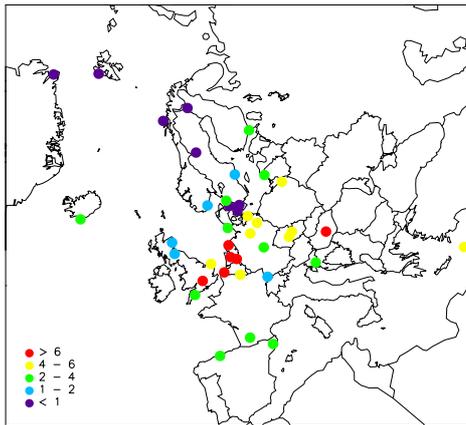


Fig. 1.5. Pb in aerosol, ng/m^3

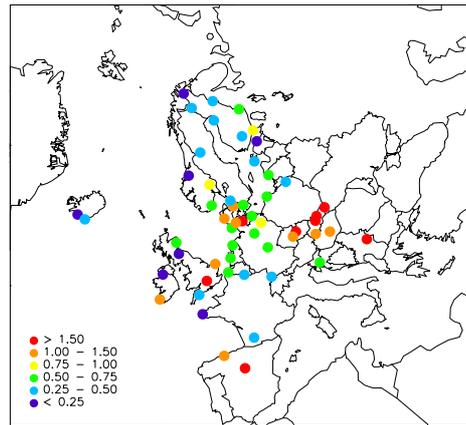


Fig. 1.6. Pb in precipitation, $\mu\text{g}/\text{L}$

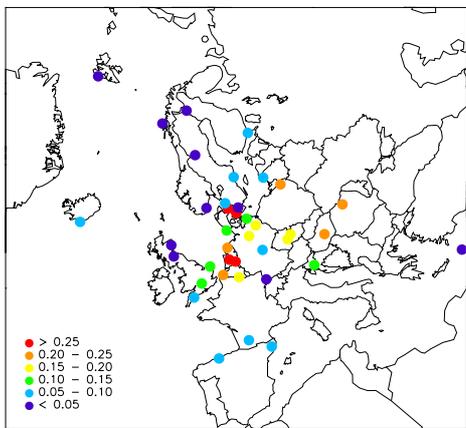


Fig. 1.7. Cd in aerosol, ng/m^3

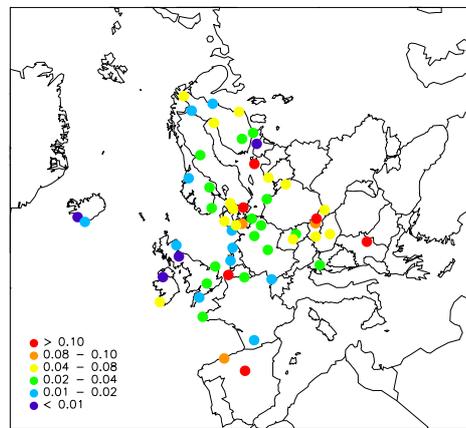


Fig. 1.8. Cd in precipitation, $\mu\text{g}/\text{L}$

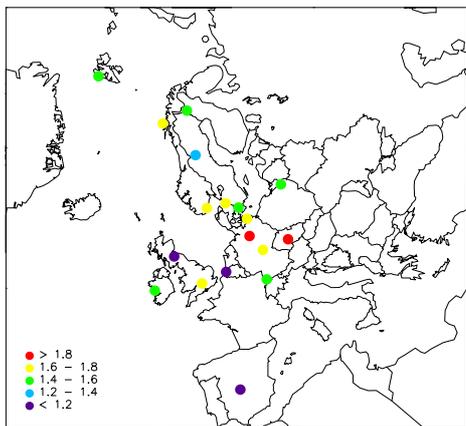


Fig. 1.9. Hg (g) in air, ng/m^3

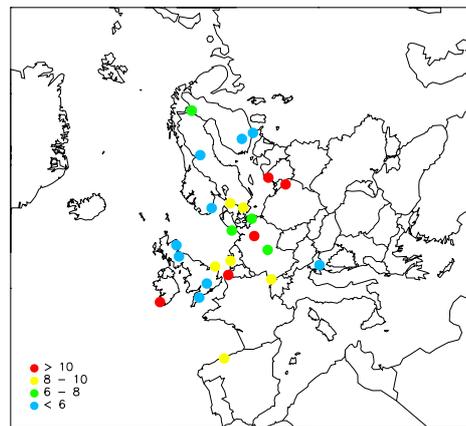


Fig. 1.10. Hg in precipitation, ng/L

1.3. Pollution levels and long-term trends for 1990 - 2011

Pollution levels of lead, cadmium and mercury declined significantly for the period from 1990 to 2011 in the EMEP region. Detailed analysis of long-term deposition trends of these metals in 1990-2010 was carried out in the previous year [Travnikov *et al*, 2012]. This section is focused on heavy metal pollution levels in 2011 and their trends between 1990 and 2011.

Decrease of deposition of lead, cadmium and mercury in the EMEP region in the period from 1990 to 2011 made up 75%, 51% and 37%, respectively (Fig. 1.11). The major decrease took place in 1990-2000, whereas since 2000 the rate of pollution reduction declined. Spatial variability is indicated in the figure by 90% (range between 5th and 95th percentiles) and 50% (range between 25th and 75th percentiles) intervals of deposition variation over the EMEP domain. In different countries the decrease of deposition can significantly differ from the values averaged over the entire EMEP region. The main driving force of the pollution decrease is the reduction of anthropogenic emissions, carried out by countries following the requirements of the Protocol on Heavy Metals. However, decline of deposition is lower than the reduction of anthropogenic emissions in the EMEP region (Fig. 1.1). The reason for this is significant contribution of secondary sources, i.e., wind re-suspension of previously deposited lead and cadmium from anthropogenic sources. The long-term decline of contribution of these secondary sources is much slower than that of anthropogenic emissions. Lower decline of mercury levels compared to emission reduction is explained by significant contribution to mercury deposition by intercontinental transport from sources located outside the EMEP domain.

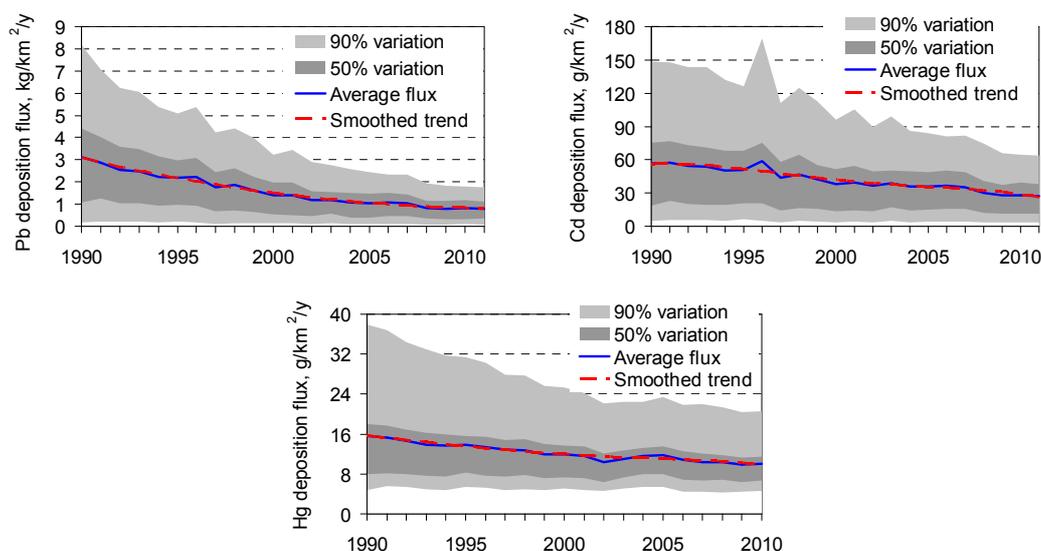


Fig. 1.11. Temporal trends of Pb, Cd and Hg deposition flux in the EMEP countries. Deposition flux: blue solid line presents area weighted average deposition flux; dashed red line is smoothed trend; dark and light grey shadowed areas show 50% and 90% variation intervals over the whole territory, respectively.

Pollution levels in the EMEP region changed significantly between 1990 and 2011 (exemplified by cadmium in Fig 1.12). Maps for all considered metals are available at the MSC-E web site (www.msceast.org). The highest deposition levels both in 1990 and 2011 are noted for Poland, Benelux region, and countries of the eastern part of Europe. Besides, relatively high deposition levels in 1990 are seen in the Balkan region and the eastern part of the Ukraine. The northern parts of Scandinavia and Russia are characterized by the lowest levels of pollution. This distribution is typical for lead and cadmium. In case of mercury there is no elevated deposition in the central part of Russia.

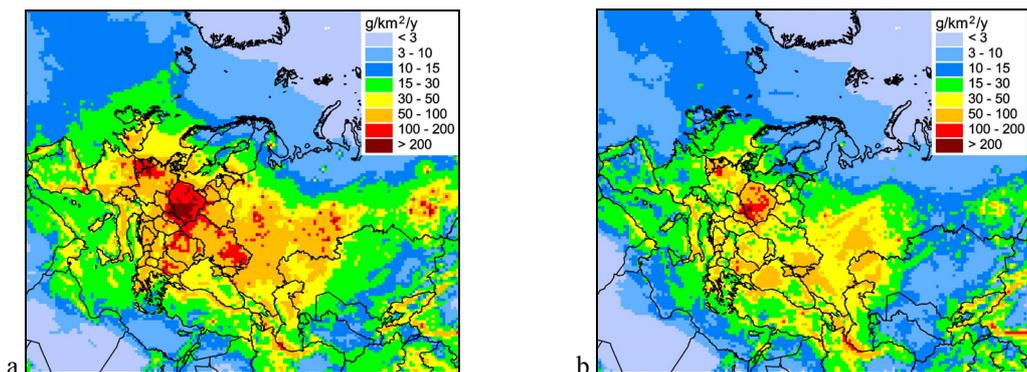


Fig. 1.12. Spatial distribution of annual cadmium deposition flux in the EMEP region in 1990(a) and 2011(b).

Country-averaged deposition of cadmium, caused by anthropogenic emissions, secondary sources and non-EMEP sources in 2011 ranges from 7.5 to 100 g/km²/y (Fig. 1.13). For lead the corresponding range of deposition is 0.25 – 7.3 kg/km²/y, and for mercury deposition varies from 4 to 23 g/km²/y. There is a group of countries where deposition of all three metals is the highest. They are Monaco, Slovakia, Poland, Belgium and Bosnia and Herzegovina. The lowest deposition of these metals is indicated in the Central Asian countries (Kazakhstan, Turkmenistan, Uzbekistan) and in the north-western part of Europe (Sweden, Ireland, Finland, and Iceland). Relatively low deposition in Central Asia is explained mainly by low precipitation amounts, while in north-western Europe it is caused by low emissions in this region.

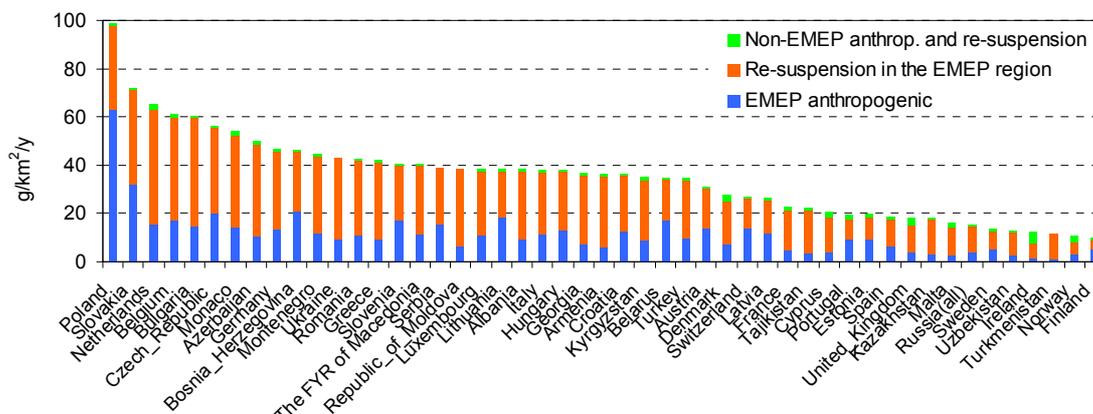


Fig. 1.13. Country-averaged deposition fluxes of cadmium from the European and Central Asian anthropogenic, natural/historical and non-EMEP sources in 2011

Large part of lead and cadmium deposition to the EMEP countries is caused by secondary sources (i.e., wind re-suspension). Its average contribution is 76% for lead, and 67% for cadmium, ranging from 35 to 90% in different countries. However, it should be noted that the uncertainty of estimates of re-suspension is high, and substantial contribution of this process to deposition may act as a compensation of uncertainties of anthropogenic emission data. The role of non-EMEP sources is about 3-4%. However, in countries located close to borders of the EMEP region (e.g., Armenia, Tajikistan, Norway etc.) this contribution can be higher and can make up 10-20%. In Iceland this contribution reaches 50%. Unlike lead and cadmium,

Pb and Cd pollution levels are caused mainly by sources in the EMEP region. Contribution of non-EMEP sources to lead and cadmium levels in the EMEP countries is relatively low ($\pm 3-4\%$). In contrast, intercontinental transport of mercury is important contributor to pollution levels in the EMEP region.

Unlike lead and cadmium, intercontinental transport of mercury is important contributor to pollution levels in the EMEP region.

contribution of secondary sources of the EMEP region to mercury deposition in the EMEP countries is not high, whereas contribution of non-EMEP sources is dominating. Non-EMEP sources are responsible for more than 50% of mercury pollution in 40 of 51 countries. The effect of intercontinental transport on mercury pollution in the EMEP countries is discussed in Chapter 2.

Deposition changes between 1990 and 2011, expressed in relative and absolute terms are illustrated by cadmium in Fig. 1.14. Positive values mean increase and negative – decrease of deposition between 1990 and 2011. Deposition went down in most part of the EMEP region by 40 – 60% within the considered period (Fig. 1.14a). However, an increase of deposition is noted in the Mediterranean region as well as in the central part of Russia. It should be stressed that the increase in these parts of the EMEP domain in absolute terms is not very high: 10-30 g/km²/y, while decline in central Europe is 30 – 100 g/km²/y (Fig. 1.14b). Map of relative changes of lead deposition is very similar to that for cadmium. However, the main difference is the fact that decline of lead deposition in Europe is much deeper than that of cadmium, and typically exceeds 60%. Besides, the increase is noted for the easternmost part of the Mediterranean region, while in the central part the deposition has declined. As for mercury, the decline of deposition in most of Europe and Central Asia is 20 – 60%, and increase of deposition takes place in the Mediterranean region and in northern Scandinavia. Unlike lead and cadmium, the increase of mercury deposition in the central part of Russia is not indicated.

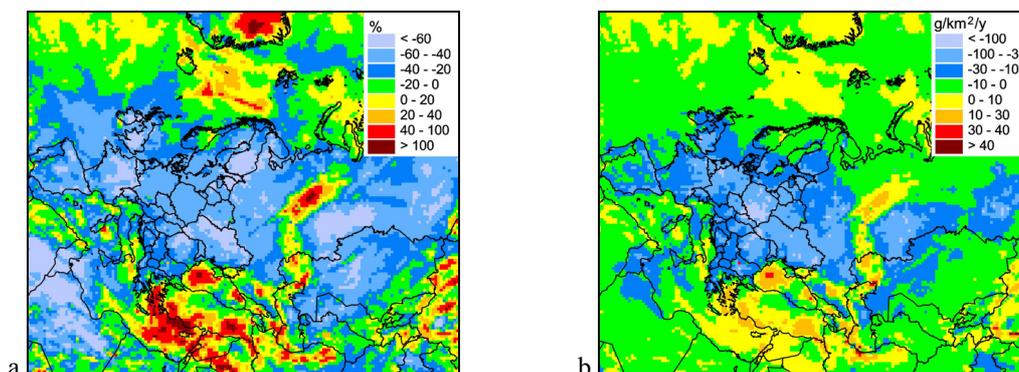


Fig. 1.14. Relative (a) and absolute (b) changes between cadmium deposition in 2011 and 1990

There are three major factors which affect the changes of deposition: emissions, meteorological variability and wind re-suspension. The effects of these three components were estimated by series of numerical tests, in which two of the components were fixed at magnitude for 1990, and the third one was chosen at level of 2011. It is important to note that meteorology and re-suspension are not fully independent components because re-suspension to large extent depends on meteorological parameters. However, this fact does not likely affect the main outcomes of the experiments.

The most obvious effect on deposition levels in the EMEP region is made by emission changes. It results to 20 – 60% (50-100 g/km²/y) decline of cadmium deposition levels over the major part of the EMEP region (Fig. 1.15a) and explains the decrease of deposition in the most part of Europe and Russia (Fig. 1.14). These regions are known for significant emissions (Fig. 1.3) and thus the effect of emission reduction is more pronounced here. Increase of deposition in some areas of the Mediterranean region, Caucasus, the Arctic seas and the northern Atlantic is explained mainly by higher precipitation amounts (Fig. 1.15b). Although the increase in these regions, expressed in relative terms, is considerable, the growth in absolute terms is not high: ±15 g/km²/y. Higher wind re-suspension in 2011 compared to 1990 favours increase of deposition in Central Asia, some regions in central Russia as well as in southern and central Europe (Fig. 1.15c). The absolute effect of re-emission is about ±15 g/km²/y over the most part of the EMEP region. In Central Asia its magnitude is much higher (> 100 g/km²/y).

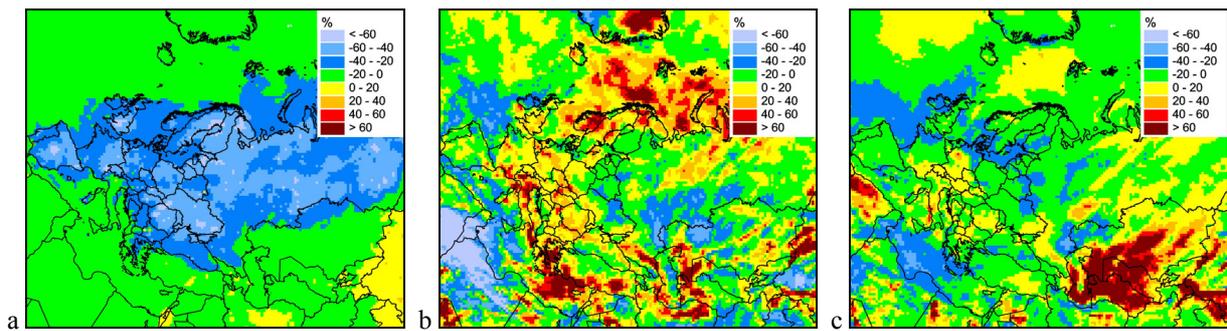


Fig. 1.15. Relative changes of Cd deposition between 1990 and 2011 due to effect of emission changes (a), meteorological variability (b) and changes of wind re-suspension (c).

In some cases the changes of deposition are explained by synergetic effect of the factors. For example, deposition in 2011 is significantly higher than that in 1990 in the central part of Russia. It is explained by higher re-suspension of cadmium in the Central Asian region (Fig. 1.15c) followed by atmospheric transport to the central part of Russia. Strong winds over dry soil in arid regions of western Kazakhstan and Turkmenistan gave rise of high suspension of soil dust in April, 2011. Predominant meridional wind flows from these regions favour transport to the central part of Russia. Strong precipitation taken place in the beginning of April caused wet scavenging of these dust particles, containing lead and cadmium, thus resulting in elevated deposition.

Changes of heavy metal pollution levels in countries depend not only on reduction of national emissions but also on other factors such as meteorological variability and wind re-suspension. In various parts of the EMEP region the effect of these factors is comparable.

For Europe as a whole the effect of anthropogenic emission changes (meteorology and re-emission fixed) is exhibited as a smooth decline between 1990 and 2011 (Fig. 1.16). The joint effect of factors, not controlled directly by humans, such as annual variability of meteorological parameters and changes of wind re-suspension, does not exceed 15% for Europe as a whole. It also follows from Fig. 1.16 that perturbations of the deposition trend are caused mostly by meteorological factors.

In individual countries the effect of meteorological variability on long-term change of pollution levels is somewhat stronger than that for Europe as a whole. In almost all countries of Europe the variability of deposition in 1990-2011 period varies from $\pm 10\%$ to $\pm 30\%$ (Fig. 1.17). It means that even if a country continues to reduce national emissions, its deposition can exhibit some increase from one year to another due to annual variability of meteorological conditions.

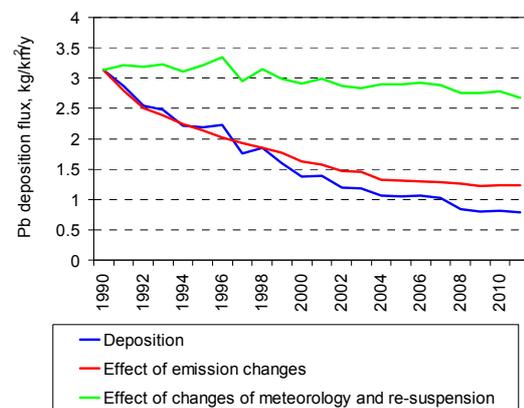


Fig. 1.16. Long-term annual deposition trends simulated under different assumptions on changes of emission, re-emission and meteorological parameters.

Another characteristic of transboundary pollution in the EMEP domain is relative and absolute contribution of emission sources of a country to long-range transport. Main contribution to long-range transport, expressed in absolute units, is made by countries with high national emissions. For lead, cadmium and mercury the leading countries are Poland, Kazakhstan, Turkey and Italy in 2011.

Fraction of mass of lead and cadmium emitted in a country and transported outside country's borders typically varies from 60 to 90% (Fig. 1.19). Mercury persists longer in the atmosphere compared to lead and cadmium. Therefore, its fraction of emissions involved in transboundary transport is rather high and ranges from 80 to almost 100%.

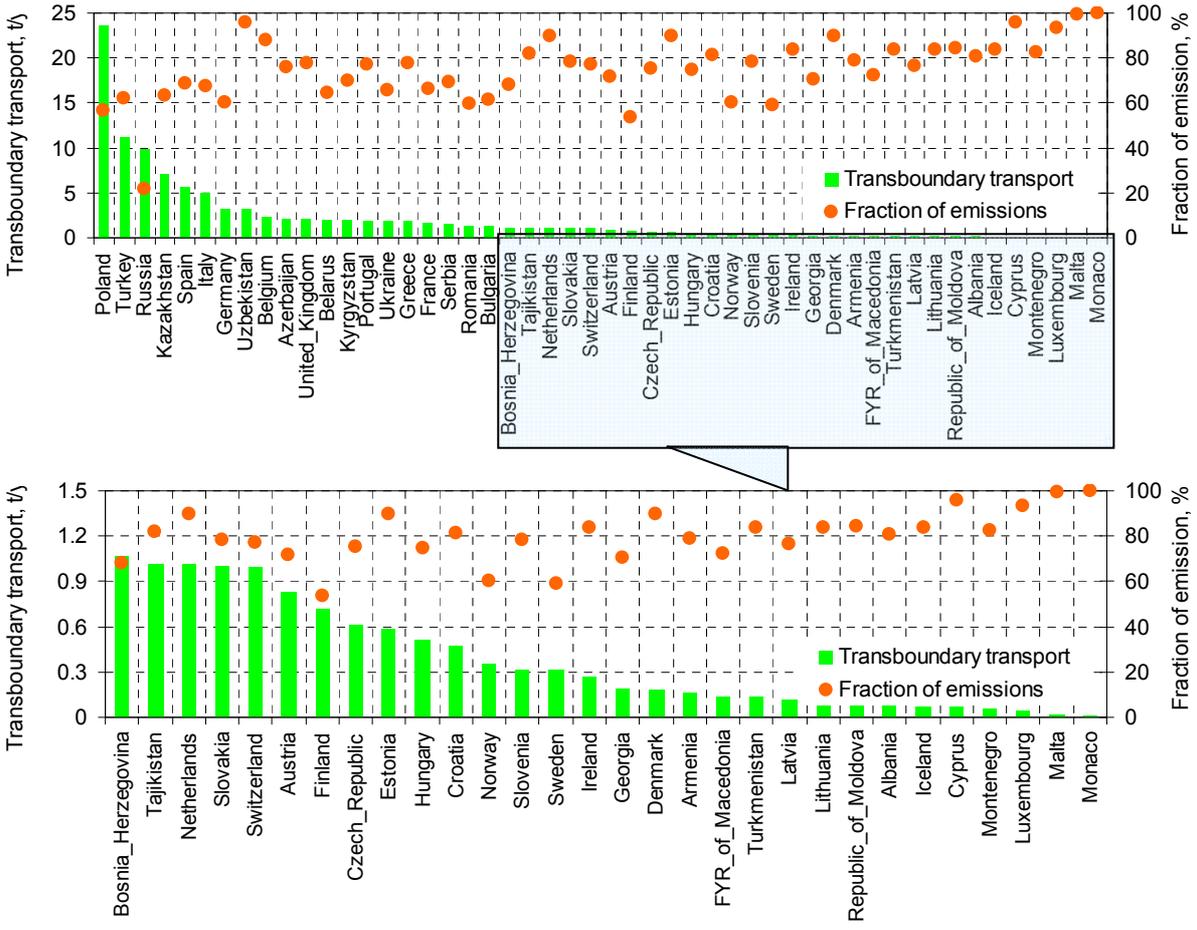


Fig. 1.19. Absolute contribution of the European and Central Asian countries to cadmium transboundary transport in 2011 and relative fraction of national emissions involved into the transboundary pollution

Changes of pollution levels in the EMEP countries for the considered period are characterized by relative decline of deposition between 1990 and 2011. This decline is caused by changes of three types of sources. The first one is deposition from national anthropogenic sources. Its change reflects effectiveness of country's efforts in reducing emissions according to the Protocol. The second type is transboundary transport. The changes in contribution of this component characterize the effect of emission reduction in neighbouring countries. The third type comprises sources which are not

Most of atmospheric emissions of heavy metals released by country's national sources (60-90% for Pd and Cd, and 80-100% for Hg) are transported and deposited outside country's territory.

controlled directly by the EMEP countries. These are secondary sources (i.e., wind re-suspension of particles containing heavy metals or natural emission of mercury) and sources located outside territories of the EMEP countries (so-called non-EMEP sources).

Changes of these three components of heavy metal pollution are demonstrated for each country of the EMEP region in Fig. 1.20. In this figure positive value of the change means decline, and negative means growth. As a rule, decline of pollution levels due to reduction of national emissions is similar to that caused by the decrease of contribution from transboundary transport. Decline of contribution of secondary and non-EMEP sources is relatively low. However, in a number of countries the specific situation may differ. In two thirds of the EMEP countries changes of transboundary pollution and of secondary and non-EMEP sources prevails over variation of domestic sources.

For example, lead deposition decline by about 80% both in the United Kingdom and Latvia. In absolute terms the decrease is also similar: 3 kg/km²/y in the United Kingdom and 2.4 kg/km²/y in Latvia. Both countries reduced their national emissions by more than 90%. However, in the United Kingdom pollution decline is mostly caused by reduction of national emissions, while in Latvia the decline is explained by decrease of deposition from transboundary transport (i.e., due to emission reduction in other countries). This difference in composition of the reductions is explained mostly by location of these two countries. The United Kingdom is located in the north-western part of Europe. The contribution of transboundary transport to this country is relatively low for the considered period due to prevailing westerly atmospheric transport. Therefore, the reduction of national emissions plays the main role in the decrease of heavy metal pollution levels.

Situation with Latvia is different. This country is surrounded by countries with significant emissions, such as e.g., Poland or Russia. For example, in 2011 around 90% of lead deposition from anthropogenic sources is caused by foreign sources (Fig. 1.18). Therefore, even significant reduction of national emissions (more than 90%) resulted to relatively small effect on decrease of pollution levels in this country. However, emission changes in neighbouring countries make major contribution to decline of pollution levels in Latvia.

Long-term decrease of deposition in EMEP countries is caused by reduction of national emissions, decrease of deposition from transboundary transport and changes of contribution from uncontrollable (non-EMEP and secondary) sources. Their relative contributions differ in different countries.

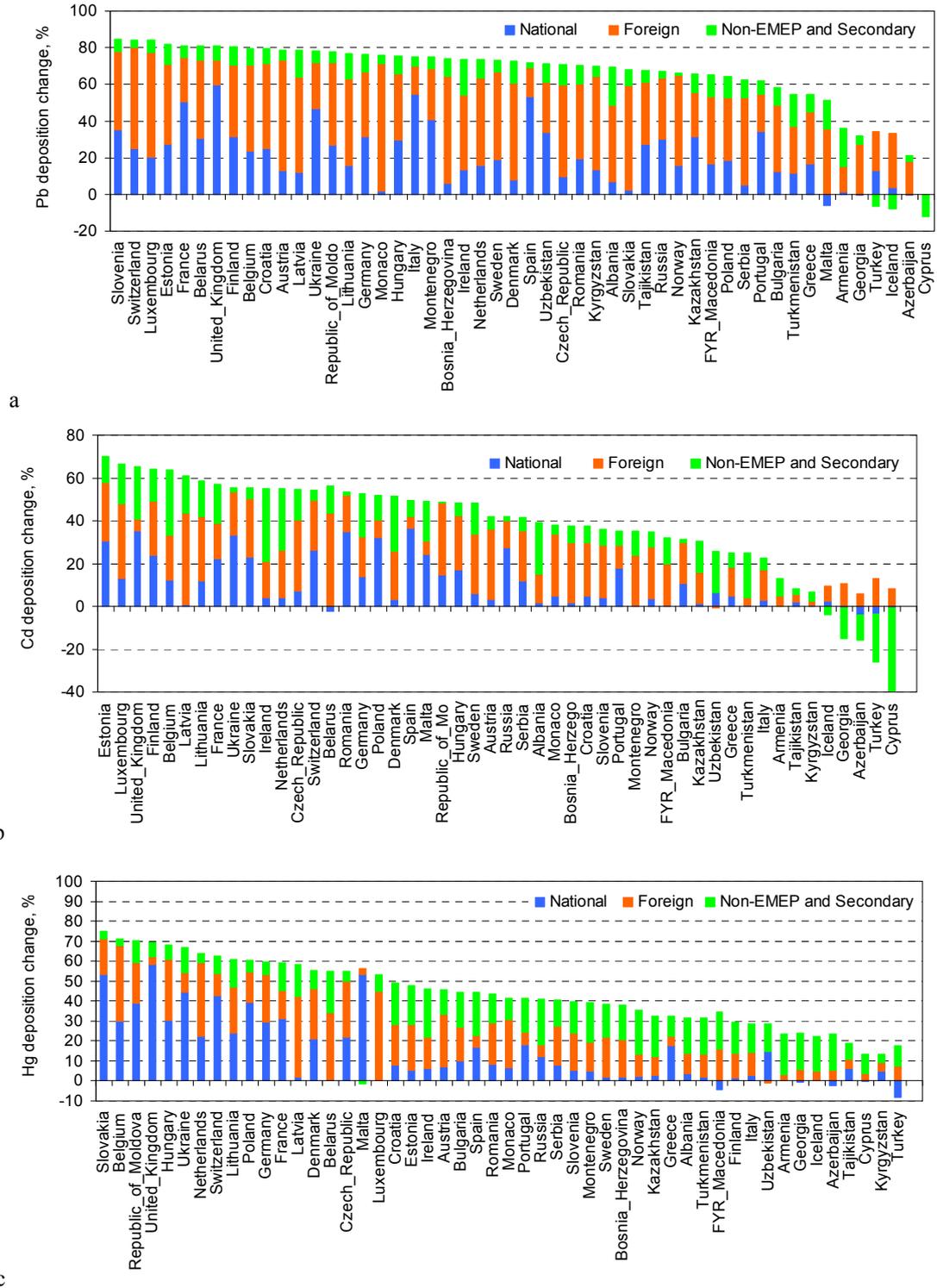


Fig. 1.20. Reduction of lead (a), cadmium (b) and mercury (c) deposition caused by changes of national, foreign and sum of secondary and non-EMEP sources between 1990 and 2011 in countries of the EMEP region. Positive value means decrease of deposition, and negative – increase.

1.5. Ecosystem-dependent deposition

Data on annual deposition of heavy metals are necessary to evaluate negative effects on human health and biota. Each ecosystem has specific threshold (so-called critical load) of heavy metal levels over which negative effects may occur. In order to understand if this critical load is exceeded, deposition fluxes are compared with the critical loads. Coordinating Centre for Effects (CCE) of the Working Group on Effects (WGE) is responsible for evaluation and mapping of critical load exceedances over the EMEP region. Critical loads of deposition differ depending on particular ecosystem. In order to support CCE with necessary information, ecosystem-dependent deposition fluxes are simulated.

Deposition fluxes of heavy metals to various ecosystems are not the same. The main reason for this is different properties of the underlying surface. For example, deposition of lead in 2011 to arable lands ranges from 0.5 – 2 kg/km²/y over the major part of the EMEP region (Fig. 1.21). Deposition to forests is markedly higher – 1-5 kg/km²/y. It means that every square km of forest surface receives on average about twice more load of heavy metals than surface with low vegetation, and thus accumulation of metals likely tends to be more significant in forest soils.

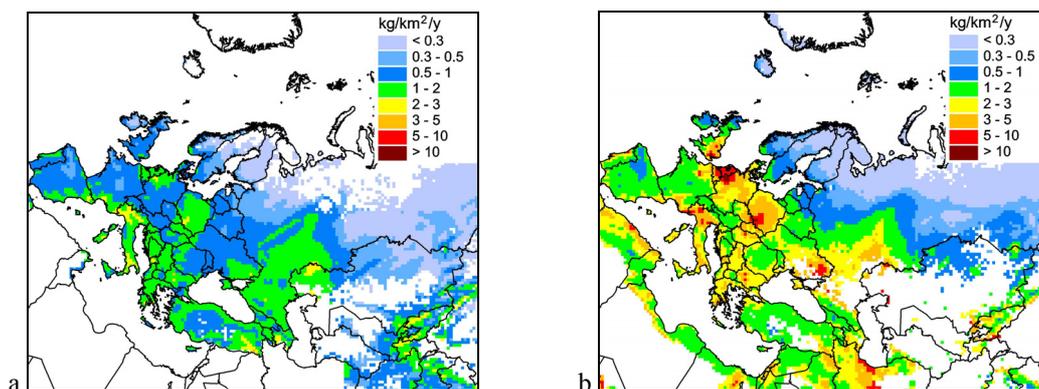


Fig. 1.21. Deposition of lead to arable lands (a) and forests (b) in 2011.

Long-term trends of ecosystem-dependent deposition differ for various types of the land-cover categories. For example, the most pronounced decline of mercury deposition (about 3 times) is noted for urban areas (Fig. 1.22). This fact is not surprising: most of emission sources are located in populated areas and hence the reduction of the emissions first of all is reflected on urban areas. For other land cover categories the decrease is smaller: deposition to forests and arable lands declined around twice, and to ocean waters - about 1.4 times. Ocean is a pathway of mercury accumulation in tissues of fish, which is consumed by humans. Low decrease of mercury atmospheric input to waters may indicate that accumulation of mercury in sea water and bioaccumulation in marine species still continues.

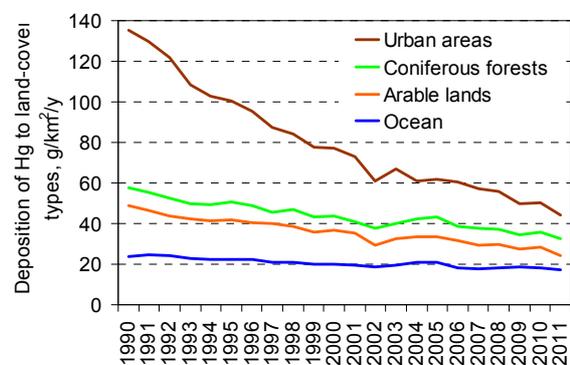


Fig. 1.22. Long-term trends of mercury deposition to selected ecosystems for 1990 – 2011 over the EMEP countries.

1.6. “Near-real time” estimates of pollution levels

Timely delivering of information on pollution and minimization of the time gap between the collection and reporting of results of pollution assessment was discussed during a number of recent meetings of EMEP TFMM. Delivery of modelling results largely depends on the reporting of official information on emissions which takes place two years in arrears. At the same time, taking into account that recent changes of emissions in the EMEP countries were relatively low, model assessment of pollution can be provided with shorter delay (e.g. a few months) performing model simulations with the emissions submitted in previous year. The aim of this activity is to support countries with the most recent information on pollution levels. This year, as a first step in this direction, MSC-E prepared preliminary information on the POP pollution levels for 2012 along with regular model assessment for the year 2011.

An example of total deposition map of lead for 2012 and relative difference between deposition in 2012 and 2011 are shown in Fig. 1.23. As seen, deposition of lead in 2012 is expected to be significantly higher than that in 2011 in central regions of France, south-eastern Europe, some areas of the Mediterranean and in the EECCA countries. The levels in the southern part of Spain and in Scandinavia in 2012 tend to be considerably lower than those in 2011. In other regions the difference between deposition in 2012 and 2011 will likely remain within $\pm 30\%$. The difference is caused by changes of meteorological conditions (precipitation amount, air temperature and wind patterns) and variation of wind re-suspension.

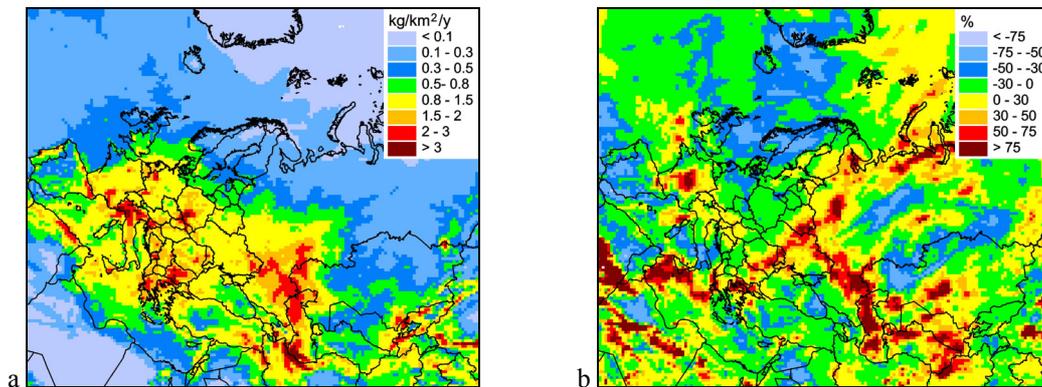


Fig. 1.23. Spatial distribution of total deposition of lead, based on emissions of 2011 and meteorological data for 2012 (a) and relative difference between deposition in 2012 and 2011 (b).

2. INTERCONTINENTAL TRANSPORT OF MERCURY

Mercury differs from other heavy metals by its ability to long-range dispersion in the atmosphere. The bulk mercury atmospheric form – gaseous elemental mercury (GEM) – which is a poorly soluble and relatively inert gas can drift in the atmosphere for months making possible transport over thousands of kilometres. After that it can be oxidized and deposited to the ground or water surfaces contributing to local pollution of terrestrial and aquatic ecosystems. Therefore, mercury pollution levels in Europe are largely affected by emission sources from other regions. On the other hand, mercury originated from European sources contributes to contamination of other regions and continents.

For evaluation of mercury intercontinental transport and contribution of distant source regions to pollution of the EMEP countries the Global EMEP Multi-media Modelling System (GLEMOS) is used. GLEMOS is a multi-scale multi-pollutant simulation platform recently developed for operational and research applications within the EMEP programme. The framework allows simulations of dispersion and cycling of different classes of pollutants (mercury and other heavy metals, POPs) in the environment with a flexible choice of the simulation domain (from global to local scale) and spatial resolution. Development of the modelling system was documented in a series of technical reports [Tarrason and Gusev, 2008; Travnikov et al., 2009; Jonson and Travnikov, 2010; Travnikov and Jonson, 2011; Jonson and Travnikov, 2012]. Recent research and updates of the modelling system performed during the last year are briefly outlined in Annex A and discussed in more details in the MSC-E Technical Report [Shatalov et al., 2013]. They are focused on improvement of the mercury chemical scheme and refinement of the model assessment.

Model simulations of mercury atmospheric dispersion on a global scale performed with GLEMOS support the idea of mercury potential to the long-range transport (Fig. 2.1a). Relatively high deposition fluxes are detected even for some remote areas of the oceans and the polar regions. Mercury deposition to Europe consists of almost equal contributions of contemporary anthropogenic emissions (97 t/y) and emissions from natural and legacy sources (96 t/y). Two thirds of the anthropogenic part (65 t/y) are contributed by domestic anthropogenic sources and the other one third is by transport from sources located in other regions. Contribution of natural and legacy sources is almost equally divided between emissions from terrestrial and oceanic surfaces. The largest external contributor to mercury anthropogenic deposition in Europe is East Asia (Fig. 2.1b). Transport from this region to Europe (15 t/y) is almost double of the reverse transport from Europe to the region (8.7 t/y). As to other regions including North and South Americas, South and Central Asia, Africa, Australia and Oceania, Europe acts as a net exporter of atmospheric mercury transporting significantly more pollution to these regions than receiving from them. The largest receptor of mercury emitted from European sources is Africa that is defined by both its close location to Europe and large area of the continent.

In the field of the intercontinental transport assessment MSC-E contributes to the work of the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). The Centre participated in the HTAP Assessment 2010 [HTAP, 2010] both providing model estimates for mercury and leading the data analysis. In current phase of the TF HTAP work MSC-E takes a lead of the task force activities on mercury in co-operation with the GMOS project (Global Mercury Observational System, www.gmos.eu).

Mercury pollution of the EMEP region is largely affected by emission sources located in East Asia. On the other hand, Europe acts as a net exporter of atmospheric mercury to other regions (North and South Americas, South and Central Asia, Africa, Australia and Oceania)

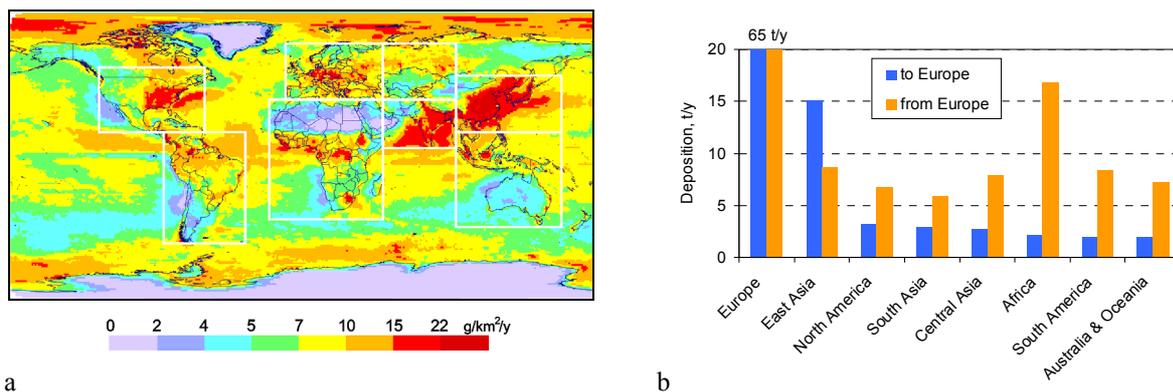


Fig. 2.1. Mercury deposition on a global scale (a) and the mutual mercury atmospheric transport between Europe and other continents (b) as simulated with GLEMOS. White rectangles on the map designate boundaries of the major source and receptor regions considered in the study.

3. POLLUTION ASSESSMENT IN THE EECCA COUNTRIES

3.1. Emission data and measurements in the EECCA countries

Countries of Eastern Europe, Caucasus and Central Asia (EECCA) are of high priority within CLRTAP. These countries include Russia, Belarus, the Ukraine, Republic of Moldova, Kazakhstan, Turkmenistan, Kyrgyzstan, Uzbekistan, Tajikistan, Armenia, Georgia and Azerbaijan. At present only two countries (Armenia and the Ukraine) have signed, and one country (Republic of Moldova) has ratified the Protocol on Heavy Metals. In order to facilitate wider ratification and implementation of the Protocol, information (in Russian and English) on heavy metal pollution levels in the EECCA countries has been produced under EMEP this year.

The main challenge for assessment of heavy metal pollution levels in the EECCA countries is limited information on emission data. Only two countries - Republic of Moldova and Belarus officially reported their long-term trends on emissions of the three metals for 1990 – 2011 (Fig. 3.1). As for other countries, their data on trends are incomplete. To fill the gaps in the emission data in these countries expert estimates are used (see Chapter 1). It should be stressed that quality of the pollution assessment strongly depends on the availability and quality of input information, first of all, on emissions. Therefore, joint efforts of experts of the EECCA countries and the EMEP bodies in the field of emission inventories are needed.

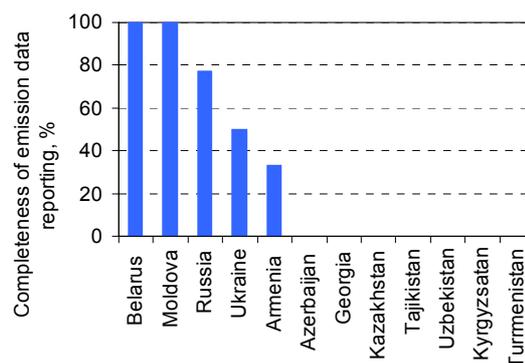


Fig. 3.1. Completeness of heavy metal emission data totals reporting in EECCA countries for 1990-2011 period.

Another issue is monitoring data. Information on heavy metal measurements in air and precipitation from the EECCA countries is not available in the CCC data base for the time being. However, measurements of heavy metal levels are included in monitoring programmes, at least, in some of the EECCA countries, and these data are often available in national research institutions or by request to national experts. Example of observed lead concentrations in air and precipitation measured at

stations of Russian integrated background monitoring network from 1990 to 2011 are shown in Fig. 3.2. These stations are located in biospheric reserves, and are likely representative for evaluation of background pollution levels. The measurements do not exhibit any distinct trend of air concentrations. This fact does not match with the declared reduction of national emission data. The same is related to concentrations in precipitation. On one hand, lack of trend of observed concentrations may be explained by significant contribution of secondary emissions. On the other hand, no information is available about quality of measurement data.

The main challenge of assessment of heavy metal pollution levels in the EECCA countries is limited information on emissions and measurements. Joint activity of EMEP and national experts in this field is needed to overcome this problem.

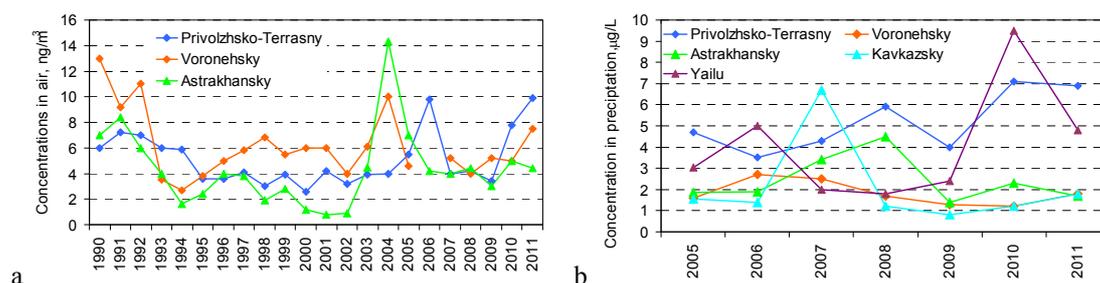


Fig. 3.2. Concentrations in air (a) and precipitation (b) of lead observed at Russian stations of integrated background monitoring network.

Some measurement information is also available from national monitoring network of Kazakhstan. There are about 40 stations measuring concentrations in precipitation and wet deposition of lead and cadmium (Fig. 3.3). Stations are distributed more or less evenly over the country's area, thus providing good spatial coverage. However, information about their representativeness and quality of measurements is not available.

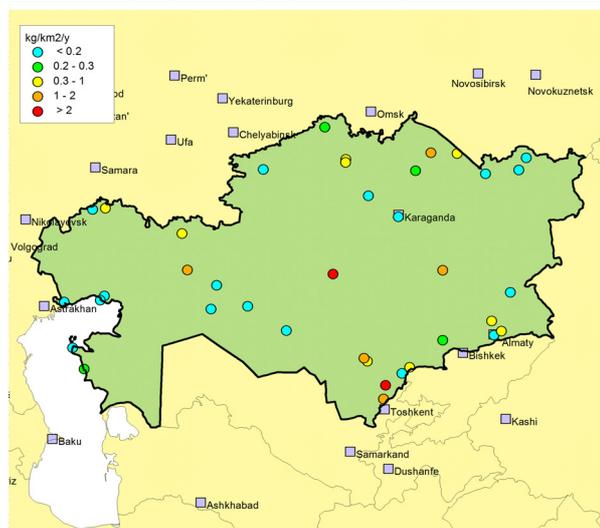


Fig. 3.3. Wet deposition of lead measured at national monitoring network of Kazakhstan in 2008

Since monitoring information for the EECCA countries is limited, assessment of pollution levels in these countries is relied entirely on modelling. However, modelling also needs measurement information for the verification purposes. Therefore, it is highly appreciated if countries could submit their national monitoring data to EMEP as a tentative solution. Besides, it would be useful if national experts from the EECCA countries start to participate in annual intercalibration procedures under supervision of CCC.

In order to increase understanding of factors controlling air pollution and to improve assessment of heavy metal pollution levels, active cooperation between national experts from the EECCA countries and EMEP bodies is highly needed. In particular, the Task Force on Measurements and Modelling (TFMM) of EMEP annually provides a forum for discussion of problems in the field of monitoring and atmospheric transport modelling.

3.2. Levels and trends

Deposition of heavy metals over the EECCA countries is distributed very unevenly (Fig. 3.4). High levels of all three metals are noted for the south-western (the Ukraine, Belarus, Caucasus region), eastern (Russia) and south-eastern (Kyrgyzstan, Tajikistan) parts of the region. Relatively high levels in these areas are caused by location of emission sources, and partly – because of transboundary transport from neighbouring countries. Besides, elevated levels of lead and cadmium deposition in 2011 are partly explained by significant contribution of dust re-suspension (see Chapter 1). The lowest levels are indicated for the Arctic regions of Russia, where emissions are low. Besides, low deposition takes place in the desert areas of Central Asia, which is explained by low precipitation.

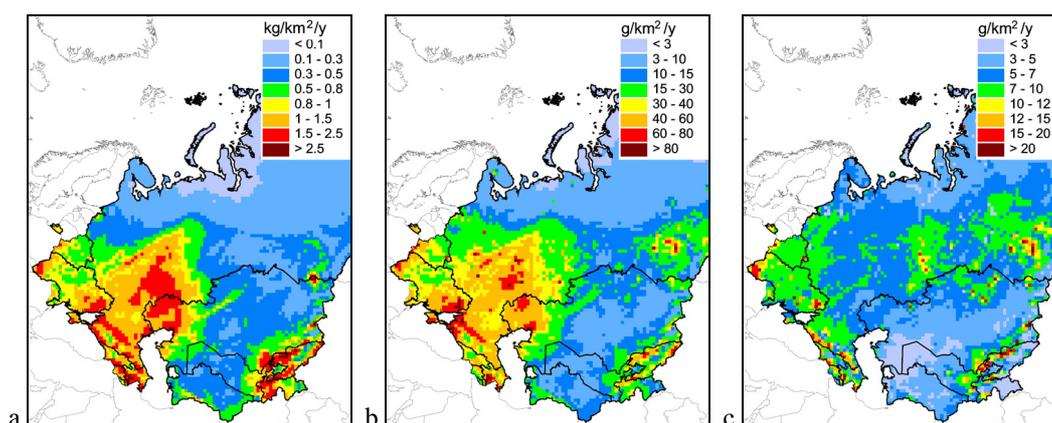


Fig. 3.4. Deposition fields of lead (a), cadmium (b) and net flux of mercury (c) in the EECCA countries in 2011

Changes of heavy metal pollution levels differ significantly among the countries. The most pronounced decline is noted for lead in countries of the western part of the EECCA region: the Ukraine, Republic of Moldova, Belarus, Russia (Fig. 3.5). The reduction of deposition is around 80% and similar to that averaged for EU27 countries. Somewhat smaller (55 - 70%) reduction is seen in the Central Asian countries. For cadmium and mercury the rate of deposition decline is distributed in the same way: the highest decrease is noted for countries from the European part of the EECCA region, then smaller decrease – in the countries of Central Asia and the lowest decline – in the Caucasus countries (Georgia, Armenia, Azerbaijan).

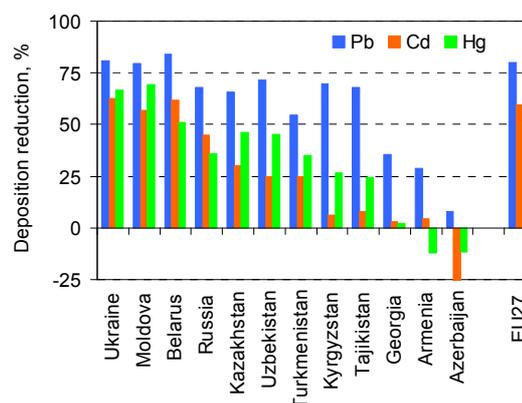


Fig. 3.5. Deposition reduction of lead, cadmium and mercury in the EECCA countries and in EU27 between 1990 and 2011.

Significant decline of deposition levels in the western and central Asian parts of the EECCA region is explained by reduction of national emissions and decrease of deposition caused by transboundary transport (Fig. 1.19). Continuous reduction of lead and cadmium national emissions and emissions in neighbouring countries resulted to almost gradual declining trend of pollution levels (Fig 3.6a), disturbed by effects of meteorological variability. Emissions in countries of the Caucasus region (Georgia, Armenia, Azerbaijan) do not vary much over the considered period, except for lead in Armenia. Besides, countries of this region are influenced by secondary sources and transport from non-EMEP sources. Therefore, the changes between 1990 and 2011 are relatively low. Since the role of reduction of national and foreign emissions for this region is not very high, long-term changes of

pollution in this region are controlled mainly by the other factors such as annual variability of secondary sources and meteorological parameters and transport from non-EMEP sources. As a result, the long-term deposition in these countries do not exhibit distinct trend (Fig. 3.6b).

The highest reduction of pollution levels between 1990 and 2011 is noted in the European part of the EECCA region, while the lowest – in the Caucasus region.

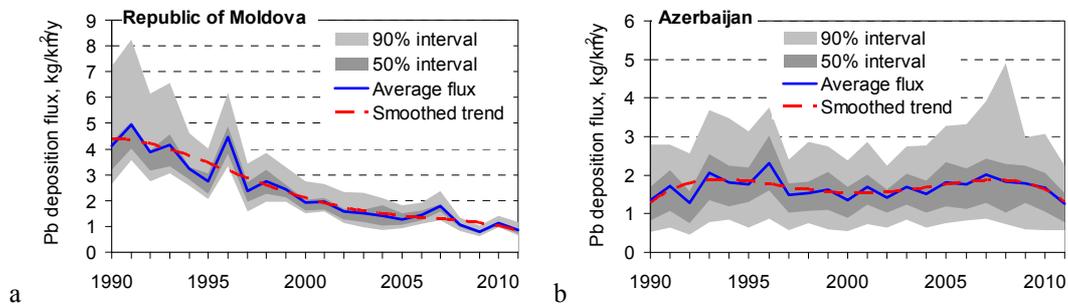


Fig. 3.6. Long-term trends of lead deposition between 1990 and 2011 in the selected EECCA countries

The difference in pollution reduction in different parts of the EECCA region is illustrated by comparison of pie charts with contributions of different emission sources to pollution in Republic of Moldova (the western part of the EECCA region) and Azerbaijan (Caucasus region) (Fig. 3.7). In 1990 the major contribution of pollution in Moldova was caused by foreign (49%) and national (27%) anthropogenic sources. By 2011 the relative role of these sources substantially declined, while the contribution of secondary sources became predominant. In Azerbaijan the role of secondary sources was predominant both in 1990 and 2011.

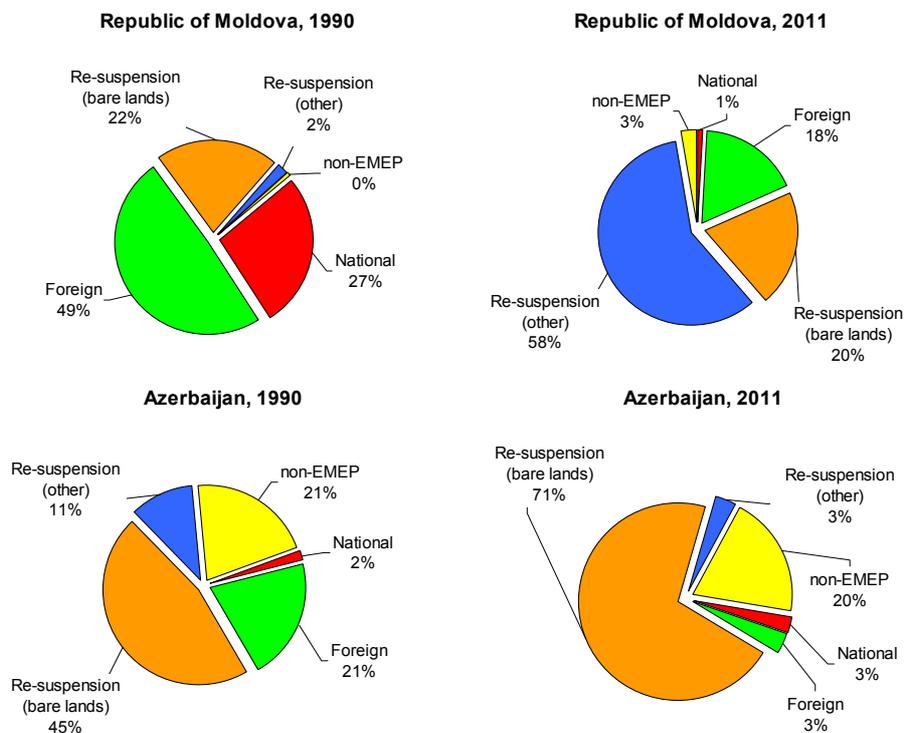


Fig. 3.7. Contribution of different types of sources to total deposition of lead in Republic of Moldova (top) and Azerbaijan (bottom) in 1990 and 2011.

Another peculiarity of the Caucasus countries is the fact that the contribution of non-EMEP sources to pollution is also much higher than that in the western part of the EECCA region. Therefore, information about emissions in non-EMEP countries presented in the EMEP domain (e.g., Iran, Syria, Iraq etc) is needed to improve calculations of pollution levels in the EECCA countries.

It is also interesting to mention, that the composition of contributions of secondary sources is highly different between two considered regions. In the European part of the EMEP region in 2011 the main contribution is denoted as 're-suspension from other sources'. In this case 'other' mostly includes re-suspension from urban lands. In case of Caucasus region, the main secondary source is re-suspension from bare lands of the Central Asian deserts. Additional research is needed to specify the role of dust suspension from bare lands. It is important not only to the EECCA region, but also for the other EMEP countries.

The contributions of anthropogenic, secondary and non-EMEP sources to pollution levels differ largely among the EECCA countries. Individual peculiarities of each EECCA country should be taken into account when assessing heavy metal pollution levels.

4. DISSEMINATION OF INFORMATION

Dissemination of the assessment results and other relevant information aimed at support of political decisions is of high importance. Annual reports containing current status of heavy metal pollution within the EMEP region are supplemented by presentation of information on the web. It provides more flexible and targeted assistance to national experts and authorities with data required for the environment protection regulations. Another important aspect is the information exchange with other international organizations and programmes.

4.1. Information on the web

A variety of relevant information on heavy metal pollution is available on the MSC-E website (<http://www.msceast.org>) (Fig.4.1). It contains model estimates of air concentration and deposition fluxes within the EMEP domain, estimates of heavy metal (first of all, mercury) dispersion on a global scale and evaluation of pollution levels on a local scale performed for selected countries in the framework of the EMEP case studies on heavy metals [Ilyin *et al.*, 2011]. In addition, description of the MSC-E chemical transport models is given along with information on input data used for modelling. Details of the research and assessment can be found in numerous EMEP reports and other publications also presented on the website.

Besides, detailed information on heavy metal pollution and other contaminants is given for each individual EMEP country. This country-specific information includes variety of data on emissions, measurements and model assessment for particular country collected in one place to make easier access to information and its analysis by national experts (as described in the next section). It should be noted that access to emission and measurement data is realized through the interlink with appropriate websites of CEIP and CCC. In addition, an experimental format of the web presentation (exemplified by the Czech Republic and Germany) has been developed to include also access from the same place to information on other pollutants (SO_x, NO_x, O₃, PM) as the interlink with the website of MSC-W. However, this approach needs further development and discussion within the Convention.

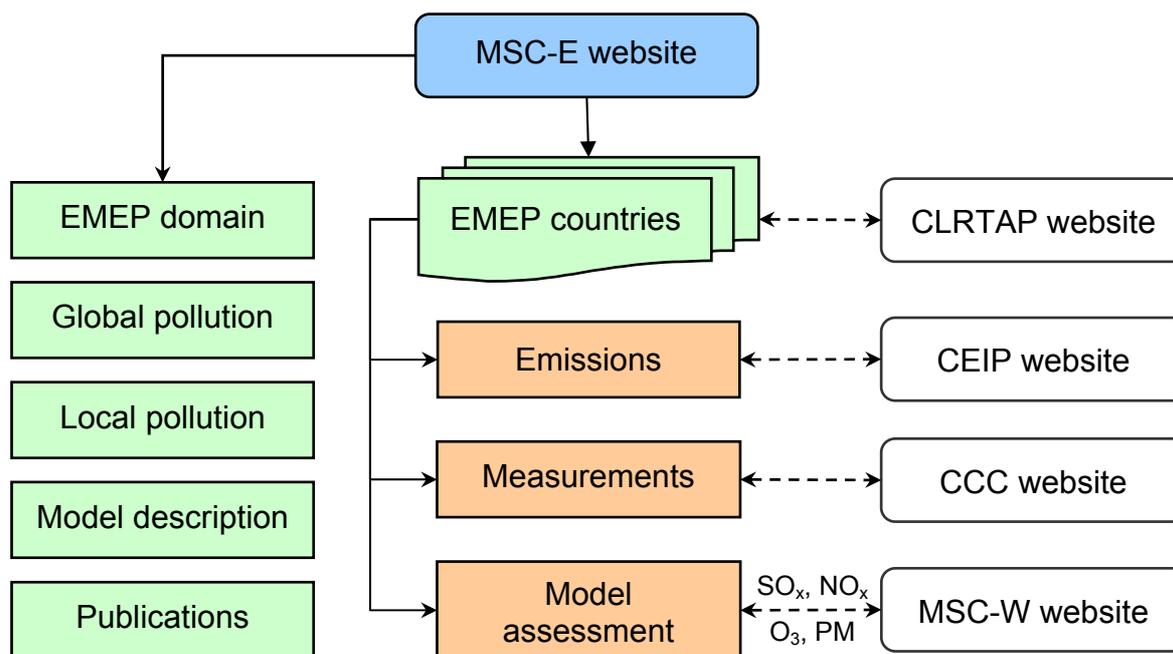


Fig. 4.1. General structure of information presented at the MSC-E website

4.2. Country-specific information

Detailed information on heavy metal pollution for each individual EMEP country is presented in the form of graphs, maps and data files on the MSC-E website (www.msceast.org). An additional work has been initiated to provide the EECCA countries with information in Russian (ru.msceast.org/). The country-specific information includes the following items:

- Emission data (spatial distribution and trends);
- Measurements (raw data, monthly and yearly statistics);
- Spatial distribution of pollution levels and their long-term changes;
- Transboundary pollution of a country;
- Contribution of national sources to transboundary transport;
- Ecosystem-specific deposition.

For each country, information on annual total deposition and mean annual concentrations in air with spatial resolution 50x50 km is presented (Fig. 4.2a). On the website a user can set gradations of deposition values. The maps are accompanied with distribution diagrams.

Information on long-term trends relates to the period 1990-2011. It includes country-averaged deposition fluxes in the considered period, smoothed trend line, and normalized deposition values in 1990, 1995, 2000, 2005 and 2010 (Fig. 4.2b). The normalized values are introduced in order to reduce the effect of annual meteorological variability on pollution levels. Besides, range between 5% lowest and 5% highest deposition fluxes over space (90% interval) and 25% lowest and 25% highest deposition fluxes (50% interval) are indicated.

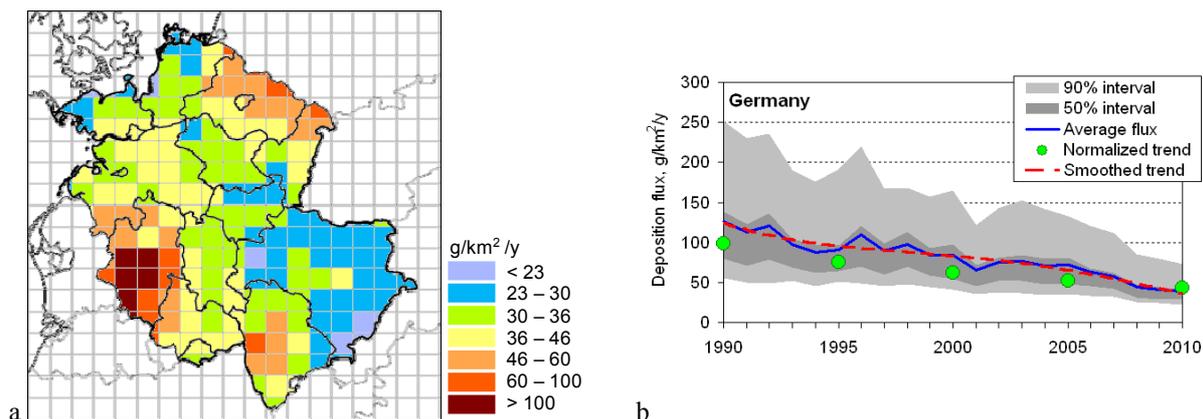


Fig. 4.2. Total deposition (a) and long-term deposition trend of cadmium (b) to Germany

Data on transboundary pollution of each country include maps of total deposition flux to a country's territory from foreign sources and their relative contribution to anthropogenic deposition. Besides, total deposition from foreign sources to a country's territory is presented (Fig. 4.3a). Only several main contributors are selected, while the inputs of the remaining countries-sources are summed in sector 'other'. Information on the contribution from all EMEP countries can be derived from source-receptor matrix also allocated on the web site.

Each country contributes to atmospheric pollution of other EMEP countries. Spatial distribution of deposition caused by country's sources is available for the entire EMEP region (Fig. 4.3b). Besides, country's total deposition to main regions-receptors (countries or seas) and its fraction of deposition within the EMEP region are indicated. Deposition to other regions is available in source-receptor matrix.

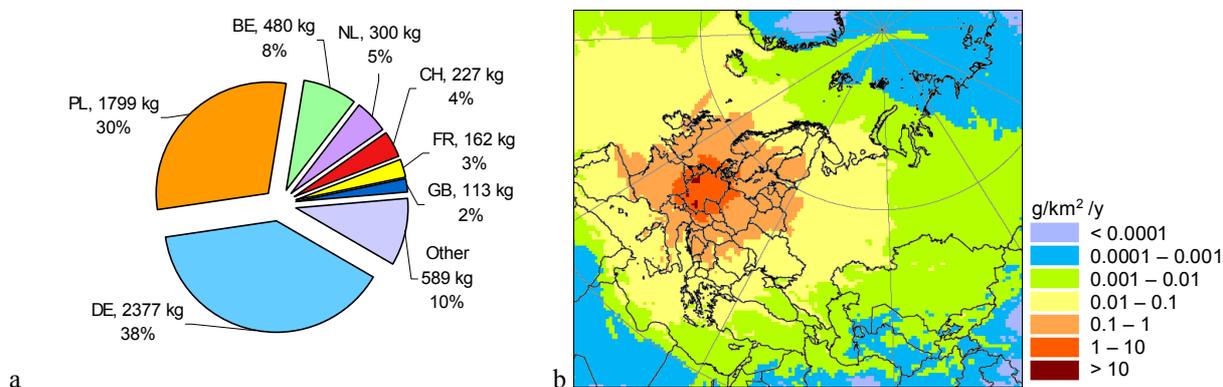


Fig. 4.3. Contribution of countries to cadmium anthropogenic deposition to Germany (a) and cadmium deposition from German national sources (b)

Deposition to different types of ecosystems is important information for the effects community. This information is used for evaluation of critical load exceedances. Country-specific maps of deposition to 17 land cover categories are available.

In addition to this, more detailed information can be provided. It can include pollution levels with fine spatial resolution, data on contamination of individual country's provinces, contribution of key source categories to pollution levels, pollution from large point sources etc. However, this information now is obtained in the framework of the EMEP country-specific case studies.

4.3. Information for international organizations and programmes

EMEP is the unique instrument within the LRTAP Convention providing regular pollution assessment and supporting Parties to the Convention with information on pollution levels (including heavy metals and POPs) in Europe and other regions. Therefore, there is a wide interest outside the Convention to the data products and analysis performed by the EMEP research Centres. 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 context of the co-operation MSC-E regularly exchanges information with different international bodies. These activities are performed in accordance with the EMEP workplan. Some aspects of this co-operation are discussed below.

A new global legally binding instrument on mercury (named Minamata Convention) was adopted in January 2013 by the Intergovernmental Negotiating Committee presenting more than 140 countries. The Minamata Convention will be opened for signature by governments at a Diplomatic Conference in October 2013 in Japan. In the course of preparation of the negotiations process a special document - Global Mercury Assessment 2013 – has been prepared. MSC-E actively participated in preparation of the Technical Background Report for the Global Mercury Assessment 2013 [AMAP/UNEP, 2013]. In particular, as a co-author of the assessment it provided model estimates and analysis of mercury atmospheric dispersion and deposition on a global scale.

An example of the analysis of intercontinental transport of mercury is illustrated in Fig. 4.4. The model estimates allow concluding that the contribution of the intercontinental transport is significant, particularly in regions with few local emission sources. In fact, the contribution of foreign anthropogenic sources to annual Hg deposition fluxes varies from 10% to 30% on average anywhere on the globe. Moreover, in areas of small domestic emissions, the contribution from domestic sources can be exceeded by contributions from foreign sources. Among the major contributors, East Asia is the most dominant source region, with annual contributions from anthropogenic sources of 10–14% to mercury deposition in other regions. In addition, 35–70% of total deposition to most regions consists of deposition contributed by global natural and secondary emissions.

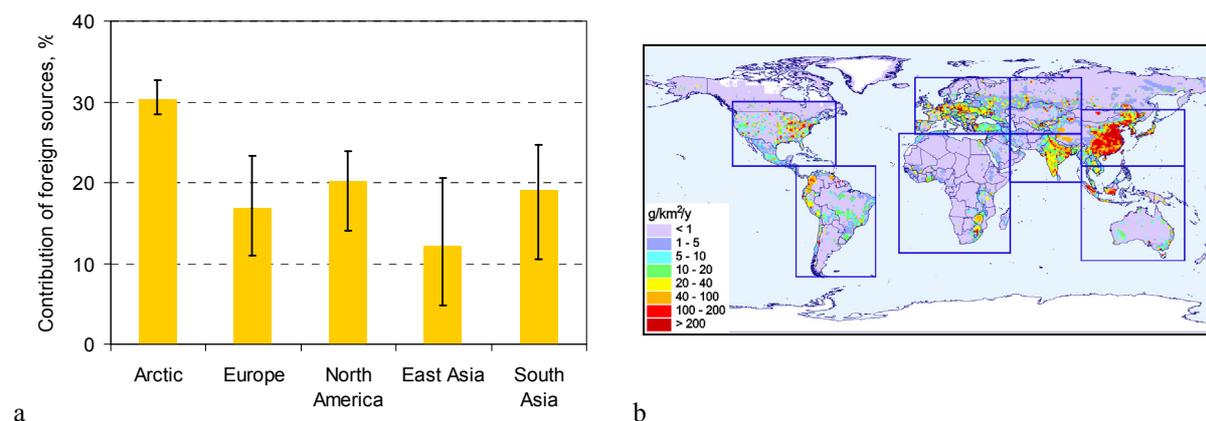


Fig. 4.4. Contribution of foreign anthropogenic sources to mercury deposition in different regions as simulated with the GLEMOS model (a). Global distribution of anthropogenic Hg emissions in 2005 and location of source/receptor regions – Europe, North America, East Asia, South Asia, Central Asia, Africa, South America, Australia and Oceania (b).

MSC-E has a long and successful experience of co-operation with the Arctic Monitoring and Assessment Programme, which includes participation in a number of joint assessments and projects. Specifically, the Centre was involved in the joint RAIPON/AMAP/GEF project focussed on the Russian Arctic pollution by persistent toxic substances [http://www.amap.no/resources/pts_project.htm; Dutchak et al., 2002], and took part in the regular AMAP Assessments of the Arctic pollution with mercury and other heavy metals [AMAP, 2005; 2011].

Fig. 4.5 illustrates the model assessment of the Arctic pollution with mercury. Spatial pattern of annual net deposition shows elevated deposition fluxes over the European and eastern sectors of the Arctic and over the Canadian Archipelago (Fig. 4.5a). The largest contribution to mercury deposition to the Arctic is made by East Asia, followed by Europe and Africa (Fig. 4.5b). It should be noted that the African contribution largely consists of mercury from natural and legacy sources. On the whole, contemporary anthropogenic sources contribute around one third of total deposition to the Arctic. Other two thirds come equally from terrestrial and oceanic sources which have both natural and legacy origin.

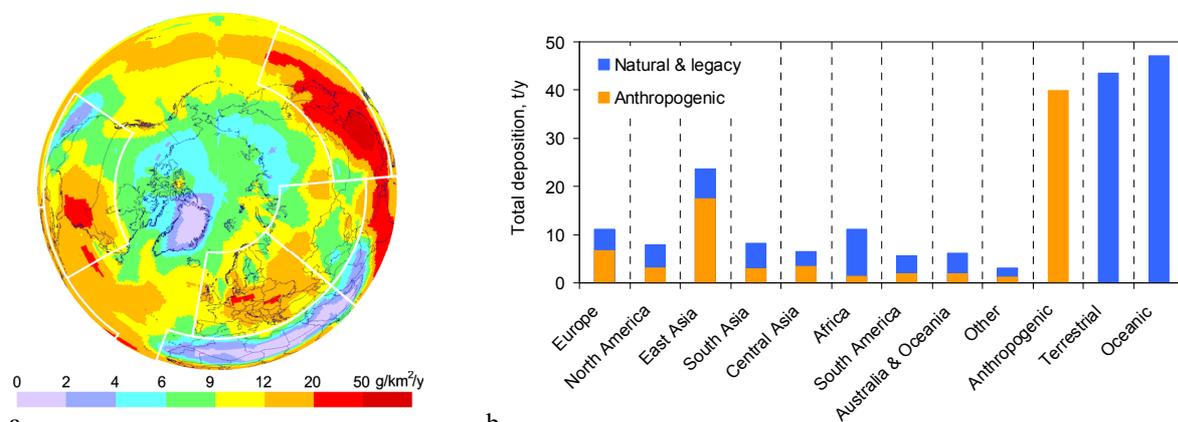


Fig. 4.5. Mercury annual net deposition flux over the Arctic and surrounding regions (a) and contribution of various source regions to mercury deposition in the Arctic (north of 66.5°N) as simulated with the GLEMOS model

Information on atmospheric pollution of marginal seas within the EMEP region is of interest for the marine conventions (e.g., HELCOM, OSPAR). In cooperation with the 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 HELCOM and the Convention and based on the long-term EMEP/HELCOM contract. Recent assessment of airborne pollution load to the Baltic Sea [Bartnicki et al., 2012] includes information on spatial distribution, long-term trends and source apportionment of lead, cadmium, and mercury deposition (Fig. 4.6a). Atmospheric load of heavy metals to the North Sea and the Northern Atlantic was evaluated by MSC-E in the framework of a number of contracts between EMEP and OSPAR (Fig. 4.6b). The assessment contained detailed study of the OSPAR maritime area pollution with lead, cadmium, and mercury, attribution of different emission categories and evaluation of the simulation results against observations from the OSPAR monitoring sites (CAMP) [Gusev et al., 2008].

It is important in future to formulate ways of working, outreach and information sharing to ensure more close long-term cooperation between the Convention and other relevant international bodies.

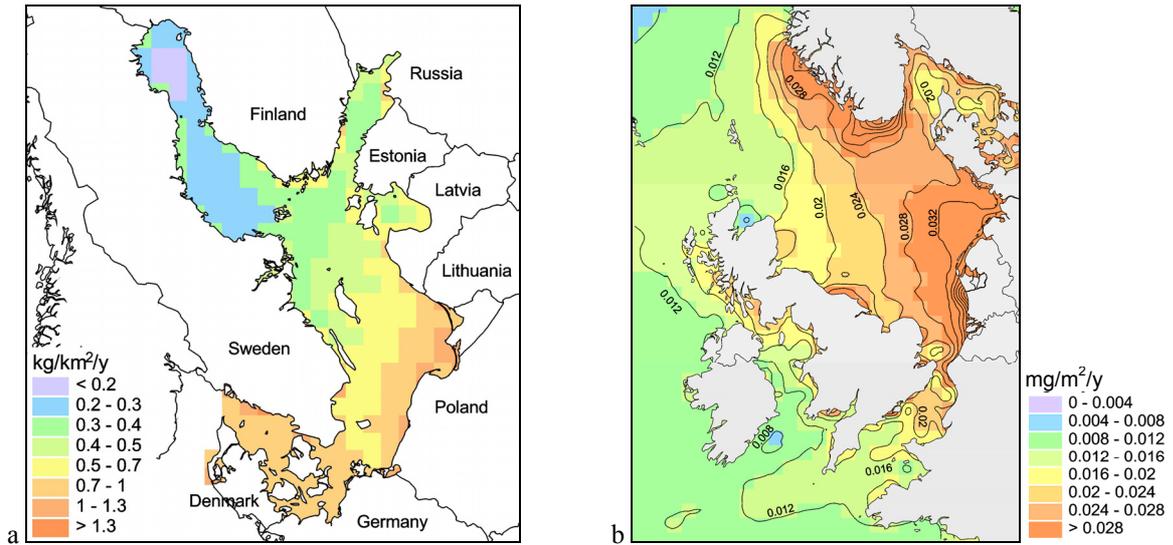


Fig. 4.6. Spatial distribution of lead deposition over the Baltic Sea (a) and cadmium deposition over the North Sea (b)

MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

Main activities of the EMEP Centres MSC-E and CCC in 2014 will be focused on the assessment of heavy metal pollution levels in the EMEP region and support of the EMEP countries with information required for implementation of the Protocol on Heavy Metals. Long-term changes of heavy metal deposition and transboundary transport in the EMEP domain will be assessed for the period 1990-2012. Particular attention will be paid to the pollution assessment in the EECCA countries.

Analysis of the results of Centres activities in 2013 allowed formulating problems and challenges affecting quality of the pollution assessment and suggesting directions of future research and improvements.

- Completeness of emission data, information on large-point sources, key emission source categories, and updated expert estimates of emissions are needed for further progress in the assessment of heavy metal pollution in the EMEP countries. It is particularly true for the EECCA countries. Tight cooperation between TFEIP and modelling community is needed to find the optimal solutions of the emission-related problems. Further development and application of the inverse modelling approach could also contribute to the analysis of emission data uncertainties.
- The EMEP monitoring network for heavy metals consists of more than sixty sites and covers significant part of the EMEP countries. However, there is still a need for better coverage in the eastern and southern parts of Europe and, especially, in the EECCA countries. Measurement data outside the EMEP region required for evaluation of global-scale modelling are also very limited. More active involvement of national data as well as data from international programmes could favour the improvement of the measurement data coverage. The situation with data coverage needs to be discussed within TFMM and TF HTAP.
- Mercury is a global pollutant. Emissions of mercury in regions outside EMEP substantially affect mercury levels in the EMEP region. For the successful evaluation of mercury in the EMEP countries emissions and monitoring data over the entire globe are needed. Besides, information on mercury speciation and historical emissions are required.
- Further development of the Global EMEP Multi-media Modelling System (GLEMOS) will be continued. It will include transition of operational calculations of heavy metals to the latitude-longitude projection and refinement of the model parameterizations of physical and chemical processes. The model program code will be distributed as an open source to support development of country-scale modelling approaches by national experts.
- In order to examine effects of transition from coarser to finer grid, special country-specific case studies will be continued with a focus on the assessment of heavy metal pollution in the Netherlands. Besides, it is planned to initiate similar work with one of the EECCA countries.
- Atmospheric speciation and chemistry of mercury are the main sources of uncertainty of modelled concentrations and deposition. In order to reduce the existing uncertainties and improve quality of the assessment it is planned to continue refinement of mercury chemistry modules. Besides, development of multi-media approach to simulation of mercury dispersion in the environment will be started with focus on the aqueous ecosystems.

- The relative contribution of secondary sources of lead and cadmium to pollution levels in the EMEP region has increased due to significant reduction of anthropogenic emissions since 1990. Investigation of the processes governing wind re-suspension of heavy metals will be continued to update parameterisations applied in the model. This work is to be performed in cooperation with TFMM and national experts.
- Development of the EMEP/MSC-E website will be continued with the aim at providing easy and handy access to all country-related information on emission, monitoring and modelling results of pollutants targeted by the Convention. Besides, a version of the website in Russian is to be developed to facilitate access to the information of Russian speaking experts from the EECCA countries.

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ASSESSMENT UNCERTAINTIES AND RESEARCH

Quality assurance is an important aspect of the pollution assessment. Any assessment results contain uncertainties imposed by restrictions of the method and inaccuracy of input data. MSC-E continuously works on both improvement of the models and update of input information required for the model assessment. Evaluation of the model results against observations and recent research activities of the Centre aimed at reduction of the assessment uncertainties are briefly discussed below. More details are available in the Technical Report [Shatalov *et al.*, 2013].

A.1. Quality of the model assessment

Assessment of pollution levels in the EMEP region is based on the information on the results of long-range transport modelling and measurement data. In regions where measurement data are available it is possible to evaluate modelling results via comparison of modelled values with the observed levels.

All components of the assessment – emissions, modelling approaches, measurement data – are subject to various uncertainties. Therefore, it is hardly possible to expect exact coincidence of modelled and measured parameters. If the difference between annual modelled and measured parameter is high (e.g., a factor of 2 or more) detailed analysis of reasons for this is needed. Comparison is carried out with two measurable parameters: concentrations in air and wet deposition fluxes. Major indicators of the comparison between modelled and measured quantities are summarized in Table A.1.

Table A.1. Main statistical indicators of agreement between annual modelled and measured levels of air concentrations and wet deposition fluxes for 2011.

| | Lead | | Cadmium | | Mercury | |
|-------------------------|------------------|---------|------------------|---------|------------------|---------|
| | C _{air} | Wet Dep | C _{air} | Wet Dep | C _{air} | Wet Dep |
| Relative bias | 30.89 | 15.75 | 12.96 | -48.80 | -1.89 | 25.30 |
| Correlation coefficient | 0.86 | 0.56 | 0.74 | 0.45 | 0.73 | 0.23 |
| NRMSE | 0.80 | 0.62 | 0.85 | 0.83 | 0.08 | 0.78 |
| F2, % | 70.59 | 72.34 | 71.88 | 56.00 | 100.00 | 73.33 |
| F3, % | 91.18 | 97.87 | 90.63 | 80.00 | 100.00 | 100.00 |

C_{air} – concentration in air

Wet Dep – wet deposition flux

NRMSE – Normalized Root Mean Square Error

F2 – share of values fitting to factor of 2 difference

F3 – share of values fitting to factor of 3 difference

As follows from the table, relatively good agreement between modelled and measured levels is noted for lead. The model performance for lead varies from between geographical regions. Over most part of Europe, covered by measurement stations, the agreement between modelled and observed concentrations in air and wet deposition fluxes is reasonable (Fig. A.1). However, observed levels were significantly (1.6 – 3.4 times) overestimated at the Dutch and Belgian stations. Similar overestimations are also noted for a few stations in other parts Europe. It is thought that the reason for this overestimation is too high contribution of wind re-suspension of lead in regions affecting these

stations. Currently the work on the improvement of wind re-suspension scheme is ongoing (see section A3).

For cadmium concentrations in air the situation is similar to that for lead: substantial (up to 2.9 times) overestimation is noted for the Dutch and Belgian stations, and also at a few stations in other parts of Europe (Fig. A.2a). This overestimation can also be attributed to the influence of wind re-suspension. However, for wet deposition the discrepancies between modelled and observed levels are higher than that for lead (Fig. A.2b). Model matches the observations relatively well at stations in the United Kingdom, Germany, Poland, Denmark, France, Slovenia. However, significant underestimation is noted for stations in Scandinavia, the Baltic region, in south-western (Spain) and some countries of central (Slovakia, the Czech Republic, Hungary) part of Europe.

Lead levels in 2011 are reproduced by the model reasonably well in the EMEP region. Special attention should be paid to overestimation of concentrations in the Benelux region, likely caused by uncertainties of wind re-suspension.

Lead and cadmium are both particulate pollutants. Therefore, their atmospheric behaviour is expected to be similar. However, the model performance for lead is significantly better than that for cadmium. Obviously, there are inherent model uncertainties, but they hardly can lead to 2-3 times deviation of modelled levels from the observed ones. Possible reason of the discrepancies could be uncertainties of the emission data. Even with significant wind re-suspension, contributing around 65-70% to modelled cadmium levels on average, its wet deposition still remain underestimated. Inverse modelling approach can be applied in order to identify regions where emissions need more detailed investigation. Theoretical background and some practical applications of this approach are described in MSC-E Technical report [Shatalov et al., 2013]. Finally, uncertainty of cadmium measurements may be higher, because of much lower levels than that of lead. For comparison, quantity of stations where measurements are below detection limit most part of the year, for cadmium is much higher than that for lead.

Although over most part of the region the model well matches the observed levels of cadmium, in some regions (Scandinavia, south-western and central part of Europe) underestimation of the observed levels takes place, likely because of emission uncertainties. Implementation of inverse modelling approach can help to identify regions where emissions need more detailed investigation.

Annual mean concentrations of mercury in air exhibit low variability in space. Both modelled and measured levels range within 1.4 – 1.8 ng/m³ (Fig. A.3a). The model tends to overestimate wet deposition fluxes of mercury (Fig. A.3b). Wet deposition depends on air concentrations of readily wet-scavenged mercury forms, which, in turn, are mostly products of atmospheric chemical transformations. Mercury chemistry is known to be highly uncertain at present. New scientific findings concerning atmospheric chemistry of mercury are analysed in detail before being included into modelling schemes. More information about it can be seen in section A.2 of this chapter as well as in the technical report.

Atmospheric chemistry of mercury is the major source of uncertainties. In order to improve model assessment of mercury levels in the EMEP countries, more research activity in this field is needed.

Annual mean concentrations of mercury in air exhibit low variability in space. Both modelled and measured levels range within 1.4 – 1.8 ng/m³ (Fig. A.3a). The model tends to overestimate wet deposition fluxes of mercury (Fig. A.3b). Wet deposition depends on air concentrations of readily wet-scavenged mercury forms, which, in turn, are mostly products of atmospheric chemical transformations. Mercury chemistry is known to be highly uncertain at present. New scientific findings concerning atmospheric chemistry of mercury are analysed in detail before being included into modelling schemes. More information about it can be seen in section A.2 of this chapter as well as in the technical report.

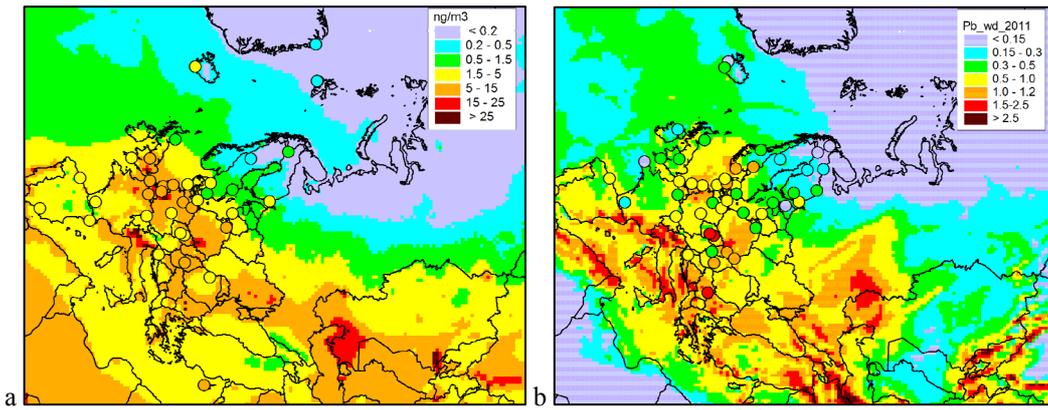


Fig. A.1. Modelled and observed air concentrations of lead air concentrations (a) and wet deposition (b) in 2011

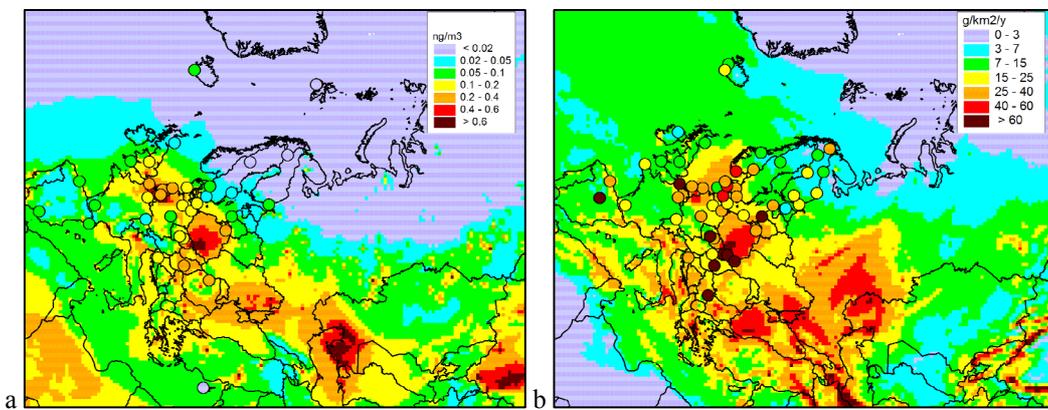


Fig. A.2. Modelled and observed air concentrations of cadmium air concentrations (a) and wet deposition (b) in 2011

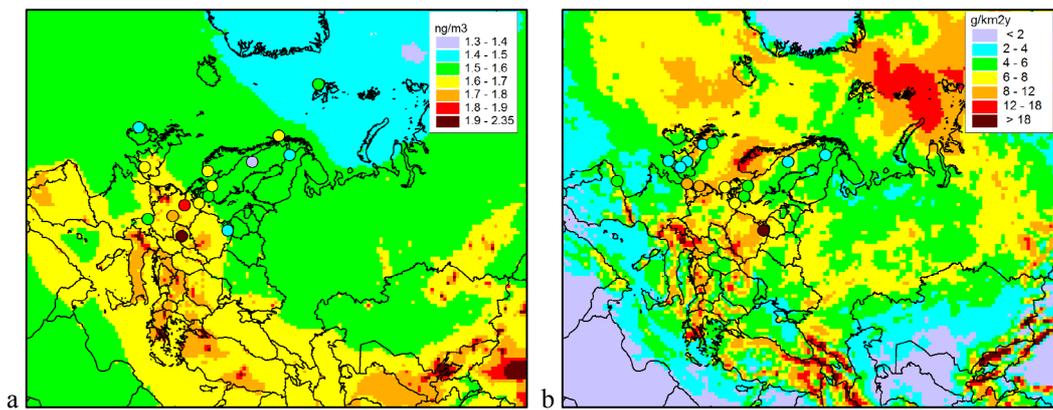


Fig. A.3. Modelled and observed air concentrations of mercury air concentrations (a) and wet deposition (b) in 2011

A.2. Atmospheric chemistry of mercury

Atmospheric chemistry plays important role in the chain of processes governing mercury fate in the environment starting from its emissions to the atmosphere and ending at ecosystems and human exposure. In particular, it defines lifetime of mercury in the atmosphere and the distance of its transport from emission sources. The MSC-E model successfully reproduces available observations of mercury concentrations and deposition fluxes. Nevertheless, there are significant gaps of knowledge on mercury oxidation and reduction processes in the atmospheres that restrict further model development. First of all, it relates to the dominating oxidation mechanisms in different atmospheric environments, kinetics and products of the reactions.

To study physical and chemical mechanisms a number of sensitivity runs were performed with various parameterizations and combinations of considered processes. The main focus of the analysis includes evaluation of speciation of Hg anthropogenic emissions; testing the major mechanisms of Hg oxidation; and examining the physical state of the oxidation products (gaseous or particulate). The simulation results were evaluated against detailed measurements of mercury concentrations in the continental boundary layer (EMEP site Waldhof, DE2). An example of the analysis is shown in Fig. A.4. Each panel of the figure presents concentration of gaseous oxidized mercury (GOM) at the Waldhof site as a product of one of the considered process: anthropogenic emissions (Fig. A.4.a), oxidation of Hg^0 by ozone (Fig. A.4.b), by OH radical (Fig. A.4.c) and by atomic Br (Fig. A.4.d).

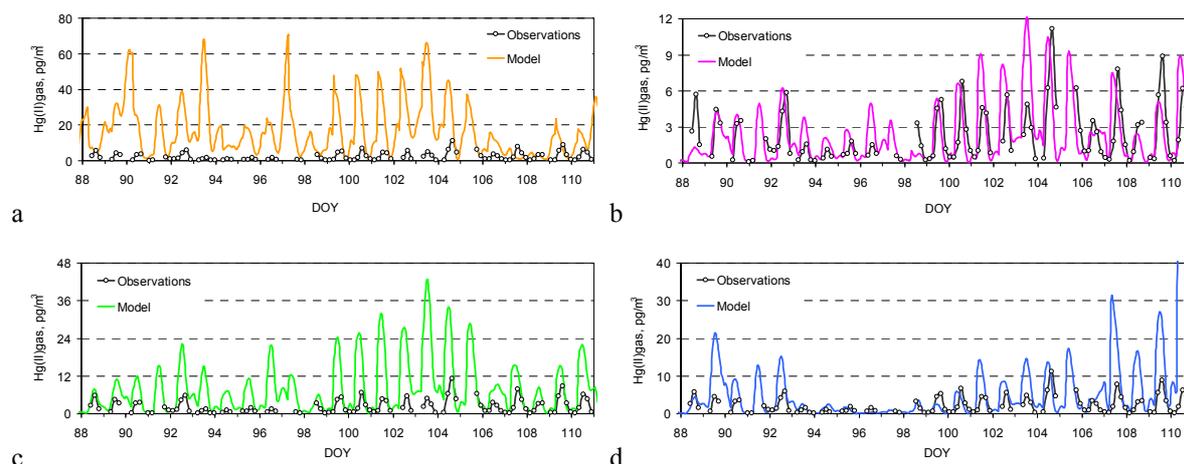


Fig. A.4. Evaluation of simulated gaseous oxidized mercury (GOM) against measurements at Waldhof (DE2) for different oxidation mechanisms: (a) – no oxidation, anthropogenic emissions of GOM; (b) – oxidation by O_3 , no GOM emissions; (c) – oxidation by OH, no GOM emissions; (d) – oxidation by Br, no GOM emissions

To evaluate mercury atmospheric chemistry in the upper troposphere the simulation results were compared with measurements performed onboard passenger Lufthansa aircrafts as a part of the CARIBIC project. Four CARIBIC flights were chosen for the analysis directed to different parts of the northern and southern hemispheres in 2009: Frankfurt-Caracas (22-23 Apr 2009), Frankfurt-Vancouver (23-24 Apr 2009), Frankfurt-Osaka (27-28 May 2009) and Frankfurt-Cape Town (27-29 Oct 2009) (Fig. A.5a). The characteristic feature of these measurements is very low concentrations of total gaseous mercury (below 0.6 ng/m^3) observed over long distances in most CARIBIC flights. These depletions characterise stratospheric parts of the flights and reflect deep oxidation of Hg^0 in the lower stratosphere.

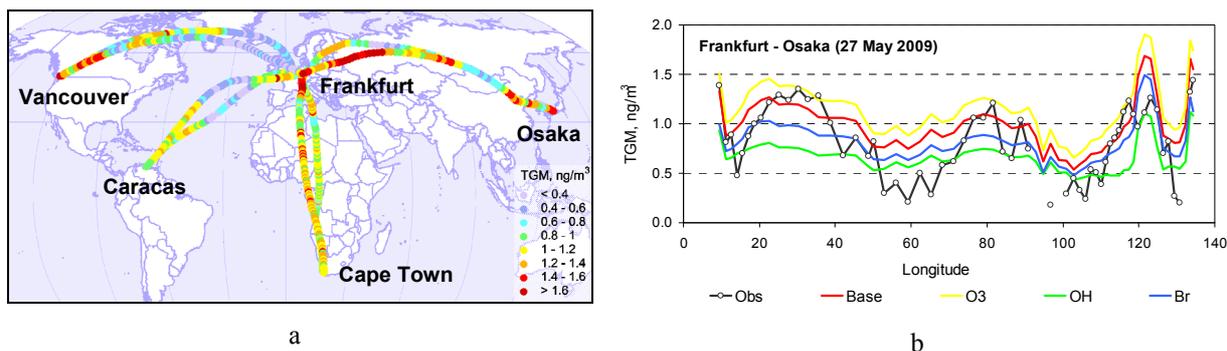


Fig. A.5. Tracks of the selected CARIBIC flights (a) and evaluation of different oxidation mechanisms for the flight to Osaka (b)

The flight to Osaka over the whole Eurasia includes a number of tropospheric and stratospheric sections with mercury concentrations dropping down to 0.2 ng/m^3 (Fig. A5b). The model successfully reproduces measured variation of total gaseous mercury during this flight; however, it underpredicts depth of the depletion in the stratosphere. Three tropospheric and four stratospheric sections are clearly seen in the simulated time series for all the sensitivity runs.

Based on the conducted analysis it is possible to conclude that available emissions inventories considerably overestimate a proportion of oxidized mercury forms in mercury anthropogenic emissions. Gaseous fraction of oxidized mercury most likely has the photochemical origin, whereas the particulate form is largely affected by direct anthropogenic emissions. Each of the treated chemical mechanisms reproduces the diurnal variation of oxidized mercury in the surface air but lead to overestimation of observations (except for oxidation by ozone). Besides, the model succeeds reproducing the spatiotemporal variation of mercury concentration in the upper atmosphere, in particular, the depletion of gaseous mercury in the lower stratosphere.

A.3. Model assessment with fine resolution: analysis of wind re-suspension

In section A1 it was demonstrated that the model overestimates air concentrations of lead at some stations, mainly located in the Benelux region. Modelled concentrations are formed by various sources, including anthropogenic emissions, wind re-suspension from land-cover categories (bare lands, urban, arable lands, and sea surfaces) and non-EMEP sources. In case of stations in Belgium and the Netherlands the major contributor is re-suspension of lead from urban territories (Fig. A.6). Most likely, the re-suspension flux from urban surfaces is overestimated in the model. One of current research activity in the MSC-E is aimed at the improvement of re-suspension flux to reduce discrepancies between modelled and observed concentrations in air.

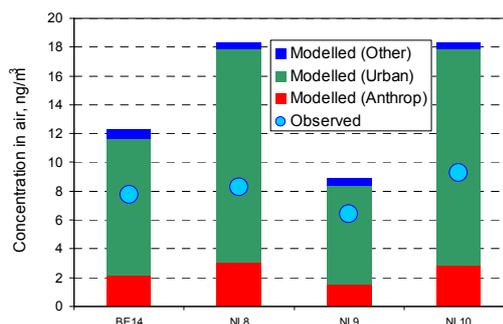


Fig. A.6. Modelled concentrations of lead in air in 2011 from different sources compared against values observed at the Dutch and Belgian stations.

Wind re-suspension of metals, as it assumed in the model, is calculated as a product of soil dust flux and soil concentration. Soil concentrations are based on data observed in background soils over Europe [http://weppi.gtk.fi/publ/foregsatlas/]. However, soil concentrations are usually measured at 10-20 cm depth, while suspension of soil dust occurs from the uppermost soil layer (roughly, first mm). In this uppermost soil layer the concentrations are typically enriched relative to those at 20-cm depth. Besides, in soils located close to areas of anthropogenic activity, metals have been accumulated over long-term history of air pollution. In order to take into account these two factors favouring increase of soil concentrations, observed soil concentration is multiplied by so-called enrichment factor.

This factor is highly uncertain. In MSC-E technical report [Shatalov et al, 2012] its value and spatial distribution was estimated, assuming proportionally to long-term anthropogenic deposition and reaching satisfactory fit between modelled and observed concentrations in air and wet deposition. Although this approach led to reasonably good agreement between modelled and measured pollution levels for EMEP region as a whole, in some areas it resulted to overestimation of the observed levels. Therefore, in some parts of the EMEP domain the enrichment factor is likely to be too high and should be corrected.

Overestimation of the observed air concentrations of lead in the Benelux region was examined in detail in the framework of the so-called EMEP country specific case studies. The main purpose of the case studies is to investigate in detail factors affecting the levels of heavy metal pollution in individual countries. Currently three countries – the Czech Republic, Croatia and the Netherlands are involved in these studies. Assessment of pollution in the framework of case studies for these countries is carried out with fine (5x5 or 10x10 km²) spatial resolution. It is important to note that scaling down from the EMEP region to the individual country allows to take into account more detailed country-specific information on emissions, measurements, land-cover etc. It is assumed that the experience reached in the studies for individual countries will be further applied for the entire EMEP domain.

In the framework of the case study work for the Netherlands pollution levels of lead with fine (5x5 km²) spatial resolution in 2007 were simulated. The transition from coarser (50x50) to finer resolution did not solve the problem of the discrepancies between modelled and observed concentrations in the Benelux region: the overestimation of measured levels remained (Fig. A.7a). Like in case of modelling over EMEP region, the main contributor to modelled air concentrations responsible for the overestimation is re-suspension from urban territories (Fig. A.7b).

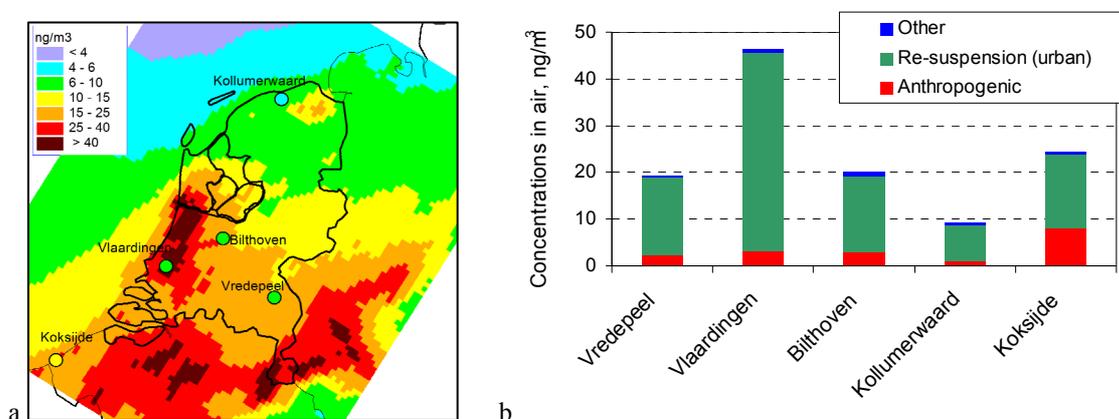


Fig. A.7. Spatial distribution of air concentrations over the Netherlands in 2007 with resolution 5x5 km², and observed concentrations at the Dutch and Belgian monitoring stations (a) and contribution of different sources to annual mean modelled concentrations at the stations (b).

Since re-suspension of metals depends on two factors (suspension flux of soil dust and concentration of lead in soil dust particles), their influence was analysed separately. In this section only main results are formulated, while the more detailed information can be found in technical report [Shatalov et al., 2013]. First of all, the dust flux was corrected via modification of soil moisture parameterization. It resulted to some reduction of modelled air concentrations and thus, better agreement between modelled and observed levels.

The further step was aimed at the correction of the enrichment factor for urban territories. It was established that the overestimation is noted mainly for the spring-time period: March and April. In these periods the overestimation makes up 2-6 times (Fig. A.8). For other periods the agreement between modelled and measured air concentrations was much better.

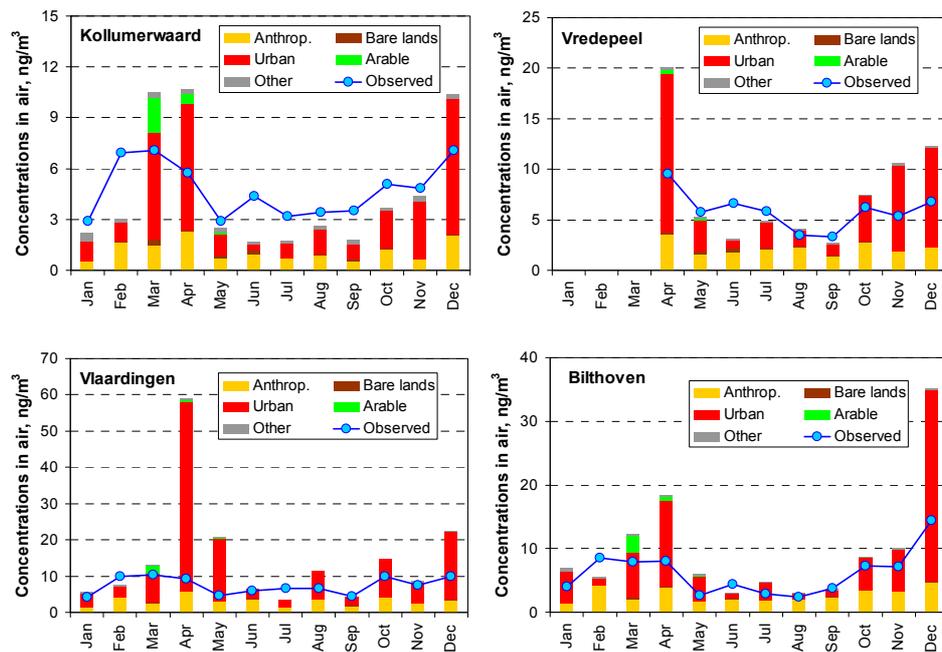


Fig. A.8. Monthly mean modelled ($5 \times 5 \text{ km}^2$) and observed concentrations of lead at the Dutch stations. Contributions of anthropogenic emissions and re-suspension from different types of land cover are identified.

In order to identify regions where enrichment factor needs correction, the method of inverse modelling was applied [Shatalov et al., 2013]. Using this approach the contributions of emission sources from individual gridcells in different periods of time were established. For example, at station Bilthoven the main contribution to air concentrations from re-suspension from urban territories in period from 7th to 19th of April comes from Belgium, central part of Germany and the United Kingdom (Fig. A.9a). On the base of combination of contribution maps for different periods and different stations a map of area where the enrichment factor needs reduction was established (Fig. A.9b).

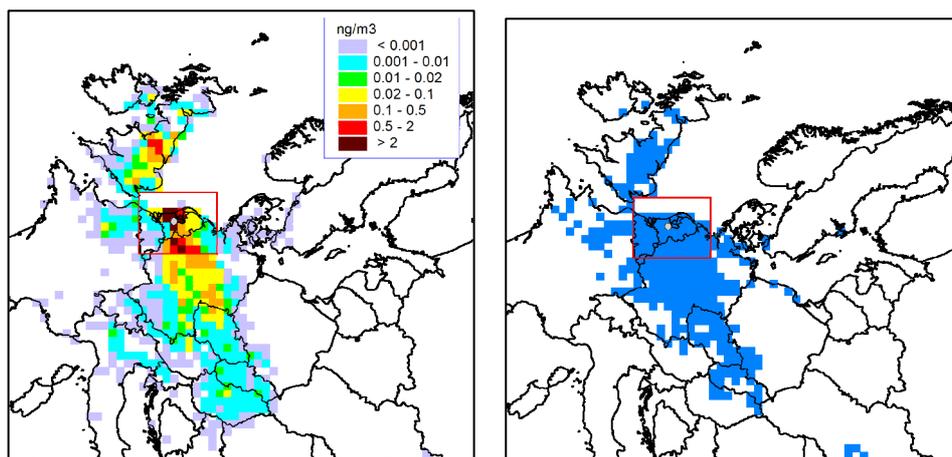


Fig. A.9. Contributions (ng/m^3) of re-suspension from urban areas to air concentrations at station Bilthoven in the period from 7 to 19th of April (a) and area, where emission factor for urban territories was reduced (b).

The correction of dust flux together with application of inverse modelling approach resulted to significant improvement of the modelled air concentrations, compared to the observed ones (Fig. A.10). In future this experience is planned to apply for the entire EMEP region.

On the base of the presented results it is possible to conclude the following:

- Formal transition to finer resolution does not always favours the improvement of modelling results. Detailed analysis of factors affecting the modelling results is needed.
- Wind re-suspension is important contributor to heavy metal levels. More detailed investigation of factors affecting re-suspension is needed (soil moisture, concentrations in soil and their enrichment)

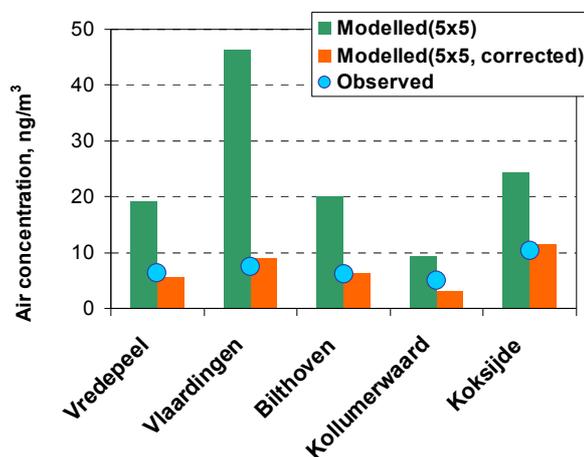


Fig. A.10. Mean annual initial and corrected modelled ($5 \times 5 \text{ km}^2$) and observed concentrations in air at stations of the Benelux region.