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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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Persistent Organic Pollutants in the Environment

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Persistent Organic Pollutants in the Environment

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EXECUTIVE SUMMARY

Persistent organic pollutants (POPs) is a group of substances characterized by toxicity, resistance to degradation, bio-accumulation in food chains, and significant potential to long-range transport. They are known to cause harmful effects to human health and ecosystems even at low concentrations and for a long time after their initial release to the environment. POPs are within the scope of the activity of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) since the adoption of the Protocol on POPs in Aarhus in 1998. Due to international cooperation and measures for pollution abatement within CLRTAP, supported by continuous scientific work on monitoring and assessment, pollution by POPs substantially decreased during the past two decades. However, levels of POP concentrations in the EMEP countries still pose risk to human health and ecosystems.

According to the Protocol on POPs the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) shall provide each annual session of the Executive Body of the Convention with information on the long-range transport and deposition of POPs within the EMEP region. Different aspects of the assessment of POP 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), and the Task Force on Emission Inventories and Projections (TFEIP).

Following the recommendations of the Bureau of the EMEP Steering Body this report is focused on major results of the EMEP Centres' activities in 2013 in support of implementation of the Protocol on POPs. It provides a short summary of the assessment of POP pollution in the EMEP region including overview of emission data used for modelling, status of the EMEP monitoring network for POPs, and analysis of pollution levels and trends based on the results of model simulations. Progress in the research and development activities was reported and discussed at the meetings of the EMEP Task Forces. Detailed description of the scientific work can be found in the MSC-E Technical report [*Shatalov et al*., 2013].

Emission data for model assessment

According to officially reported data complemented by expert estimates, emissions of PAHs, PCDD/Fs, and HCB to the atmosphere in the EMEP countries have been essentially reduced since 1990. The most significant decline of emissions in the period 1990 – 2011 is indicated for HCB (~85%) followed by PCDD/Fs (~60%) and PAHs (~35%).

Completeness and consistency of POP emission data provided by the EMEP countries are gradually improving, though official information on emission time-series, spatial distribution, and coverage of emission source categories still requires refinement. This is especially important in the process of forthcoming transition of emission reporting to finer spatial resolution.

Quality of POP pollution assessment significantly depends on the reduction of uncertainties in currently reported POP emissions. Uncertainties of official data along with the ways of their reduction were the subject of discussions at the recent EMEP meetings of TFMM and TFEIP with participation of the EMEP modelling Centres. Importance of complete time-series of emissions and coverage of the EMEP region by the gridded emission data was emphasized. Besides, significance of the data on chemical speciation of emissions, their seasonal variations, historical emissions, and emissions to the media other than the atmosphere (e.g. soil and water compartments) was reflected. This information is

currently obtained from available expert estimates, most of which requires updating. To analyze existing uncertainties in the emission data inverse modelling can be applied that is capable to indicate their potential reasons and areas where they most likely occur.

For some of the POPs non-EMEP emission sources essentially contribute to the pollution of the EMEP countries. Thus, to improve evaluation of their intercontinental transport there is a need to refine estimates of global emissions, which can be done in the framework of cooperation with international organizations and national experts.

The work on the refinement of emissions within the EMEP region, performed under the EMEP TFEIP, is essentially important for the improvement of the quality of model assessment of pollution.

Monitoring of POPs

The EMEP monitoring network included 29 sites in 2011 measuring various POPs either in air or in precipitation. Among other POPs, PAHs are more frequently measured within EMEP. The number of sites measuring HCB concentrations was considerably smaller. It should be mentioned that regular measurements of PCDD/F concentrations are currently not implemented at the EMEP monitoring sites. At the same time, a lot of measurements of PCDD/Fs were made by national monitoring campaigns. Collection of these data in the EMEP database and their analysis are highly desirable for the assessment of PCDD/F pollution.

Another important issue for the assessment of POP pollution is spatial coverage of the EMEP region by measurements. Though the number of sites is gradually increasing, spatial coverage is not sufficient yet. Most of the sites of the EMEP monitoring network for POPs are located in the northern and western parts of Europe, while the eastern and southern parts as well as the Central Asian countries are still poorly covered. In this respect passive sampling campaigns could essentially complement monitoring of POPs in the EMEP domain.

Monitoring of POPs in other regions of the globe is important for the evaluation of POP intercontinental transport. A lot of measurement data on POPs exist in various international organizations and programs (e.g. the Arctic Monitoring and Assessment Programme (AMAP), OSPAR Commission, United Nations Environment Programme (UNEP)), however not all of them are freely available due to different restrictions. Thus, it would be reasonable to consider possibilities for simplifying the access to such measurements for scientific purposes through the EMEP database.

Since POPs are multi-media chemicals, collection of available national monitoring data on their concentrations in the environmental media other than the atmosphere is of importance for the evaluation of pollution levels and harmful effects on biota and human health.

Questions of further development of the EMEP monitoring network for POPs, collection of additional POP measurements, and their accessibility through the EMEP database are proposed to be discussed at the TFMM and TF HTAP meetings.

Assessment of POP pollution in the EMEP region

Assessment of POP pollution was performed for PAHs, PCDD/Fs, and HCB for the period 1990 – 2011. Results of the assessment characterize contemporary levels of pollution and their changes in the EMEP countries in comparison to the base year (1990).

Pollution of the EMEP region is formed by different sources of POPs, including anthropogenic emissions of the EMEP countries, non-EMEP (global) emission sources, and secondary emissions. Particularly, pollution by PAHs is resulted mainly from anthropogenic emissions within the EMEP domain, while contribution of non-EMEP emission sources is relatively small. For PCDD/Fs and HCB there is a need to take into account intercontinental transport and secondary emission sources. Long-term variations of pollution levels in the EMEP domain for considered POPs are controlled by both changes of emissions and variability of meteorological conditions. The latter one can alter pollution levels from year to year in some regions by almost 30%.

Polycyclic Aromatic Hydrocarbons (PAHs)

Model assessment indicates that PAH pollution levels in the EMEP region decreased by 30% from 1990 to 2011. The highest decline is indicated for the EU countries (almost 40%). Reduction of PAH pollution levels was more significant during the 1990s. In particular, B[a]P air concentrations in the EU countries dropped by 38% from 1990 to 2000. However, after 2000 the decreasing trend was almost levelled off and during the recent years B[a]P air concentrations in more than a half of the EU countries tended to increase following the growth of their emissions.

Changes of PAH pollution levels in other EMEP countries, especially in the eastern part of Europe and in the Central Asian countries, were comparatively small. According to model estimates and available national measurements, levels of B[a]P in air in some areas of these countries were significant and thus could pose risk to ecosystems and human health.

Transboundary transport of PAHs continued to play significant role in the period from 1990 to 2011. Though the total deposition of B[a]P in the EMEP countries decreased from 300 to 185 tonnes per year during that period, the relative contribution of transboundary transport remained essential accounting for about 50%.

Results of model simulations of PAH pollution levels reasonably well agree with observed annual mean concentrations. At the same time, more significant deviations between the modelled and measured air concentrations are indicated for intra-annual variations. Further improvement of the evaluation of PAH pollution on seasonal level can be achieved by the refinement of seasonal variations of PAH emissions and the application of refined parameterizations of PAH gas-particle partitioning to and degradation on different aerosol components, particularly, elemental and organic carbon.

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)

PCDD/Fs are characterized by essential persistence in the environment. They remain to be hazardous to human health and ecosystems even at low concentrations long after the moment they were originally emitted. Complex assessment of PCDD/F pollution in the EMEP countries is currently hampered by the absence of regular measurements of PCDD/Fs at the EMEP monitoring sites. Thus, model evaluation of PCDD/F long-range transport and fate pollution is the only source of the information on their pollution levels in the EMEP countries.

According to the results of model assessment PCDD/F pollution levels in the EMEP region decreased from 1990 to 2011 by 55%. Similar to PAHs, the highest decline is indicated for the EU countries (75%). Pollution of the EMEP region by PCDD/Fs is controlled by both the primary anthropogenic and secondary emission sources (re-volatilization from the soil and water compartments to the atmosphere). Secondary emissions can contribute more than 50% to total PCDD/F deposition.

Model simulations indicate that more than a half of PCDD/F deposition from anthropogenic emission sources within the EMEP domain is contributed by transboundary transport between the EMEP countries. Additional contribution is also expected from the non-EMEP emission sources. At the same time, data on emissions outside of the EMEP region are currently available only for a few countries. Thus, to refine estimates of PCDD/F intercontinental transport global inventory of PCDD/F anthropogenic emissions is needed.

Major uncertainties in the evaluation of PCDD/F pollution levels can be associated with incompleteness of data on anthropogenic emissions and poor knowledge on contribution of the secondary emissions. Particularly, officially reported anthropogenic emissions most likely do not cover all potential source categories, which is pointed out in a number of PCDD/F emission inventories. Further, secondary emissions to the atmosphere can be affected by the direct releases of PCDD/Fs to soil and water compartments due to various activities including heat and power generation, open burning of biomass, disposal of wastes, etc. Experimental model simulations with the emissions to the atmosphere and soil indicate reasonable agreement of model results with available measurements of PCDD/F concentrations in air and soil made by national campaigns. Thus, to improve evaluation of PCDD/F pollution in the EMEP countries there is a need to improve official emission inventories and to take into account emissions to soil and water.

Hexachlorobenzene (HCB)

HCB is a pollutant of global dispersion with significant potential to the cycling between the environment compartments. Modelling of HCB global transport and accumulation in the environment, performed on the basis of the scenario of historic and contemporary HCB emissions, indicated significant decline of HCB pollution within the EMEP region from 1990 to 2011 (by almost 90%). As primary emissions decreased the variability of HCB air concentrations in the EMEP region reduced substantially, leading to their relatively uniform spatial distribution.

Contemporary pollution of the EMEP region by HCB is dominated by the secondary emission sources of the EMEP countries and non-EMEP emission sources, while contribution of anthropogenic HCB emissions is comparatively small. According to modelling results, contribution of secondary HCB emissions to the pollution levels accounts for more than 60%. Elevated levels of HCB concentrations can be seen in the areas of its historic production and use for agricultural purposes, resulted in substantial accumulation of this pollutant in soil and sediments. Taking into account significant resistance of HCB to degradation, future levels of air pollution by HCB can be supported for quite a long time by secondary emissions.

Intercontinental transport of HCB substantially contributes to the pollution levels in the EMEP region (by almost 35%). High levels of HCB air concentrations in some regions outside of the EMEP domain, particularly, in Eastern and Southern Asia, indicate the presence of essential emission sources, which can contribute to the pollution of the EMEP countries.

Further improvement of the evaluation of HCB pollution levels requires the refinement of global historical and contemporary emissions of HCB, including also improvement of officially reported data within the EMEP region. Besides, to study the cycling of HCB in the environmental compartments, it is necessary to update the information on various characteristics of the surface media (e.g. organic matter content) and parameterizations of the degradation and phase partitioning processes, which influence the accumulation of HCB in media.

New substances

POPs are included into the work of many international organizations (LRTAP, the Stockholm, Basel, and Rotterdam Conventions, the European Union (EU), AMAP, OSPAR, Helsinki Commission (HELCOM), etc.) representing thus wide concern of POP pollution in most of the regions of the globe. New substances continue to be studied with respect to their harmful effects on human health and ecosystems and be identified as candidates to POPs as at the national level and under different international agreements (CLRTAP, the Stockholm Convention). Environmental observations along with modelling can play important role in the screening process and evaluation of the fate and potential effects of new substances. In this respect the European Community Regulation REACH (Registration, Evaluation, Authorisation and Restriction of Chemical substances) may be a source of information on their physical-chemical properties, emissions, and the results of risk assessment.

POP pollution in the EECCA countries

Providing information on POP pollution to the EECCA countries to facilitate their joining to the Protocol on POPs is one of the priority goals of the LRTAP Convention. This goal is clearly stated in the revised version of the Action Plan to involve the EECCA countries in the work of the Convention [ECE/EB.AIR/WG.5/2007/17]. At present more than a half of the EECCA countries started reporting their national inventories of POP emissions. Monitoring of POP air concentrations is initiated at two EMEP sites in Republic of Moldova and Kazakhstan. In line with measurements of these sites, national data on POP concentrations in the EECCA countries can be used for the evaluation of POP pollution levels (for example, monitoring of POPs in the Russian and Ukrainian cities and reserves).

Model assessment indicated reduction of POP pollution in the most of the EECCA countries from 1990 to 2011. Particularly, the highest decline of air concentrations is obtained for HCB, while for PCDD/Fs and PAHs it is relatively small. It should be noted that decrease of POP pollution levels in this region do not follow the decline in the EU countries. Comparable levels of pollution reduction can be seen in the western part of the region, which is mostly affected by the emission changes in the EU countries.

Further improvement of the evaluation of POP pollution levels in the EECCA region requires the refinement of information on their emissions. In this respect it is necessary to assist these countries in preparation of their national emission inventories. Additional efforts are required to increase the number of monitoring sites and involve national measurement data on POPs obtained within the EECCA countries. To support the work on the pollution assessment in these countries specific case studies on POPs can be organized with the participation of national experts in emissions, monitoring and modelling as well as with the support from the EMEP Centres. Special attention needs to be paid also to the development of Russian version of the EMEP/MSC-E website to assist the EECCA countries with relevant information.

Dissemination of information on POP pollution

Dissemination of POP assessment results and other relevant information is of high importance for the environment protection regulations. This information can be accessed as through annual reports and web presentation (http://www.msceast.org), which provides more flexible and targeted support to experts and authorities with the required data. Relevant information on POP pollution levels includes country-specific data for particular EMEP countries as well as estimates of pollution for the EMEP region and intercontinental transport of POPs.

An experimental format of the web presentation, exemplified by the Czech Republic and Germany, has been proposed. Along with the information on POPs and HMs it includes access from the same place to information on other pollutants (SO_x, NO_x, O_3, PM). At the same time, this format requires further development with the participation of other EMEP Centres and discussion within the Convention.

Exchange of information on POPs with various international organizations and programmes is highly desirable. Information on POP levels and trends within the EMEP region, POP deposition to marginal seas, and intercontinental transport of POPs, generated by EMEP, is of interest for different international organizations (e.g. AMAP, HELCOM, OSPAR, and the Stockholm Convention). To support further cooperation and exchange with this information it would be reasonable to consider establishing of specific agreements with international organizations and programmes dealing with the assessment of POP pollution and impact.

CONTENTS

INTRODUCTION

Persistent organic pollutants (POPs) is a group of semi-volatile toxic substances that resist to degradation and bio-accumulate in food chains causing harmful effects to human health and ecosystems. Particularly, they can be accumulated in soil, water bodies, vegetation, and sediments and then be re-emitted back to the atmosphere. Pollution by POPs substantially reduced during the past two decades. However, levels of their concentrations in the environment still pose risk to human health and ecosystems.

POPs are within the scope of the activity of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter the Convention) since the adoption of the Protocol on POPs in Aarhus in 1998. According to the Article 9 (3) of the Protocol, EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) provides the Convention with information on the long-range transport and deposition of POPs within the EMEP region. Different aspects of the assessment of POP pollution are covered by the EMEP Scientific Centres: Centre on Emission Inventories and Projections (CEIP), Chemical Co-ordinating Centre (CCC), Meteorological Synthesizing Centre – East (MSC-E). The work of the Centres is conducted in collaboration with the Task Force on Measurements and Modelling (TFMM), the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), and the Task Force on Emission Inventories and Projections (TFEIP).

The report is focused on major results of the EMEP Centres' activities in 2013 in support of implementation of the Protocol on POPs. It presents a short summary of the assessment of POP pollution in the EMEP region including overview of emission data used for modelling, status of the EMEP monitoring network for POPs, and analysis of pollution levels and trends based on the results of model simulations. Model assessment contains characteristics of long-term changes of air concentrations of PAHs, PCDD/Fs and HCB, transboundary transport between the EMEP countries, including contributions of secondary sources, and information on POP pollution that can be used for the evaluation of effects (Chapter 1). Additionally, intercontinental transport of POPs on a global scale is characterized in the context of influence of other regions and continents on the pollution in the EMEP region as well as contamination of other regions by the emission sources of the EMEP countries (Chapter 2).

Particular attention is paid to the assessment of POP pollution levels in the EECCA countries (Chapter 3). Assessment of pollution in this region is rather uncertain due to significant gaps in information on emissions and lack of monitoring data. Available estimates show that pollution changes in these countries do not generally follow the overall European reduction. Therefore, involvement of the EECCA countries into the active co-operation is one of the priority tasks within the Convention.

Dissemination of the scientific information aimed at support of political decisions is of high importance (Chapter 4). Along with 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 significant aspect is information exchange with other international organizations and programmes, which broadens the dissemination scale and strengthens the EMEP status on the international level.

In line with the information on pollution levels and their changes from 1990 to 2011, main challenges and needs for further investigations directed to the refinement of pollution assessment are highlighted for each of the considered POPs. They are, inter alia, lack of monitoring data on PCDD/Fs under EMEP, evaluation of contamination of soil and water compartments due to both accumulation of pollutants from the atmosphere and direct emissions of POPs to these media (PCDD/Fs and HCB),

and interaction of the pollutants (PAHs and PCDD/Fs) with atmospheric aerosol components (including elemental and organic carbon). In this context, significant efforts are undertaken for updating and improving modelling approaches and input information to enhance the assessment quality. Regular activities in this direction include, but are not restricted to, evaluation of the modelling results against measurements, development of the model parameterizations, and development of inverse modelling approach for the analysis of possible emission uncertainties. These issues can be topics of scientific discussion at the EMEP Task Forces (TFMM, TFHTAP, and TFEIP). Detailed description of the research activities can be found in the EMEP/MSC-E Technical Report [*Shatalov et al.*, 2013].

1. POLLUTION OF THE EMEP REGION

In accordance with the requirements of the POP Protocol, EMEP should perform assessment of POP pollution within the EMEP domain and provide information on deposition and transboundary transport for the EMEP countries.

Assessment of POP pollution was performed for PAHs, PCDD/Fs, and HCB for the period 1990-2011 to characterize contemporary levels of pollution and their long-term changes in the EMEP countries in comparison to the base year. Model simulations were carried out using the latest version of the MSCE-POP model on the basis of gridded emission data reported by the EMEP countries complemented by available expert estimates. Assessment of pollution includes analysis of long-term trends based on both measurement data of the EMEP monitoring network and modelling results. Transboundary transport of the considered POPs is characterized along with the evaluation of contributions of secondary sources of POPs and intercontinental transport.

1.1. Emission data for model assessment

Completeness and consistency of POP emission data provided by the EMEP countries have been essentially improved in the recent years. Thus in 2013, emissions of POPs at least for one year in the period 1990-2011 were reported by 41 countries. Data for the base year 1990 and the year 2011 were provided by 33 countries and 28 countries submitted gridded emissions of the considered POPs. At the same time POP emissions officially reported by countries are still the subject of uncertainties and require further improvements [*Mareckova et al.*, 2012].

To perform model assessment of POP pollution levels and trends (1990-2011) within the EMEP domain gridded emission data on PAHs, PCDD/Fs, and HCB were generated by CEIP and MSC-E. Particularly, datasets of annual gridded emissions for 2011 with spatial resolution 50x50 km^2 were provided by CEIP. MSC-E prepared gridded emission data for the year 1990 using official emissions submitted by countries in the current reporting year. Gaps in the temporal and spatial distribution of official emissions were filled by the emission expert estimates worked out by TNO [*Denier van der Gon et al.*, 2005] and MSC-E.

In order to evaluate contributions of intercontinental transport and secondary sources (re-volatilization to the atmosphere) scenarios of global HCB and PCDD/F emissions (including historical emissions) were prepared. Additionally, for PCDD/Fs importance of consideration of emissions to media other than the atmosphere (soil, water bodies) was evaluated on the basis of inventory of PCDD/F emissions to soil and water compartments compiled in the framework of the EU project on dioxins [*Wenborn et al.*, 1999].

Changes of POP emissions in the EMEP countries in the period 1990-2011

Long-term changes of POP pollution in the EMEP domain depend on changes of anthropogenic emissions of the EMEP countries as well as on the variations of secondary emissions (re-volatilization) which are determined by the accumulation of pollutants in surface media (soil, water bodies).

Considering the changes of anthropogenic emissions, it can be seen that PAH, PCDD/F, and HCB emissions to the atmosphere in the EMEP countries have essentially changed during the two recent decades. The most significant decline of emissions in this period is indicated for HCB (~85%) followed

by PCDD/Fs (60%) and PAHs (~35%). More substantial decrease of emissions took place in the western and central European countries of the EMEP domain, while in the EECCA countries emissions did not changed much or even increased.

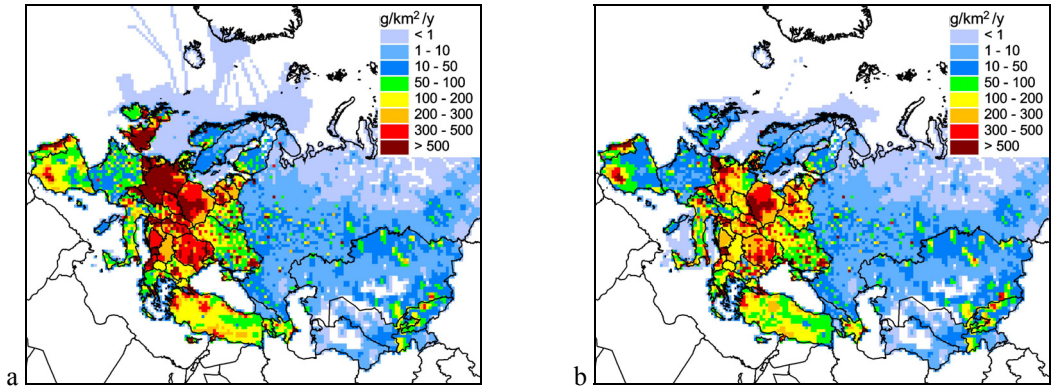


Fig. 1.1. *Spatial distribution of emissions of the sum of 4 PAHs in the EMEP countries in 1990 (a) and in 2011 (b) with resolution 50x50 km².*

PAH emissions in the EMEP countries declined from 2470 tonnes in 1990 down to 1570 tonnes in 2011 (Fig. 1.1). Compared with 1990, lower PAH emissions for 2011 can be seen in 34 EMEP countries. The largest decrease of PAH emissions is noted for the UK (96%), the Netherlands (81%), Cyprus (76%), and Republic of Moldova (73%). At the same time in Denmark, Belarus, Estonia, and Italy PAH emissions increased in comparison to their levels in 1990 by 91%, 18%, 17%, and 14%, respectively.

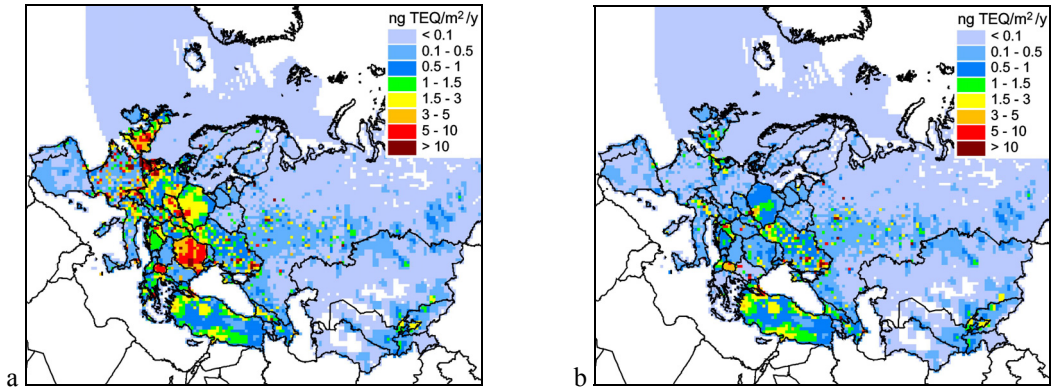


Fig. 1.2. *Spatial distribution of PCDD/F emissions in the EMEP countries in 1990 (a) and in 2011 (b) with resolution 50x50 km².*

Emissions of PCDD/Fs dropped from about 15 kg TEQ* in 1990 to 6.4 kg TEQ in 2011 (Fig. 1.2). According to the reported information PCDD/F emissions were lower in 2011 in comparison to 1990 in 38 EMEP countries. The most significant decline of emissions took place in Luxembourg (41 times), the Netherlands (24 times), France (19 times), and Iceland (18 times). In several countries, namely, in Belarus, Bulgaria, Albania, and Lichtenstein, PCDD/F emissions in 2011 were higher by 63%, 59%, 22%, and 12% comparing to their levels in 1990.

* - Toxicity of PCDD/Fs is expressed according to the NATO toxic equivalents scheme (I-TEQ)

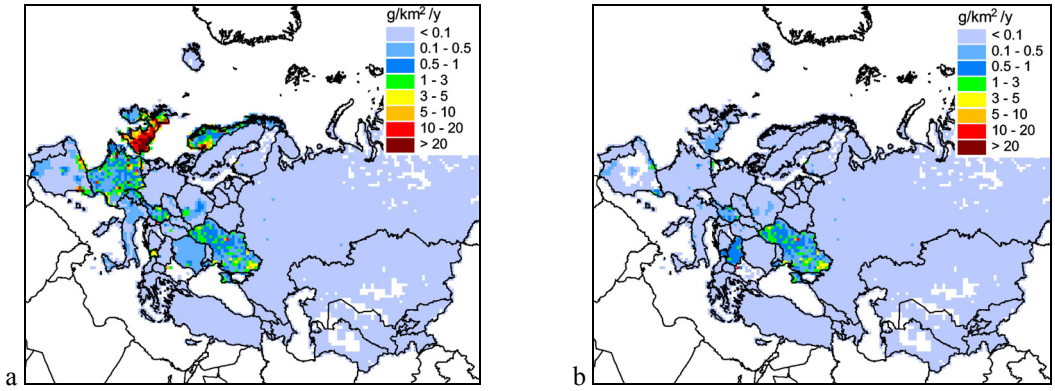


Fig. 1.3. *Spatial distribution of HCB emissions in the EMEP countries in 1990 (a) and in 2011 (b) with resolution 50x50 km².*

The degree of reduction of HCB emissions from 1990 to 2011 comparing to PAHs and PCDD/Fs is essentially higher (Fig.1.3). In that period HCB emissions decreased in 31 EMEP countries changing from 7.2 tonnes in 1990 to 0.98 tonnes in 2011. Among particular countries the largest decrease was reported by the United Kingdom (about 130 times) followed by Slovenia (94 times) and France (76 times). At the same time, some countries reported increase of HCB emissions in that period, particularly, Serbia (more than 500 times), Estonia (2.9 times), the Netherlands (2.9 times), and Luxembourg (2.7 times).

Officially reported emission data indicate inhomogeneous changes of emissions within the EMEP domain in the period 1990-2011 (Fig. 1.4). Particularly, the decrease of emissions was more intensive in the beginning of the period, while during the recent years some of the EMEP countries reported increasing of their emissions. Besides, the rate of emission changes was different in different parts of the EMEP domain. To evaluate the effect of these changes on the variations of POP pollution levels model assessment of long-term trends was performed based on the emission data for 1990-2011.

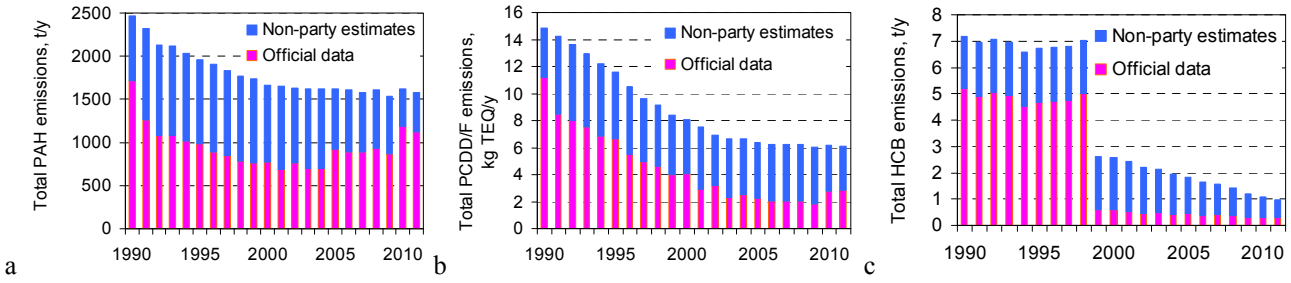


Fig. 1.4. *Temporal changes of PAHs (a), PCDD/Fs (b), and HCB (c) emissions in the EMEP countries from 1990 to 2011*

Key source categories of POP emissions

Information on POP emissions from individual emission sectors is important for preparation of temporal variations of emissions and their congener composition for complex mixtures (e.g. PCDD/Fs). Temporal variations of B[a]P emissions for the EMEP domain were constructed on the basis of source sector distribution of emissions using the approach described in the EMEP Status report [*Shatalov et al.*, 2012a]. The use of temporally resolved B[a]P emissions led to noticeable improvement of model assessment of pollution. This approach is planned to be applied for the evaluation of temporal variations of PCDD/F pollution levels.

Uncertainties of POP emissions

To characterize the quality of pollution assessment the information on the level of uncertainties of input data, particularly, emissions, is required. Currently only small part of the EMEP countries provides uncertainties of their officially reported POP emissions. According to this information, the level of uncertainties of reported POP emissions is quite substantial. To take this into account model assessment of pollution on the basis of official emission data can be complemented with scenario model simulations to indicate uncertainty range of modelling results.

Such scenario can be worked out on the basis of analysis of discrepancies between measurements and modelling results performed with the help of inverse modelling. Application of inverse modelling approach for the investigation of environmental problems is described in many scientific publications [*Villani et al.,* 2010; *Carouge et al.,* 2010 a,b and others]. At present the simplified version of adjoint model for B[a]P [*Shatalov et al.,* 2013] has been applied for construction of emission scenario refining the agreement between measured and modelled B[a]P air concentrations. For the analysis ten EMEP measurement sites located in the north-western part of Europe were chosen.

The improvement of the agreement between observed and modelled B[a]P air concentrations including seasonal variations resulting from the constructed scenario is demonstrated by the example of data for the EMEP monitoring sites DE09 and PL05 (Fig. 1.5).

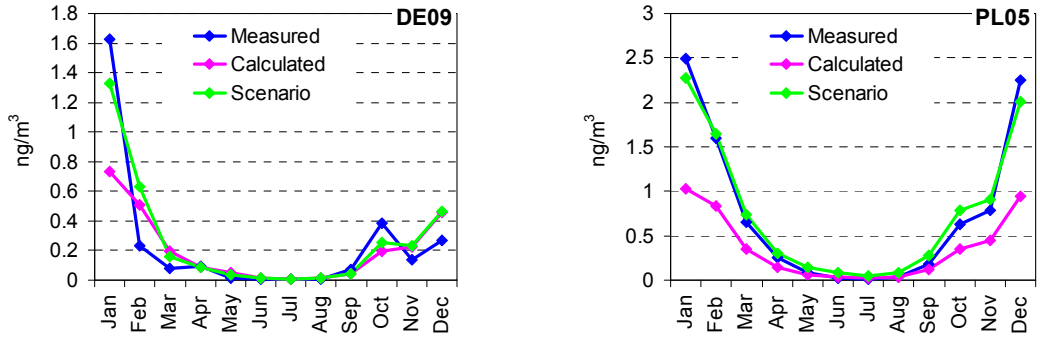


Fig. 1.5. *The comparison of B[a]P air concentrations measured at DE09 and PL05 (monthly averages, ng/m^3) with model calculations based on official emission data and constructed emission scenario.*

Inverse modelling allows indicating areas within which emission uncertainties most likely occur.

The analysis of measurement/modelling agreement shows the necessity of further investigation of B[a]P emissions in some regions of Central Europe with participation of national emission experts. The results of this activity can be discussed at the EMEP Task Forces on Emission Inventory and Projections and on Measurements and Modelling.

Further improvement of POP emissions

Completeness, consistency and uncertainties of officially reported POP emissions were the subject of discussions at the recent meetings of the EMEP Task Forces, namely, TFMM in Zagreb and TFEIP in Istanbul (May, 2013). Particularly, the meetings addressed the quality of currently available official emissions and considered the ways of their further improvement. Following the request of the Bureau of the EMEP Steering Body and TFEIP, EMEP modellers were invited to present their thoughts and

wishes concerning reported emissions, and how these data might be improved 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 the EMEP countries were one of the most significant characteristics for model assessment of POP 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.

Data on large point sources and their characteristics were also recognized as important information for modelling. 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.

Quality of POP pollution assessment and its improvement essentially depends on the reduction of incompleteness and uncertainties in POP emissions.

Further work in this direction should be continued in close cooperation between the EMEP modellers and experts in emissions in the framework of the EMEP TFEIP.

Discussing the wishes of modellers with regard to emission data and their characteristics, importance of the information on chemical speciation of emissions, seasonal variations, historical emissions, and emissions to the media other than atmosphere (e.g. soil, water bodies) for model assessment was indicated.

1.2. Monitoring of POPs

EMEP measurements of POPs in 2011

POPs were included in the EMEP's monitoring program in 1999. However, earlier data has been available and collected, and the EMEP database thus also includes older measurements (see http://ebas.nilu.no). A number of countries have been reporting POPs within the EMEP area in connection with different national and international programmes such as HELCOM, AMAP and OSPAR CAMP. Detailed information about the sites, the measurement methods used in 2011, and the annual averages and statistics are found in EMEP/CCC's data report on heavy metals and POPs [*Aas and Breivik*, 2013].

In 2011 there were thirteen sites measuring POPs in both compartments (air and precipitation), and altogether there were twenty nine measurement sites reporting POPs in either air or precipitation, which was the same number of sites as in 2010 (Table 1.1). There is one additional site in the UK and one in Spain. But two sites reporting data last year in Republic of Moldova and Kazakhstan provided only campaign data for that year. Furthermore there are four additional sites measuring campaign-like data for PAHs covering three-four months of the year. There has been an increase in EMEP sites measuring PAHs the last years because these measurements are required according to the EU air quality directive [*EU*, 2004]. Benzo[a]pyrene (B[a]P), which is a by-product of incomplete combustion processes, is the most frequently measured POP component in EMEP.

Table 1.1. *Measurement sites and program in 2011*

Country	Code	Name	POPs in air and aerosol	POPs in precipitation
Belgium	BE0013R	Houtem		PAHs
	BE0014R	Koksijde		PCBs, pest., HCHs
Cyprus	CY0002R	Ayia Marina	PAHs	
Czech rep.	CZ0003R	Kosetice	PAHs, PCBs, pest., HCB, HCHs	PAHs, PCBs, pest., HCHs
Germany	DE0001R	Westerland	PAHs, PCBs, pest., HCHs	PAHs, PCBs, pest., HCB, HCHs
	DE0003R	Schauinsland	PAHs	PAHs
	DE0008R	Schmücke	PAHs	PAHs
	DE0009R	Zingst	PAHs, PCBs, pest, HCB, HCHs	PAHs, PCBs, pest., HCB, HCHs
Denmark	DK0010G	Nord, Greenland	PAHs, pest., HCB, HCHs	
Spain	ES0001R	San Pablo		PAHs (total dep, 4 month campaign)
	ES0006R	Mahón	PAHs (3 month campaign)	PAHs (total dep, 4 month campaign)
	ES0007R	Víznar	PAHs (3 month campaign)	PAHs (total dep, 4 month campaign)
	ES0008R	Niembro	PAHs	PAHs (total dep, 4 month campaign)
	ES0014R	Els Torms		PAHs (total dep, 4 month campaign)
Finland	FI0036R	Pallas/Matarova	PAHs, PCBs, pest., HCHs	PAHs, PCBs, HCHs
Great-	GB0014R	High Muffles	PCBs, PAHs	
Britain	GB0036R	Harwell	PAH	
Iceland	IS0091R	Storhofdi	PCBs, pest., HCB, HCHs	PCBs, pest., HCB, HCHs
Latvia	LV0010R	Rucava	PAHs	
Netherlands	NL0009R	Kollumerwaard	PAHs	
	NL0091R	De Zilk	PAHs	γ-HCH
Norway	NO0042G	Spitsbergen	PAHs, PCBs, pest., HCHs, HCB	
	NO0002R	Birkenes	PAHs, PCBs, pest., HCHs, HCB	PCBs, PAHs,HCB, HCHs
	NO0090R	Andøya	PAHs, PCBs, pest., HCHs, HCB	
Poland	PL0005R	Diabla Gora	PAHs	PAHs
Sweden	SE0011R	Vavihill	PAHs	PAHs (total dep.)
	SE0012R	Aspvreten	PAHs, PCBs, pest., HCHs	PAHs, PCBs, HCHs (total dep.)
	SE0014R	Råö	PAHs, PCBs, pest., HCHs	PAHs, PCBs, HCHs (total dep.)
Slovenia	SI0008R	Iskrba	PAHs	PAHs

Measurements of PCDD/F concentrations in air and other media

Regular measurements on PCDD/Fs within the EMEP monitoring network are not currently performed. Therefore MSC-E carried out the work on collection of measurement data for PCDD/Fs from national monitoring campaigns. A set of measurements is used for evaluation of the quality of model results. Literature search performed during last two years allowed collecting over 1000 individual measurements of PCDD/F total toxicity made in 18 European countries (Austria, Belgium, Croatia, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, the Netherlands, Poland, Portugal, Spain, Sweden, Switzerland and the UK) within the period from 1988 to 2010.

Collection and evaluation of measurement data on PCDD/F concentrations in various environmental media from national campaigns is highly important.

Collected measurement data were analyzed from the viewpoint of their usage for the comparison with modelling results. Particularly, data on PCDD/F concentrations in rural and background regions were selected, while measurements for urban and industrial regions were not used for the comparison. Further, the set of measurements was evaluated statistically to exclude outliers. Additionally, for the verification of model estimates of PCDD/F intercontinental transport measurements made outside of the EMEP region were compiled as well.

To evaluate model estimates of PCDD/F content in the soil compartment, collection of measurement data on soil concentrations is initiated. These data are of importance since soil concentrations of PCDD/Fs are subject to national regulation in a number of Parties to the Convention (e. g. Canada and Italy). Collection of measurement data on PCDD/Fs concentrations in the atmosphere and other media under EMEP and evaluation of these data can essentially refine the evaluation of environmental pollution levels in the EMEP region.

1.3. Pollution levels and long-term trends

This year pollution levels of PAHs, PCDD/Fs, and HCB were evaluated for 2011 and compared with their levels in the reference year (1990). This section presents a short summary of model assessment results including analysis of transboundary transport and long-term changes of pollution.

Pollution by POPs is formed by different sources including anthropogenic emissions of the EMEP countries, distant emissions outside of the EMEP domain, and secondary emissions as within the EMEP region and beyond its boundaries. Relative contributions of these groups of sources are different for the considered pollutants. Particularly, pollution of the EMEP countries by PAHs is mostly determined by anthropogenic emissions within the EMEP domain, while contribution of non-EMEP (global) emission sources is relatively small. Therefore, model assessment of PAH pollution is focused on the emission sources of the EMEP countries neglecting the intercontinental transport. At the same time, for PCDD/Fs and HCB this contribution is more essential which leads to the necessity of evaluation of their intercontinental transport.

Polyaromatic hydrocarbons (PAHs)

According to the Protocol on POPs, four indicator PAH compounds (benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F) and indeno[1,2,3-cd]pyrene (IP)) were included into the model assessment of pollution. Spatial distribution of 4 PAHs annual mean air concentrations within the EMEP domain for 1990 and 2011 is shown in Fig. 1.6.

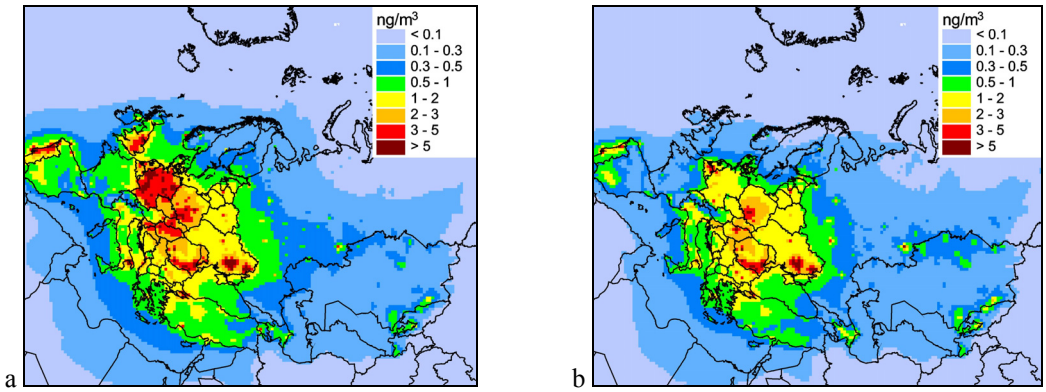


Fig. 1.6. *Spatial distribution of annual mean air concentrations of 4 PAHs in the EMEP domain for 1990 (a) and 2011 (b), ng/m^3.*

Results of model simulations indicate that PAH air concentrations in the majority of the EMEP countries tend to decrease in the period from 1990 to 2011. Particularly, average reduction of PAH concentrations is accounted for 30%.

PAH pollution levels in the EMEP region decreased from 1990 to 2011 by 30% on the average. The highest decline is indicated for the EU countries (39%).

The decrease of PAH air concentrations is not homogeneous over the EMEP domain. The highest decline is indicated for the EU countries (39%), with the most essential changes in the western and central parts of Europe. At the same time, decrease of PAH pollution in the southern and eastern parts of Europe as well as in the Central Asian countries is much smaller.

Air concentrations of B[a]P in the EU countries dropped by 38% from 1990 to 2000. After 2000 the decreasing trend was almost levelled off and B[a]P air concentrations tended to increase in the most of the EU countries.

To perform more detailed analysis of PAH pollution changes during the two recent decades, inter-annual variations of air concentrations were evaluated on the example of B[a]P. Relative changes of annual mean B[a]P air concentrations in the EU and other EMEP countries are shown in Fig.1.7. The rate of pollution decrease was different during the period from 1990 to 2011 as well as the direction of the changes varied in some countries from the decreasing to increasing. Thus, reduction of pollution levels was more significant during the 1990s. In particular, B[a]P air concentrations in the EU countries dropped by 38% from 1990 to 2000. However, after 2000 the decreasing trend was almost levelled off and during the recent years B[a]P air concentrations in more than a half of the EU countries tended to increase following the growth of their emissions.

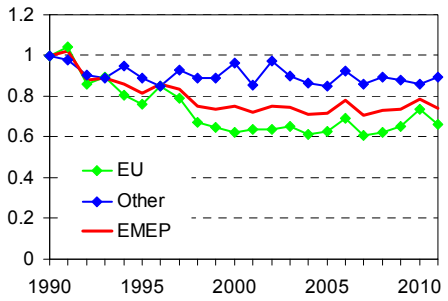


Fig. 1.7. *Relative changes of B[a]P air concentrations in the EU and other EMEP countries in the period 1990-2011.*

Different tendencies in temporal variations of B[a]P air concentrations in the EMEP countries are illustrated in Fig. 1.8. Particularly, significant decrease of B[a]P air concentrations in Germany can be seen in the 1990s, which is almost levelled off after 2000 (Fig. 1.8a). Opposite tendencies in the dynamics of B[a]P pollution levels in Denmark can be seen in the periods 1990-1999 and 2000-2011 (Fig. 1.8b).

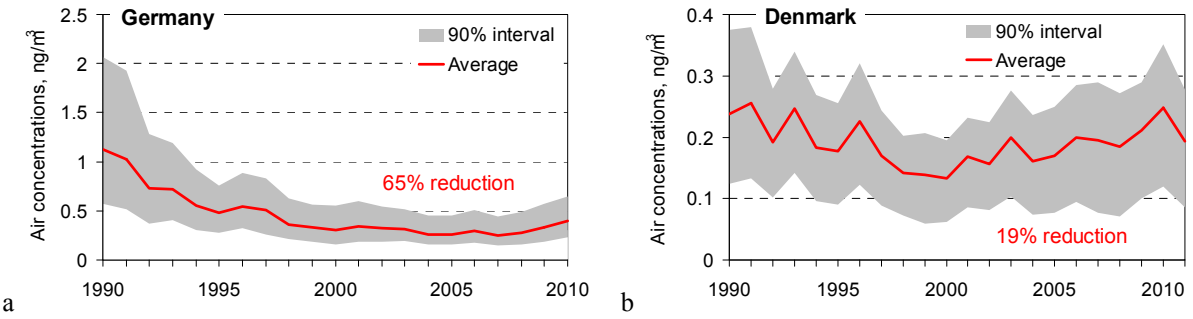


Fig. 1.8. *Trends of annual mean B[a]P air concentrations in the period 1990- 2011 for Germany and Denmark. Grey strip shows the range of concentrations over the country.*

Inter-annual variations of B[a]P pollution levels depend on changes of emissions and variability of meteorological conditions (precipitation, prevailing atmospheric flows, and temperature). The effect of the latter factor is illustrated in the Fig. 1.9 on the example of the changes of pollution levels between the two years 2010 and 2011.

Inter-annual variability of meteorological conditions can lead to essential changes of pollution levels (e.g. ±30% for annual mean B[a]P air concentrations)

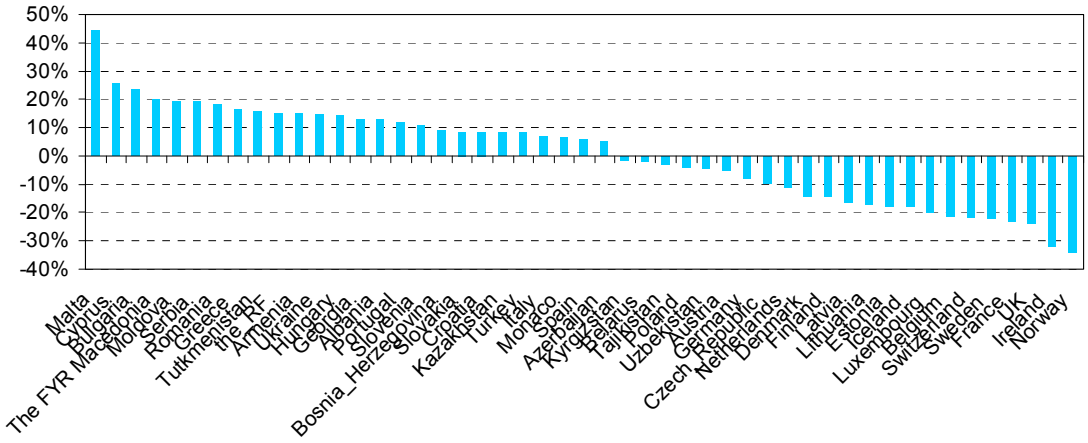


Fig. 1.9. *Changes of annual mean B[a]P air concentrations in the EMEP countries between 2010 and 2011 due to variability of meteorological conditions. Negative values reflect decreasing of pollution levels, and positive ones – increasing.*

Model simulations for these years were carried out with the same emissions for the year 2010 and different meteorology. It is seen that variability of meteorological parameters can lead to considerable changes in annual mean B[a]P air concentrations (from – 30% to+ 30%) which should be taken into account in the interpretation of long-term pollution changes.

The comparison of model predictions of annual mean B[a]P air concentrations with measurements of the EMEP monitoring network for the period 1990-2011 revealed that over 70% of modelled values agreed with observed ones within a factor of 2 with the relative bias about 15%. Besides, spatial distribution of modelled air concentrations reasonably well correlated with the observed one (correlation coefficient amounted to 0.7). At the same time, the comparison of monthly mean B[a]P concentrations in air indicated more substantial discrepancies. Particularly, it was shown that the model tended to underestimate observed winter time air concentrations of B[a]P and overestimate summer time ones by more than a factor of 2 (Fig. 1.10).

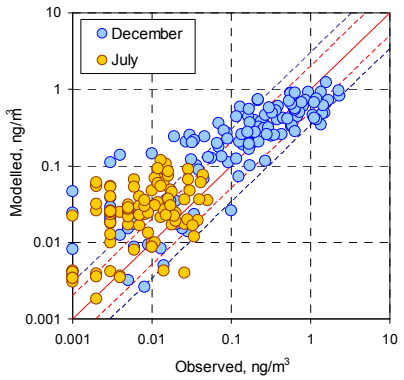


Fig.1.10. *Comparison of monthly mean calculated B[a]P air concentrations with measurements of the EMEP monitoring sites for the period 1990 – 2011, ng/m^3*

Possible improvement of the agreement between the modelled and observed monthly mean air concentrations can be associated with the application of parameterization of POP gas-particle partitioning to different components of aerosols (e.g. elemental carbon (EC), organic carbon (OC), and other components) (Figs. 1.11, 1.12).

A number of recent studies indicated that the rates of degradation of B[a]P, sorbed on EC, OC, and inorganic components, could be different and these fractions of B[a]P could have essentially different half-lives. Thus, to improve the agreement of model predictions and measurements there is a need to refine the description of degradation process for B[a]P fractions sorbed on aerosol particles.

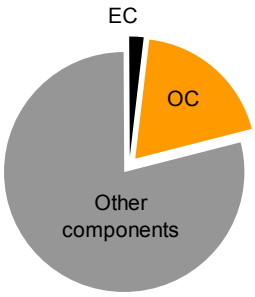


Fig. 1.11. *Relative content of EC and OC in atmospheric aerosol of background regions [Lohmann and Lammel, 2004]*

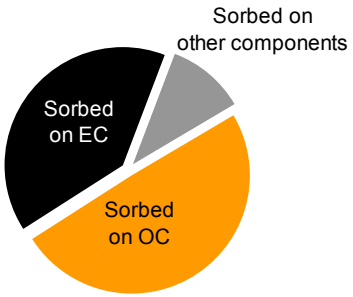


Fig.1.12. *Estimates of B[a]P sorption to EC, OC, and other components of atmospheric aerosols for background regions*

Polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs)

In comparison to PAHs, reduction of PCDD/F pollution in the EMEP domain is more significant. Spatial distribution of annual mean PCDD/F air concentrations in 1990 and 2011 within the EMEP region is demonstrated in Fig. 1.13. It can be seen that the most essential changes took place in the western and central European countries.

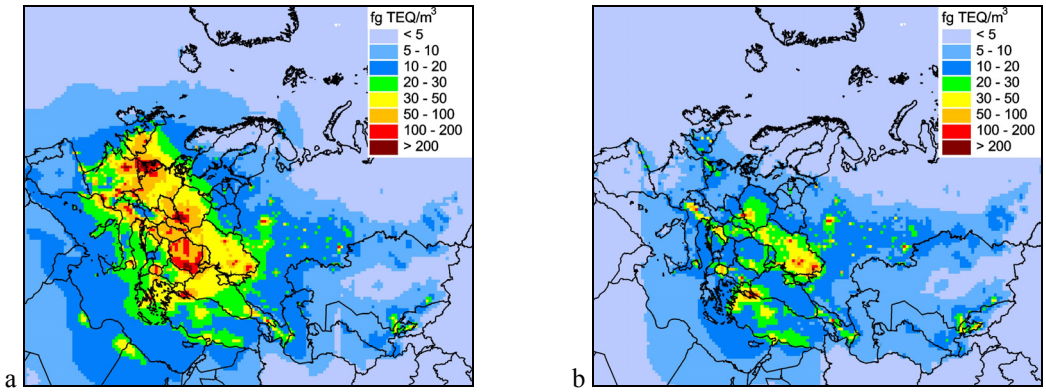


Fig. 1.13. *Spatial distribution of annual mean concentrations of PCDD/Fs in the EMEP domain for 1990 (a) and 2011 (b), fg TEQ/m^3.*

PCDD/F pollution levels in the EMEP countries have decreased from 1990 to 2011 by 55%. The highest decline is indicated for the EU countries (75%).

PCDD/F pollution levels in the EMEP countries declined from 1990 to 2011 by 55% on average. The rates of pollution changes were different in different parts of the EMEP region (Fig. 1.14). The most significant changes took place in the EU countries, where PCDD/F air concentrations decreased on average by 75% from 1990 to 2011. Less significant reduction of pollution can be seen in other EMEP countries (by about 20%).

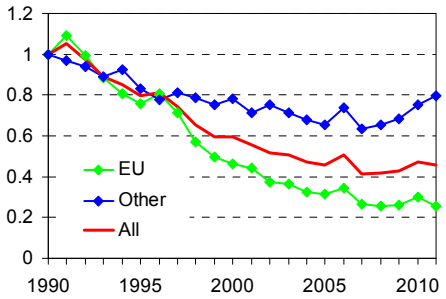


Fig. 1.14. *Relative changes of PCDD/F air concentrations in the EU and other EMEP countries in the period 1990-2011.*

Examples of long-term variations of PCDD/F air concentrations, presented in Fig. 1.15, illustrate different character of changes in the EMEP countries. In particular, the highest decrease of PCDD/F air concentrations among the EU countries is indicated for the Netherlands (by 93%) followed by Luxembourg (by 89%), and France (by 87%). Moderate changes of PCDD/F pollution levels are illustrated on the example of Croatia where the decline in the period 1990-2011 is amounted to 43%.

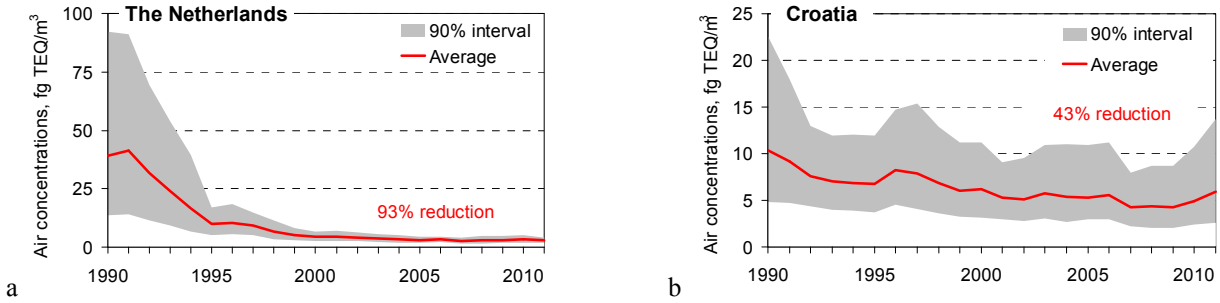


Fig. 1.15. *Trends of annual mean PCDD/F air concentrations from 1990 to 2011 for the Netherlands and Croatia. Grey strip shows the range of concentrations over the country.*

Similarly to PAHs variability of meteorological parameters can lead to significant changes in annual mean PCDD/F air concentrations (from –30% to +20%) which should be taken into account in the interpretation of long-term pollution changes.

Major uncertainties in the evaluation of PCDD/F pollution levels can be associated with incompleteness of data on anthropogenic emissions and poor knowledge on contribution of the secondary emissions. Particularly, officially reported anthropogenic emissions most likely do not cover all potential source categories, which is pointed out in a number of PCDD/F emission inventories [*Fiedler et al.*, 2007].

Results of model simulations indicate that PCDD/F pollution levels can be controlled not only by the anthropogenic emissions, but also secondary sources (re-volatilization of PCDD/Fs from the soil and water compartments to the atmosphere). The contribution of secondary sources to PCDD/F pollution was evaluated as more than 50% due to long-term accumulation of PCDD/Fs in soil and seawater (see Section 1.4).

Secondary emissions to the atmosphere can be affected by the direct releases of PCDD/Fs to soil and water compartments due to various activities including heat and power generation, open burning of biomass, disposal of wastes, etc. The inventory of PCDD/F emissions to air, soil, and water for 17 European countries was carried out in the framework of the EU project "Releases of Dioxins and Furans to Land and Water in Europe" [*Wenborn et al.*, 1999]. According to this inventory releases of PCDD/Fs to soil can substantially exceed their emissions to the atmosphere (Fig. 1.16). Experimental modelling of PCDD/F pollution levels on the basis of the data of this EU project permitted to obtain good agreement between the model results and available measurements of PCDD/F air concentrations from national campaigns. As shown in the Fig. 1.17 the model reasonably well reproduced spatial and temporal variations of PCDD/F air concentrations observed within the EMEP region.

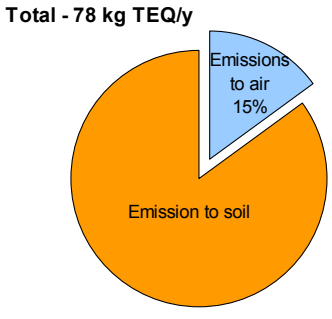


Fig.1.16. *Relative contributions of PCDD/F releases to air and soil compartments to total emission based on the data of the EU project [Wenborn et al., 1999].*

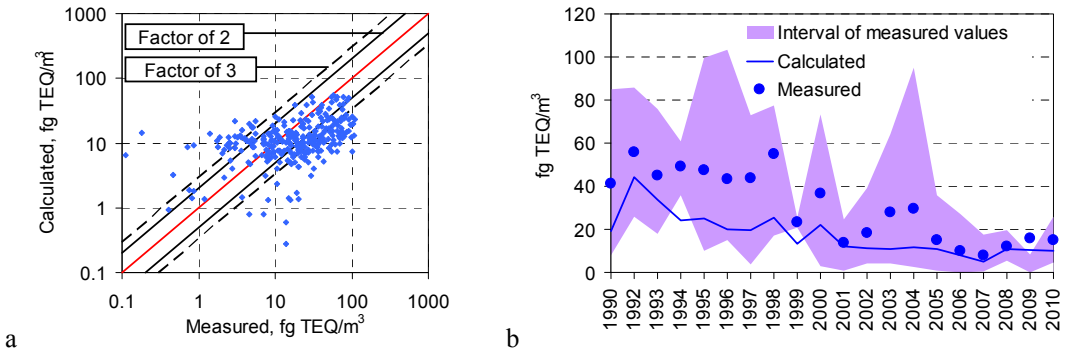


Fig. 1.17. *Comparison of modelled vs. measured total concentrations of PCDD/Fs in the atmosphere for the period 1990-2010: (a) scatter plot with the comparison of individual measurements and modelled values, (b) temporal variations of annually averaged modelled and observed total PCDD/F concentrations (fg TEQ/m^3).*

To improve evaluation of PCDD/F pollution in the EMEP countries there is need to take into account emissions of PCDD/Fs to soil and water compartments

To improve evaluation of pollution levels of PCDD/Fs in the EMEP countries further refinement of the data on their emissions to soil and water compartments (at least at the level of expert estimates) is desirable.

It should be mentioned that at present no regular measurements of PCDD/F content in air is performed under the EMEP monitoring network. As a consequence, model assessment together with the results of national monitoring campaigns is the only source of the information on PCDD/F contamination in the EMEP region.

Hexachlorobenzene (HCB)

HCB is characterized by essentially high potential to long-range transport and persistence in the environment (half-life in air ~3 y, half-life in soil ~6 y). Levels of HCB in the atmosphere in recent decade are mostly controlled by secondary emissions (re-volatilization from surface media) while contribution of anthropogenic emissions in the European region is relatively small [*Barber et al.*, 2005]. Thus, to evaluate its levels of pollution within the EMEP countries there is need to take into account not only contemporary anthropogenic emissions, but also historic emissions, which significantly contributed to the accumulation of HCB in the environment. Importance of secondary HCB emissions was considered in several previous studies performed by MSC-E [*Shatalov et al.*, 2010; *Gusev et al.,* 2011]. It was concluded that further improvement of the evaluation of HCB pollution levels in the EMEP countries could be achieved by the refinement of official data on HCB emissions and construction of scenarios of historical emissions of HCB to describe its global distribution and accumulation in the environment.

Model assessment of HCB pollution in the EMEP region was carried out for the period 1990-2011 using official emission data reported by the EMEP countries complemented by available expert estimates.

Model simulations permitted to describe reasonably well the spatial and temporal variations of HCB pollution levels within the EMEP region. Spatial distributions of annual mean HCB air concentrations in the EMEP domain in 1990 and 2011 are illustrated in Fig. 1.18. Comparison of modelling results against available long-term measurements at the EMEP monitoring sites revealed reasonable agreement between the modelled and observed concentrations (Fig. 1.19).

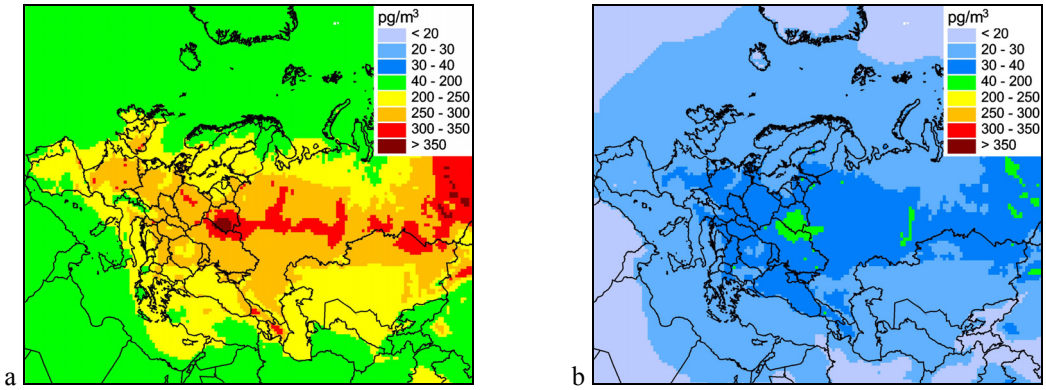


Fig. 1.18. *Spatial distribution of HCB annual mean concentrations in the EMEP region for 1990 (a) and 2011 (b), pg/m^3.*

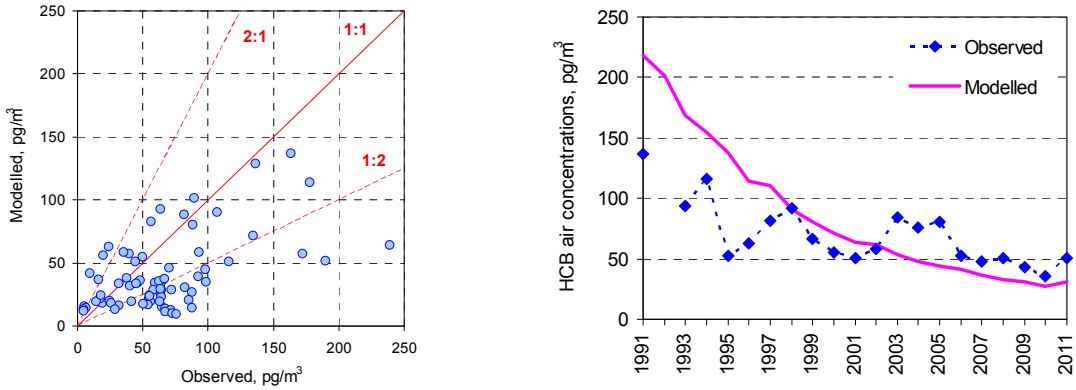


Fig. 1.19. *Comparison of annually averaged modelled annual HCB air concentrations with long-term measurements of the EMEP monitoring sites for the period 1991-2011, ng/m^3.*

Following the elimination of HCB application in the agriculture and the reduction of its other anthropogenic emissions the levels of HCB pollution in the EMEP countries have substantially reduced (by almost 90%) from 1990 to 2011 (Fig. 1.20). As primary emissions decreased the variability of HCB air concentrations in the EMEP region has reduced accounting for several times in recent years, which agrees with available measurements [*Jaward et al.*, 2004; *Halse et al.,* 2011], while air concentrations of other two POPs considered in the report vary by orders of magnitude.

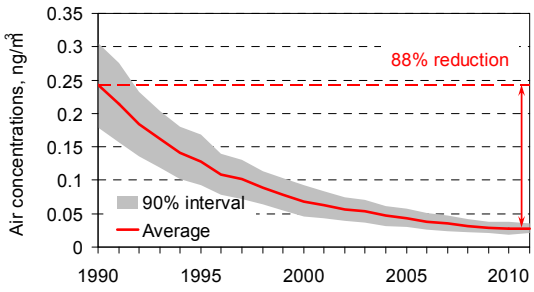


Fig. 1.20. *Long-term changes of HCB air concentrations from 1990 to 2011 in the EMEP region.*

According to modelling results the decrease of HCB pollution in the EMEP countries is nearly homogeneous in contrast to PAHs and PCDD/Fs. Examples of the trends of HCB air concentrations in two EMEP countries, namely, Norway and Switzerland, are presented in Fig. 1.21. As shown, levels of HCB concentrations in these countries have dropped on average by 90%. This illustrates similar rates of decline of HCB pollution in different parts of the EMEP region.

HCB pollution levels within the EMEP region have declined by almost 90% from 1990 to 2011.

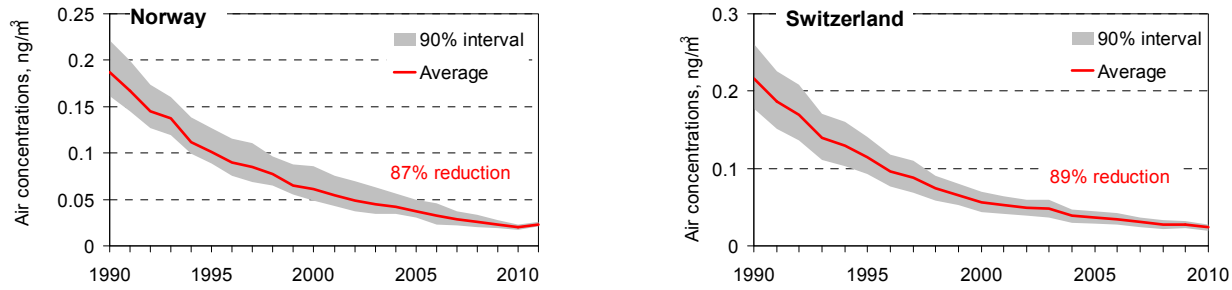


Fig. 1.21. *Trends of annual mean HCB air concentrations in the period from 1990 to 2011 for Norway and Switzerland. Grey strip shows the range of concentrations over the country.*

To improve evaluation of HCB pollution levels refinement of global historical and contemporary emissions of HCB is needed.

Uncertainties in the evaluation of HCB pollution levels in European countries are mostly caused by the incompleteness of the information on historical emissions of HCB as for the EMEP region and for the global scale. Recent studies of HCB pollution indicate quite uniform levels of its concentrations in the atmosphere and essential influence of secondary emission sources, while contribution of current anthropogenic emissions is comparatively small. Thus, to provide more accurate description of HCB pollution levels there is a need to further work on the refinement of scenarios of global historical emissions of HCB including also refinement of officially reported emissions. Besides, to study the cycling and accumulation of HCB in the environmental compartments it is necessary to refine the information on various characteristics of the surface media (e.g. organic matter content) and parameterizations of degradation and phase partitioning processes which influence the accumulation of HCB in media.

Information on the characteristics of surface media and parameterizations of HCB degradation and phase partitioning in media require further updating.

1.4. Transboundary transport

Transboundary transport of PAH, PCDD/F, and HCB within the EMEP region was carried out for 1990 and 2011 on the basis of officially reported emission data complemented by available expert estimates. Short summary presents analysis of major changes of transboundary transport between the EMEP countries in this period including changes of contributions of anthropogenic, secondary and non-EMEP emission sources.

Polyaromatic hydrocarbons (PAHs)

Evaluation of PAH transboundary transport within the EMEP region in 1990 and 2011 was performed for B[a]P, which is used as the indicator compound for PAHs. B[a]P presents in the atmosphere predominantly in particulate phase and its residence time in the atmosphere amounts up to several days depending on the meteorological conditions. B[a]P pollution in the EMEP region is mostly formed by the anthropogenic emission sources, thus the analysis of transboundary transport is focused on the changes in contributions of emissions originated from the anthropogenic sources of the EMEP countries. The contributions of non-EMEP emissions and secondary sources are comparatively low and not considered here.

Results of model assessment indicate that transboundary transport of B[a]P essentially contributed to the pollution of the EMEP region in the period from 1990 to 2011. Particularly, on average about half of B[a]P deposition in the EMEP countries originated from the emission sources outside their territories. At the same time, contributions of transboundary transport to the pollution of the EMEP countries vary in wide range (from about 5% to almost 99%).

Contribution of transboundary transport to total B[a]P deposition in the EMEP countries is essential and accounted for about 50%.

Relative changes of B[a]P total deposition over the individual EMEP countries from the base year 1990 to 2011, caused by changing national emissions and variations of transboundary transport, are presented in Fig. 1.22. It can be seen that in the majority of the EMEP countries total deposition of B[a]P has considerably decreased from 1990 to 2011.

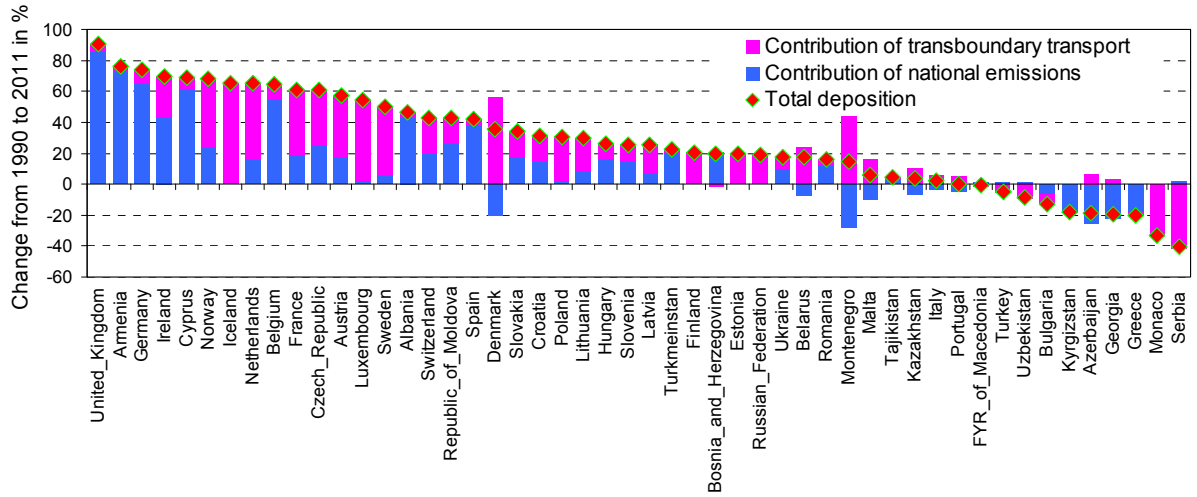


Fig. 1.22. *Relative changes of B[a]P total deposition in the EMEP countries from 1990 to 2011 as well as changes in contributions of national emission sources and transboundary transport. Positive value means decrease of deposition, and negative – increase.*

Temporal changes of B[a]P total deposition in the EMEP countries are caused by different reasons. A number of countries are characterized by significant reduction of national emissions which essentially contributes to total deposition reduction (e.g. in the UK, Armenia, Germany, Belgium). For some of the countries (e.g. Norway, Iceland, the Netherlands, and Sweden) changes of total deposition strongly depend on transboundary transport.

Model assessment of transboundary transport provides detailed information on changes of total deposition for particular countries. The effect of reduction or growth of national anthropogenic emissions on relative importance of transboundary transport is illustrated on the example the UK and Denmark. Both countries are characterized by the decreasing total deposition of B[a]P from 1990 to 2011.

In the UK levels of B[a]P total deposition substantially decreased (by almost 90%) due to reduction of national B[a]P emissions and transboundary transport from the other EMEP countries (Fig.1.23). It is seen that the effect of reduction of national emissions is much more significant. Deposition from national emissions of the UK decreased by almost 14 times, which led to the increase of relative contribution of transboundary transport from 9% in 1990 to 43% in 2011. At the same time absolute values of B[a]P deposition originated from the emissions of the other EMEP countries declined from 1.4 to 0.6 tonnes.

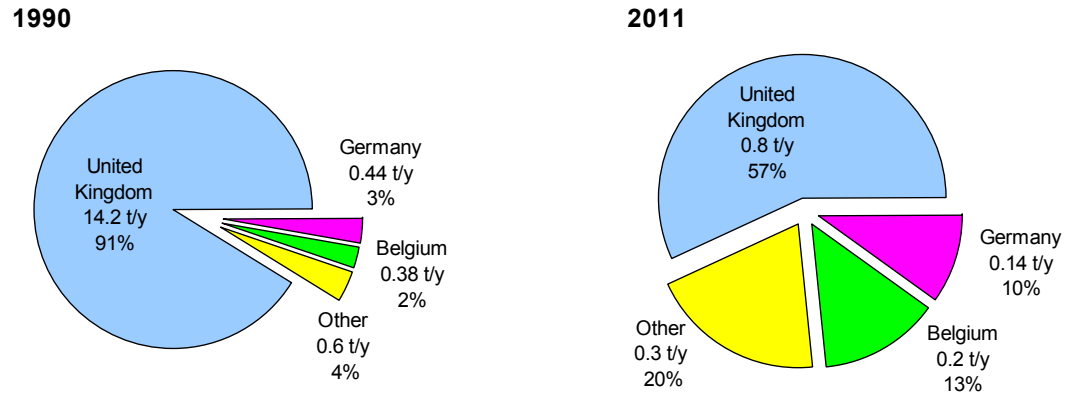


Fig. 1.23. *Changes in relative contributions of national emission sources and transboundary transport to total B[a]P deposition in the UK from 1990 to 2011, t/y.*

Contrary to that, in Denmark opposite tendencies in the changes of deposition from national emissions and transboundary transport led to essential redistribution of their contributions (Fig. 1.24). Particularly, deposition of B[a]P from national emissions increased almost twice from 1990 to 2011. At the same time, contribution of transboundary transport in that period declined nearly nine times. This resulted in the decrease of relative importance of transboundary transport in Denmark (decline of contribution from 82% down to 41%).

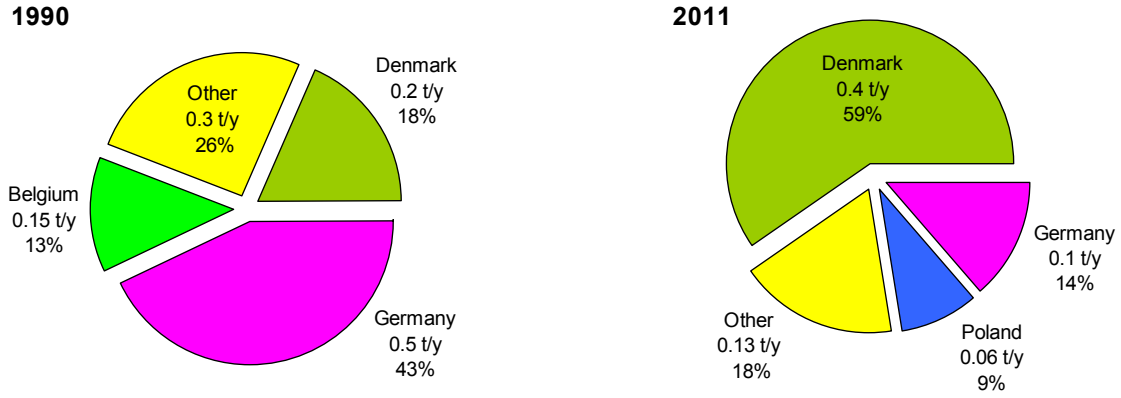


Fig. 1.24. *Changes in relative contributions of national emission sources and transboundary transport to total B[a]P deposition in Denmark from 1990 to 2011, t/y.*

Polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs)

Pollution by PCDD/Fs in the EMEP domain is mostly controlled by both primary anthropogenic and secondary emission sources. Secondary emissions can contribute more than 50% to total PCDD/F deposition.

Model assessment of PCDD/F transboundary transport within the EMEP region was carried out for 1990 and 2011 to evaluate changes in pollution levels and contributions of different sources. PCDD/Fs are characterized by significant persistence in the environment and relatively high long-range transport potential. To take this into account, the evaluation of their transboundary transport requires consideration of contributions of secondary emissions (re-volatilization from the environmental media other than the atmosphere). Additional contribution is also expected from the non-EMEP emission sources. At the same time, data on emissions outside of the EMEP region are currently available only

for a few countries. Thus, to refine estimates of PCDD/F intercontinental transport global inventory of PCDD/F anthropogenic emissions is needed.

Relative contributions of contemporary anthropogenic emissions of the EMEP countries, secondary emissions, and non-EMEP emission sources, evaluated using model simulations for 1990 and 2011, are shown in the diagrams in Fig. 1.25.

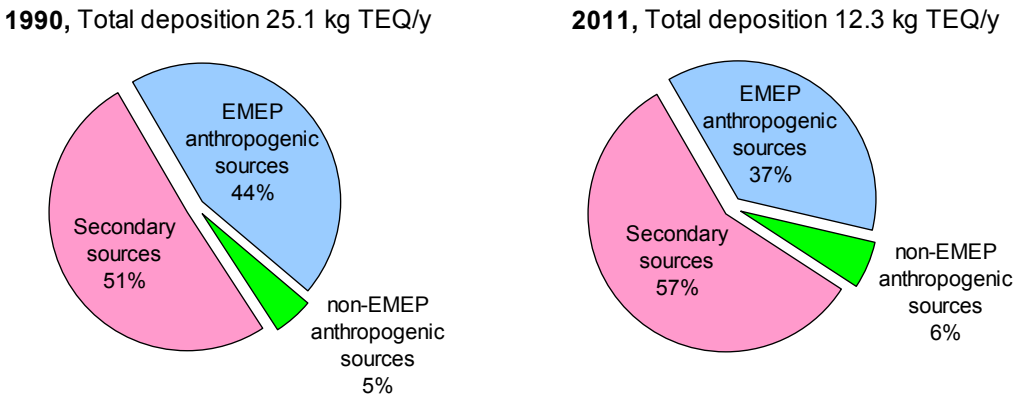


Fig. 1.25. *Contributions of contemporary EMEP anthropogenic emission sources, secondary emissions and non-EMEP emission sources to total deposition to the EMEP countries in 1990 and 2011.*

Total deposition of PCDD/Fs to the EMEP countries reduced more than twice during this period (from 25.1 kg TEQ/y in 1990 to 12.3 kg TEQ/y in 2011). At that relative fraction of secondary emission sources increased from 51% to 57%, while the fraction of anthropogenic emission sources decreased from 44% to 37%.

Similar to B[a]P, contribution of transboundary transport to PCDD/F deposition in the EMEP countries was not changed significantly from 1990 to 2011. Particularly, model simulations indicate that more than a half of PCDD/F deposition from anthropogenic emission sources within the EMEP domain is contributed by the transboundary transport between the EMEP countries.

Contribution of transboundary transport to PCDD/F deposition from anthropogenic emission sources within the EMEP region in 2011 exceeds 50%.

Relative changes of total PCDD/F deposition in the EMEP countries, as well as changes in contributions of national emission sources, transboundary transport, secondary emissions, and non-EMEP emission sources from the base year 1990 to 2011, are presented in the Fig. 1.26. Most of the EMEP countries are characterized by substantial reduction (more than 40%) of total PCDD/F deposition from 1990 to 2011. In more than a half of the countries PCDD/F deposition were reduced due to the decrease of contributions of secondary emissions (re-volatilization) and transboundary transport.

At the same time, changes of total PCDD/F deposition in a number of countries, namely, in the Netherlands, the Czech Republic, Romania, the UK, France and some others, are caused by considerable reduction of national emissions.

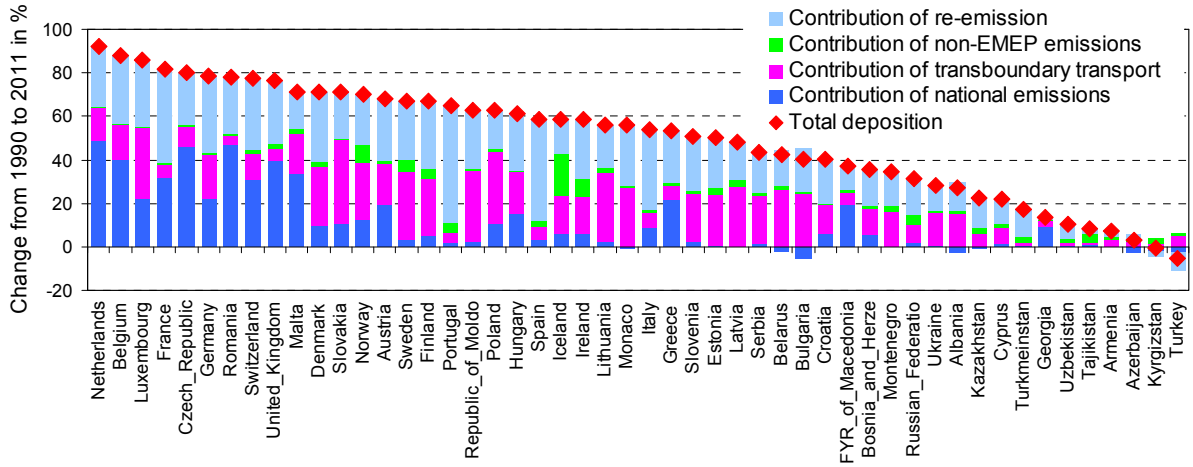


Fig. 1.26. *Relative changes of total PCDD/F deposition in the EMEP countries from 1990 to 2011 as well as changes in contributions of national anthropogenic emissions, transboundary transport, non-EMEP emissions, and re-emission. Positive value means decrease of deposition, and negative – increase.*

For example, deposition from national emissions in France decreased by approximately 20 times, which led to the increase of relative contribution of transboundary transport from 18% in 1990 to 47% in 2011 (Fig. 1.27). Inhomogeneous reduction of national emissions in surrounding countries resulted in the redistribution of their contribution to deposition over France. Particularly, major contributions to transboundary PCDD/F deposition in France in 1990 were made by Belgium and Germany. More substantial reduction of their PCDD/F emissions, comparing to other countries, led to the decrease of their relative contributions. Thus, in 2011 two main contributors to transboundary PCDD/F deposition over France were changed to Italy and Spain.

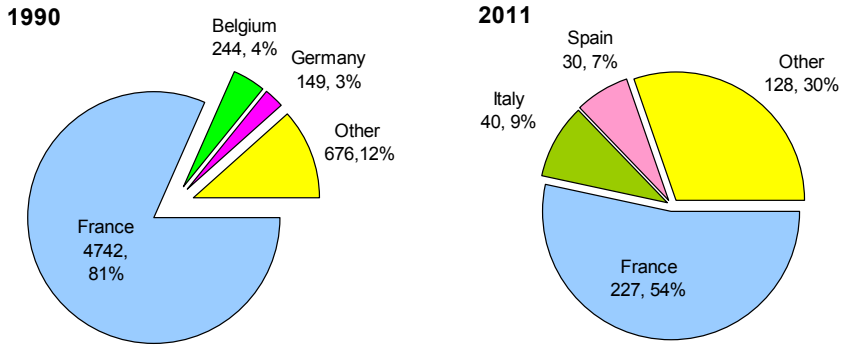


Fig. 1.27. *Changes in relative contributions of national emission sources and transboundary transport to total PCDD/F deposition in France from 1990 to 2011, g TEQ/y.*

Hexachlorobenzene (HCB)

Contemporary levels of HCB pollution in the EMEP countries are largely determined by non-EMEP and secondary emission sources.

Contemporary levels of HCB pollution in the EMEP countries are largely determined by secondary emission sources, particularly, re-volatilization of HCB previously accumulated in the surface media (soil, water bodies). Essential reduction of anthropogenic HCB emissions during several recent decades, as in the EMEP region and in the North America, has led to substantial decrease of their contribution to the pollution levels. Thus,

anthropogenic emissions provide relatively small contribution to HCB pollution [*Shatalov et al.*, 2012b]. At the same time, contributions of secondary emissions and intercontinental transport to the pollution of the EMEP countries have considerably increased.

Spatial distribution of HCB air concentrations in 2011 originated from non-EMEP emission sources, secondary and anthropogenic emissions within the EMEP is given in Fig. 1.28. It can be seen that dominating contribution to HCB pollution levels in 2011 belongs to secondary emissions and transboundary transport from non-EMEP sources (see Chapter 2). Though the contribution of anthropogenic sources to HCB pollution in the EMEP domain, on the average, is relatively small, it can be significant in particular countries with considerably high emissions and in the industrial regions with significant sources of HCB.

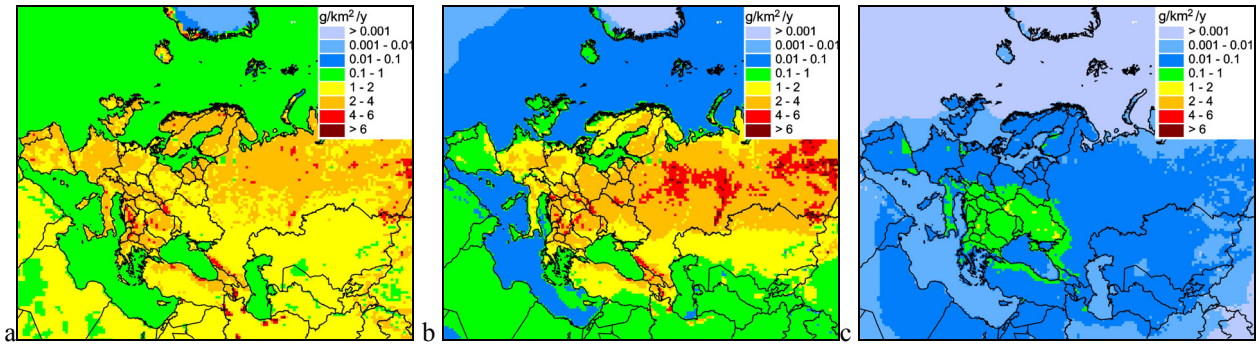


Fig. 1.28. *Spatial distribution of HCB annual deposition fluxes from non-EMEP emission sources (a), re-emission (b), and contemporary anthropogenic emissions of the EMEP countries (c) for 2011, g/km²/y.*

Levels of HCB pollution in the EMEP countries substantially decreased from 1990 to 2011. At the same time, relative contributions of non-EMEP emission sources, secondary emissions, and EMEP anthropogenic emissions did not change much (Fig. 1.29). Particularly, contribution of secondary sources within the EMEP region has slightly decreased, while for the contributions of intercontinental transport and EMEP anthropogenic emissions some increase is indicated.

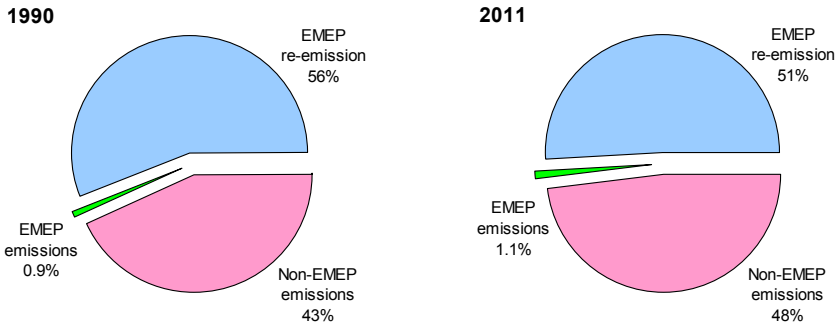


Fig. 1.29. *Contributions of anthropogenic emissions of the EMEP countries, re-emission and non-EMEP emission sources to HCB pollution in 1990 and 2011.*

Temporal trends of anthropogenic emissions and re-emission fluxes, obtained on the basis of model simulations, are compared in Fig. 1.30. Both anthropogenic emissions and re-emission flux have been reduced from 1990 to 2011 nearly to the same extent. It also can be seen that re-emission flux essentially exceeds anthropogenic emissions.

Model assessment of HCB pollution indicates that in the most of the EMEP countries reduction of HCB deposition was caused by the decrease of contributions of secondary and non-EMEP emission sources (Fig.1.31). Contributions of the changes in the anthropogenic emissions within the EMEP region are comparatively small.

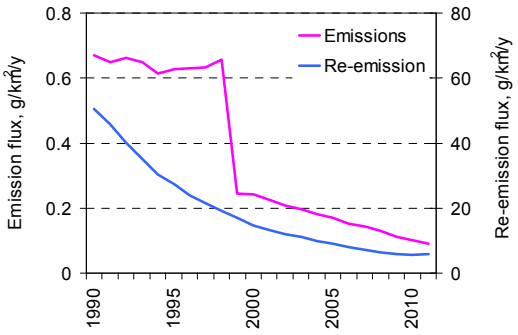


Fig. 1.30. *Temporal trends of anthropogenic HCB emissions and re-emission fluxes in the EMEP countries from 1990 to 2011. Note that the scales for the emission and re-emission fluxes are different!*

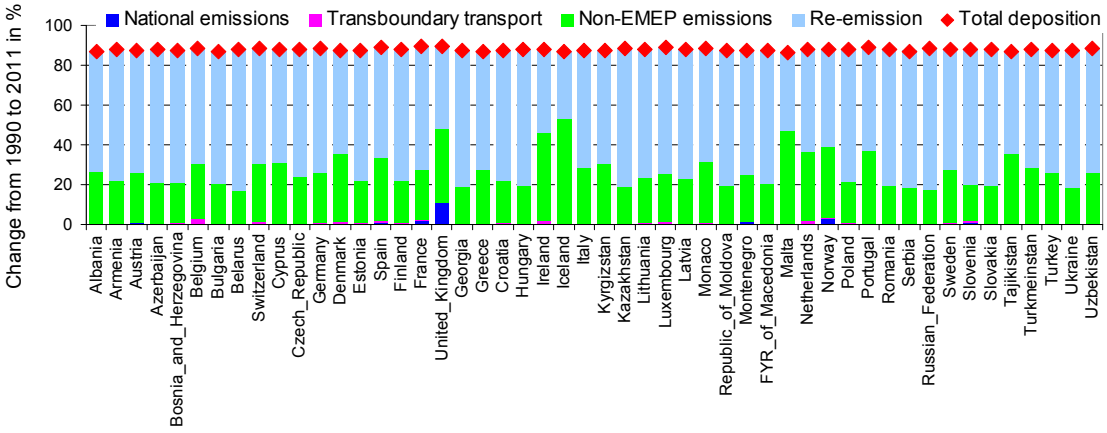


Fig. 1.31. *Relative changes of total HCB deposition in the EMEP countries from 1990 to 2011 as well as changes in contributions of national anthropogenic emissions, transboundary transport, non-EMEP emissions, and re-emission.*

However, it should be mentioned that anthropogenic HCB emissions reported by the EMEP countries are subject of uncertainties and incompleteness [*Mareckova et al.*, 2012] and thus their contribution can be underestimated. Therefore, to improve the quality of estimates of HCB pollution of the EMEP countries there is a need to refine the quality and completeness of official information on HCB emissions.

1.5. Evaluation of effects

Calculations performed in the framework of routine EMEP activities allows generate an information that can be used by subsidiary bodies to the Convention and its Parties for evaluation of possible hazards for human health and ecosystems.

Deposition to ecosystems. As indicated in the Annual Report 2010/2011 of the International Cooperative Programme on

Information on deposition fluxes to various types of land cover can be used for the analysis of effects of POP pollution on ecosystems.

Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) under Working Group on Effects (WGE) [*Harmens et al.*, 2011], mosses can potentially be used as bio-monitors of POPs. Despite the content of POPs in mosses was often analyzed to indicate levels of pollution and their spatial variations, only few studies examined the relation between POP air concentrations and/or deposition fluxes and concentrations in mosses. Most of these studies were focused on PAHs. EMEP can contribute to these investigations providing information on temporal and spatial variations of POP deposition fluxes to various types of vegetation cover. As an example, the fluxes of B[a]P to areas covered by grass and coniferous forest for 2011 are shown in Fig. 1.32.

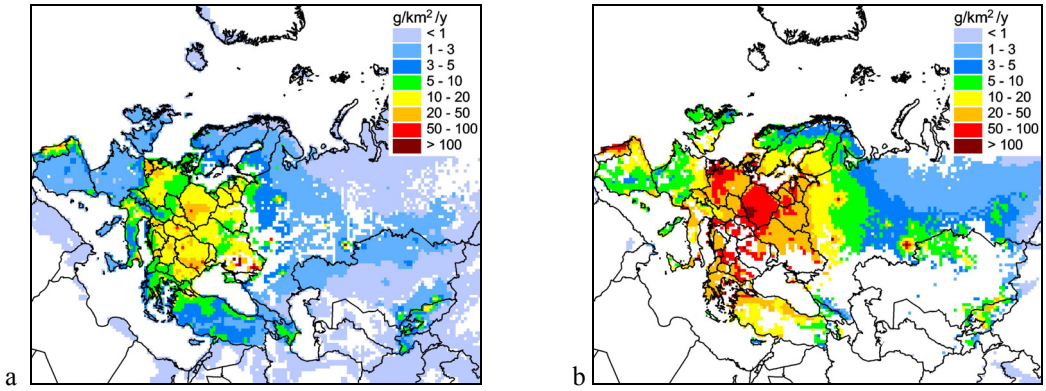


Fig. 1.32. *Deposition fluxes of B[a]P to areas covered by grass (a) and coniferous forest (b) for 2011, g/km^2/y*

It can be seen that deposition flux of B[a]P to different types of underlying surface may differ several times. Particularly, deposition fluxes to forest-covered areas are several times higher than to grass. Besides, fluxes to vegetation-covered areas are subject to strong spatial variations.

Soil concentrations. Thresholds levels for PCDD/F soil concentrations are established in a number of national legislations for characterization of risk from PCDD/Fs to human health and the environment. For example, threshold level for PCDD/F soil concentrations in Canada is set to 4 ng TEQ/kg (Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health). In Italy Soil Acceptable Concentration Limit (SACL) for PCDD/Fs in soil amounts to 10 ng TEQ/kg (D.Lgs 152/06).

Model assessment of POP pollution allows evaluating soil contamination levels in the EMEP domain. For example, modelled concentrations of PCDD/Fs in top 5 cm of soil for 2011 are shown in Fig. 1.33.

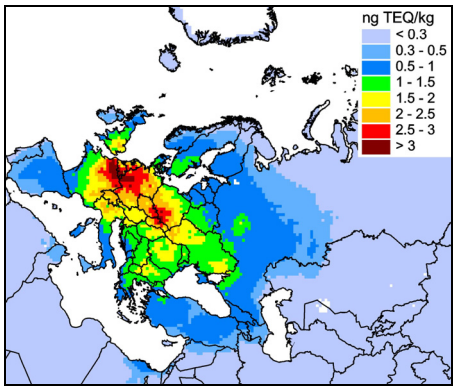


Fig. 1.33. *Concentrations of PCDD/Fs in top 5 cm soil layer in 2011, ng TEQ/kg*

PCDD/F concentrations in soil within the EMEP region vary from 0.1 to more than 3 ng TEQ/kg. The highest soil contamination is indicated for the essential part of Central and Western Europe. Monitoring of soil concentrations showed quite similar concentration levels. In particular, measured soil concentrations in the period from May to October 2004 in Austria were between 0.17 ng TEQ/kg and 9 ng TEQ/kg with median 0.77 ng TEQ/kg. Higher levels of soil concentrations were observed in the UK ranging from 0.16 ng TEQ/kg to over 100 ng TEQ/kg with the median of 2 ng TEQ/kg. In the Piedmont region (northern Italy) in 2003 measured PCDD/F soil concentrations were equal to 1.4 ± 0.7 ng TEQ/kg for agricultural soils and 3.0 ± 2.6 ng TEQ/kg for natural soils [*Fabietty et al.,* 2010]. Measurements of soil concentrations in Switzerland

made in 2002 in the framework of Swiss national monitoring network (NABO) showed values between 1 ng TEQ/kg and 11 ng TEQ/kg [*Schmid et al.,* 2005].

Modelling results well agree with median values of the above measurements. At the same time, maximum values of observed concentrations can be substantially higher than modelled ones since the model represents average soil concentrations over 50×50 km grid cells. Besides, high local contamination can be conditioned by the direct emissions of PCDD/Fs to soil. The ranges of measured soil concentrations show that these concentrations can vary several times within one and the same region.

Modelled POP concentrations in various media (air, soil) can be of use to indicate areas with exceedances of nationally accepted threshold levels.

Air concentrations. Levels of PAHs air concentrations are subject to national legislations. Particularly, in the EU countries the target value (TV) for annual mean B[a]P air concentrations is set to 1 ng/m^3. Besides, the upper and lower assessment thresholds (UAT and LAT) equal to 0.6 and 0.4 ng/m^3 are established. Here UAT is a level below which a combination of measurements and modelling techniques may be used to assess ambient air quality, and LAT is a level below which sole use of modelling or objective estimation techniques can be sufficient. Similar threshold levels of air concentrations are accepted in some of the EECCA countries.

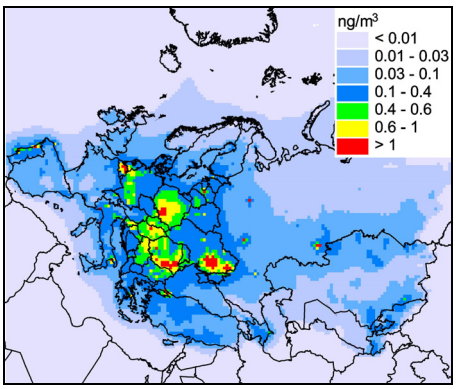


Fig. 1.34. *B[a]P air concentrations in 2011, ng/m^3, annual averages. Areas where TV is exceeded are marked red.*

Modelled B[a]P air concentrations can be used to determine the areas where exceedances over the threshold levels take place. As an example, spatial distribution of B[a]P air concentrations obtained for 2011 is shown in Fig. 1.34. The areas where the TV is exceeded are marked by red, areas with exceedances of UAT and LAT are marked by yellow and green, respectively.

Human and animal tissues. Due to high ability of POPs for bioaccumulation along the food chain, they can pose risk to animals and human health. A lot of studies on levels of various POPs in animals, fish and humans were recently performed. As an example, the BalticPOPs research program funded by the Swedish Environment Protection Agency (SEPA) can be mentioned, which was focused on the investigation of levels of PCDD/Fs in Baltic edible fish. From this viewpoint modelling results on POP concentrations in the environmental media can be used for the evaluation of hazards for humans and animals.

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. several 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.

Examples of such preliminary model simulations for 2012 carried out using emission data for 2010 are given in Fig. 1.35. The maps illustrate the levels of pollution of B[a]P and PCDD/Fs. This information can be helpful for national environmental agencies and experts to describe recent variations of pollution and effect of changing meteorology.

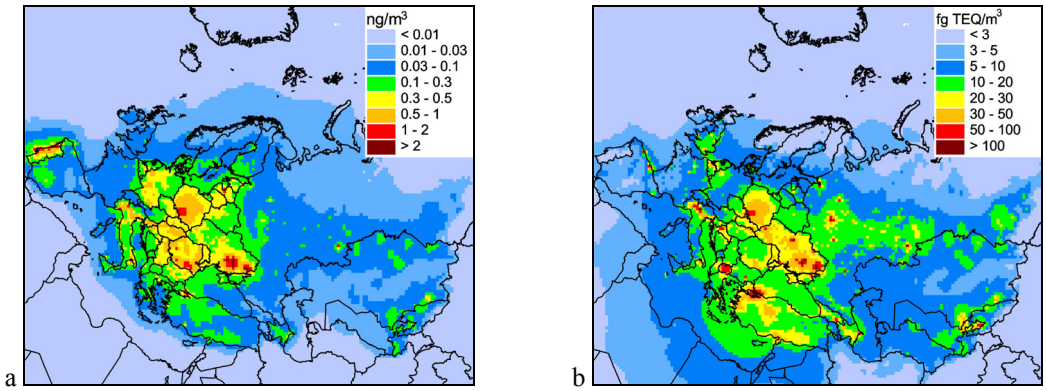


Fig. 1.35. *Spatial distribution of B[a]P (a) and PCDD/F (b) annual mean air concentrations for 2012.*

2. INTERCONTINENTAL TRANSPORT OF POPS

Intercontinental transport of long-lived POPs significantly contributes to the pollution of the EMEP region. Due to their physical-chemical properties these POPs tend to disperse globally and accumulate in surface media (soil, sediments, and water bodies) which subsequently can act as secondary emission sources and contribute to the pollution.

To evaluate intercontinental transport and contributions of global POP emission sources to the pollution of the EMEP region the Global EMEP Multi-media Modelling System (GLEMOS) is applied. GLEMOS is an Eulerian type multi-pollutant multi-media modelling system being developed for operational and research applications within EMEP. Main features of the GLEMOS modelling system include modular structure and flexible choice of modelling domain (from global to local scale) and spatial resolution. The model describes cycling of POPs in four environmental media (the atmosphere, soil, vegetation, and the ocean). Development of the modelling system was documented in a series of MSC-E/MSC-W joint technical reports [*Tarrason and Gusev*, 2008; *Travnikov et al*., 2009; *Jonson and Travnikov*, 2010; *Travnikov and Jonson*, 2011; *Jonson and Travnikov*, 2012]. Recent updates of the global modelling system are described in the MSC-E Technical Report [*Shatalov et al*., 2013].

This year GLEMOS has been applied for the evaluation of global scale transport, fate and contributions of secondary emission sources to the pollution of the EMEP region. Results of model simulations for 2011 are exemplified for two POPs characterized by significant potential to the long-range transport, namely, HCB and PCB-153.

HCB is a pollutant of global dispersion with significant potential to the cycling between the environment compartments. Experimental modelling of HCB global transport and accumulation in the environment,

performed on the basis of the scenario of historic and contemporary HCB emissions constructed by MSC-E [*Shatalov et al.*, 2013], permitted to estimate contributions of anthropogenic and secondary emission sources to the pollution of the EMEP countries. Spatial distribution of HCB annual mean air concentrations for 2011 illustrates the dispersion of HCB in the atmosphere from major emission sources (Fig. 2.1a). Elevated levels of HCB air concentrations can be seen in the EMEP region and in Southeast Asia which indicates importance of their emissions for the pollution of other regions.

Intercontinental transport of HCB significantly affects pollution of the EMEP region contributing about 35% to levels of air concentrations.

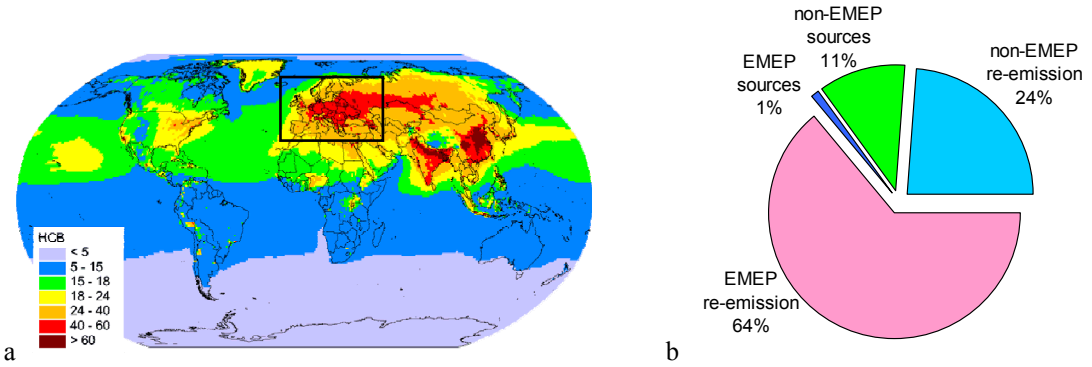


Fig. 2.1. *Spatial distribution of annual mean HCB air concentrations (pg/m^3) for 2011 (a). Contributions of various emission sources (EMEP anthropogenic emissions, non-EMEP anthropogenic emissions, secondary emission sources within the EMEP region and beyond) to the pollution of the EMEP region (b).*

According to the modelling results pollution of the EMEP region by HCB is dominated by the secondary emission sources, while contribution of anthropogenic emissions is comparatively small. The intercontinental transport of HCB significantly influences the pollution levels in the EMEP region. Particularly, the share of non-EMEP emission sources accounts for 35% (Fig. 2.1b) from which 24% is contributed by the non-EMEP secondary emissions and 11% by the non-EMEP anthropogenic emissions. Among the non-EMEP anthropogenic emissions the largest contribution is made by the sources of Southeast Asia (8%). HCB emissions of the EMEP region essentially contribute to the pollution of other regions. Particularly, about 50% of HCB emitted within the EMEP countries is transported outside of the region.

Spatial distribution of PCB-153 annual mean air concentrations for 2011 is illustrated in Fig. 2.2a. Comparing to HCB, PCB-153 is less volatile and tend to accumulate in the environmental compartments (e.g. soil, seawater). Dispersion of PCB-153 in the atmosphere reflects relative importance of major emission sources. According to the global inventory of PCB emissions [*Breivik et al.*, 2007] applied in model simulations more than 60% of global PCB-153 emission belongs to the EMEP region. Following this distribution of global emissions pollution of the EMEP region is dominated by the anthropogenic and secondary emission sources of the EMEP countries. Besides, these emissions significantly contribute to the pollution of other regions (Fig. 2.2b). As shown in the Figure, 26% of PCB-153 emitted by the EMEP sources is transported outside of the EMEP region.

Essential efforts for the evaluation of intercontinental transport of various pollutants, including POPs, were recently undertaken by the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). The work of the Task Force and experts from various international and national organizations supported the preparation of the HTAP Assessment 2010 [*HTAP*, 2010], which summarized current state of the knowledge on the POP intercontinental transport. Major results of this assessment and recommendations for further work are outlined in [*Gusev et al.*, 2012]. MSC-E participated in this

activity and contributed to the assessment carrying out experimental model simulations of POP global transport and performing their analysis. It is planned to continue this activity in the framework of the next phase of the TF HTAP work for 2012-2016 [http://www.htap.org].

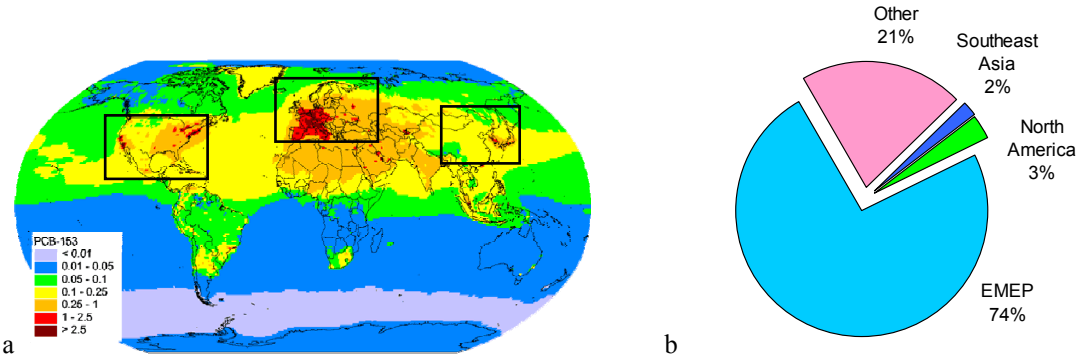


Fig. 2.2. *Spatial distribution of annual mean PCB-153 air concentrations (pg/m^3) for 2011 (a). Contributions of PCB-153 emissions of the EMEP region to pollution levels in various regions of the globe (b).*

3. POLLUTION ASSESSMENT IN THE EECCA COUNTRIES

3.1. Emission and monitoring data in the EECCA countries

Countries of the Eastern Europe, Caucasus and Central Asia (EECCA) are of high priority within CLRTAP. In particular, at the fortieth session of the WGSR revised version of the Action Plan was elaborated to involve the EECCA countries into the work of the Convention (ECE/EB.AIR/WG.5/2007/17). This Action Plan envisaged the raise of the political profile of the Convention activities in this region and creation of awareness of potential health and environment problems in the EECCA countries by compiling national and international expert reports.

It should be noted that only two of the EECCA countries (Armenia and the Ukraine) have signed, and one country (Republic of Moldova) – ratified the Protocol on POPs. To facilitate further ratification and implementation of the Protocol, information (in Russian and English) on POP pollution levels in the EECCA countries has been produced under EMEP this year.

At present more than a half of the EECCA countries started reporting of their national inventories of POP emissions (see Fig. 3.1). Among them, two countries (the Ukraine and Belarus) reported information on spatial distribution of emissions. To fill gaps in the reported emission data expert estimates are applied [*Shatalov et al.*, 2013].

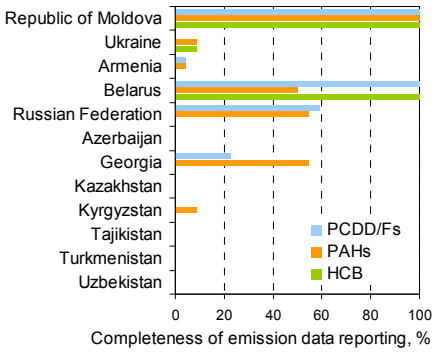


Fig. 3.1. *Completeness of official emission data for POPs reported by the EECCA countries for 1990-2011.*

Monitoring of POP air concentrations is initiated at two EMEP sites located in Republic of Moldova and Kazakhstan (KZ01 and MD13). Along with measurements of the EMEP monitoring network, a lot of national data on POP pollution levels in the EECCA countries are available. In particular, regular monitoring of B[a]P in air is carried out in the urban areas of Russia

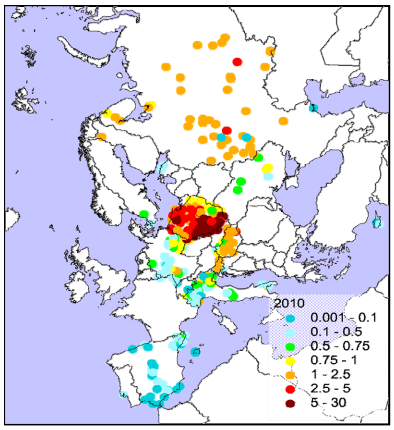


Fig.3.2. *National measurements of B[a]P air concentrations in Russia, the Ukraine, and in the EU countries for 2010, ng/m^3.*

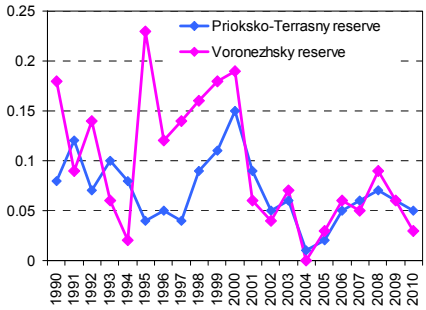


Fig. 3.3. *B[a]P concentrations in air measured at two Russian reserves for the period from 1990 to 2010, ng/m^3*

and the Ukraine. National measurements of these countries for 2010 are illustrated in Fig. 3.2 along with measurements made in the EU countries (obtained from the EEA AirBase database). Elevated levels of B[a]P air concentrations, exceeding the EU target value 1 ng/m^3, can be seen in Russia and Eastern Europe. Long-term measurements of B[a]P in air obtained at the stations of Russian integrated background monitoring network reflect gradual decreasing of concentrations (Fig. 3.3). These data can complement the measurements of the EMEP sites.

Further improvement of the evaluation of POP pollution levels in the EECCA countries requires the refinement of information on their emissions. In this respect it would be reasonable to assist these countries in preparation of national emission inventories. Additional efforts are required to increase the number of monitoring sites in the EECCA countries and involve their national measurement data on POPs into the assessment of pollution. In this case active cooperation between national experts from the EECCA countries and the EMEP bodies is highly appreciated. In particular, available information on POP pollution levels in these countries, including national data, can be discussed at the annual meetings of the EMEP TFMM, which provides scientific forum for consideration of various problems in the field of pollution assessment.

To refine information on POP emissions in the EECCA countries there is a need to assist them in preparation of national emission inventories. National monitoring in the EECCA countries is important source of information for the assessment of POP pollution in this region.

3.2. Levels and trends

Results of model assessment of POP pollution levels in the EECCA countries and their changes in the period from 1990 to 2011 are summarized below. More detailed information for each EECCA country can be found in the Internet (http://www.msceast.org).

Polycyclic Aromatic Hydrocarbons (PAHs)

Pollution by PAHs significantly varies over the EECCA countries (Fig. 3.4a). High levels of PAH air concentrations are noted for the western part of the EECCA region (the Ukraine, Belarus, and the western part of the Russian Federation). Elevated levels can be seen in some areas in the northern part of Kazakhstan and in the southern part of the EECCA region.

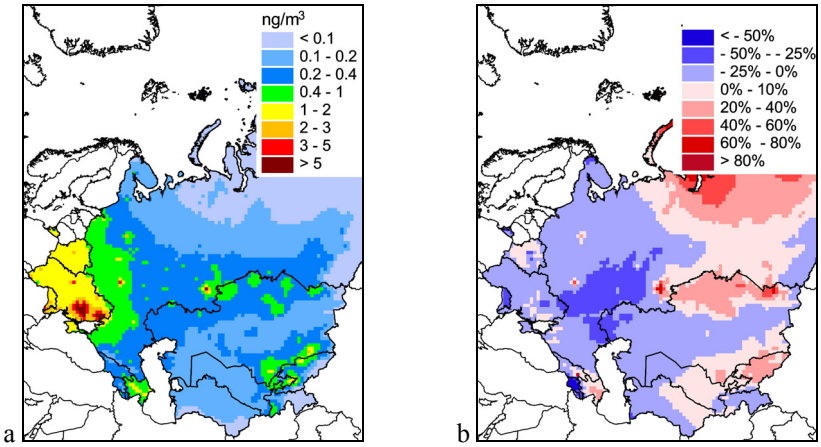


Fig. 3.4. *Spatial distribution of PAH air concentrations in the EECCA countries (four indicator PAHs) in 2011, ng/m^3 (a) and changes of concentrations from 1990 to 2011, % (b). Positive values of the change mean increase, and negative – decrease of air concentrations.*

Reduction of PAH levels of pollution in the EECCA countries from 1990 to 2011 is illustrated in Fig. 3.4b. Maximum decrease of air concentrations is obtained for Republic of Moldova, Armenia, the Ukraine and some areas in the Russian Federation. The decrease took place mostly in the western part of the region whereas in the eastern part PAH air concentrations slightly increased.

Temporal variations of PAH pollution levels during the period from 1990 to 2011 were analyzed on the example of B[a]P. Particularly, long-term changes of B[a]P air concentrations in Belarus and Republic of Moldova are shown in Fig. 3.5.

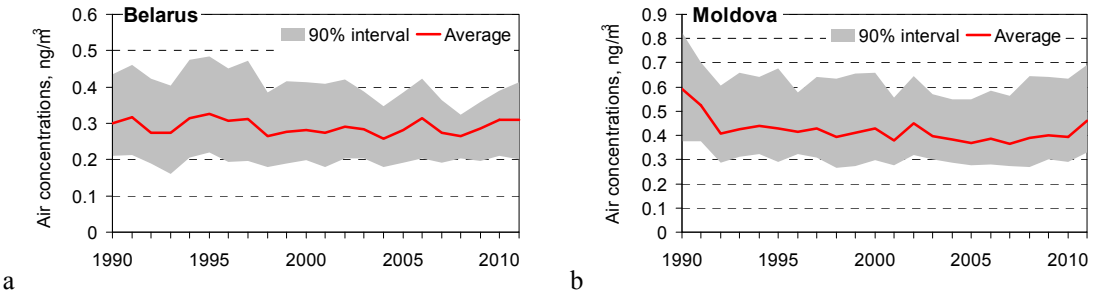


Fig. 3.5. *Temporal trends of B[a]P air concentrations from 1990 to 2011 in Belarus (a) and Republic of Moldova (b), ng/m^3. Grey strip shows the range of concentrations over the country.*

It is seen that the levels of air concentrations in Belarus did not change much. For Republic of Moldova the decrease in the beginning of the period was almost levelled off starting from 1993. At the same time, both of these countries are characterized by the slight increase of B[a]P concentrations near the end of the considered period, which can be connected with similar increase of concentrations in the EU countries (see Chapter 1).

Transboundary transport continued to play essential role in the pollution of the EECCA countries. Changes of relative contributions of national emissions and transboundary transport from other EMEP countries from 1990 to 2011 to the pollution of Belarus are illustrated in Fig. 3.6.

Decrease of PAH pollution took place mostly in the western part of the EECCA region whereas in the eastern part levels of pollution slightly increased.

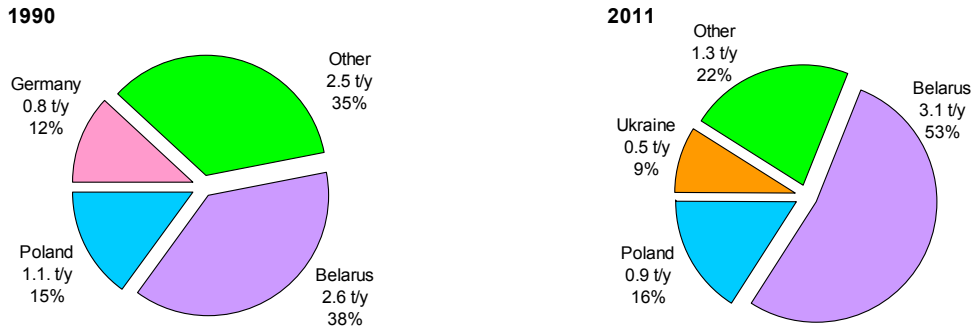


Fig. 3.6. *Contributions of the EMEP countries to B[a]P deposition to Belarus for 1990 and 2011.*

It is seen that the contribution of transboundary transport to the pollution of Belarus decreased from 62% in 1990 to 47% in 2011. This can be explained by both the reduction of pollution levels in the EU countries and the growth of national emissions in Belarus. While transport of pollution from the EU countries is decreasing, the contribution of national emission sources in the EECCA countries becomes more important.

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)

For PCDD/Fs, the decrease of air concentrations in the EECCA region is more significant comparing to PAHs. Spatial distribution of PCDD/F air concentrations in the EECCA countries in 2011 and the reduction from 1990 to 2011 are illustrated in Fig. 3.7. Relatively more significant decrease can be seen in the western part of the region (20% – 60%), particularly, in Republic of Moldova, Belarus, and the Russian Federation, while changes in other parts of the region are comparatively smaller.

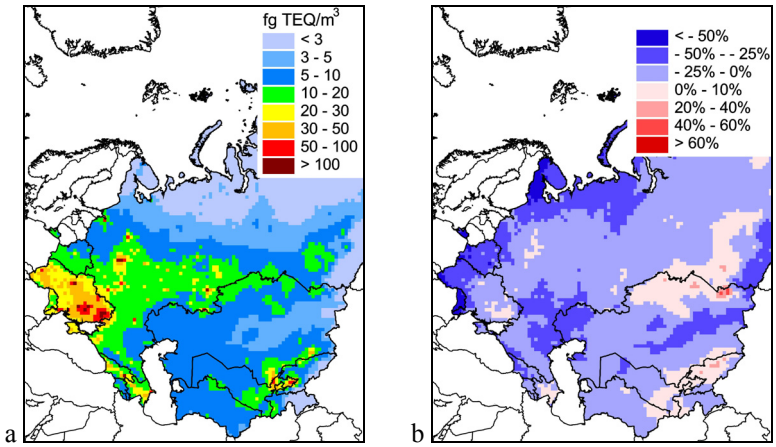


Fig. 3.7. *Spatial distribution of PCDD/F air concentrations in the EECCA countries in 2011, fg TEQ/m^3 (a) and change of concentrations from 1990 to 2011, % (b).*

Long-term variation of PCDD/F air concentrations in the EECCA countries are presented in Fig. 3.8 on the example of Republic of Moldova. Particularly, levels of air concentrations in the country decreased by almost 65% from 1990 to 2011. It can be seen also that reduction of maximum levels of concentrations is more significant accounting for about 75%. Reduction of PCDD/F air concentrations in the EECCA countries was essentially influenced by the declining PCDD/F pollution in the EU countries.

Reduction of PCDD/F air concentrations in the EECCA countries was essentially influenced by the declining PCDD/F pollution in the EU countries.

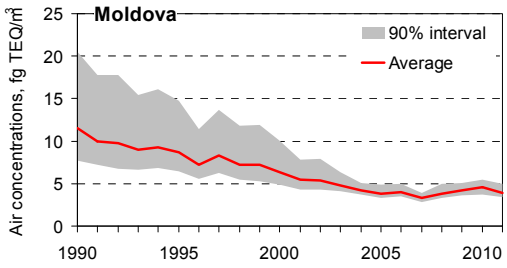


Fig. 3.8. *Temporal trends of PCDD/F air concentrations from 1990 to 2011 in Republic of Moldova, fg TEQ/m^3. Grey strip shows the range of concentrations over the country.*

Hexachlorobenzene (HCB)

Similar to other EMEP countries contemporary levels of HCB pollution in the EECCA countries are to considerable extent determined by the secondary emission sources (re-volatilization from soil and water bodies). Spatial distribution of annual mean HCB air concentrations in these countries for 2011 is shown in Fig. 3.9. Elevated levels of concentrations can be seen in the areas of historic emissions which led to substantial accumulation of HCB in soil compartment.

In most of the EECCA countries levels of HCB concentrations dropped by almost 90%.

According to modelling results the decrease of HCB pollution in the EECCA region is nearly homogeneous. In most of the EECCA countries levels of HCB concentrations dropped by almost 90%. Example of long-term changes of HCB air concentrations from 1990 to 2011 for Belarus is presented in Fig. 3.10.

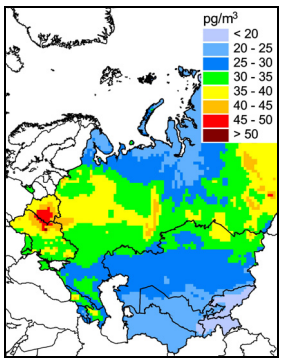


Fig. 3.9. *Spatial distribution of HCB air concentrations in EECCA countries in 2011, ng /m^3.*

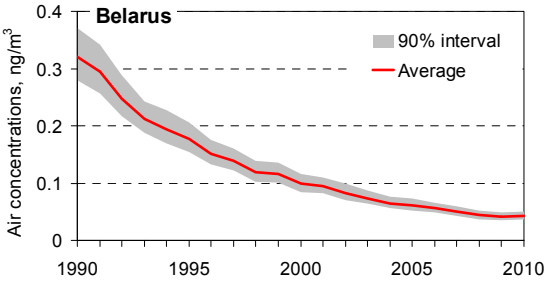


Fig. 3.10. *Temporal trends of HCB air concentrations from 1990 to 2011 in Belarus, ng/m^3.*

The above analysis of the contamination in the EECCA region is at present of the preliminary character since official emission data are available not for all countries in this region. For more reliable assessment of pollution in this region further refinement of the information on POP emissions in the EECCA countries is desirable. To facilitate the work on the pollution assessment in these countries specific case studies on POPs can be organized with the participation of national experts in monitoring and modelling as well as with the support from the EMEP Centres. Special attention needs to be paid also to the development of Russian version of the EMEP/MSC-E website to support the EECCA countries with relevant information.

To facilitate the work on the pollution assessment in the EECCA countries specific case studies on POPs can be organized with participation of national experts in emissions, monitoring and modelling as well as with the support from the EMEP Centres.

4. NEW SUBSTANCES

The hazard of chemical substances for human health and the environment is in the focus of a number of international organizations (World Health Organization (WHO), United Nations Environment Programme (UNEP), Food and Agriculture Organization (FAO)) and various Conventions, such as the Stockholm Convention on Persistent Organic Pollutants (SC), the Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal, the Rotterdam Convention on the Prior Informed Consent Procedure for Certain Hazardous Chemicals and Pesticides in International Trade, and the Convention on Long-Range Transboundary Air Pollution (CLRTAP). Most of these Conventions make provisions for the inclusion of new substances.

Sound management of hazardous substances is one of the priority activities of a number of international organizations (WHO, UNEP, FAO) and Conventions (CLRTAP, the Stockholm Convention, the Basel Convention, the Rotterdam Convention).

The necessity of cooperation between Conventions in the field of evaluation of chemicals is noted in the Report [*Global Chemicals Outlook*, 2012] prepared by experts from UNEP and WHO. The conference jointly organized by the Parties to Stockholm, Basel and Rotterdam Conventions (ExCOPs-2, 28 April – 10 May 2013) decided to widen the cooperation and strengthen the coordination with Strategic Approach to International Chemicals Management. In the framework of this decision the Persistent Organic Pollutants Review Committee (POPRC) of the Stockholm Convention and the Chemical Review Committee (CRC) of the Rotterdam Convention also recognized the necessity of exchange with information and hold joint meetings.

The production of chemicals in the world is permanently growing. However, the growth of production volumes is different in different regions. According to the data of the European Chemical Industry Council (CEFIC) Asian chemical production surpasses that of the rest of the world. Along with the growing of production volumes the nomenclature of chemical production is widened. It causes the necessity of scientifically justified selection of substances that can be considered as persistent organic pollutants (POPs) [*Gama et al*., 2012; *Brown et al*., 2012; *Breivik*, 2012; *Sailaukhanuly et al*., 2013].

Evaluation of chemicals from the point of view of their persistence, bioaccumulation and toxicity (PBT substances) in Europe is subject of the European regulation REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) which came into force on 1 June 2007. At present European database on industrial chemicals ESIS (European Chemical Substances Information System) contains about 95000 chemicals, among which around 3% can be considered as PBT [*Strempel et al.,* 2012; *Öberg and Iqbal*., 2012]. Up to the moment the information on almost 9000 industrial chemicals registered under REACH is contained in registration dossiers. According to the REACH requirements, registration of hazardous substances should include a set of data on their physical-chemical properties, emissions, etc. Thus, the data from REACH Registration Dossiers can be used as one of possible sources of information for evaluation of hazard of chemicals for human health and the environment. In addition, approaches to risk assessment worked out in the framework of the REACH legislation can also be of use under CLRTAP.

The EU Regulation REACH may be a source of information on physical-chemical properties and emissions of industrial chemicals. The approaches to risk assessment worked out under REACH can be used for the evaluation of environmental risk of POPs for human health and the environment.

The European Chemical Industry Council (CEFIC), being a coordinator of activities of chemical industry in Europe, organizes and conducts research on investigation of PBT properties of substances in the framework of Cefic Long-range Research Initiative. In the period from 2008 to 2011 project ECO3A.3-

USTO “Evaluation of the predictability and reversibility of environmental accumulation for substances for POP/PBT-like properties” was realized. One more CEFIC project to be mentioned is ECO13 “Development, application and evaluation of model-based screening procedures for PBT chemicals and POPs” (SCREEN-POP). The first step in the project was to assemble a list of about 12600 individual chemicals from high and low production volume chemicals reported by different jurisdictions. The physical-chemical properties of these chemicals were estimated using different Quantitative Structure-Permeability Relationship methods, largely from the EPI Suite software package. Emissions during production, formulation and use were estimated with the help of information on quantities entering commerce combined with physical-chemical property data (vapour pressure and water solubility) [http://www.cefic-lri.org/projects/].

The necessity of evaluation of chemicals for their inclusion to international legislations initiated a lot of scientific research on evaluation of new substances. In particular, CEFIC organizes and conducts research on determination of PBT properties of chemicals.

The necessity of evaluation of chemicals for their inclusion to international legislations initiated a lot of scientific research studies conducted in various countries with the following main directions:

- investigation of physical-chemical properties and fate in the environment of substances for evaluation of environmental life-time of new POPs and degradation processes;
- revealing source types of these substances and evaluation of emissions at various levels (global, country, local);
- determination of levels of the substances in various components of the environment including animals and humans;
- modelling fate of the substances in the environment for determination of their dispersion in the atmosphere, seawater, and soil and evaluation of exchange processes in these media and accumulation in them.

Large part of recent scientific publications on new substances is focused on the investigation of polybrominated ethers (PBDE), perfluorooctane sulfonates and their derivatives and short-chain chlorinated paraffins (SCCP). The research on PBDE and SCCP was directed mainly to the determination of environmental levels of these substances [*Cai et al*., 2012 a,b; *Yu et al*., 2013; *Zareitalabad et al.*, 2013; *Möller et al*., 2011; 2012; *Koblizkova et al*., 2012; *Benskin et al*., 2012] and others. At the same time a lot of research on PFOS is aimed at the investigation of the behaviour of the substances in the environment, particularly of degradation process. As an example, investigations of the behaviour of various salts of perfluorooctanesulfonic acid and its derivant spirits and ethers (so-called precursors) in marine environment, soil and air were performed in [*Taniyasu et al*., 2013; *Dasu et al*., 2013; *Wong et al*., 2012; *Xie et al*., 2013; *Bendig and Vetter*, 2013]. The evaluation of the content of perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), perfluorodecanoic acid (PFDA), and perfluorooctanesulfonic acid (PFOS) in the Baltic Sea was presented in [*Filipovic et al*., 2013]. This study took into account the income of these substances to the Baltic Sea from riverine discharge, atmospheric deposition, coastal wastewater discharges, and transport from the North Sea based on recently published monitoring data.

Model approaches can be used for evaluation of environmental persistence and long-range transport potential of substances considered for the inclusion to various legislations.

EMEP was involved in the investigation of new substances from the viewpoint of their long-range transport potential and persistence in the environment using model approaches. Sufficient experience and information for application of mathematical modelling methods is gained. Further development of these methods will result in the possibility of evaluation of pollution levels of new substances in the environmental media both in the EMEP region and on the global scale.

5. DISSEMINATION OF INFORMATION

Dissemination of the results on POP pollution assessment is of high importance for the environment protection regulations on a country and international levels. This information can be accessed as through the annual reports and web presentation, which provide more flexible and targeted support to experts and authorities with the required data. Another important aspect is the information exchange with other international organizations and programmes.

5.1. Information on the web

Wide range of relevant information on POP pollution levels within the EMEP region and on the global scale is currently available on the MSC-E website (Fig.5.1). It contains modelled air concentration and deposition fluxes within the EMEP domain and estimates of POP dispersion on a global scale. In addition, description of the MSC-E chemical transport models applied for the assessment is given along with information on input data used for the modelling. Details of the research and assessment can be found in the EMEP technical reports and other publications also presented on the website (http://www.msceast.org).

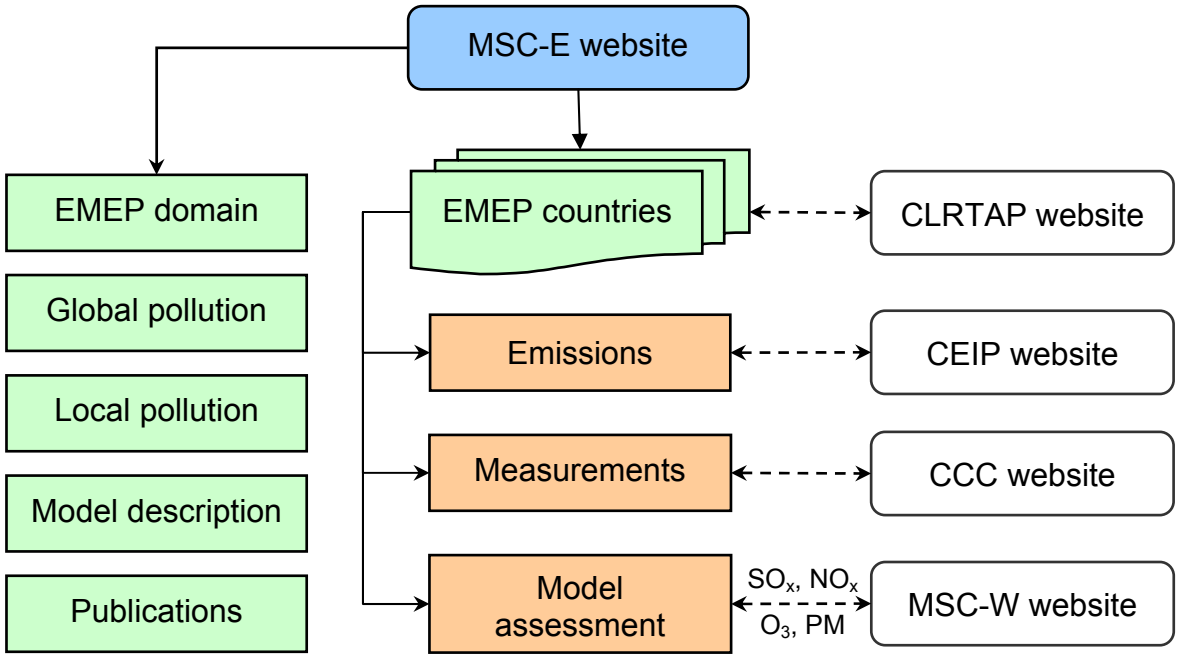


Fig. 5.1. *General structure of information presented at the MSC-E website*

Information on pollution by POPs and other contaminants is given for each individual EMEP country. This country-specific information comprises variety of data on emissions, measurements, and model assessment for a 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 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, O_3, PM) as the interlink with the website of MSC-W. However, this approach needs further development and discussion within the Convention.

5.2. Country-specific information

Detailed information on POP pollution for each individual EMEP country is presented in the form of graphs, maps and data files on the MSC-E website. An additional work has been initiated to provide the EECCA countries with information in Russian (ru.msceast.org/). 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 in the environmental media;
- Transboundary pollution of a country;
- Contribution of national sources to transboundary transport;
- Ecosystem-specific deposition.

For each country, data on mean annual concentrations in air, soil and vegetation with spatial resolution 50x50 km are presented (Fig. 5.2). On the website user can set chart scales of concentrations. The maps are accompanied with distribution diagrams.

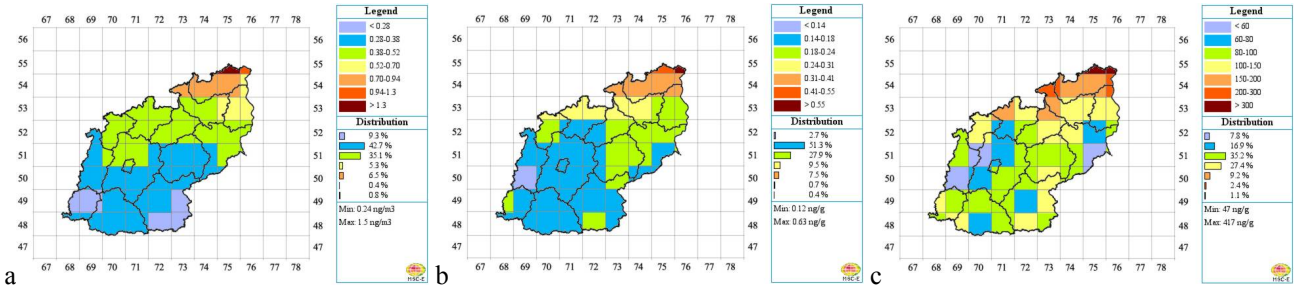


Fig. 5.2. *Spatial distribution of concentrations of B[a]P in the Czech Republic: (a) – air concentrations, ng/m^3, (b) – soil concentrations, ng/g d.w., (c) – concentration in vegetation, ng/g d.w.*

Detailed information on transboundary pollution includes maps of total deposition flux to a country's area and relative contributions of foreign sources to anthropogenic deposition. Besides, total deposition from national and foreign sources to a country's territory is presented (Fig. 5.3). Only several main contributors are selected, while 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.

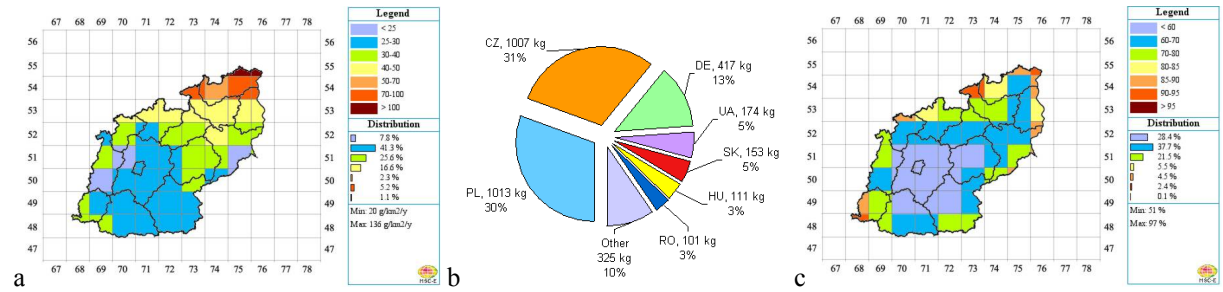


Fig. 5.3. *Characteristics of transboundary transport of B[a]P to the Czech Republic: (a) – total deposition flux, (b) – main contributors to deposition to the Czech Republic, (c) – relative contributions of foreign sources.*

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. 5.4). Besides, distribution of country's total deposition to main regions-receptors (countries or seas) within the EMEP region is indicated. Values of deposition to other regions are available in source-receptor matrix.

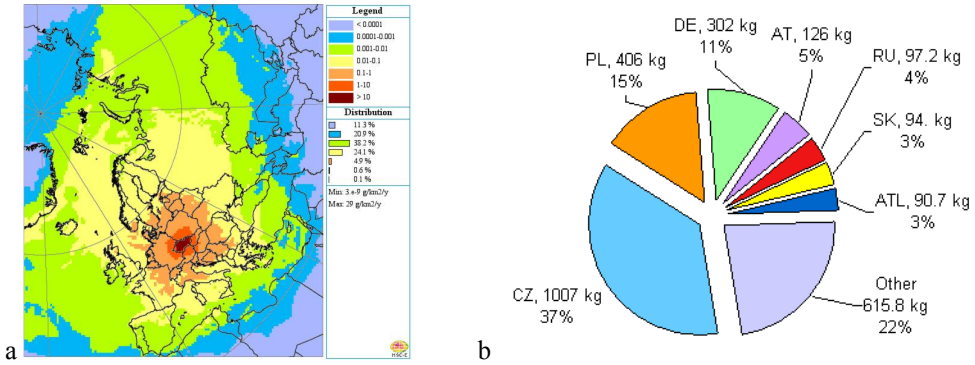


Fig. 5.4. *Characteristics of transboundary transport of B[a]P from the Czech Republic: (a) – spatial distribution, (b) – contributions to countries-receptors.*

Deposition to different types of ecosystems is important information for the effect community, particularly, for the assessment of harmful effects for human health and the environment. Country-specific maps of deposition to main land cover types are available. In addition, more detailed information on POP pollution 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.

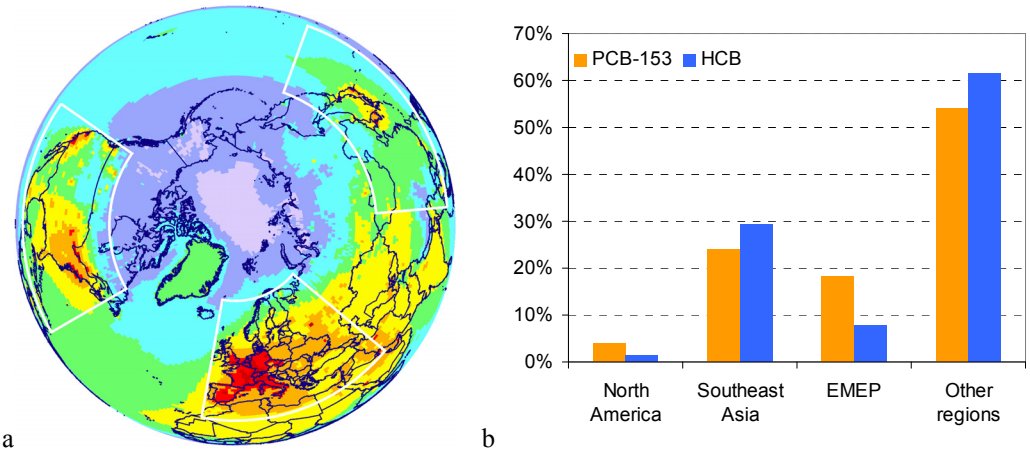
5.3. Information for international organizations and programmes

EMEP is the unique instrument within the LRTAP Convention performing regular assessment of pollution and supporting Parties to the Convention with information on POP pollution levels in Europe and other regions. There is a wide interest to the data products and analysis performed by the EMEP Centres outside of the Convention. In the context of the co-operation with international bodies regular exchange with this information is carried out. These activities are performed in accordance with the EMEP work-plan. Some aspects of this co-operation are discussed below.

To support further cooperation and exchange with information it would be reasonable to consider establishing of specific agreements with international organizations and programmes dealing with assessment of POP pollution.

POP pollution of the Arctic

MSC-E has a long and successful experience of co-operation with the Arctic Monitoring and Assessment Programme (AMAP), which included participation in a number of joint assessments and projects. Particularly, 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]. Fig. 5.5 illustrates the model assessment of the Arctic pollution by HCB and PCB-153. Spatial distribution of HCB pollution levels in the Arctic and surrounding regions calculated by the GLEMOS system is illustrated in Fig. 5.5a. Significant contribution to the pollution of the Arctic by HCB and PCB-153 was made by the emission sources of Southeast Asia and the EMEP countries (Fig. 5.5b).



a b

Fig. 5.5. *Spatial distribution of PCB-153 annual mean air concentrations in the Arctic and surrounding regions, ng/m^3 (a). Contribution of selected source regions to PCB-153 and HCB pollution levels in the Arctic (north of 66.5ºN) (b)*

Trends in measured POP concentrations in the Arctic

Long-going environmental regional monitoring programs, like EMEP, HELCOM, AMAP, IADN (US Great Lake Integrated Atmospheric Deposition Network) etc., are interested in estimating temporal trends of the POPs environmental concentrations, in order to evaluate the effectiveness of environmental regulations. Large number of monitoring stations operating under EMEP offers a unique dataset, which can give important insight about the trends of POPs in Europe.

To illustrate some of the ongoing work being carried out in CCC, examples of analysis of preliminary long-term temporal trends of PCBs, PAHs and organochlorine pesticides (HCB) from the Zeppelin station are presented (Fig. 5.6). Temporal trends were analyzed for PCBs, for PAHs, and for HCB for the periods 1998-2012, 1994-2012, and 1994-2012, respectively. These results represent part of the work which is going to be included in the 2014 UNEP Global Monitoring Plan report.

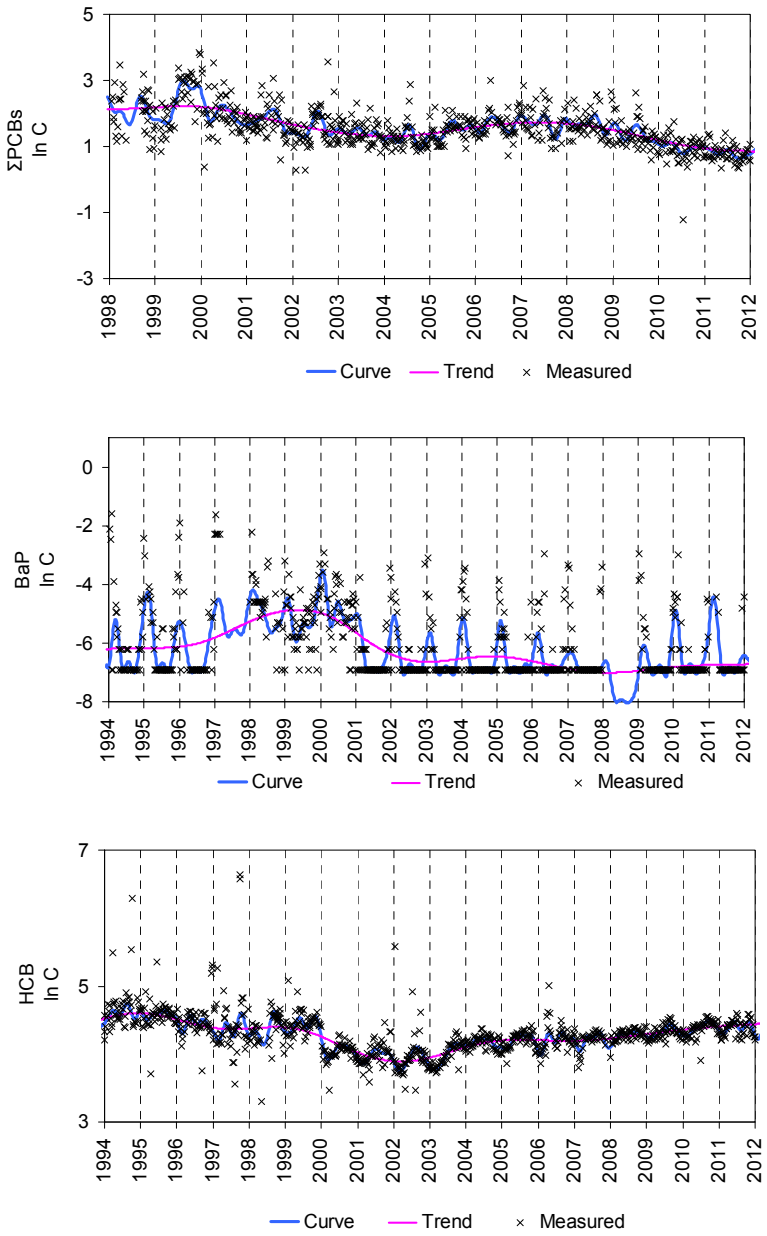


Fig. 5.6. *Long-term temporal trends and seasonal cycles of observed PCB (sum of PCB-28, 52, 101, 118, 138, 153 and 180), B[a]P, and HCB air concentrations at the EMEP monitoring site Zeppelin (NO0042) (air concentrations are expressed as pg* m^{-3}*).*

POP pollution of marginal seas

Information on atmospheric pollution of marginal seas within the EMEP region is of interest for various international marine organizations. In particular, in cooperation with other EMEP Centres, MSC-E performs regular model assessments of atmospheric pollution of the Baltic Sea by different pollutants including POPs. This work is carried out in accordance with the Memorandum of Understanding between the Baltic Marine Environment Protection Commission (HELCOM) and the United Nations Economic Commission for Europe.

Recent assessment of airborne pollution load to the Baltic Sea comprised the information on long-term trends in the PCDD/F deposition and evaluation of contributions of anthropogenic and secondary

PCDD/F emissions, originated from the EMEP countries, to the pollution levels. Results of this assessment are published in the joint annual report of the EMEP Centres [*Bartnicki et al*., 2012]. Short summary of information on the Baltic Sea pollution by PCDD/Fs is available also in a form of indicator fact sheets published in the internet on the HELCOM web site [http://www.helcom.fi]. Annual atmospheric deposition fluxes of PCDD/Fs over the surface of the Baltic Sea have decreased in the period 1990-2010 by 52%. Spatial distribution of annual PCDD/F deposition to different sub-basins of the Baltic Sea is exemplified in Fig.5.7a.

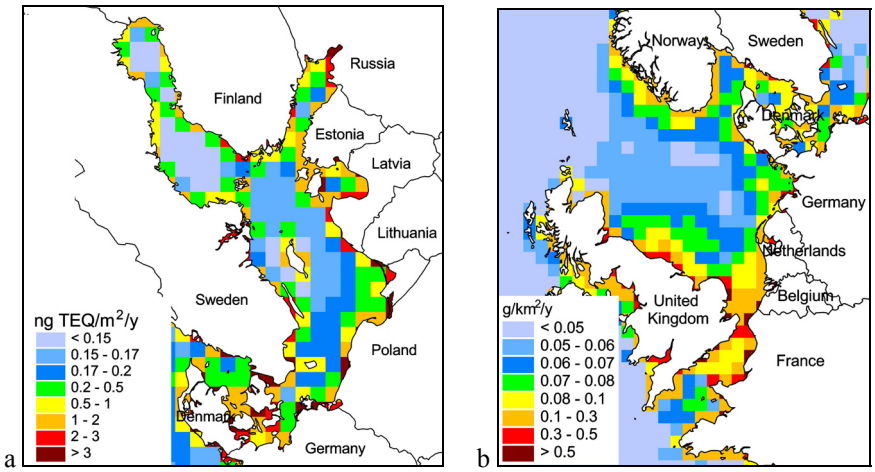


Fig. 5.7. *Spatial distribution of PCDD/F deposition over the Baltic Sea, ng TEQ/km^2/y (a) and PCB-153 deposition over the North Sea, g/km^2/y (b)*

Atmospheric load of POPs to the North Sea and the Northern Atlantic (Fig. 5.7b) was evaluated by MSC-E in the framework of a number of contracts between EMEP and OSPAR. The assessment contained evaluation of long-term pollution of the OSPAR maritime area pollution by γ-HCH and PCB-153 and comparison of modelling results against observations from the OSPAR monitoring sites (CAMP) [*Gusev et al.*, 2008].

MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

The activities of the EMEP Centres MSC-E and CCC in 2014 will be directed to the assessment of POP pollution in the EMEP region and to support the EMEP countries with information required for the implementation of the Protocol on POPs. Long-term changes of POP pollution levels and transboundary transport in the EMEP domain will be evaluated for the period 1990-2012. Particular attention will be paid to the assessment of POP pollution in the EECCA countries.

Main challenges and directions of future research aimed at the improvement of the quality of pollution assessment are formulated below.

- Completeness and consistency of POP emission data provided by the EMEP countries are gradually improving, though official information on emissions time-series, spatial distribution, and coverage of emission source categories still require refinement. Particular attention should be paid to the inventories of POP emissions in the EECCA countries. For the analysis of existing uncertainties in emission data further development and application of inverse modelling approach is necessary.
- Secondary emissions (re-volatilization and re-suspension from environmental media) can essentially contribute to the pollution for some of the considered POPs (PCDD/Fs, HCB). To improve the assessment of their pollution levels, inventories of historical emissions and direct releases to the media other than the atmosphere can be of particular importance. Evaluation of POP intercontinental transport requires updating of global emission inventories in cooperation with international organizations and national experts.
- The work on the refinement of emissions within the EMEP region, performed under the EMEP TFEIP in cooperation with modelling community, is of particular importance for the improvement of the quality of model assessment of pollution.
- Despite the number of sites in the EMEP monitoring network for POPs has increased, there is certain lack of measurements in the southern and eastern parts of the EMEP domain, particularly in the EECCA region. In this respect passive sampling campaigns and national measurements could essentially complement monitoring of POPs in the EMEP region. Measurements of POPs outside of the EMEP region are needed for the evaluation of POP intercontinental transport.
- Regular measurements of PCDD/F concentrations are not currently implemented at the EMEP monitoring sites. Thus, to improve the assessment of PCDD/F pollution, collection of measurements from national campaigns in the EMEP database is highly appreciated.
- Information on POP concentrations in soil and water bodies is significant for the evaluation of POP accumulation in media and secondary emission sources. To support model assessment of POP accumulation in environmental compartments collection of such measurements under EMEP seems reasonable.
- Improvement of the EMEP monitoring network for POPs, collection of additional POP measurements, and their accessibility through the EMEP database need further discussion at the TFMM and TF HTAP meetings.
- Further development of the Global EMEP Multi-media Modelling System (GLEMOS) will include refinement of the model parameterizations of physical and chemical processes for POPs and transition to operational calculations of POP pollution on the latitude-longitude projection. Besides, it is planned to provide open access to the GLEMOS program source code to support the development of national modelling approaches for pollution assessment.

- To improve evaluation of POP fate in the atmosphere further refinement of model parameterizations of gas-particle partitioning to and degradation on different aerosol components, particularly, elemental and organic carbon is needed.
- For the refinement of influence of secondary emission sources (re-volatilization and re-suspension processes) it is necessary to update the information on various characteristics of the surface media (e.g. organic matter content) and parameterizations of the degradation and phase partitioning processes which influence POP accumulation in media.
- Development of the EMEP/MSC-E website needs to be continued to provide more flexible access to various types of information on pollution, including country-specific information on emission, monitoring and model assessment for the pollutants considered by the Convention. Besides, a version of the website in Russian is to be developed to facilitate access to the information for Russian speaking experts from the EECCA countries.

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