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Assessment of spatial and temporal trends of POP pollution on regional and global scale

METEOROLOGICAL SYNTHESIZING CENTRE - EAST

A.Gusev, O.Rozovskaya, V.Shatalov

CHEMICAL COORDINATING CENTRE

W.Aas, P. Nizzetto



MCS-E

Meteorological Synthesizing Centre - East

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



CCC

Norwegian Institute for Air Research (NILU)

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

EXECUTIVE SUMMARY

Persistent organic pollutants (POPs) are semi-volatile persistent toxic substances that are characterized by significant potential to long-range transport and accumulation in the environmental media. Despite the measures to reduce their usage, they are still found in the environment and 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, 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.

This report summarizes recent achievements of the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) in the field of the assessment of POP pollution. It provides information on pollution levels of PAHs, PCDD/Fs, HCB, and PCBs for 2013 based on both modelling results and measurements including also information on anthropogenic emissions. Estimates of transboundary transport of pollution have been presented including information on contributions of secondary emission sources and intercontinental transport. Particular attention is paid to the analysis of long-term trends of POP pollution in the EMEP countries to support preparation of the CLRTAP Assessment Report. The present report describes further development of the Global EMEP Multi-media Modelling System (GLEMOS). Along with this, much attention is given to the exchange of information between MSC-E and subsidiary bodies to the Convention, as well as dissemination of output information to national and international organizations.

Evaluation of long-term trends of POP pollution in the EMEP countries was performed for the period 1990-2012 based on POP emission data reported by the EMEP countries and available expert estimates. According to this information anthropogenic emissions in the EMEP countries have decreased from 1990 to 2012 by more than 80% for HCB and PCB-153, by 60% for PCDD/Fs, and by 40% for PAHs. Changes of POP emissions in the EMEP domain during this period were inhomogeneous. The most significant decline of POP emissions took place in the EU28 countries, while the lowest decrease of POP emissions was seen in the Eastern European, Caucasus, and Central Asia (EECCA) countries.

Long-term monitoring of POP pollution was carried out at eleven EMEP monitoring sites including Zeppelin and Birkenes (Norway), Pallas (Finland), Aspvreten and Råö (Sweden), Storföldi (Iceland), and Kosetice (Czech Republic) measuring air concentrations and Westerland and Zingst (Germany), Pallas (Finland), Storföldi (Iceland), Lista/Birkenes (Norway), Rörvik/Råö and Aspvreten (Sweden) measuring concentrations in precipitation of PCBs, HCB, and PAHs. Analysis of temporal changes of measured POP concentrations indicated significant variations between the sites and POPs and absence of overall consistent trend.

Trend analysis based on model simulation results and available measurement data showed that maximum overall reduction took place for HCB (over 90%), followed by PCBs, PCDD/Fs, and B[a]P for which minimum reduction (about 30%) was estimated. For all considered POPs maximum reduction of contamination occurred in 1990-s, whereas during the recent years the rates of pollution reduction declined or almost leveled off. Evaluation of trends in B[a]P concentrations indicated that for most of the EU28 countries the decline of pollution levels in the end of the period (2008-2012) was changed to growth which was also confirmed by measurements of the EMEP monitoring sites.

Seasonal variations were found to be a valuable factor affecting contamination levels. For example, B[a]P air concentrations in winter could exceed summer concentrations by an order of magnitude. It should be noticed that both modelling results and measurements indicated low rate of reduction (1% –

2%) of maximum B[a]P air concentrations occurred in winter time for the whole period and tended to grow in the end of the period (2012). The analysis also indicated importance of consideration of global emissions and historical releases of POPs in course of the analysis of trends in POP contamination in the EMEP region.

Model assessment of POP pollution of the EMEP region for 2013 is based on multi-media multi-scale modelling approach, which takes into account peculiarities of POP transport and fate in the environment (e.g. phase partitioning, degradation in media, multi-hop transport, re-emission). Anthropogenic emission sources of the EMEP countries continue providing significant contribution to the transboundary pollution of the EMEP region. Particularly, substantial contribution of transboundary transport to deposition from anthropogenic emission sources over the EMEP countries is estimated that accounts for about 50% for PAHs and PCDD/Fs, 60% for PCB-153, and 70% for HCB.

For long-lived POPs it is important to apply modelling on global and regional scales to evaluate contributions of non-EMEP emission sources and secondary emissions. According to modelling results on PCDD/Fs, PCB-153, and HCB for 2013, the contribution of secondary emission sources can be about 50% and more. Non-EMEP emission sources can contribute to the pollution of the EMEP countries about 10-25%. At the same time, estimated influence of emission sources outside the EMEP region can be underestimated due to uncertainties of applied global emission inventories.

MSC-E has continued the development of GLEMOS in order to improve quality of the assessment of pollution levels and trends for the EMEP countries. The main directions of research and developments include the preparation of the GLEMOS modelling system for the transition of the EMEP operational modelling to the new EMEP grid, testing and refinement of GLEMOS capabilities to perform multi-scale model simulations for POPs, incorporation of gridded data on aerosols and atmospheric reactants provided by various chemistry transport models, as well as further refinement of the GLEMOS multi-media modelling approach for POPs.

Potential influence of climate changes on future POP pollution and exposure was recently received significant attention. In addition to previously made studies, examining potential climate-induced changes on POP pollution, the Centre prepared a brief literature review of recent modelling experiments aimed at evaluation of the influence of projected climate changes on future POP releases and concentrations. In general, obtained multimedia modelling results suggest that the effect of considered climate changes on POP transport, fate, and bioaccumulation is within a factor of two comparing to the baseline results. One of important issues mentioned in these studies is that uncertainties in model predictions are likely to be substantially larger than the level of obtained climate-induced changes of future pollution levels. Thus, further work is required to refine the knowledge on POP fate in media, hosting the most part of POPs released into the environment, and to refine estimates of future emissions of POPs.

Cooperation with the subsidiary bodies to the Convention has been continued. In particular, results of research and development activities of the Centre are presented and discussed at the EMEP Task Force on Measurements and Modelling (TFMM) meetings (<http://www.msceast.org/index.php/presentations>). Besides, MSC-E in cooperation with the TFMM elaborated a special methodology for the analysis of long-term trends applicable for wide range of pollutants including POPs (<https://wiki.met.no/emep/emep-experts/start>). In framework of collaboration with the Working Group on Effects (WGE) the Centre prepared the information on long-term trends of POP pollution in the EMEP region as contribution to the WGE Assessment Report on Trends.

Information exchange with other international organizations and programmes broaden dissemination of scientific and policy oriented information generated within EMEP and strengthen the status of the program on international level. In the framework of co-operation with the UNEP Stockholm Convention

(SC) MSC-E performed studies of global scale transport of POPs with application of the data on emissions and measurements compiled within the Stockholm Convention. In particular, pilot modelling of PCDD/F transport and fate was carried out using experimental emission scenario based on the global emission inventory of the UNEP SC. Analysis of spatial and temporal trends in global scale pollution by PCBs, PCDD/Fs, and HCB was performed using modelling results and measurement data collected in the SC Global Monitoring Programme (GMP) Data Warehouse. In addition, information on POP pollution load to the Baltic Sea and its trends as well as source apportionment of PCDD/F deposition in 2012 were submitted to the Baltic Marine Environment Protection Commission (HELCOM).

Future directions of MSC-E research will be focused on assessment of POP pollution levels in the EMEP region and support of the EMEP countries with information required for the implementation of the Protocol on POPs. The main challenges of POP pollution assessment, which need particular attention and further research in 2016-2017 have been formulated in accordance with the Key Messages of the CLRTAP Assessment Report. Evaluation of long-term trends of POP pollution during the two past decades will be completed to support the CLRTAP Assessment Report. Development of the GLEMOS modelling system for POPs will be focused on further elaboration of the multi-media approach to improve knowledge on occurrence and fate of POPs in relevant environmental compartments.

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INTRODUCTION

Persistent organic pollutants (POPs) are semi-volatile persistent toxic substances that are characterized by significant potential to long-range transport and accumulation in the environmental media. Despite the measures to reduce their usage, they are still found in the environment and pose risk to human health and ecosystems. Reduction of POP releases and of pollution levels is subject 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.

Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) annually provides the Executive Body for the Convention with information on effectiveness of emission control measures and on changes of pollution levels of the substances targeted by the Convention Protocols including POPs. Assessment of POP pollution is performed 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), the Task Force on Emission Inventories and Projections (TFEIP), and the Working Group on Effects (WGE).

This report summarizes recent progress in the activities of the EMEP Centres in 2015 in the field of the evaluation of POP pollution in the EMEP region. It provides a short overview of POP emission data applied for model assessment, status of the EMEP monitoring network for POPs, and information on spatial and temporal variations of POP pollution levels in the EMEP countries based on modelling results and measurements. Particular attention is paid to the analysis of long-term trends of POP contamination in the EMEP countries to support preparation of the CLRTAP Assessment Report. The present report describes further development of the Global EMEP Multi-media Modelling System (GLEMOS). Along with this much attention is given to the exchange of information between MSC-E and subsidiary bodies to the Convention, as well as dissemination of output information to national and international organizations.

Anthropogenic emissions of priority POPs, including PAHs, PCDD/Fs, HCB, and PCBs, have been considerably reduced in the majority of the EMEP countries since entering the POP Protocol into force. Reduction of emissions has led to substantial decline of POP pollution levels. Analysis of long-term changes of POP pollution levels supports the evaluation of effectiveness of the environmental policies implemented in the EMEP countries and, in particular, of the POP Protocol. Analysis of long-term trends in POP pollution of the EMEP region has been performed for the period 1990-2012 on the basis of the methodology developed by MSC-E to take into account the non-linear character of POP pollution changes in the EMEP countries. Main factors affecting the long-term changes of pollution levels in the EMEP countries have been analysed.

POP contamination of the environmental compartments is formed by various emission sources, including anthropogenic emissions to the atmosphere, direct releases of POPs to other media (e.g. soil, water bodies), and secondary emissions (re-mobilization from surface media to the atmosphere). Particular attention in the assessment of POP pollution levels is provided to the evaluation of contributions of secondary emission sources and non-EMEP emissions due to their noticeable influence on levels of POP pollution. Estimates of influence of these sources on the pollution of the EMEP countries are obtained using the global scale modelling on the basis of scenarios of contemporary and historic emissions.

Evaluation of interaction between climate variability and POP pollution exhibits a challenging task due to complexity of POP cycling in the environment. Recently made studies regarding this interaction

conclude that climate change has the potential to affect all pathways of POPs in the environment and may increase vulnerability for health impacts. To continue the work on the evaluation of potential effects of climate-induced changes on POP pollution, the Centre prepared a brief literature review of modelling experiments performed recently to quantify response of future POP releases and concentrations. Results of the studies suggest that the effect of considered climate changes on POP transport, fate, and bioaccumulation is within a factor of two comparing to the baseline results. However the uncertainties in model predictions are likely to be substantially larger than the level of obtained climate-induced changes of future pollution levels indicating the need of further refinement of information on POP transport and fate in the environmental compartments as well as estimates of future emissions of POPs.

MSC-E has continued the development of the GLEMOS model in order to improve quality and reliability of output information submitted to the EMEP countries. The main directions of research and developments include the preparation of the GLEMOS modelling system for the transition of the EMEP operational modelling to the new EMEP grid, testing and refinement of GLEMOS capabilities to perform multi-scale model simulations for POPs, incorporation of gridded data on aerosols and atmospheric reactants provided by various chemistry transport models, as well as further refinement of the GLEMOS multi-media modelling approach for POPs.

Specific attention has been paid to cooperation with subsidiary bodies to the Convention. Results of research and development activities are presented and discussed at the EMEP Task Force on Measurements and Modelling (TFMM) (<http://www.msceast.org/index.php/presentations>). Along with this, contribution to the WGE Assessment Report on trends in POP pollution was prepared. The information exchange with other international organizations and programmes (e.g. Helsinki Commission, the UNEP Stockholm Convention (SC) on POPs, the Arctic Monitoring and Assessment Programme (AMAP), etc.) broaden dissemination of the scientific and policy oriented information generated by the EMEP and strengthen the status of the program on international level. In addition to the annual Status reports a variety of information on POP pollution levels in the EMEP region, individual countries, marginal seas etc. is distributed via the Internet at the MSC-E website (www.msceast.org).

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

1. EMISSION DATA FOR MODEL ASSESSMENT

Model assessment of POP pollution levels in the EMEP domain was carried out using gridded emission data on PAHs (benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-cd]pyrene), PCDD/Fs (17 toxic congeners), and HCB generated by CEIP and MSC-E. For the assessment of PCB pollution in the EMEP countries MSC-E constructed gridded emission data using officially reported emissions and available expert estimates of Breivik et al. [2007].

Emission data for evaluation of long-term trends of POP pollution (1990-2012)

Evaluation of long-term trends of POP pollution in the EMEP countries was performed for the period 1990-2012. *According to officially reported data and expert estimates anthropogenic emissions in the EMEP countries have decreased from 1990 to 2012 by 84% for HCB and PCB-153, by 61% for PCDD/Fs, and by 39% for PAHs* (Fig.1.1).

Changes of POP emissions in the EMEP region during this period were inhomogeneous. Substantially different rates of changes can be seen in the EU28, EECCA, and the other EMEP countries. *The most significant decline of POP emissions took place in the EU28 countries. Particularly, emissions of HCB, PCB-153, PCDD/Fs, and PAHs dropped by 96%, 86%, 83%, and 50% respectively.* Marked change of anthropogenic HCB emissions from 1998 to 1999 was conditioned by sharp decrease of releases reported by the UK.

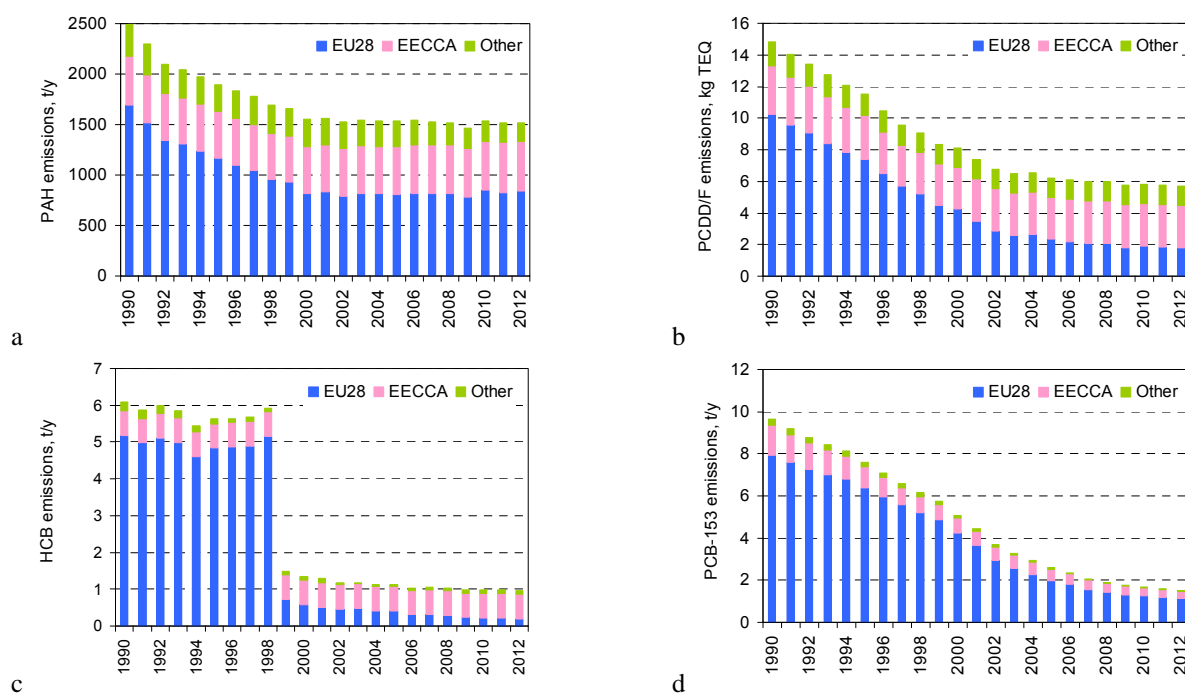


Fig. 1.1. Temporal changes of the sum of 4 PAH (a), PCDD/F (b), HCB (c), and PCB-153 (d) emissions in the EU28, EECCA, and other EMEP countries from 1990 to 2012

The lowest decrease of emissions can be seen in the EECCA countries for PCDD/Fs, PAHs, and HCB. In particular, it varies from about 10% for PCDD/Fs to almost no decrease for PAHs and HCB. At the same time, for PCB-153 the decrease of emissions in the EECCA countries is almost similar to the EU28 countries according to available expert estimates. The other EMEP countries are characterized by moderate changes. However, it should be noted that time-series of POP emissions in

most of the EECCA countries are subject of significant gaps. Thus, more realistic evaluation of temporal changes of pollution in the EMEP region requires further refinement of emission data.

Emission data for assessment of POP pollution in 1990 and 2013

Model simulations of POP pollution in the EMEP domain for 1990 and 2013 were performed using the emission data reported in the year 2014 complemented by expert estimates of emissions. Selection of previous submission of emissions was conditioned by the change of the deadline for submitting gridded emission data to the 1st of May in accordance with the revised Reporting Guidelines [ECE/EB.AIR/125]. The new reporting deadline is later in comparison with the previous years, therefore model runs for 2013 have been performed using emission data for 2012 under the assumption of equal emissions of POPs for the years 2012 and 2013. Modelling results for 2013 and 1990 will be updated using the most recent reported emission data and will be available in the internet at the MSC-E web site in September 2015.

Spatial distribution of emissions from anthropogenic sources in the EMEP domain, used in model simulations for 1990 and 2013, is shown in Figs 1.2 – 1.5. Considering changes of spatial variations of emissions during this period it can be noted that the largest decrease take place in the western and central parts of Europe. At the same time, changes of POP emissions in the EECCA countries do not show significant decline.

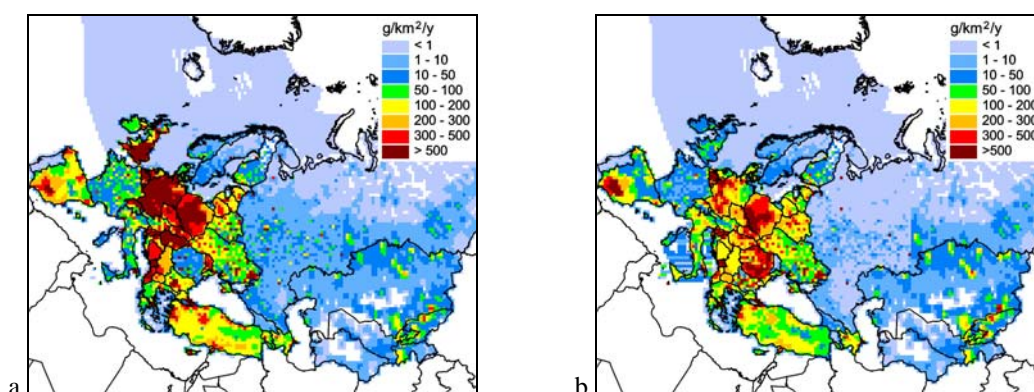


Fig. 1.2. Spatial distribution of emissions of the sum of 4 PAHs in the EMEP countries in 1990 (a) and in 2013 (b) with resolution 50x50 km².

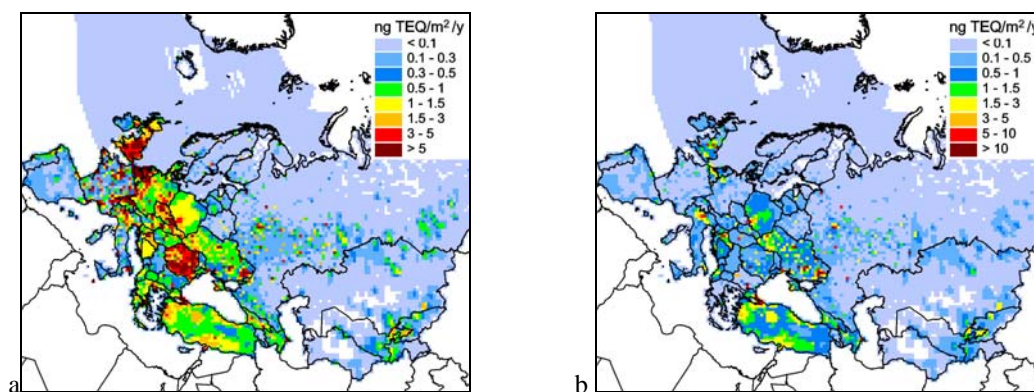


Fig. 1.3. Spatial distribution of PCDD/F emissions in the EMEP countries in 1990 (a) and in 2013 (b) with resolution 50x50 km².

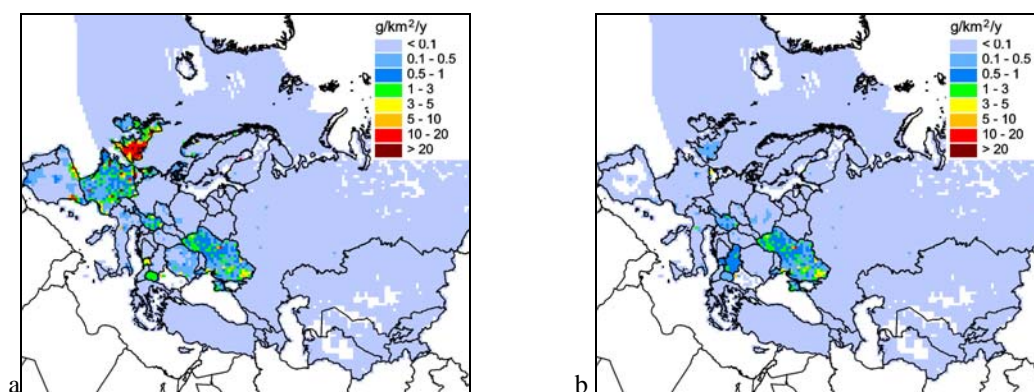


Fig. 1.4. Spatial distribution of HCB emissions in the EMEP countries in 1990 (a) and in 2013 (b) with resolution 50x50 km².

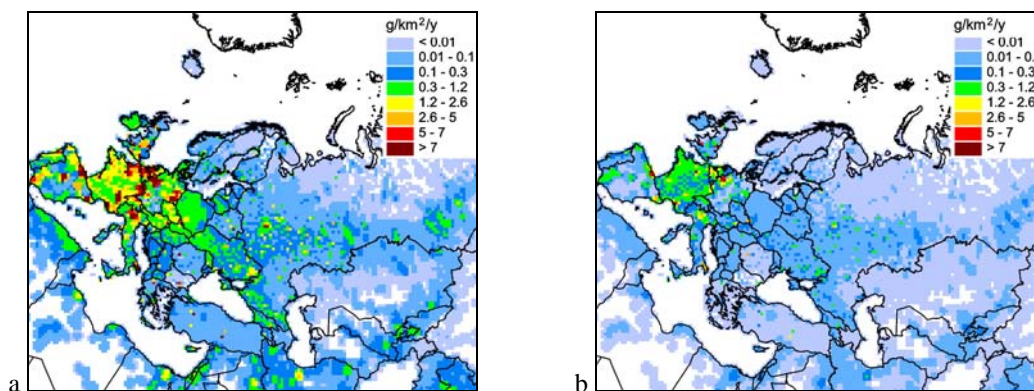


Fig. 1.5. Spatial distribution of PCB-153 emissions in the EMEP domain in 1990 (a) and in 2013 (b) with resolution 50x50 km².

POP emissions on a global scale

Long-term variations of POP pollution in the EMEP region depend on temporal changes of releases from anthropogenic and secondary emission sources. Secondary emissions are represented by the re-mobilization of POPs from surface media (e.g. soil, water bodies), where they have been previously accumulated due to legacy (historical) emissions. Significant contribution to the pollution of the EMEP countries can be made by non-EMEP anthropogenic and secondary emission sources, taking place in other regions outside the EMEP domain. In order to evaluate relative contributions of these sources scenarios of global HCB, PCDD/F and PCB emissions (including historical emissions) were updated and used for the evaluation of POP global transport and fate.

PCDD/F emissions

Modelling of global scale transport and fate of PCDD/Fs has been carried out using experimental emission scenario constructed on the basis of the data of the UNEP SC inventory. This inventory is being compiled under SC using the methodology of the UNEP Standardized Dioxins Toolkit [Fiedler, 2007; Fiedler et al., 2012; UNEP, 2013]. The inventory comprises data on national emissions of dioxins and furans reported by 68 countries characterizing the level of emissions during the recent decade. For other countries, which did not provide information on their emissions, releases of

PCDD/Fs were estimated using the analysis of compiled national inventories carried out by *Cao et al.* [2013]. More detailed description of this experimental scenario can be found in the MSC-E Technical Report [*Shatalov et al.*, 2014].

Spatial distribution of PCDD/F emissions to air and soil with resolution $1^\circ \times 1^\circ$, which was used in model simulations, is shown in Fig.1.6.

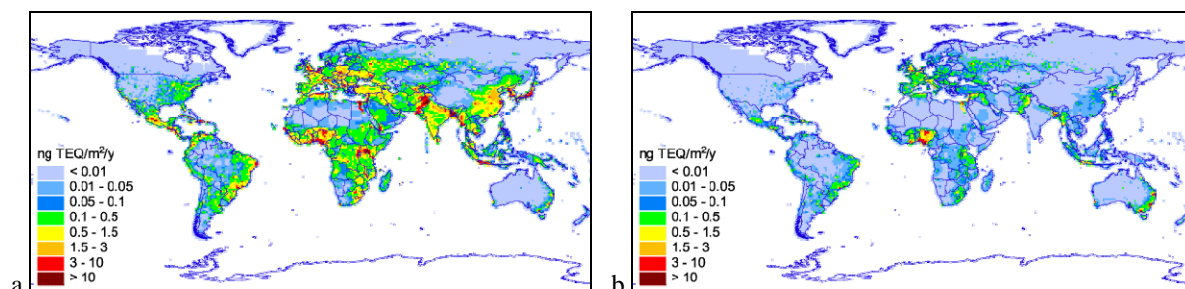


Fig. 1.6. Spatial distribution of annual PCDD/F emissions (ng TEQ/m²/y) to the atmosphere (a) and to soil (b) constructed on the basis of UNEP global PCDD/F emission inventory

HCB emissions

Similar to PCDD/Fs model evaluation of HCB global transport and fate was performed on the basis of experimental emission dataset, which described contemporary levels of HCB releases as well as historical emissions for the period from 1945 to 2013. Three scenarios of historical HCB emissions (maximum, average and minimum), elaborated previously [*Shatalov et al.*, 2010], were updated and used for modelling of long-term accumulation of the pollutant in the environmental media. Model simulations carried out for this report were made applying the maximum emission scenario, which allowed to get better agreement with available measurements of HCB air concentrations.

Spatial distributions of global HCB emission fluxes for 1990 and 2013 are shown in Fig. 1.7. According to prepared scenario, global HCB releases declined significantly during the considered period. The largest HCB emission fluxes took place in Southern and Eastern Asia. Releases of HCB to the atmosphere in the EMEP region were relatively lower.

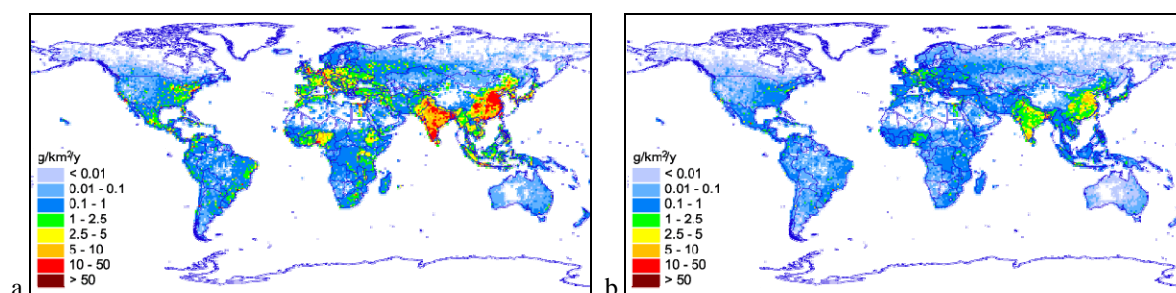


Fig. 1.7. Spatial distribution of HCB emissions in 1990 (a) and in 2013 (b) over global domain with resolution $1^\circ \times 1^\circ$

PCB emissions

Assessment of global scale transport and fate of PCBs was made on the basis of the inventory of global PCB emissions [*Breivik et al.*, 2007] and emissions officially reported by the EMEP countries. The inventory of *Breivik et al.* [2007] provided consistent set of historical and future emissions of 22

individual PCB congeners from 1930 up to 2100. It included three scenarios of emissions, namely, minimum, average, and maximum, which represented the range of emission variations. For the evaluation of pollution levels maximum scenario of emissions was chosen since it permitted to obtain modelling results with more reasonable agreement with measurements comparing to other scenarios. Modelling of pollution levels for 1990 and 2013 was carried out for indicator congener PCB-153.

The spatial distribution of PCB-153 emissions used for global modelling with resolution $1^\circ \times 1^\circ$ is illustrated in Fig. 1.8. It can be seen that considerable levels of PCB-153 emission fluxes are the characteristic of the EMEP region. Other regions are characterised by comparatively lower annual emissions.

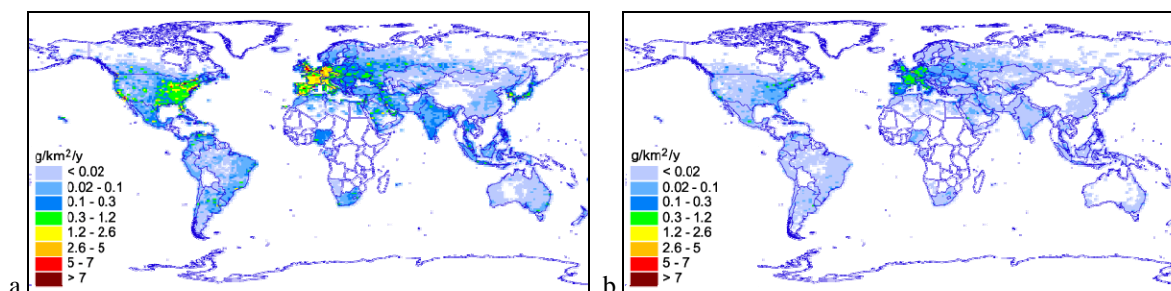


Fig. 1.8. Spatial distribution of PCB-153 emissions in 1990 (a) and in 2013 (b) over global domain with resolution $1^\circ \times 1^\circ$

2. EMEP MONITORING NETWORK OF POPS – LONG-TERM MONITORING AND TRENDS

Measurements of persistent organic pollutants (POPs) were included in the EMEP's monitoring program in 1999 but data on POPs are also available from the beginning of 1990s for some stations in the EMEP database (<http://ebas.nilu.no>). The length of the monitoring data sets varies between sites and type of POP compound (Table 2.1). In general, long-term data of POPs in air are available from Zeppelin and Birkenes in Norway, Pallas in Finland, Aspvreten and Råö in Sweden, Storfaldi in Iceland, and Kosetice in Czech Republic while long-term data of POPs in precipitation are available from Westerland and Zingst in Germany, Pallas in Finland, Storfaldi in Iceland, Lista/Birkenes in Norway, Rörvik/Råö and Aspvreten in Sweden.

Table 2.1. Long-term data on POPs in air and precipitation at EMEP sites showing the starting year.

Site (code)	Air						Precipitation				
	PCBs	HCB	HCHs	DDTs	CHLs	PAHs	PCBs	HCB	DDTs	HCHs	PAHs
Zeppelin (NO42)	1998	1993	1993	1994	1993	1994					
Birkenes (NO02/99)	2004	1991	1991	2010	2010	2009	2004	1991		1991	
Pallas (FI36)	1996		1996	1996		1996	1996			1996	1996
Storfaldi (IS91) *	1995	1995	1995	1995	1995		1995	1995	1995	1995	
Aspvreten (SE12)	1995		1995	1995		1995	1996			1995	1995
Råö/Rörvik (SE02/14)	1994		1994	1996		1994	1995			1995	1995
Kosetice (CZ03)	2002	2006	1999	1999		1999	2003		2003	2006	2003
Westerland (DE01)							1996	1996	1996	1996	1996
Zingst (DE09)							1999	1999	1999	1999	1999

* Stopped measuring POPs after 2012

Detailed information about the sites, measurement methods and results for 2013 can be found in EMEP/CCC's data reports on heavy metals and POPs [Aas and Bohlin-Nizzetto, 2015]. All data are also available at the EMEP database (<http://ebas.nilu.no>).

Long-term trends of POPs in air and precipitation measurements

An overview with trend analysis of EMEP POP data has previously been done by Tørseth *et al.* [2012], and a new trend assessment is being prepared by the EMEP TFMM, which is to be published during spring 2016. In addition, similar trend assessments are also being carried out under the Stockholm Convention on POPs whereby EMEP POP data form an important empirical basis for Europe and Arctic. Herein we illustrate selected temporal trends of long-term monitoring data in air and precipitation for two hexachlorocyclohexane (HCH) isomers; α - and γ -HCH, two polychlorinated biphenyl (PCB) congeners; PCB-52 and -153, one DDT homologue; p,p'-DDT, hexachlorobenzene (HCB), and one polycyclic aromatic hydrocarbon (PAH) compound; benzo(a)pyrene (B[a]P). For B[a]P, spatial pattern of annual mean concentrations in air in 2013 are also presented.

It should be noticed that comparing data from different sampling sites and laboratories is a complicating factor when interpreting POP measurements due to differences in sampling and analytical methodologies that might affect the comparability [Schlabach, 2011 and Melymuk, 2014]. In

addition, data for precipitation is reported as concentration or as deposition resulting in further complications when comparing POP data in precipitation.

No overall consistent temporal trends are observed for the POP compounds at all the stations. Instead the trend varies between individual compounds and sites.

Overall, the largest declining trends are observed for HCHs. Both HCH isomers show significant reductions in annual mean concentrations in air over the time period included (Fig. 2.1). Over the last decade, concentrations in air of α -HCH and γ -HCH tend to be fairly homogenous at the sites but α -HCH is still significantly higher than γ -HCH in northern Europe (FI36 and NO42). α -HCH is the predominant isomer in technical HCH which exceeds γ -HCH, both in terms of long-range atmospheric transport potential as well as propensity for re-volatilization.

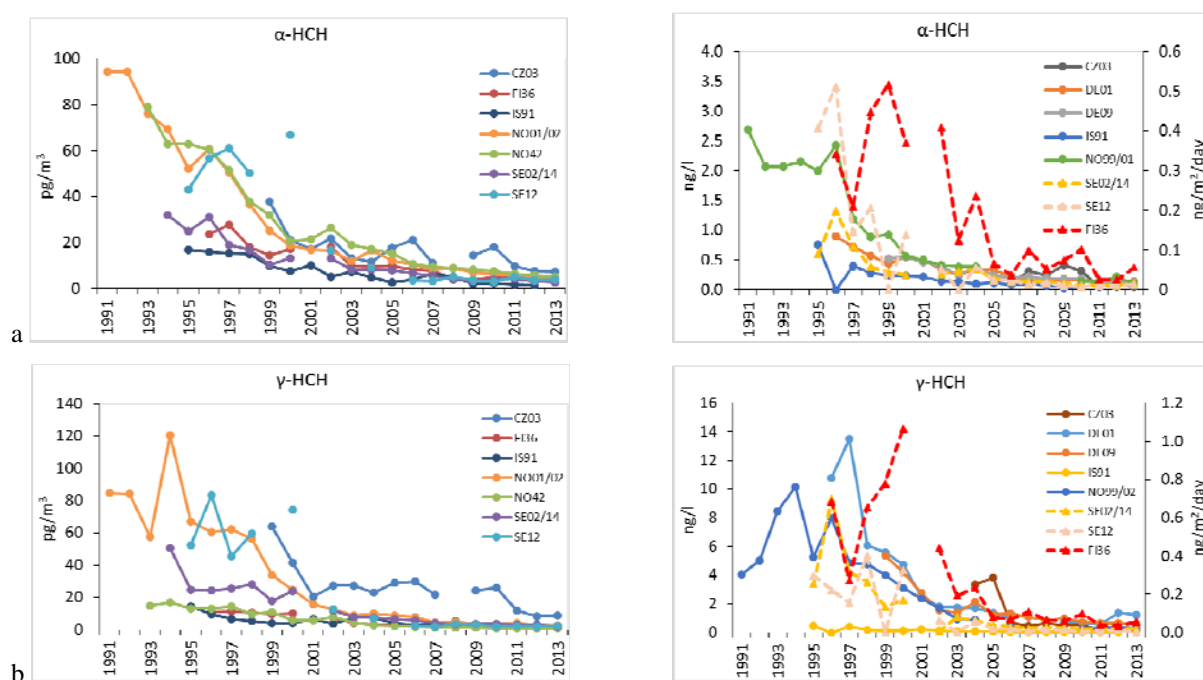
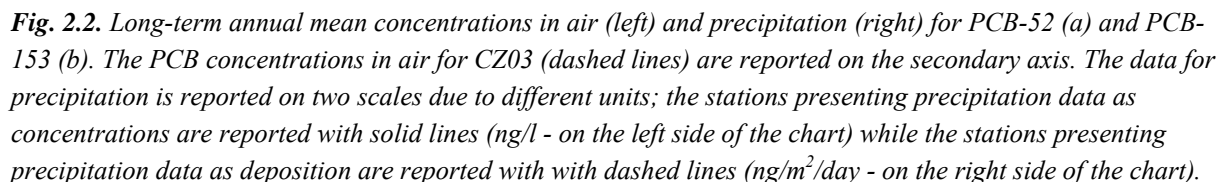


Fig. 2.1. Long-term annual mean concentrations in air (left) and precipitation (right) for α -HCH (a) and γ -HCH (b). The data for precipitation is reported on two scales due to different units; the stations presenting precipitation data as concentrations are reported with solid lines (ng/l - on the left side of the chart) while the stations presenting precipitation data as deposition are reported with dashed lines (ng/m²/day - on the right side of the chart).

The concentrations of PCBs generally show slow declining trends at all sites with a few exceptions; a large reduction at Zeppelin (NO42) during the beginning of 2000s, no clear temporal trend at Råö, Sweden (SE02/14), while an increase in PCB-concentrations in air (especially PCB-52) are observed at Storhofdi, Iceland (IS91) during 2000s (Fig.2.2). The concentrations in air over the time-period included were highest at Kosetice (CZ03) but after a sharp decline in the beginning of 2000s the levels are now more homogenous at all sites.



a

p,p'-DDT

pg/m³

Legend: E59b, E591, NO03/02, NI047, St07/St14, St17, CZ03

b

p,p'-DDT

ng/l

Legend: C/133, nfo1, I33P3, E591

Long-term data for HCB is only available for four sites. The data for Zeppelin, Norway (NO42) shows a decreasing trend during the 1990s while an increasing trend during the last decade (Fig. 2.4). For the other sites no clear trend is seen.

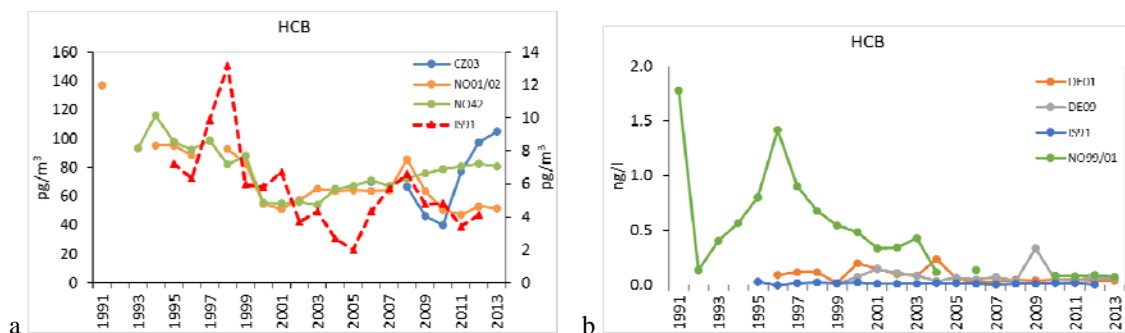


Fig. 2.4. Long-term annual mean concentrations in air (a) and precipitation (b) for HCB. The HCB concentrations in air for CZ03 (dashed lines) are reported on the secondary axis.

In addition to the Aarhus protocol on POPs, the PAHs (represented by B[a]P) are also regulated by the European Union. The measured annual mean concentrations in air of B[a]P at all stations are generally one to two orders of magnitude below the European Air Quality Standard (1 ng/m^3), defined by the 4th daughter directive [EU, 2004]. In general, highest levels of B[a]P are observed at Kosetice (CZ03) in Central Europe while lowest levels are observed at Zeppelin (NO42) in the Arctic (Fig. 2.5). The annual mean concentration of B[a]P in air at Kosetice (CZ03) in 2013 were the highest ever measured and detailed data shows levels above the European Air Quality Standard during January-March in 2013. The air concentrations of B[a]P show no clear time-trend at any station. In contrast, small declining trends are seen for B[a]P in precipitation at few sites (i.e. FI36, SE02/14, SE12). The spatial pattern of annual mean concentrations of B[a]P in air in 2013 are presented in Fig. 2.6. This shows highest concentrations to be found in central Europe while the lowest in northern Europe.

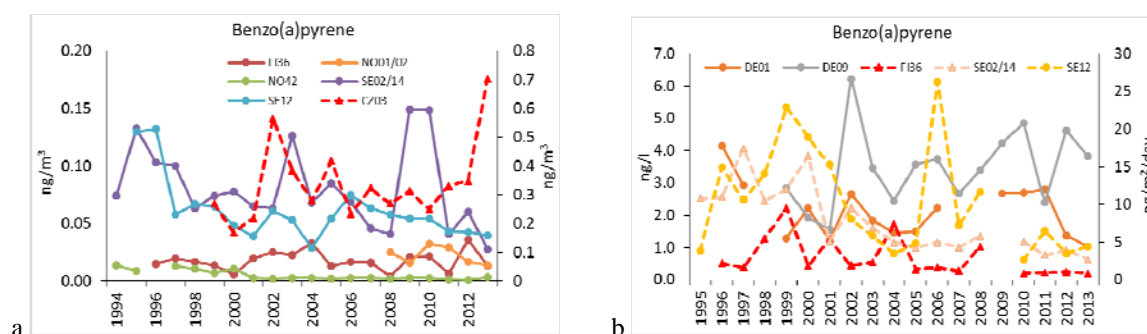


Fig. 2.5. Long-term annual mean concentrations in air (a) and precipitation (b) for B[a]P. The B[a]P concentrations in air for CZ03 (dashed lines) are reported on the secondary axis. The data for precipitation is reported on two scales due to different units; the stations presenting precipitation data as concentrations are reported with solid lines (ng/l - on the left side of the chart) while the stations presenting precipitation data as deposition are reported with dashed lines ($\text{ng/m}^2/\text{day}$ - on the right side of the chart).

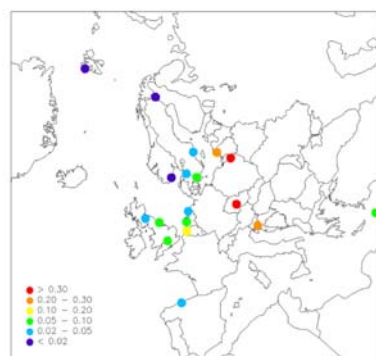


Fig. 2.6. Annual mean concentrations of B[a]P in air in 2013. Note that Cyprus is outside the map domain and presented in a dislocated point.

3. LONG-TERM TRENDS OF POP POLLUTION IN THE EMEP COUNTRIES FOR 1990-2012

This section presents the outcome of the analysis of long-term trends in the contamination of the EMEP region by selected POPs: PAHs, PCDD/Fs, PCBs, and HCB. According to the recommendations of the TFMM Workshop, trend analysis has been performed for the period from 1990 (reference year) to 2012. Analysis of long-term variations of POP pollution and main tendencies is performed based on the modelling results and measurements of the EMEP monitoring network. Below the following information on temporal changes of POP contamination is presented:

- long-term trends of POP contamination in the EMEP region and individual EMEP countries obtained on the basis of model calculations;
- long-term trends of POP concentrations in particular grid cells on the basis of modelling results and measurements;
- factors, contributing to the long-term variations of POP pollution levels in the EMEP region: temporal changes of EMEP anthropogenic emissions, non-EMEP emissions and releases from secondary sources;
- relevant information on trends in POP pollution for the evaluation of exposure and effects.

3.1. Approach to the trend analysis

Analysis of trends in pollution levels is based on the methodology elaborated by MSC-E. Short description of the methodology can be found in the Annex A and on the TFMM wiki page (<https://wiki.met.no/emep/emep-experts/start>). According to the methodology, initial time series (annual and/or monthly means of POP concentrations) are decomposed to the regular component (*trend*) and irregular component (*residue*). In turn, regular component is split into *main component* describing general decrease/increase of air concentrations during the considered period and *seasonal component* describing intra-annual variations of air concentrations (Fig. 3.1).

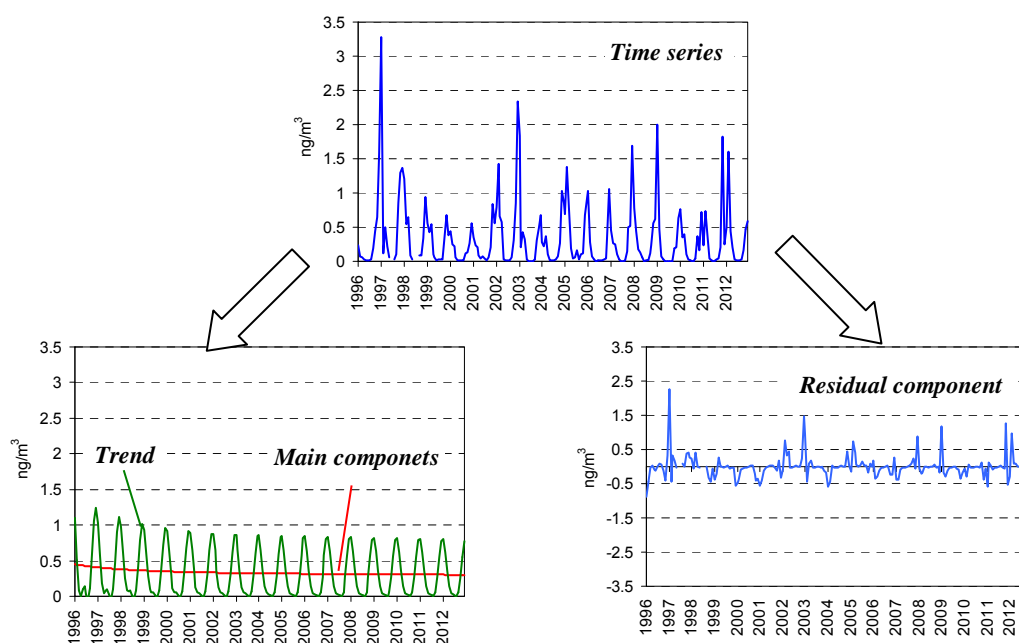


Fig. 3.1. Decomposition of time series to trend and residual component exemplified by measurements of B[a]P air concentrations at EMEP site CZ3

The residues may be conditioned by short-term and/or random perturbations of the investigated time series. The trend represents the considered time series free from these perturbations (in particular, year-to-year meteorological variability is thus excluded from the time series). For POPs (as well as for HMs) main component normally occurs to be *non-linear* (see lower left plot in Fig. 3.1). Thus, for the description of trends for these pollutants multi-exponential approach with harmonic components is applied.

The decomposition allows evaluating the following trend parameters: *total reduction for the entire period*, *average annual reduction*, *reduction in the end of the considered period*, and *seasonality*. Reduction in the end of the period is of interest since it shows contemporary reduction rate that is normally less than average annual reduction. Seasonality is defined as amplitude of seasonal component normalized by main component averaged over the whole period. This parameter expresses relative changes of main component due to seasonal variations.

3.2. POP pollution trends in the EMEP region and individual EMEP countries

Analysis of trends in POP pollution of the EMEP region and individual EMEP countries for two recent decades was performed using the results of model simulations. Reduction of pollution levels in the period from 1990 to 2012 occurred at different rates. Comparison of annual reduction rates, evaluated for this period, for the selected POPs is shown in Fig. 3.2. The diagram presents both average reduction rates of POP air concentrations for the whole period and rates of reduction for the last year of the period (2012). Estimates of rates for the year 2012 illustrate current state of contamination changes (decrease or increase). The most significant average reduction rates are seen for HCB and PCB-153 (about 10% per year), while estimated rates for PCDD/Fs and B[a]P are lower (about 2-3% per year). It is seen that levels of B[a]P air concentrations in 2012 tended to increase.

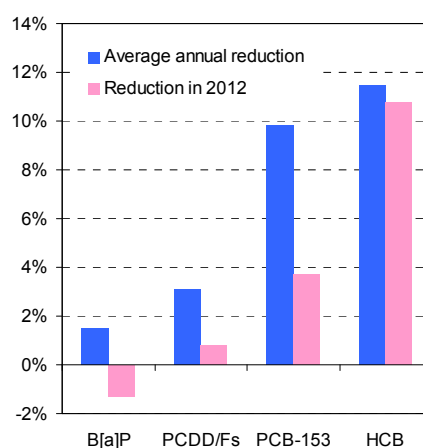


Fig. 3.2. Reduction rates of air concentrations of the considered pollutants for the EMEP region. Negative values denote growth of pollution

Trends of average POP air concentrations in the EMEP region for the period from 1990 to 2012 are shown in Fig. 3.3. To describe general tendencies in changes of air concentrations, trends of annual averages of pollution (not taking into account seasonal variations) are considered.

For the comparison, linear trends of contamination are also displayed in plots in Fig. 3.3. Linear trend does not capture general tendencies contamination changes, especially at the end of the period. In particular, for HCB it predicts even negative values of concentrations in the end of the period, for B[a]P it does not capture the growth of concentrations from 2005 to 2012.

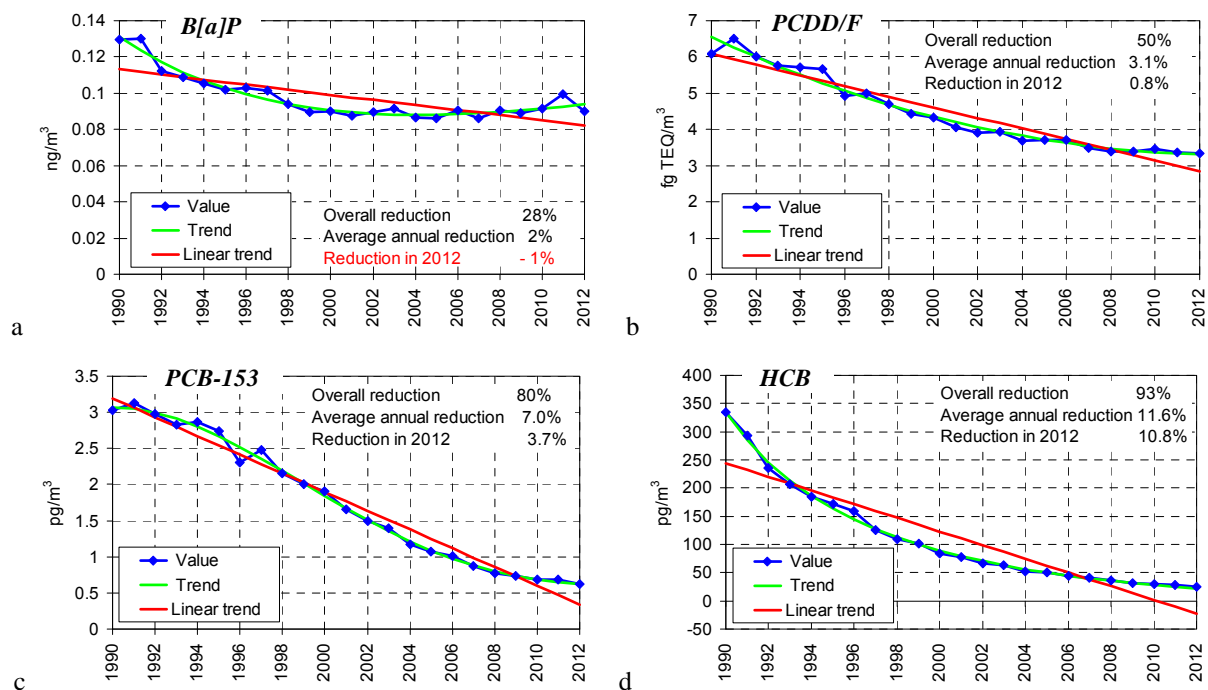


Fig. 3.3. Trends in annual mean air concentrations of (a) B[a]P, (b) PCDD/Fs, (c) PCB-153 and (d) HCB in the EMEP region for the period from 1990 to 2012. Negative reduction denotes increase of air concentrations.

Comparing trends for selected POPs it is seen that maximum reduction occurred in the beginning of the period, whereas at the end of the period reduction rate diminished or even was replaced by growth. Maximum overall reduction is obtained for HCB (over 90%), and minimum – for B[a]P (about 30%). For B[a]P increase of pollution in 2012 was obtained (about 1%), and for PCDD/Fs contamination reached stationary level, being about 0.8% only. It is noted that the growth of B[a]P air concentrations in the EMEP region from 2005 to 2012 occurred to be statistically significant at 90% confidence level.

Seasonal variability of concentrations for some POPs is important characteristic in the evaluation of long-term trends. In particular, intra-annual variations of PAH concentrations can reach an order of magnitude that needs to be considered in course of trend analysis. Developed approach for the analysis of trends takes into account seasonal variations of pollution and provides approximation of changes on the level of monthly mean air concentrations. As an example, results of the analysis of long-term trends in seasonal variations of B[a]P air concentrations in the EMEP region are shown in Fig. 3.4. Due to substantial seasonal variability B[a]P air concentration in cold period of year can exceed several times the annual average value.

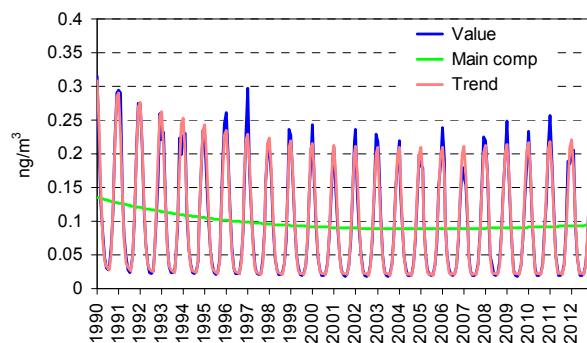


Fig. 3.4. Trends of monthly means of air concentrations of B[a]P in the EMEP region for the period from 1990 to 2012.

Analysis of trends shows that seasonal variations are significant in most of the EMEP countries being on average about 100% and ranging from 50% to 150%. Differences of seasonal variations among the

countries can be explained by their geographical location (the influence of seasonal variations is higher in continental countries than in coastal ones).

Along with changes of POP pollution in the EMEP region as a whole the analysis of trends was performed also for individual EMEP countries. To exemplify this information, results of trend analysis for B[a]P air concentrations in particular EMEP countries are given below.

Average annual reduction rates for the period from 1990 to 2012 and reduction rates in the last year of the period (2012) for all EMEP countries are shown in Fig. 3.5 (a – c). For the analysis of long-term changes of pollution the EMEP countries were divided into three groups: EU28, EECCA, and remaining countries specified as the group 'other'.

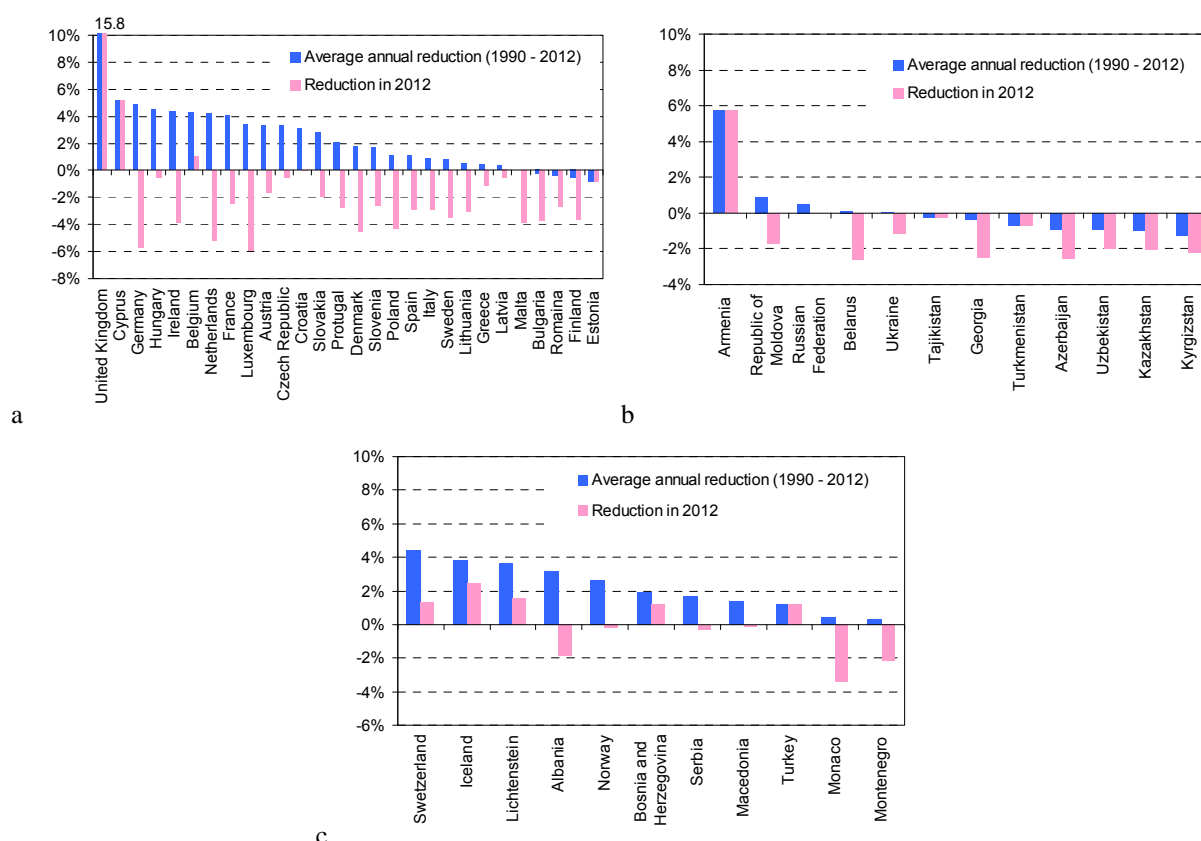


Fig. 3.5. Estimated average annual reduction rates of B[a]P air concentrations for the period from 1990 to 2012 and reduction rates in the last year (2012) of the period in the EMEP countries: a) the EU28 countries, b) the EECCA countries, c) other EMEP countries. Negative values denote growth of pollution.

Results of the analysis indicate that for most of the EU28 countries reduction of B[a]P pollution levels in the end of the considered period was changed to growth. For most of the EECCA countries negative rates of changes are obtained that indicates the increase of air concentrations on the average. Other countries are generally characterized by slower decline of contamination levels from 1990 to 2012 comparing to the EU countries.

Examples of trends for two EMEP countries, namely, for Germany and Belgium, with different types of temporal variations of B[a]P air concentrations are displayed in Fig. 3.6. Along with large total reduction (68%), the growth of contamination in Germany in the end of the period (from 2005) is obtained, reaching 6% in the last year 2012. It should be noted that this growth of B[a]P air

concentrations from 2005 to 2012 is statistically significant. Increase of B[a]P pollution levels can also be seen in measurements of B[a]P content in precipitation made at German monitoring sites (Fig. 2.5). In contrast, for Belgium, though total reduction is slightly lower, decline of contamination took place throughout the entire period being less in the end of the period (1%) than in the beginning (about 6%).

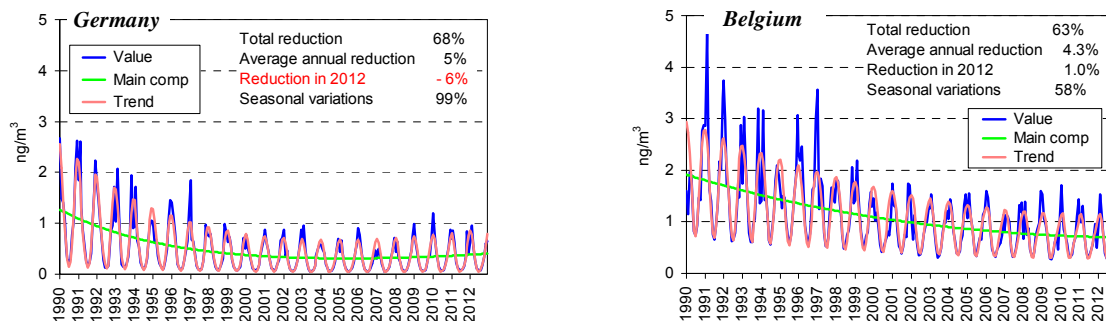


Fig. 3.6. Trends of B[a]P air contamination in the two EMEP countries.

For the comparison, calculated trends for B[a]P emissions in the two considered countries are shown in Fig. 3.7. It is seen that trends of air concentrations and emission totals (on the level of annual averages) well agree with one another. Differences in values of reduction parameters for modelled air concentrations and emissions are explained by transboundary transport.

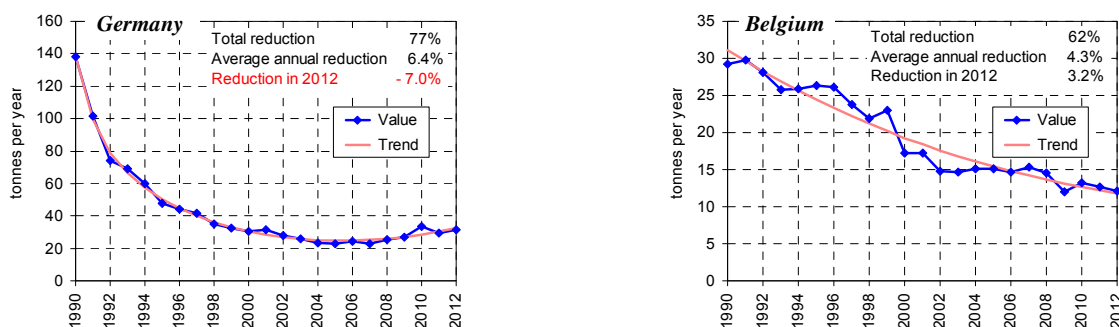


Fig. 3.7. Trends of B[a]P emission totals in the two EMEP countries.

3.3. Trends in observed and modelled levels of pollution in particular grid cells

Model simulations provide information on changes of pollution levels for the whole EMEP region. In the particular grid cells of modelling domain, where the long-term measurements of POP concentrations were carried out, it was possible to perform combined analysis of trends in the pollution levels. It should be taken into account that measurement data describe pollution levels at particular locations whereas model calculations provide values averaged over the 50×50 km grid cells.

Measurements of B[a]P, PCB-153 and HCB air concentrations for sufficiently long period of time were available for the following EMEP monitoring sites: CZ3 (Kosetice), FI96 (Pallas), IS91 (Storhofdi), NO42 (Zeppelin), NO99 (Lista) SE2/14 (Rörvik/Råö) and SE12 (Aspvreten). Location of these sites is shown in Fig. 3.8. Since time periods of measurements for different sites are different, average annual reduction rates are used to evaluate decline of pollution levels.

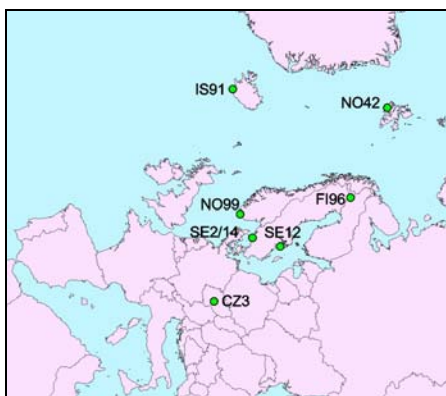


Fig. 3.8. Location of EMEP sites chosen for the trend analysis for POPs

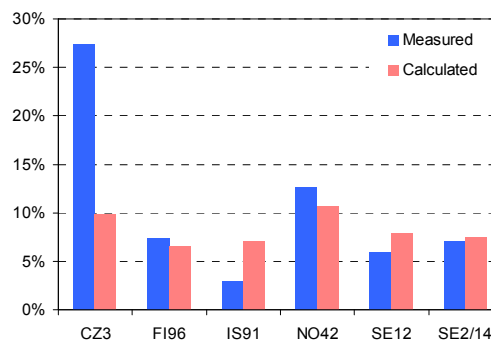


Fig. 3.9. Average annual reduction rates of PCB-153 calculated by trend analysis of measurement and modelling data

Average annual reduction rates of PCB-153 air concentrations estimated on the basis of combined trend analysis of measurements and modelling results are shown in Fig. 3.9. It is seen that measurements and model results indicate similar reduction rates of PCB-153 for all the sites except for CZ3 (Kosetice). Difference in the reduction rates for CZ3 site is caused by disagreement between modelled and measured values of PCB-153 air concentrations in the period 1997-1999 (see Fig. 3.10). It may be conditioned by several factors, including uncertainties in the emission data applied in model simulations as well as changes in measurement techniques during the monitoring period. This issue should be further analyzed with joint participation of experts in modelling, measurements and emissions.

Statistics of average annual reduction rates obtained on the basis of trend analysis of modelled and observed PCB-153 air concentrations is shown in Fig. 3.11a. It can be concluded that annual average reduction of concentrations is about 8% with possible variations from 3% to 12%. These figures agree with the estimate of average annual reduction within the EMEP domain (7%) obtained with the help of model calculations (see Fig. 3.3 above). Recalculation of reduction from annual average to the entire period (from 1990 to 2012) results in overall decline about 85%.

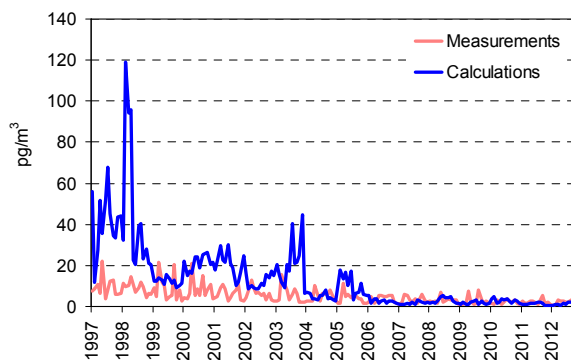


Fig. 3.10. Comparison of modelled and measured concentrations of PCB-153 at site CZ3 (Kosetice)

Average level of seasonality of PCB-153 air concentrations estimated on the basis of both measurements and model calculations is about 60%. At the same time, there is marked difference between the seasonality of observed and modelled concentrations for particular monitoring sites. The reason of these differences requires further analysis. It can be mentioned also, that for better description of seasonality of PCB air concentrations refinement of information on seasonal variations of PCB emissions is needed.

Average annual reduction rate of PCB-153 is about 8%. Recalculation of reduction from annual average to the period from 1990 to 2012 results in overall decline about 85%. The analysis of both measurement and calculation data shows that seasonal variations can change pollution levels of PCB-153 by about 60%.

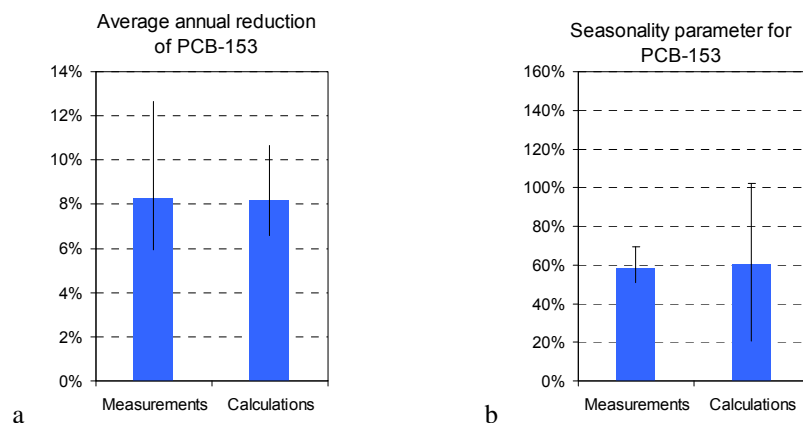


Fig. 3.11. Average annual reduction of PCB-153 contamination (a) and seasonality parameter (b). The whiskers indicate the variation of the parameter between site locations.

For B[a]P the analysis was carried out for four sites, namely, CZ3, FI96, SE2/14 and SE12 (Fig. 3.12). Measurements of the site NO42 were not included since levels of concentrations at this site might be affected by the emission sources located outside the EMEP domain, which were not taken into account in model simulations.

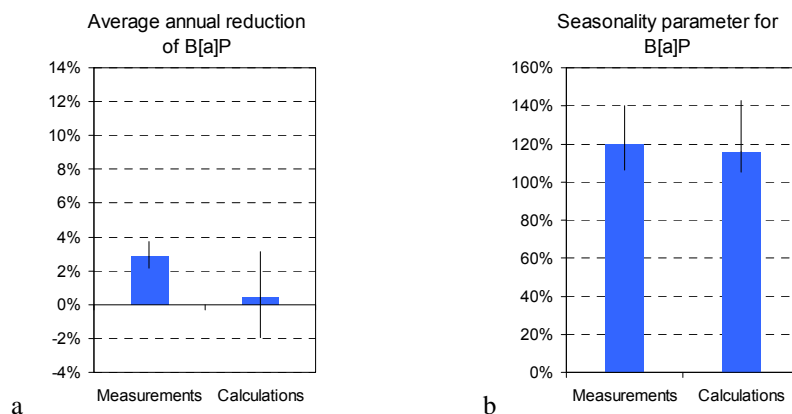


Fig. 3.12. Comparison of average annual reduction of measured and modelled B[a]P air concentrations (a) and seasonality parameter (b). The whiskers indicate the variation of the parameter between site locations.

According to obtained results *average annual reduction rate of B[a]P is rather low, namely, 0.5% and 3% based on model results and measurements, respectively. Recalculation of reduction from annual average to the period from 1990 to 2012 results in total reduction from 10% to 50%.*

Analysis of both measurement and calculation data shows that seasonal variations of B[a]P air concentrations can change pollution levels up to several times from annual average value (Fig. 3.13). The results of the analysis of maximum B[a]P concentrations at location of the EMEP measurement sites are displayed in Fig. 3.14, where average annual reduction of air concentrations and their reduction in the last year of the period (2012) are given.

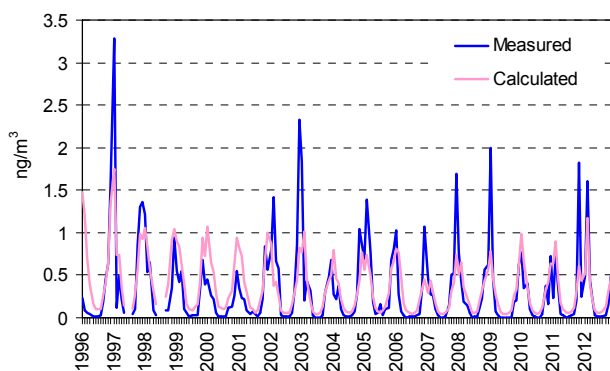


Fig. 3.13. Modelled and measured B[a]P concentrations at CZ3.

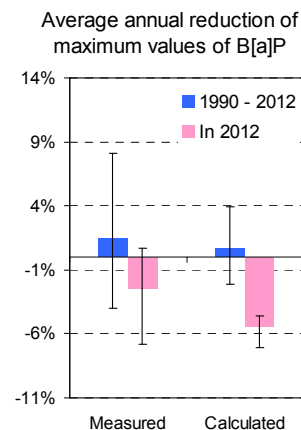


Fig. 3.14. Reduction of maximum (winter time) observed and modelled air concentrations of B[a]P (average for the period 1990-2012 and in the year 2012).

The analysis shows that *both model calculations and measurements indicate low rate of reduction of B[a]P air concentrations in winter time for the whole period (1% – 2%) and show the growth of B[a]P concentrations in the end of the period (2012).*

For HCB, long enough time series of measured air concentrations were available for the following EMEP monitoring sites CZ3, NO42, NO99 and IS91. The results of the analysis of trends for the sites CZ3 and NO99 are summarized in Fig. 3.15, where average annual reduction for the entire period and reduction in the last year of the period are displayed. Similar to PCB-153 *average annual reduction of HCB is high (9% – 10% per year). The reduction in 2012 is lower for all sites than average reduction.*

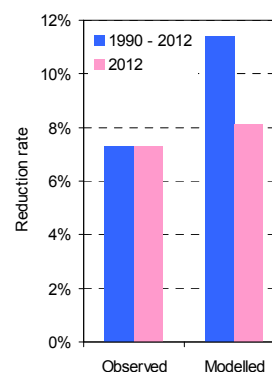


Fig. 3.15. Reduction of observed and modelled annual mean HCB air concentrations (average for the period 1990-2012 and in the year 2012)

Analysis of temporal variations of HCB air concentrations for the two remote sites, NO42 and IS91, indicates larger difference in estimates of trends. Particularly, model overestimates annual average reduction for the period 1990-2012 and does not capture the growth of HCB air concentrations in the end of the period. The reason of the differences is connected with that the levels of pollution at these sites are likely influenced by the emission sources outside the EMEP domain. Thus refinement of description of global scale HCB emissions and their temporal changes in various regions of the world is needed.

Regular measurements of dioxins and furans air concentrations at the EMEP monitoring sites are not currently performed. At the same time, it is possible to use available long-term measurements of PCDD/F air concentrations in the EMEP countries reported in literature for the analysis of long-term changes of pollution. A number of studies provided results of long-term measurements of dioxins and furans concentrations in air. Significant decrease of PCDD/F air concentrations (about 70%) was observed in Spain in the period 1994-2004 [Abad *et al.*, 2007]. Monitoring of PCDD/F air concentrations in the UK at urban and rural sites indicated sharp decline in early 1990s and smaller decline in later period of time [Katsoyiannis *et al.*, 2010]. Pronounced decrease of PCDD/F air concentrations and deposition (about a factor of 5) was also observed in Germany from 1988-1992 to 2005 [Bruckmann *et al.*, 2013] which was almost leveled off since 2005. In general, results of model simulations are in line with available measurements and demonstrate similar decline of PCDD/F pollution in the EU countries from 1990 to 2012 (about 80%).

3.4. Factors affecting long-term trends of POP pollution in the EMEP region

Long-term trends of POP pollution levels in the EMEP countries depend on changes of anthropogenic and secondary emissions as well as other factors like long-term variations of meteorological conditions, changes of land-use and land-cover characteristics, etc. Temporal variations of anthropogenic and secondary emissions are likely the major factor influencing changes of POP pollution in the countries. In this section evaluation of contributions of various groups of emission sources to the contamination of the EMEP region and their temporal changes are briefly discussed. Considered groups of emission sources include anthropogenic emissions of the EMEP countries, secondary emissions and emissions of non-EMEP sources. By secondary emissions we mean re-volatilization of POPs from the environmental media polluted previously.

For a particular EMEP country, contributions of contemporary anthropogenic emissions can be split into contributions of national emissions and of transboundary transport from the rest EMEP countries. Relative changes of PCDD/F deposition in the EMEP countries, together with changes of the contributions of national emission sources and transboundary transport from the year 1990 to 2012 are illustrated in the Fig. 3.16.

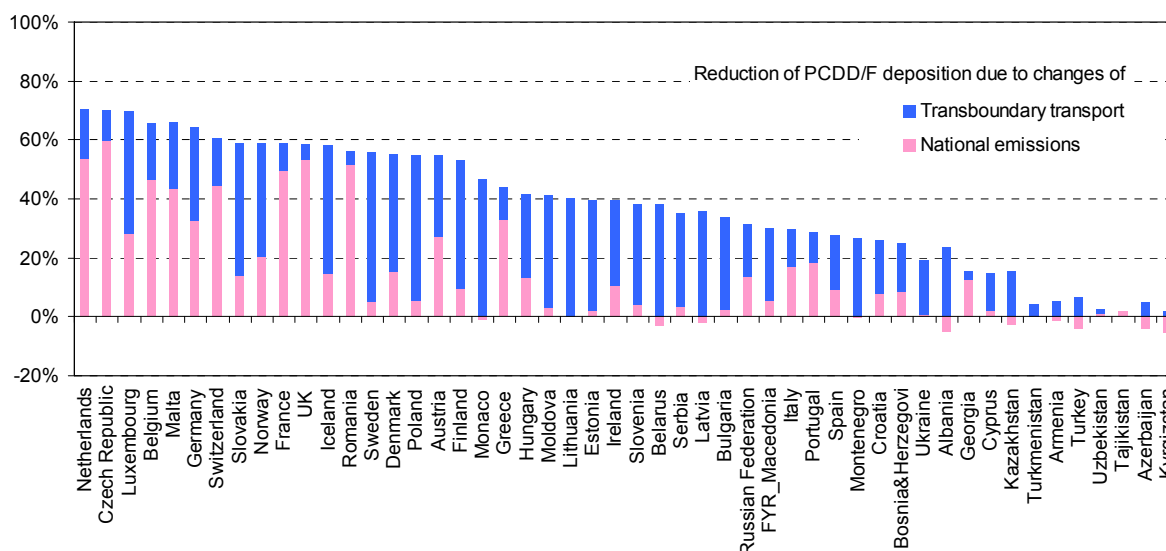


Fig. 3.16. Reduction of anthropogenic PCDD/F deposition in the EMEP countries caused by the changes of national emissions and changes of transboundary transport between 1990 and 2012. Positive value means decrease of deposition, and negative – increase.

It is seen that *in most of the EMEP countries reduction of pollution was largely influenced by the decrease of contribution of transboundary transport*. Examples of dynamics of contamination in two EMEP countries (the Netherlands and Republic of Moldova) for the period 1990-2012 and changes of contribution of national emissions and transboundary transport are shown in Fig. 3.17.

The analysis indicated that for the Netherlands reduction of deposition flux from 1990 to 2012 occurred mainly in the beginning of the period and was influenced by the reduction of national emissions. On the opposite, reduction of contamination in Republic of Moldova was conditioned by the decline of transboundary transport.

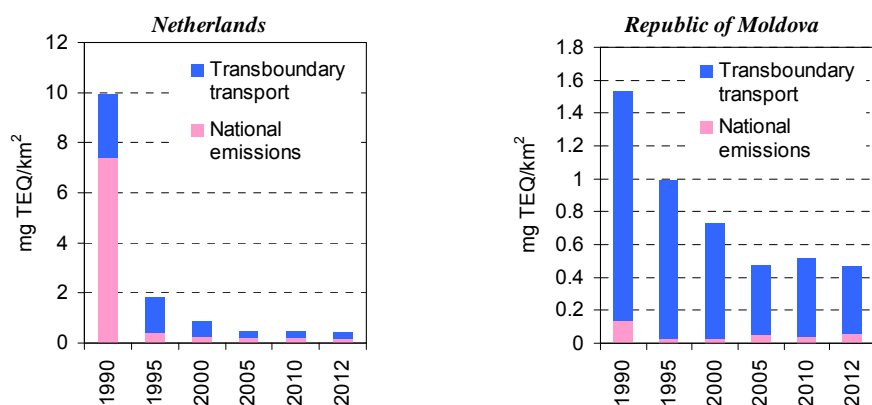


Fig. 3.17. Reduction of anthropogenic PCDD/F deposition in the Netherlands and Republic of Moldova caused by the changes of national emissions and changes of transboundary transport between 1990 and 2012.

Another factor, influencing the trends of POP pollution in the EMEP region, is temporal variations of contribution of non-EMEP emission sources, which varies between the considered POPs. *Noticeable contribution of non-EMEP emission sources to the pollution levels is estimated for PCBs and HCB, while for selected PAHs it is considerably low.* Comparison of variations of contributions of EMEP and non-EMEP sources to PCB-153 air concentrations in mixing layer in the considered period is shown in Fig. 3.18. Estimates of contributions of these two types of sources were obtained using global scale model simulations based on the global emission inventory of Breivik *et al.* [2007]. Decline of non-EMEP emissions was relatively faster comparing to the EMEP emissions according to the emission inventory. This led to faster decrease of contribution of non-EMEP emissions to the pollution of the EMEP region. At the same time, the study of Breivik *et al.* [2011] suggests that levels of PCB concentrations in the atmosphere in some regions (e.g. in Africa, India, and Asia) can be affected by contemporary e-waste destruction activities, which can provide additional contribution to non-EMEP emissions and their slower decline. Besides, the study of Cui *et al.* [2013] indicated increasing of emission of unintentionally produced PCBs in China starting from 1980s due to developing cement production and steel industries in China, especially during recent decade. Taking into account the outcome of these studies, the contribution of non-EMEP contemporary emissions to PCB pollution levels in the EMEP countries can grow in time.

Secondary emissions of POPs from the environmental compartments (e.g. soil, water) can provide substantial contribution to contamination in the EMEP region along with contemporary anthropogenic emissions. Due to historical emissions surface media have accumulated significant amount of POPs, which is re-emitted to the atmosphere and supports levels of air concentrations. Relation between the contributions of contemporary and historical emissions to annual mean air concentrations of PCB-153 in the period 1990-2012 is shown in Fig. 3.19. Contemporary emissions denote the releases of PCB-153 of the particular years shown in the diagram, and historical emissions denote releases during all preceding years. From model calculations, it is seen that secondary emissions due to historical releases can noticeably contribute to the levels of pollution. Moreover, model calculations indicate that relative contribution of historical emissions grows with time (from 35% in 1990 to 45% in 2012). It should be mentioned that these estimates should be considered as preliminary due to uncertainties in the evaluation of historical emissions from 1930 to present, model description of inter-media exchange, etc.

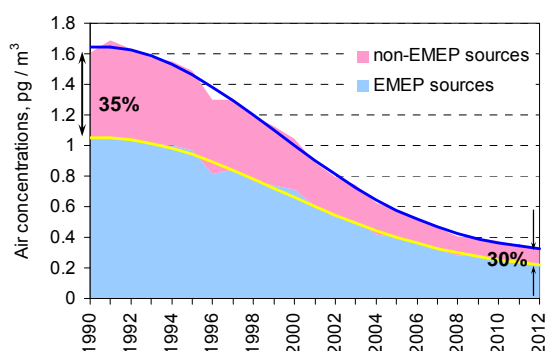


Fig. 3.18. Comparison of contributions of EMEP and non-EMEP sources to PCB-153 air concentrations in mixing layer over EMEP domain and their trends.

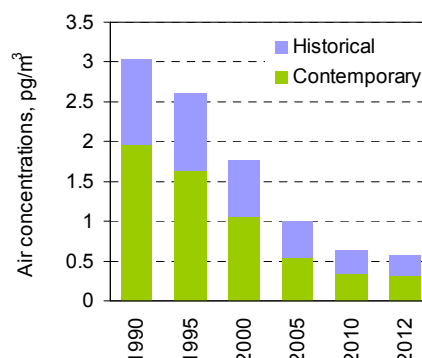


Fig. 3.19. Contributions of contemporary and historical emissions to PCB-153 air concentrations in Europe (ground layer)

3.5. Information for the evaluation of effects of POP pollution

The information obtained from the trend analysis can be of use for the evaluation of harmful effects for human health and the environment.

Air concentrations of POPs can vary in high extent between different parts of a country. From this viewpoint it is interesting to evaluate percentiles of the distribution of contamination over a country. This evaluation can be performed by the following two ways:

- Area-based approach. Here the change of concentration range for, say, 10% of the country area with highest contamination is determined.
- Population-based approach. The changes of concentration range for 10% of country population living under highest concentrations is evaluated.

Examples of such evaluation for Germany are presented in Fig. 3.20.

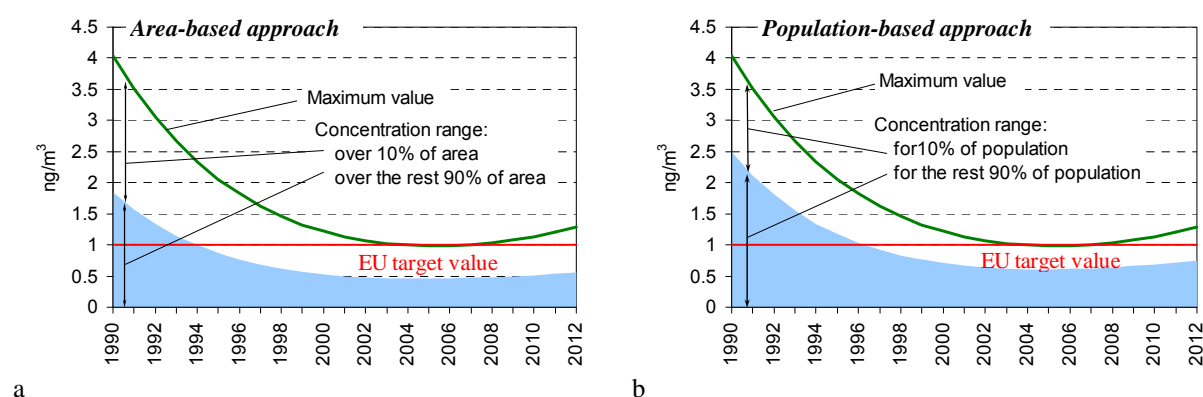


Fig. 3.20. Percentiles of the distribution of B[a]P contamination over Germany: (a) area-based, (b) population-based. Red point shows the value of trend of lower limit of air concentrations for 10% of population in 2012.

The analysis shows that concentration range for 10% of population is higher than that over 10% of the country area. It is conditioned by the fact that contamination is higher in the regions with high population density. Hence, *population-based approach of evaluation of percentiles might be preferable*.

The value of trend for 2012 (red point on the diagram Fig 3.20b shows the value of lower limit of annual mean concentration level for 10% of population (free of irregular variations) in the end of the considered period.

Results of population-based evaluation are demonstrated in Fig. 3.21. It is found that for 4 EMEP countries expected values of annual mean air concentrations for 10% of population exceed EU target value. For the rest countries in the diagram exceedance of monthly means of air concentrations for 10% of population over the EU target value for can be expected. The diagram in Fig. 3.21 shows also calculated range of decrease/increase of air concentrations obtained by trend analysis. For countries with large increase rate further enlargement of lower limit of concentrations for 10% of population can be expected.

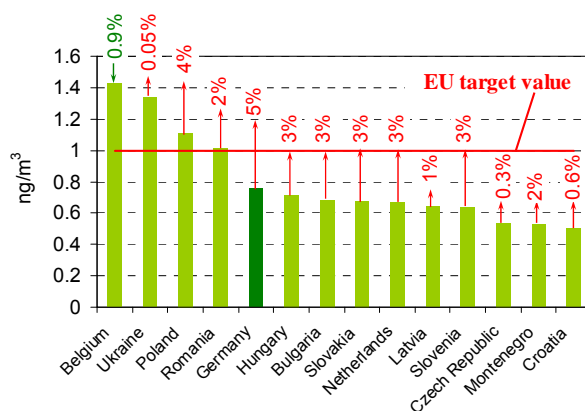


Fig. 3.21. Expected value of lower limit of annual mean concentration level for 10% of population. Arrows show calculated increase rates in the end of the considered period (2012) evaluated on the basis of trend analysis.

Model assessment of trends allows evaluating trends of deposition to various land-use types. As an example of such information, trends of B[a]P deposition to deciduous forest and arable lands for the period from 1990 to 2012 are shown in Fig. 3.22. It is seen that deposition flux to arable lands is slightly larger than that to the deciduous forests. Total reduction of deposition fluxes was estimated to 25% approximately. However, recent years of the period were characterized by the increase of deposition fluxes. Specifically, B[a]P deposition fluxes to deciduous forests and arable lands increased in 2012 by 1.2% and 0.6% respectively. The growth of these deposition fluxes was found to be statistically significant at 90% confidence level.

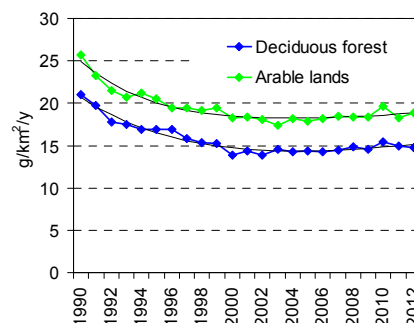


Fig. 3.22. Trends of annual B[a]P deposition to deciduous forests and arable lands within the EMEP region

Similar information on trends of deposition fluxes to various ecosystems (deciduous forest, coniferous forest, grass, cropland) can be generated for all POPs included into MSC-E modelling activities (PAHs, PCDD/Fs, PCBs, and HCB) and for all EMEP countries on request.

4. POLLUTION LEVELS AND TRANSBOUNDARY TRANSPORT OF POPS IN 2013

Model assessment of POP pollution levels in the EMEP domain is based on multi-media modelling approach, which takes into account peculiarities of POP transport and fate in the environment (e.g. phase partitioning, degradation in media, multi-hop transport). For long-lived POPs it is important to evaluate global scale transport and contribution of non-EMEP sources to the pollution levels in the EMEP countries. For this purpose modelling approach for POPs comprises the use of multi-scale model simulations (global and regional). Global scale modelling provides initial and boundary conditions for regional scale modelling within the EMEP domain and characterizes intercontinental transport of pollution from non-EMEP emission sources.

4.1. Polycyclic aromatic hydrocarbons (PAHs)

Modelling of PAH pollution levels in the EMEP domain was performed for the four indicator congeners (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)) included into the Protocol on POPs. Model simulations were carried out for 1990 and 2013. Assessment of PAH pollution levels consists of analysis of long-term changes, evaluation of transboundary transport between the EMEP countries, and comparison of obtained modelling results with available measurements.

Pollution levels in the EMEP region

Model predictions of PAH annual mean air concentrations in the EMEP region for 1990 and 2013 are presented in Fig. 4.1. *Levels of four PAH air concentrations in the EMEP countries decreased since 1990 by 33% on average. At the same time, in some of the EMEP countries the level of PAH pollution in 2013 was still significant or tended to increase.*

In particular countries changes of PAH concentrations in air from 1990 to 2013 vary from almost 90% decrease up to 30% increase. *The most significant decline of air concentrations was estimated for the United Kingdom (87%), followed by Armenia (72%) and Germany (69%).* In thirteen countries levels of PAH pollution dropped more than twice. The lowest decline (below 2%) was obtained for Estonia, Turkmenistan, Malta, and Tajikistan. *For some of the EECCA countries model predictions indicated increase of PAH air concentrations (namely, for Kyrgyzstan, Kazakhstan, Azerbaijan, and Uzbekistan).*

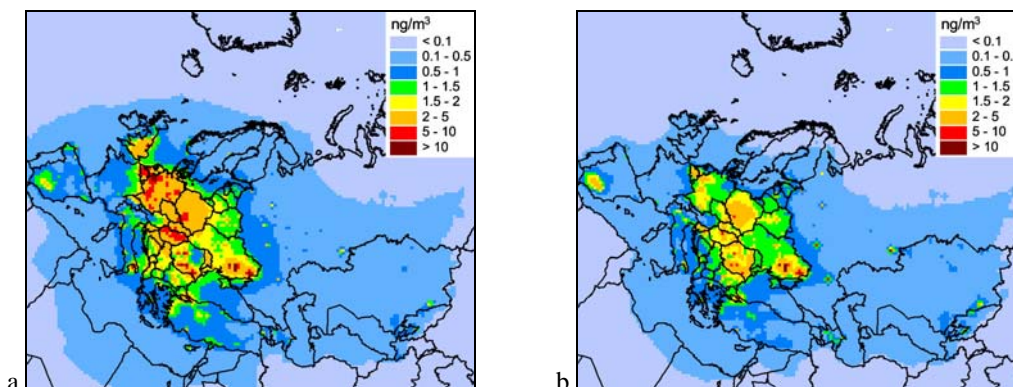


Fig. 4.1. Spatial distribution of calculated annual mean air concentrations of 4 PAHs in the EMEP domain for 1990 (a) and 2013 (b), ng/m³.

Inter-annual variations of PAH pollution levels during the two recent years, 2012 and 2013, are displayed in the Fig. 4.2. To estimate changes of pollution in the EMEP countries model simulations were performed with meteorological data for years 2012 and 2013 and with the same emission dataset for 2012. Thus modelling results illustrate the effect of variability of meteorological conditions (precipitation, prevailing atmospheric flows, temperature, etc.) between the years 2012 and 2013.

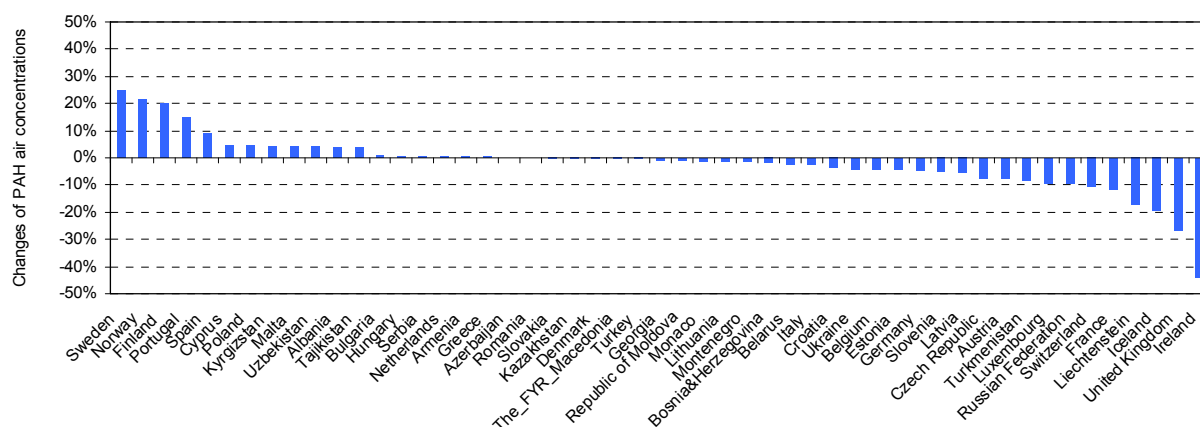


Fig. 4.2. Changes of annual mean PAH air concentrations in the EMEP countries between 2012 and 2013 due to variability of meteorological conditions. Negative values denote increase of pollution levels, and positive ones – decrease.

It is seen that variations of meteorological parameters can lead to considerable changes in the annual mean PAH air concentrations (from about 25% decrease to 40% increase). The most significant decline of concentrations (by 20-25%) took place in countries of Northern Europe, namely, Sweden, Norway, Finland. The largest increase of air concentrations was estimated for Ireland, the UK, and Iceland (by 20-40%).

Comparison with measurements

Results of model simulations for PAH congeners were evaluated against the measurements of air concentrations made at the EMEP monitoring sites in 2013 (Fig. 4.3). Modelled air concentrations of sum of the 4 PAHs well agree with observed pollution levels. In particular, for all monitoring sites difference between the modelled and observed concentrations is within a factor of two (Fig. 4.3a). Model generally captures the spatial distribution of PAH air concentrations with correlation about 0.9. At the same time, there is a tendency to underestimate concentrations in the Czech Republic and Poland.

Comparison of model predictions with measurements for particular PAHs is shown in Fig. 4.3b. It is seen that most part of modelled concentrations is within a factor of two in comparison with measured values. Highest agreement between the model and measurements is obtained for air concentrations of IP, followed by B[a]P, B[b]F, and B[k]F. For all four considered PAHs spatial correlation of modelled and measured values is in the range 0.7-0.9.

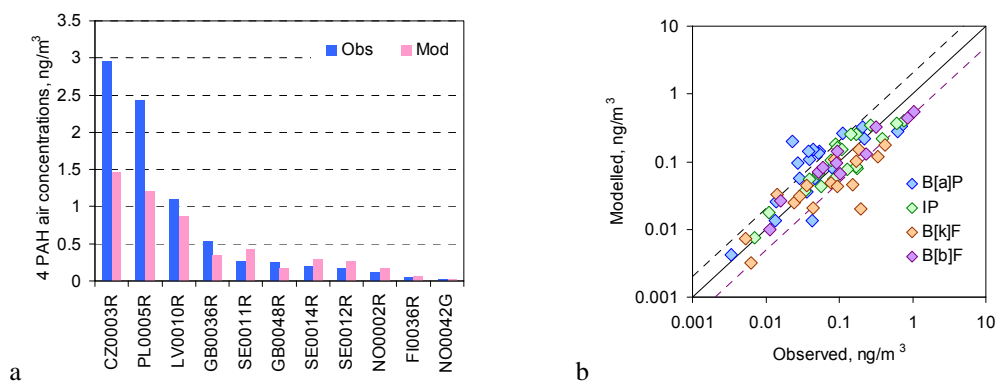


Fig. 4.3. Comparison of modelled concentrations of 4 PAHs (a) and of B[a]P, B[b]F, B[k]F, and IP (b) with measurements of the EMEP monitoring sites for 2013. Dashed lines denote the area of agreement between the modelled and measured values within a factor of two.

Transboundary transport within the EMEP region

Transboundary transport of four indicator PAHs within the EMEP region was evaluated for 1990 and 2013. Pollution levels of the selected PAHs are mostly formed by the anthropogenic emission sources, while contributions of non-EMEP emissions and secondary sources are comparatively low. For this reason analysis of transboundary transport is mainly focused on the anthropogenic emission sources of the EMEP countries.

Model estimates of total annual deposition of 4 PAHs to the EMEP countries in 2013 are presented in Fig. 4.4. *The largest contribution to total deposition of 4 PAHs belongs to B[b]F (37%) followed by B[a]P (26%) and IP (21%), while the lowest one is made by B[k]F (16%).*

Elevated levels of PAH deposition fluxes in the EMEP countries, more than 200 g/km²/y, are indicated for Belgium, Slovenia, Romania, Montenegro, and Slovakia. Relatively low intensity of deposition is obtained for the EECCA countries and regions located far from major European emission sources (e.g. Northern Europe).

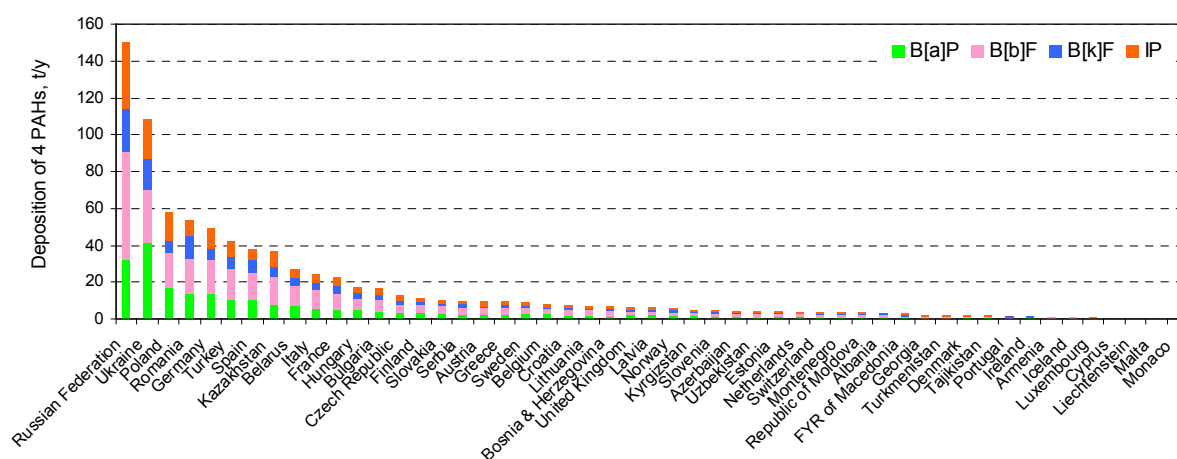


Fig. 4.4. Annual deposition fluxes of 4 PAHs (B[a]P, B[b]F, B[k]F, and IP) in the EMEP countries calculated for 2013, g/km²/y

Model assessment of PAH pollution in the EMEP region demonstrates that transboundary transport provides substantial contribution to the contamination of the EMEP countries. *For most of the countries (almost 75%) the deposition of 4 PAHs over their territories in 2013 was mostly caused by the transboundary transport of pollution exceeding contributions of their national emissions* (Fig. 4.5). On average, transboundary transport between the EMEP countries contributed to total deposition of 4 PAHs over their territories about 50%.

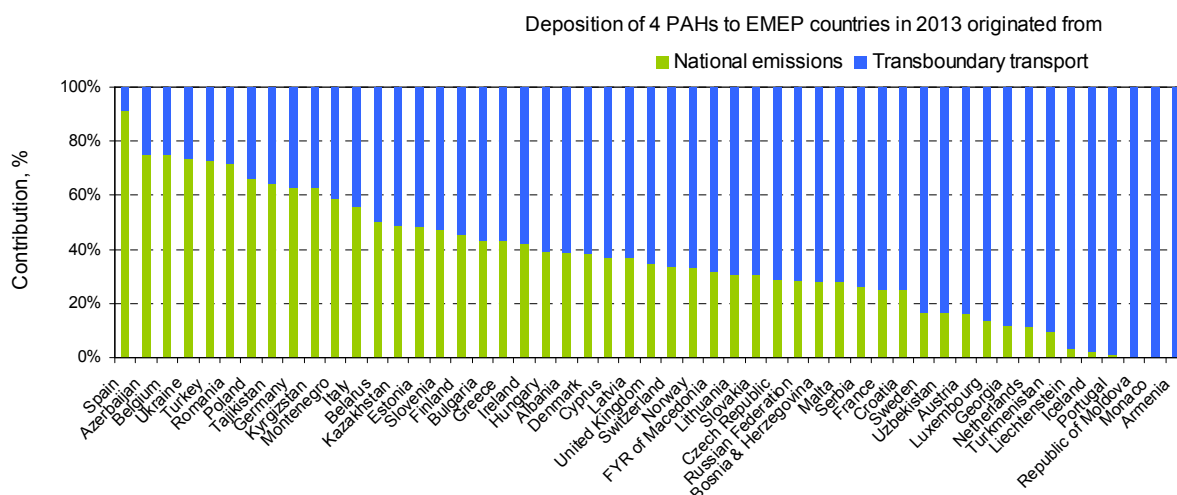


Fig. 4.5. Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of 4 PAHs over the EMEP countries in the 2013.

Transboundary transport of pollution from a particular country to other countries can also be characterised by the fractions of national emission, deposited over its own territory and outside its boundaries. *For 48 countries the fraction of PAHs emitted by the national emission sources and deposited to other EMEP countries was higher than the fraction of emission deposited to the country itself.* Almost half of total annual deposition of 4 PAHs within the EMEP region in 2013 was provided by the emission sources of five countries, namely, Ukraine, Poland, Romania, Germany, and the Russian Federation.

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

Polychlorinated dibenzo(p)dioxins and dibenzofurans are unintentional by-products released into the environment during various combustion processes (e.g. including industrial processes, waste incineration, and open burning of wastes and biomass). Along with anthropogenic emissions, additional contribution to the pollution levels can be made by the secondary emission sources, formed by long-term accumulation of PCDD/Fs in the surface media and direct emissions to soil and water bodies. PCDD/Fs are characterized by significant persistence in the environmental media like soils and sediments. Thus their content in these media can pose risk to the ecosystems and human health.

Similar to the previous year the evaluation of PCDD/F pollution levels was performed with application of experimental emission scenarios for regional and global modelling. Model assessment of pollution in the EMEP region for 1990 and 2013 was based on the developed approach for the construction of PCDD/F emission scenarios [Shatalov et al., 2012]. Along with this, the study of global scale transport and fate of PCDD/Fs was continued using emission scenario based on the UNEP SC inventory of dioxins and furans releases to different media. Results of the assessment of PCDD/F pollution levels include analysis of long-term changes of pollution and evaluation of transboundary transport between

the EMEP countries. Model predictions were compared against available measurements of PCDD/F air concentrations. Short overview of the outcome of these studies is presented below. Detailed information on emission scenarios, results of model simulations, and their analysis is presented in the MSC-E Technical Report [Shatalov et al., 2015].

Pollution levels in the EMEP region

Comparison of model estimates of PCDD/F air concentrations in 1990 and 2013 is given in Fig. 4.6. *Levels of PCDD/F pollution in the EMEP countries declined during the two recent decades by 60% on average.* The most substantial decrease of pollution is seen in the countries of the European Union. Changes of pollution in the EECCA countries and the other EMEP countries during this period are generally smaller.

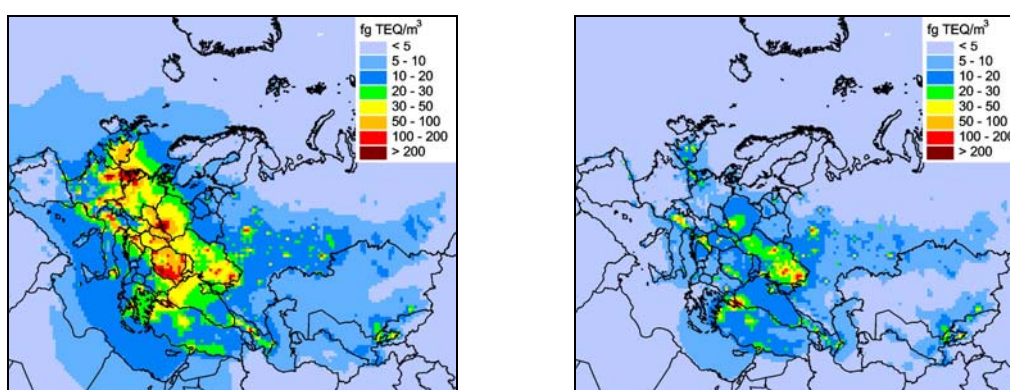


Fig. 4.6. Spatial distribution of calculated annual mean air concentrations of PCDD/Fs in the EMEP domain for 1990 (a) and 2013 (b), fg TEQ/m³.

Model simulations indicate that decline of PCDD/F air concentrations in the EMEP countries in the period 1990-2013 vary from more than 90% to 30%. *The largest drop of air concentrations was estimated for the Netherlands (93%), followed by Luxembourg (91%) and France (89%).* In 39 countries levels of PCDD/F concentrations decreased by more than 50%. The lowest decline (about 30%) was obtained for Latvia and Ukraine. *Among the EECCA countries the largest decrease indicated for the Republic of Moldova (70%) and Turkmenistan (60%), while for the other countries of this group reduction of pollution was about 40% on average.*

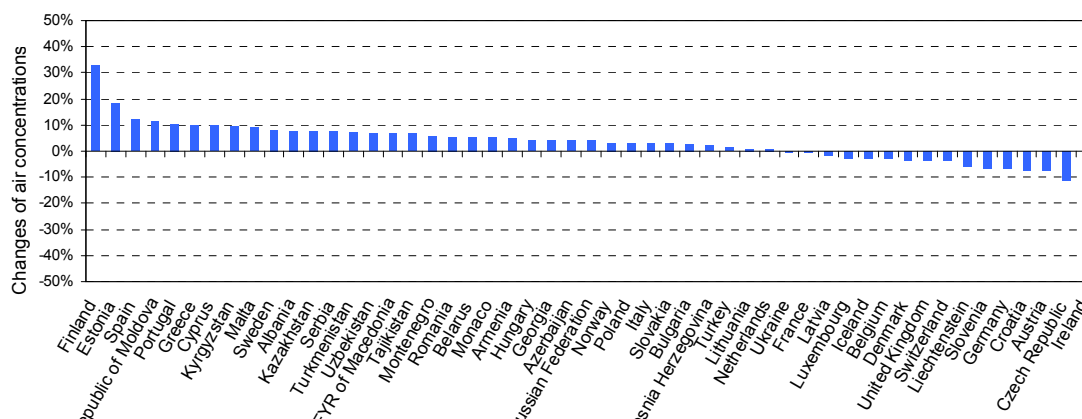


Fig. 4.7. Changes of annual mean PCDD/F air concentrations in the EMEP countries between 2012 and 2013 due to variability of meteorological conditions. Negative values denote increase of pollution levels, and positive ones – decrease.

Levels of annual mean PCDD/F air concentrations in the EMEP countries varied from 2012 to 2013 from about 30% decrease to 25% increase (Fig. 4.7). Model simulations for these years were performed using meteorological data for the corresponding years and the same emission data for the year 2012. Thus, differences between the model estimates for the considered two years indicated the effect of inter annual variability of meteorological parameters (temperature, precipitation amount, atmospheric transport pathways, etc.). The largest decline of air concentrations took place in Finland (by 30%), Estonia (by 20%), and Spain (by 12%), while for Ireland and the Czech Republic increase of air concentrations was estimated by about 25% and 12%, respectively. The other countries were characterized by moderate changes of PCDD/F air concentrations in the range from -10% to 10%.

Transboundary transport within the EMEP region

Transboundary transport of dioxins and furans within the EMEP region was evaluated for 1990 and 2013 taking into account contributions of different types of emission sources (anthropogenic and secondary). The contribution of non-EMEP emission sources was estimated similar to the approach, described in the EMEP Status Report [Gusev *et al.*, 2013]. Particularly, data on emissions outside the EMEP region were defined for North America and Eastern Asia. These data on non-EMEP emissions represented only part of potential emission sources located outside the EMEP region. Therefore, there is ongoing work on the construction of global emission scenario, covering larger set of the countries based on the data on the dioxins and furans emission inventories available in the UNEP SC. This set of emissions can permit to reduce uncertainties of model estimates of intercontinental transport on further stages of the work. Results of global scale modelling with the experimental emission scenario are described below.

Levels of annual deposition of PCDD/Fs in the EMEP countries in 1990 and 2013 originated from different emission sources are shown in Fig. 4.8. *During the two recent decades PCDD/F deposition declined more than twice. Most significant decrease (about 60%) was obtained for deposition from anthropogenic emission sources.* Changes of deposition from secondary emissions and from non-EMEP sources were lower, by 44% and 40%. Thus, relative contribution of the EMEP secondary sources was increased from 51% in 1990 to 58% in 2013. Similarly, relative contribution of non-EMEP emissions was also tended to grow slightly during the period (from 5% to 6%).

Model estimates of contributions of national emission sources and transboundary transport to PCDD/F anthropogenic deposition in the EMEP countries in 2013 are presented in the Fig. 4.9. According to modelling results *for almost 70% of the EMEP countries the contributions of transboundary transport to PCDD/F deposition from anthropogenic emission sources exceed the contributions of national emission sources.*

Dioxins and furans emitted by the sources of a particular country are deposited to its own territory and to the territories of other countries. Relative amount of pollutant deposited outside the territory of a country can be used as a characteristic of transboundary transport of pollution. *For 18 countries the fraction of PCDD/Fs emitted by the national emission sources and deposited outside their territories was higher than the fraction deposited over the country itself.* About 60% of total annual deposition of PCDD/Fs within the EMEP region in 2013 were provided by the emission sources of four countries, namely, Ukraine, the Russian Federation, Turkey, and Poland.

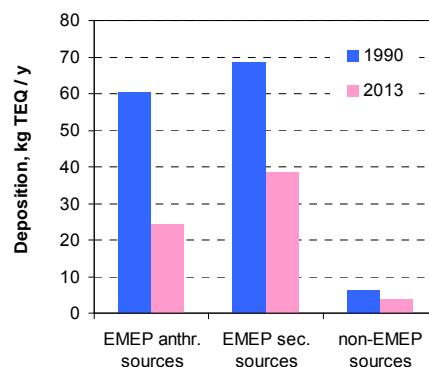


Fig. 4.8. Deposition of PCDD/Fs to the EMEP countries in 1990 and 2013 from anthropogenic and secondary emission sources of the EMEP region as well as from non-EMEP emissions.

Deposition of PCDD/Fs to EMEP countries in 2013 originated from:

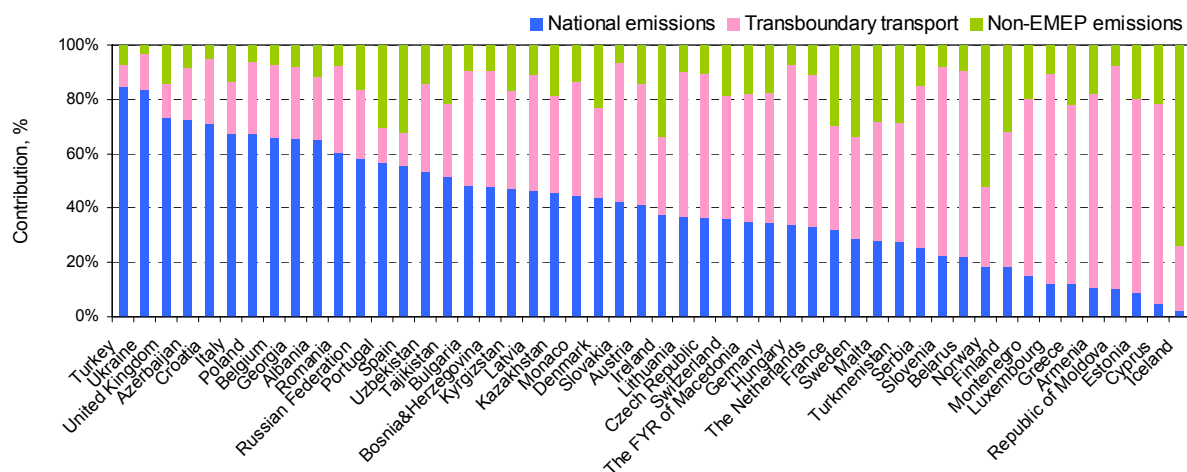


Fig. 4.9. Relative contributions of national emission sources and transboundary transport (including transport from non-EMEP emission sources) to PCDD/F anthropogenic deposition over the EMEP countries in the 2013.

Global scale levels of pollution

This year model evaluation of global scale transport and fate of dioxins and furans was continued on the basis of the experimental emission scenario based on the emission inventory of the UNEP SC described in [Gusev *et al.*, 2014b; Shatalov *et al.*, 2014]. Model simulations were performed with the meteorological data for 2012 and spatial resolution $1^\circ \times 1^\circ$. Initial conditions to evaluate pollution levels in 2012 were obtained using long-term spin-up model run for the period from 1930s to present time. Spatial distribution of annual mean PCDD/F air concentrations for 2012 is presented in Fig. 4.10.

Results of model simulations indicated elevated levels of PCDD/F air concentrations (> 10 fg TEQ/m³) in Africa and South and East Asia. Lower levels of pollution were characteristic of Europe, North and South America, and Australia. Comparison of model predictions with measurements of PCDD/F air concentrations in Europe, North America, and East Asia showed reasonable agreement between modelled and observed air concentrations [Gusev *et al.*, 2014].

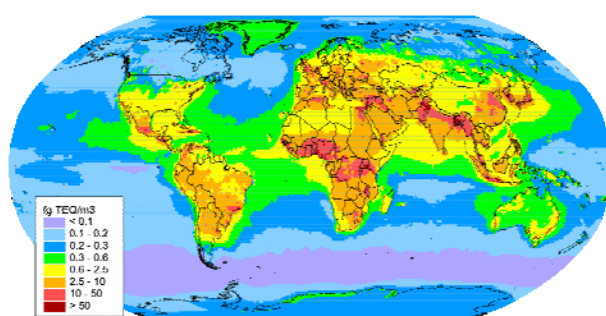


Fig. 4.10. Spatial distribution of modelled annual mean PCDD/F concentrations in air (fg TEQ/m³) obtained for 2012.

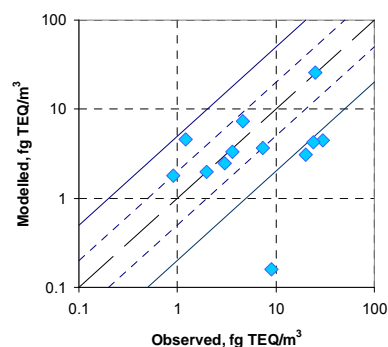


Fig. 4.11. Comparison of modelled annual mean PCDD/F air concentrations for 2012 (fg TEQ/m³) with measurements performed in South America region. Dashed lines denote the area of agreement between modelled and measured values within a factor of 2, solid lines – within a factor of 5.

In 2015 results of global scale model simulations were evaluated against the measurements of dioxins and furans air concentrations carried out in South America [Schuster *et al.*, 2015]. Monitoring of PCDD/Fs in air was performed in the period 2011-2013 using a passive air sampling network established on the basis of Global Atmospheric Passive Sampling (GAPS) sites and six additional sites in the Group of Latin American and Caribbean Countries region (GRULAC).

Model predictions obtained for 2012 are in reasonable agreement with measured air concentrations (Fig. 4.11). For more than a half of the sites, modelled values are within a factor of two with measurements and are found to correlate with observed levels. For some of the urban and rural sites the model predictions are lower than observed air concentrations, which is likely caused by the uncertainties in the spatial distribution of PCDD/F emissions, which is based on the density of population. Along with this comparison, there is ongoing work on the evaluation of modelling results for PCDD/Fs using the measurements collected in the UNEP SC GMP Data Warehouse. Results of this activity will be reported on further stages of the work.

To improve evaluation of PCDD/F pollution further co-operation with relevant activities on the assessment of PCDD/F emissions and monitoring of their content in the atmosphere, performed under the UNEP SC, is of importance.

4.3. Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) are toxic chemicals intentionally produced for various technical applications since 1929. Similar to dioxins and furans, PCBs are characterized by significant persistence in the environment and tend to accumulate in soil and sediment compartments. Though their production and use is banned in many countries PCBs are still detected in various regions of the world and continue posing risks to human health and ecosystems.

Model assessment of PCB pollution levels in the EMEP countries was carried out using combination of global and regional scale modelling. Model simulations were made for the indicator congener PCB-153. Transboundary transport of PCB-153 between the EMEP countries was evaluated using the regional model simulations for the EMEP domain with initial and boundary conditions obtained from the global modelling results. Model predictions were compared with measurements of the EMEP monitoring sites and observations collected in the UNEP SC GMP Data Warehouse.

Global scale levels of pollution

Global distributions of annual mean PCB-153 air concentrations evaluated for 1990 and 2013 are presented in Fig. 4.12. The figure illustrates significant decline of global scale pollution levels from 1990 to 2013. According to available data on PCB emissions about 90% of PCBs during the period of its production and use were released into the environment in the Northern Hemisphere. It is seen that major emission sources of PCBs are located in the European region and in North America.

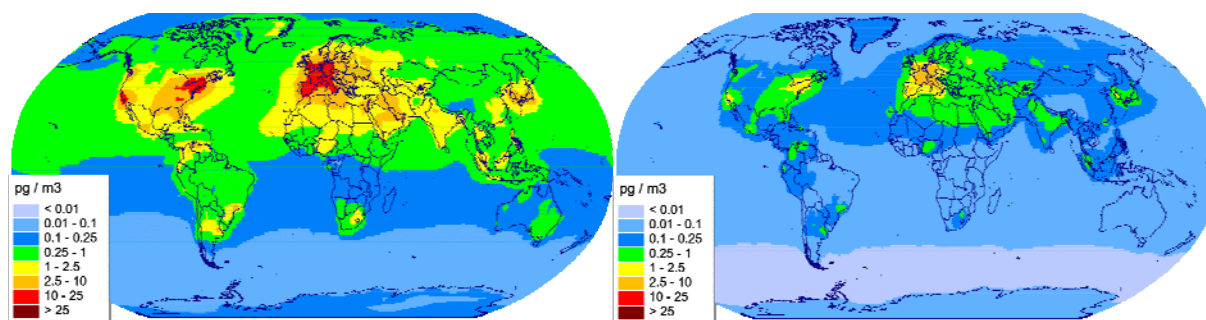


Fig. 4.12. Spatial distribution of global annual mean PCB-153 air concentrations (pg/m^3) for 1990 (a) and 2013 (b) with spatial resolution $1^\circ \times 1^\circ$.

In 2013 elevated air concentrations are seen in Europe ($1\text{--}10 \text{ pg}/\text{m}^3$) and North America ($0.25\text{--}2.5 \text{ pg}/\text{m}^3$), while the other regions are characterized by low air concentrations ($0.01\text{--}0.1 \text{ pg}/\text{m}^3$) which is generally confirmed by measurements.

Results of model simulation for PCB-153 were compared with available measurements performed from the beginning of 1990s up to 2013 in various regions of the world in the remote, background, and rural areas (Fig. 4.13). Measurement data were obtained from the UNEP SC GMP Data Warehouse and included measurements made using active and passive sampling of POP air concentrations.

Comparison indicates that more than 70% of model predictions the difference between modelled and observed air concentrations is within a factor of five and for about 40% of them it is within a factor of two. In general, the model closely describes temporal variations of observed pollution levels during the two recent decades. The spatial correlation of model predictions and measurements is better for the European region, while for other regions the level of correlation is lower.

Comparison of model predictions and measurements of PCB-153 air concentrations in several regions of the world is illustrated in Fig. 4.14. In general, the model predictions capture the spatial distribution of observed levels of pollution. *Model results reasonably well agree with measurements carried out in the European countries as well as in the countries of Central Asia and the Caucasus region. At the same time, more significant differences between the modelling results and measurements are obtained for South America, Africa, Asia, and Australia.* It is seen that about a half of model predictions for these regions are in reasonable agreement with measurements, while some of the measurements are significantly underestimated by the model exhibiting the tendency to underpredict levels of PCB-153 air concentrations in the Southern Hemisphere. This underprediction can be attributed to larger uncertainties in the emissions for these regions comparing to the Northern Hemisphere.

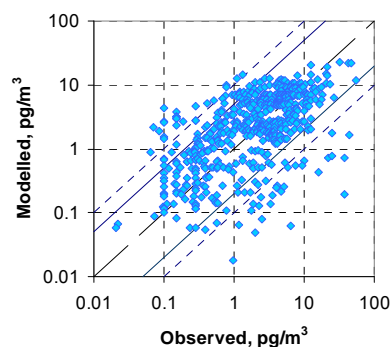


Fig. 4.13. Comparison of annual mean modelled PCB-153 air concentrations with measurements made in the period 1995-2013. Solid lines denote the area of agreement within a factor of 5, and dashed line – a factor of 10

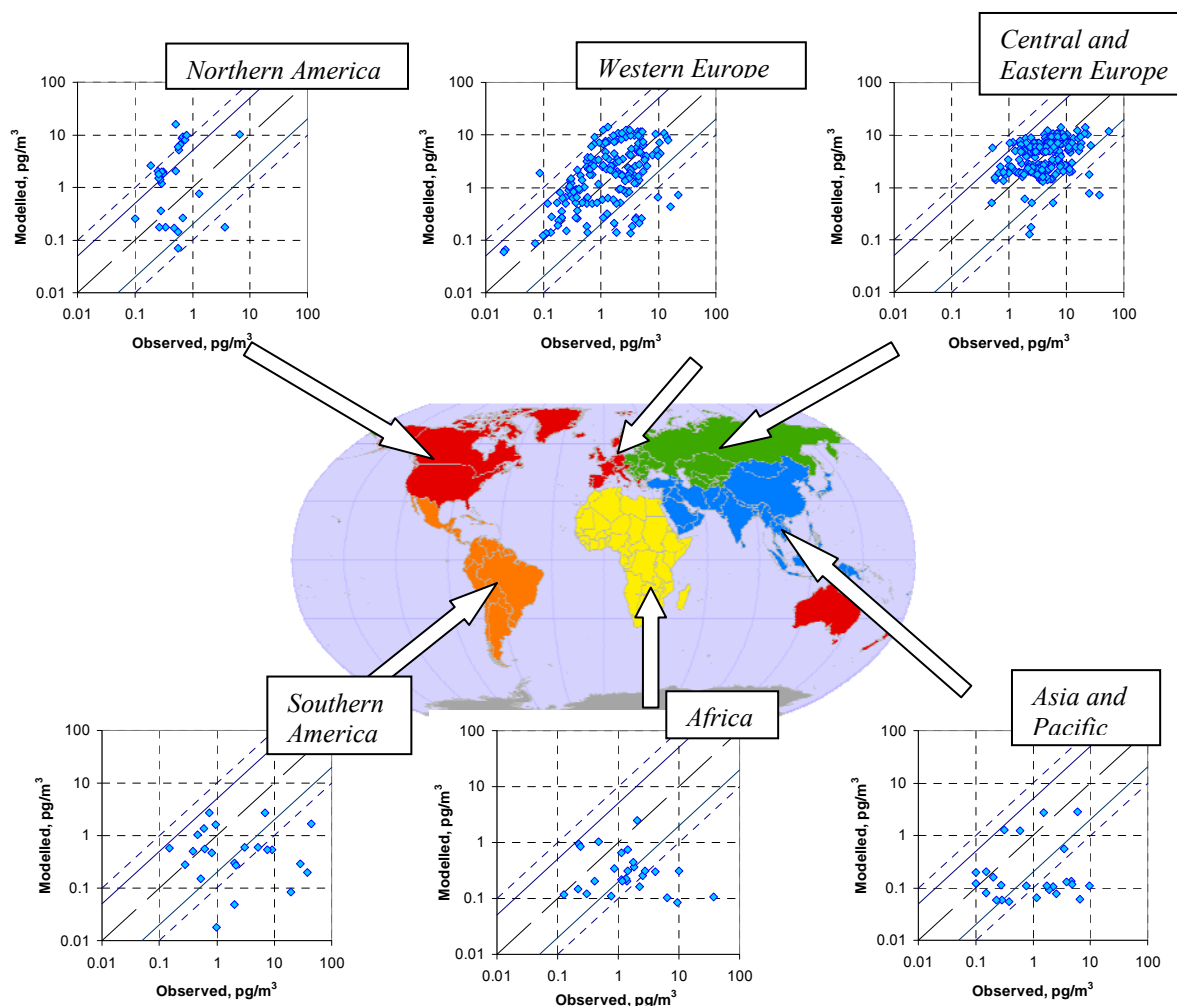


Fig. 4.14. Comparison of annual mean modelled PCB-153 air concentrations with measurements made in different regions in the period 2003-2013.

Pollution levels in the EMEP region

Model estimates of PCB-153 pollution levels in the EMEP region for 1990 and 2013 are presented in Fig. 4.15. *Modelling results indicate decrease of PCB-153 concentrations in the EMEP countries during this period by 80% on average.* In particular countries decline of pollution levels in the considered period varies from 90% to 70%. The largest change of air concentrations was obtained for Monaco (89%), the United Kingdom (89%), and Slovakia (86%), while the lowest was estimated for Belgium (72%).

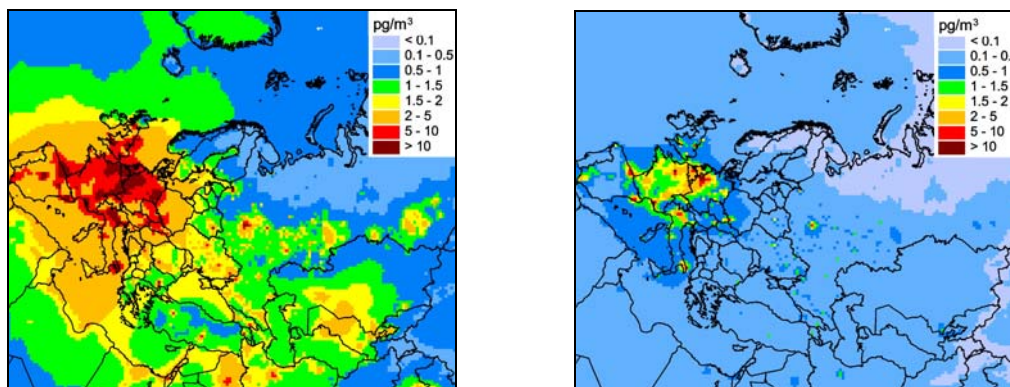


Fig. 4.15. Spatial distribution of calculated annual mean air concentrations of PCB-153 in the EMEP domain for 1990 (a) and 2013 (b), ng/m^3 .

Modelling results for the EMEP region were evaluated against measurements of the EMEP monitoring sites for 2013 (Fig. 4.16). Differences between the modelled annual mean air concentrations and measurements are mostly within a factor of two for all considered monitoring sites with the exception of FI36R. The lowest discrepancies (10%-25%) are found for Norwegian sites (NO90, NO42) and Swedish site SE12. For the other monitoring sites the differences are somewhat higher (30%-50%). Modelled PCB-153 air concentrations reasonably well captures spatial trend of concentrations (the correlation coefficient is 0.97). It is seen that in general the model tends to slightly underestimate levels of PCB-153 air concentrations in 2013.

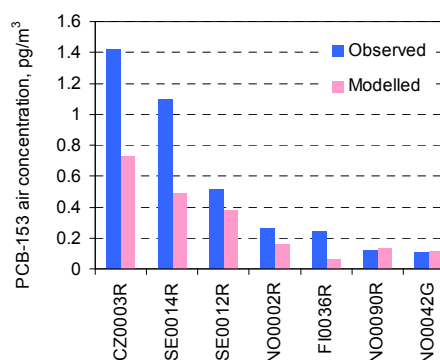


Fig. 4.16. Comparison of annual mean modelled PCB-153 air concentrations with measurements of the EMEP monitoring sites for 2013, pg/m^3 .

Transboundary transport within the EMEP region

Evaluation of PCB-153 transboundary transport within the EMEP region was carried out for 1990 and 2013. Results of model simulations were used to estimate long-term changes of transboundary fluxes and changes of contributions of main groups of emission sources to the pollution of particular EMEP countries. As seen from Fig 4.17 *the most significant contribution to PCB pollution in the EMEP countries is made by secondary emission sources (about 50%)*. The contribution of the other two groups of emission sources, namely, anthropogenic emissions of EMEP countries and non-EMEP emissions, is lower (about 40% and 10%, respectively).

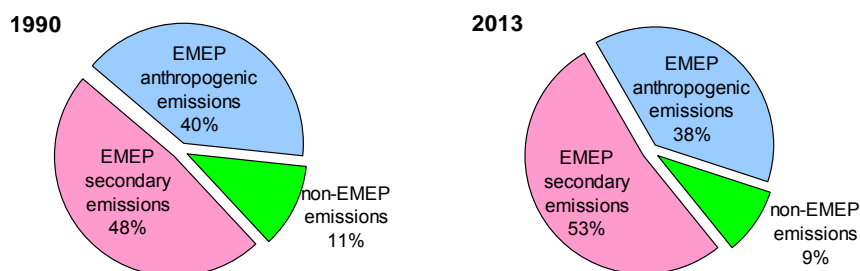


Fig. 4.17. Relative contributions of EMEP anthropogenic and secondary emission sources as well as non-EMEP emissions to total deposition of PCB-153 to the EMEP countries in 1990 and 2013.

Comparing relative contributions of these three groups of emissions to the pollution in 1990 and 2013, it is seen that contribution the secondary sources has increased by 5%, while contributions of EMEP anthropogenic emissions and non-EMEP emissions have decreased to similar extent.

Changes of PCB-153 deposition over the EMEP countries from 1990 to 2013 and contributions to the reduction of pollution due to decline of national emission sources, transboundary transport, secondary emissions, and non-EMEP emission sources are presented in the Fig. 4.18. *Model simulations show that annual deposition has decreased mostly due to declining of releases of PCB-153 from secondary emission sources.* For particular countries comparable contribution to the reduction of pollution can be made by the decline of anthropogenic emissions of the EMEP countries (for example, the United Kingdom, Germany, and Spain) and non-EMEP emissions (for example, Iceland, Ireland, Norway, and Sweden, Kirgizstan, Tajikistan).

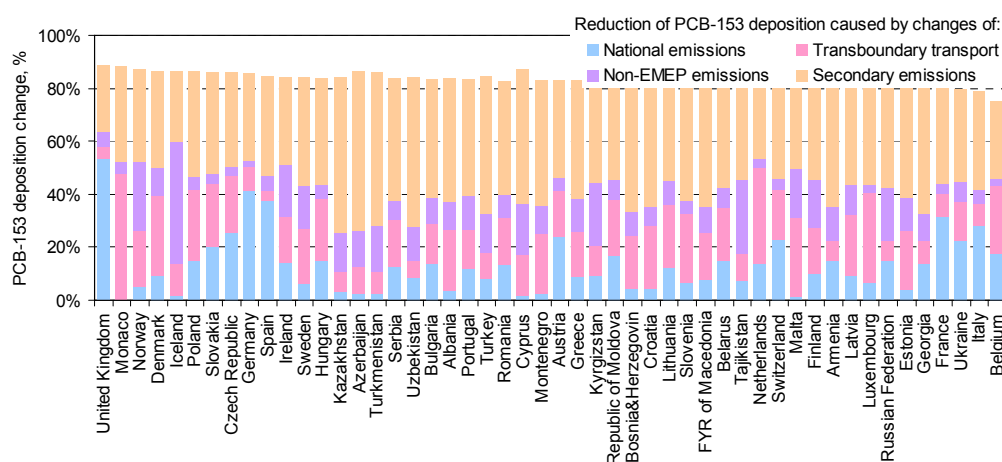


Fig. 4.18. Reduction of PCB-153 deposition in the EMEP countries caused by the changes of national, secondary and non-EMEP emissions as well as changes of transboundary transport between 1990 and 2013.

To further improve the estimates of PCB pollution levels in the EMEP region the information on congener composition of PCB emissions in the EMEP countries is needed. At present, officially reported emission data provide the total releases of PCBs without information on contribution of particular congeners. Specification of congener profiles for PCB emissions and inclusion of this information into national emission inventories of the EMEP countries is of importance for the evaluation of total PCB concentrations and deposition fluxes.

4.4. Hexachlorobenzene (HCB)

Hexachlorobenzene is highly persistent toxic chemical which was widely used as fungicide from the beginning of 1940s. HCB was also made as unintentional by-product in the production of various chlorinated compounds. After restrictions and banning of HCB usage as a fungicide/pesticide in many countries starting from 1970s its atmospheric concentrations significantly declined. Contemporary levels of HCB pollution are likely supported to significant extent by the secondary releases from the environmental media previously polluted due to agricultural or industrial activities.

Evaluation of HCB pollution of the EMEP region was performed using nested global and regional scale model simulations. Experimental scenario of global contemporary and historical HCB emissions was updated taking into account official emission data reported by the EMEP countries for the period 1990-

2012. Global dispersion of HCB and its accumulation in the environmental media were evaluated. Transboundary transport of pollution between the EMEP countries was assessed in course of regional model simulations for the EMEP domain with initial and boundary conditions obtained from the global modelling results. Modelled air concentrations of HCB were evaluated against measurements of the EMEP monitoring sites and observations collected in the UNEP SC GMP Data Warehouse.

Global scale levels of pollution

Estimates of spatial distribution of annual mean HCB concentrations in air obtained for 1990 and 2013 are illustrated in Fig. 4.19. As seen from the figure, levels of HCB air concentrations substantially decreased from 1990 to 2013.

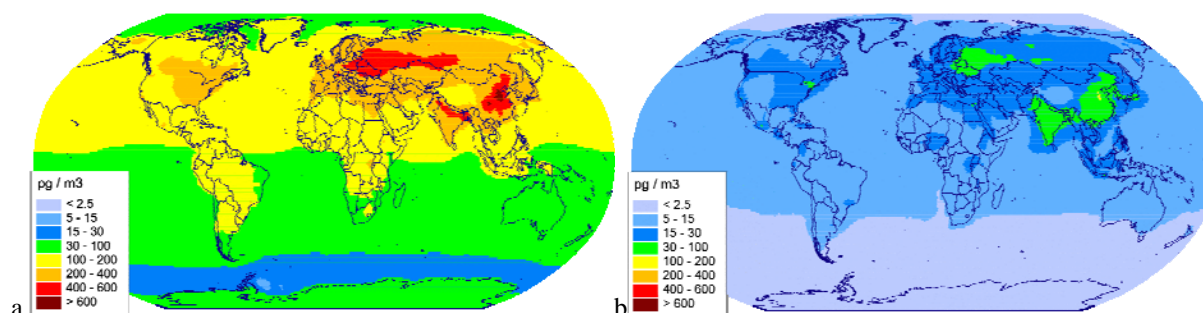


Fig. 4.19. Spatial distribution of global annual mean HCB air concentrations (pg/m^3) for 1990 (a) and 2013 (b) with spatial resolution $1^\circ \times 1^\circ$.

According to modelling results major sources of HCB pollution include Southeast Asia, Europe, Central Asia, and North America. In 2013 elevated air concentrations can be seen in Southeast Asia and Eastern Europe ($30\text{--}100 \text{ pg/m}^3$), while the other regions are characterized by relatively lower concentrations ($5\text{--}30 \text{ pg/m}^3$) which is consistent with observed levels.

To evaluate modelling results available measurements of HCB air concentrations carried out in the period 1990–2013 were used. Observed concentrations were obtained from the UNEP SC GMP Data Warehouse. Measurements performed in the remote, background, and rural areas using active and passive sampling were selected. Comparison of modelled and observed HCB annual mean air concentrations is shown in Fig. 4.20. Evaluation of model predictions shows that almost 90% of modelled concentrations agree with observed air concentrations within a factor of five and for about 45% of them the agreement is within a factor of two. In general, the model captures spatial distribution of observed HCB pollution levels.

Level of agreement between the modelled and measured HCB air concentrations in several regions of the world is shown in Fig. 4.21. It is seen that model predictions reasonably agree with measurements made in most of the regions. At the same time, the model slightly underestimates observed air concentrations

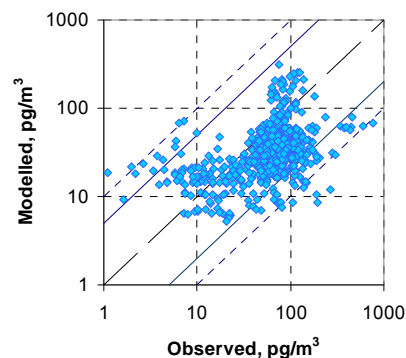


Fig. 4.20. Comparison of annual mean modelled HCB air concentrations with measurements made in the period 1990–2013. Solid lines denote the area of agreement within a factor of 5, and dashed line – a factor of 10

in North America, Western Europe, Central and Eastern Europe. The underestimation of observed HCB air concentrations is likely caused by the uncertainties of emission scenario applied in model simulations and requires further analysis in cooperation with experts in the field of POP emissions and measurements.

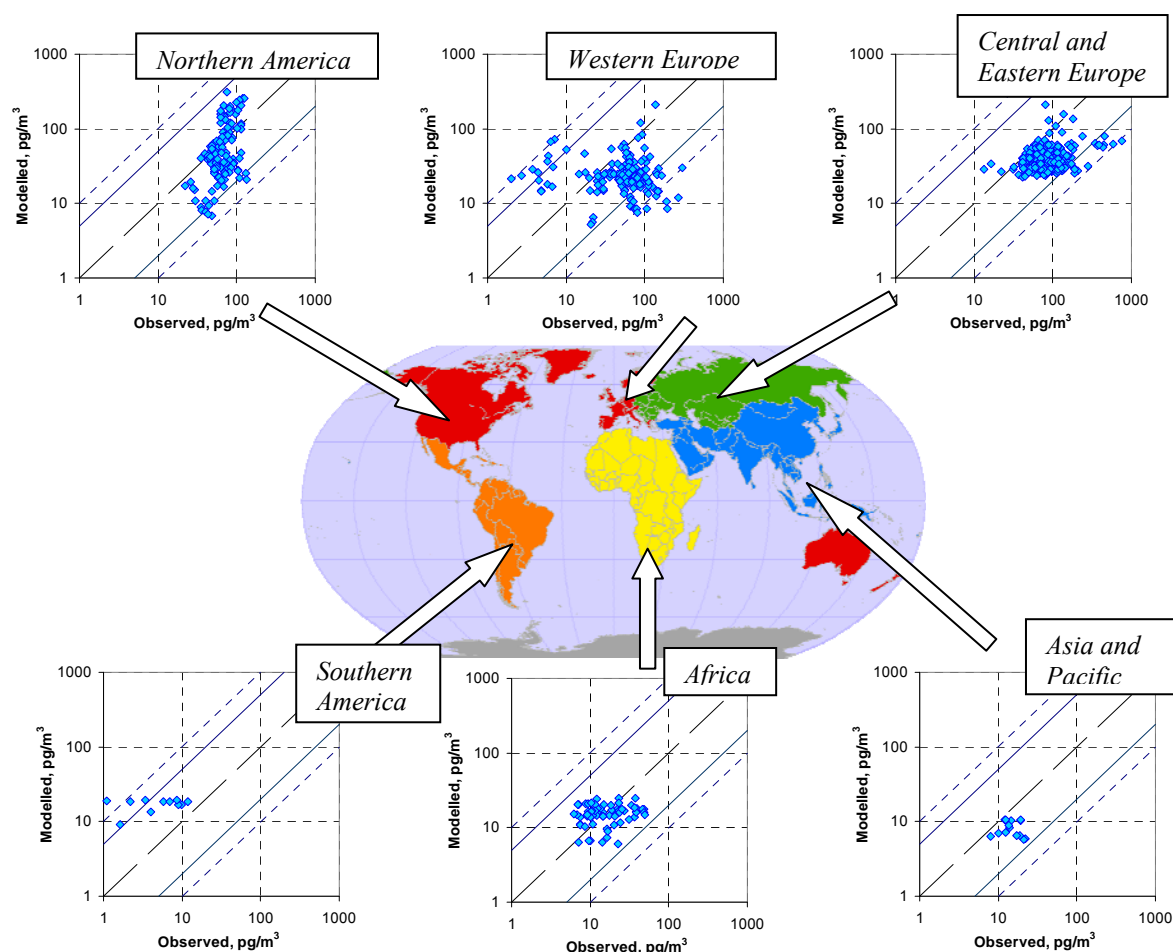


Fig. 4.21. Comparison of annual mean modelled HCB air concentrations with measurements made in different regions in the period 1990-2013.

Pollution levels in the EMEP region

Levels of HCB pollution in the EMEP region obtained by regional scale modelling for 1990 to 2013 are given in Fig. 4.22. *According to modelling results annual mean air concentrations of HCB in the EMEP countries decreased in the considered years by 90% on average.* As it is seen from the figure changes of pollution levels during this period were mostly homogeneous.

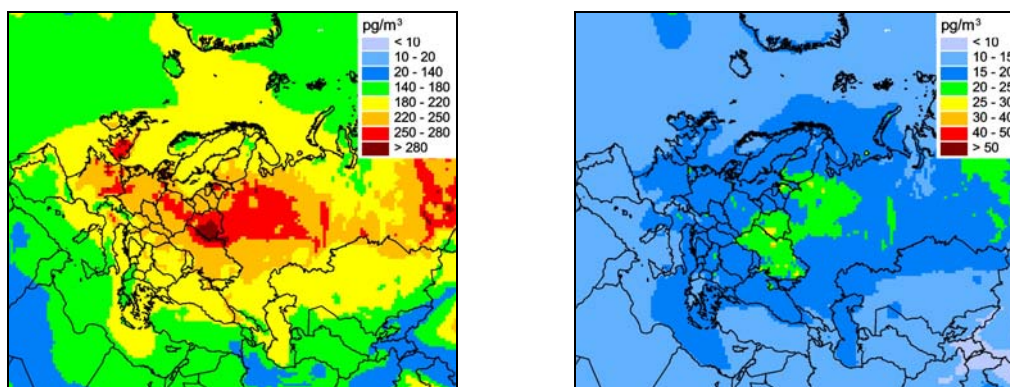


Fig. 4.22. Spatial distribution of calculated annual mean air concentrations of HCB in the EMEP domain for 1990 (a) and 2013 (b), ng/m^3 .

Modelling results for the EMEP region were evaluated against measurements of the EMEP monitoring sites for 2013 (Fig. 4.23). Deviations between the modelled annual mean HCB air concentrations and measurements are within a factor of two for five sites, namely, Swedish sites (SE12, SE14), German site DE1, Finnish site (FI36), and Norwegian site (NO90). For two sites, CZ3 and NO2, larger differences between modelled and measured values are obtained, which can be explained by incompleteness and uncertainties of anthropogenic and secondary emissions for these regions. Measurements of HCB concentrations for the CZ3 show significant inter-annual variations, especially in the periods 2003-2006 and 2010-2013 as shown in the Fig. 4.24. It is seen that the model does not capture variations of observed concentrations in these periods. At the same time, it reasonably well describes long-term changes of HCB air concentrations observed at this site. To further analyze the reasons of these differences application of fine resolution modelling and cooperation with national experts are of importance.

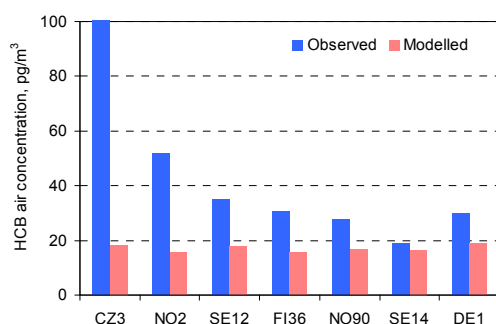


Fig. 4.23. Comparison of annual mean modelled HCB air concentrations with measurements of the EMEP monitoring sites for 2013, pg/m^3 .

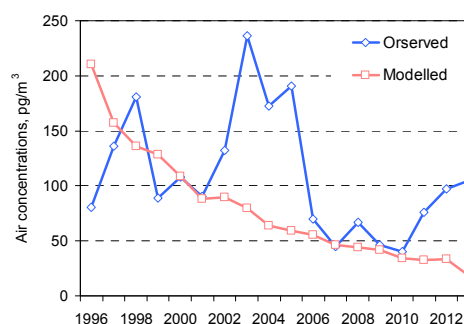


Fig. 4.24. Comparison of long-term changes of modelled and observed HCB annual mean air concentrations for the site CZ3, pg/m^3 .

Transboundary transport within the EMEP region

To evaluate changes of HCB transboundary transport within the EMEP region results of regional scale model simulations for 1990 and 2013 were analyzed. According to model estimates *secondary emission sources substantially contributed to HCB pollution in the EMEP countries. Both for 1990 and 2013 their contribution was estimated to about 70%.* Along with this significant contribution was obtained for non-EMEP emissions (about 27%). At the same time, the share of anthropogenic emission sources was comparatively low being about 1%. Despite marked declining of HCB pollution in the EMEP countries in the considered period changes of relative contributions of these three groups of emission sources were rather small.

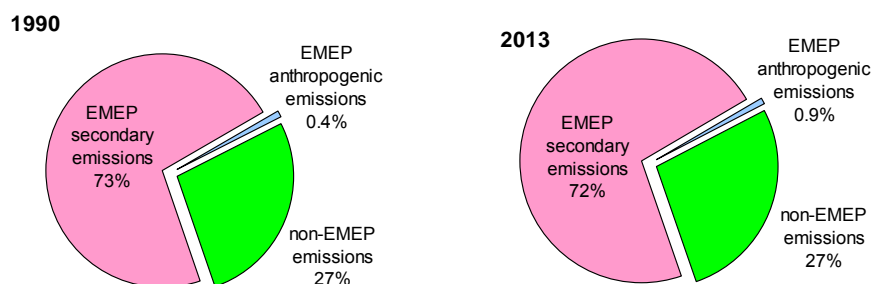


Fig. 4.25. Relative contributions of EMEP anthropogenic and secondary emission sources as well as non-EMEP emissions to total deposition of HCB to the EMEP countries in 1990 and 2013.

Decrease of HCB deposition in the EMEP countries from 1990 to 2013 along with changes of contributions of national emission sources, transboundary transport, secondary emissions, and non-EMEP emission sources are shown in the Fig. 4.26.

Evaluation of long-term changes of HCB pollution showed that *reduction of deposition in the EMEP countries was connected with the decreasing contributions of secondary and non-EMEP emission sources, while influence of changes of national emissions of the EMEP countries was less important.* However, due to uncertainties and incompleteness of anthropogenic HCB emissions reported by the EMEP countries their contributions can be underestimated. To improve assessment of HCB pollution of the EMEP region there is a need to refine national inventories of HCB emissions and the estimates of global contemporary and historical emissions.

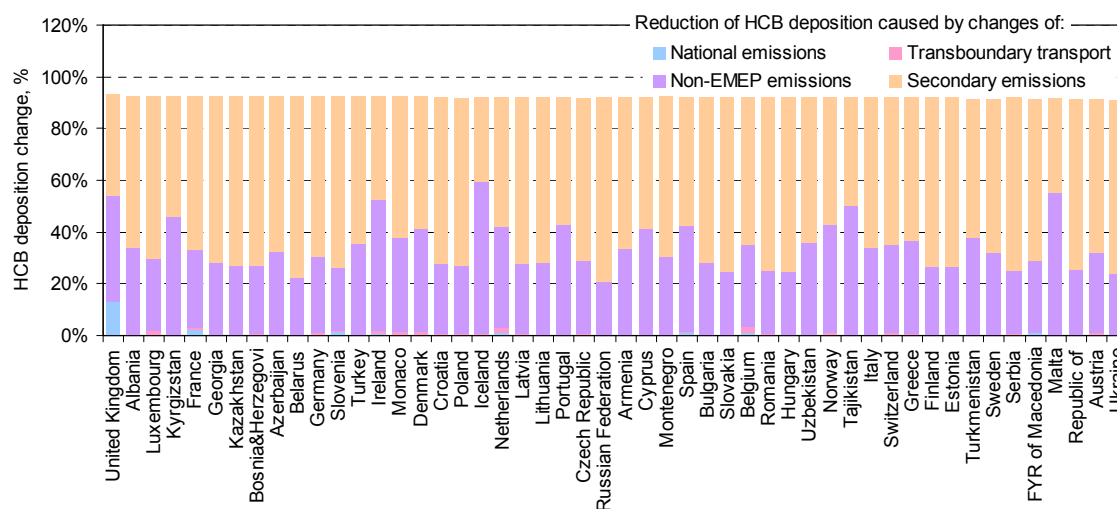


Fig. 4.26. Reduction of HCB deposition in the EMEP countries caused by the changes of national, secondary and non-EMEP emissions as well as changes of transboundary transport between 1990 and 2013.

5. FURTHER DEVELOPMENT OF GLEMOS MODELLING SYSTEM

This year MSC-E has continued development of the Global EMEP Multi-media Modelling System (GLEMOS) for HMs and POPs in several directions. Further work was carried out to prepare the GLEMOS for the transition of the EMEP operational modelling of HM and POP pollution to the new EMEP grid. It included testing and refinement of GLEMOS capabilities to perform multi-scale model simulations. In particular, modelling of global scale dispersion of POPs using geographic (latitude-longitude) projection was carried out to provide initial and boundary conditions for the regional scale modelling within the EMEP domain. Results of nesting of global and regional modelling are described in Chapter 4 of the report.

Ongoing developments include further work on application of the Weather Research and Forecasting (WRF) model in order to prepare meteorological input data for multi-scale model simulations of HM and POP pollution. Particularly, the recent version of the WRF model is being adapted to perform the pre-processing of required meteorological parameters for the GLEMOS model and to provide consistent set of meteorological data for global and regional model calculations in the new EMEP grid. Different parameterizations of the WRF model were tested to provide spatially and temporally resolved meteorological data consistent with available measurements.

Modelling of gas-particle partitioning of POPs and their degradation in the particle-bound form requires information on distribution of atmospheric reactants and aerosols as well as information on chemical composition of aerosol particles. At the previous stages of the work model parameterizations of gas-particle partitioning and degradation processes for POPs were elaborated. There is ongoing work on the incorporation of gridded data on aerosols and atmospheric reactants based on the output of chemistry transport models. In particular, global data on concentrations of atmospheric reactants and aerosol particles with high temporal resolution obtained by the MOZART model [Emmos *et al.*, 2010] are being applied to describe interaction of POPs with aerosols and degradation in the atmosphere. On further stages it is planned to develop simplified chemical modules for the GLEMOS modelling system, which can provide necessary information on aerosols and reactants in course of modelling of POP transport and fate.

Development and refinement of the GLEMOS multimedia modelling approach for POPs is being continued. Current activities in this direction include collection of measurements obtained in various campaigns aimed at parallel monitoring of POP concentrations in the atmosphere and water and soil compartments. Particularly, data on POP content in soil compartment have been collected from available publications in literature and projects. Besides, results of monitoring of PCBs, HCB, and other POPs performed in several cruises in the Atlantic, Pacific, Indian, and Arctic oceans [Huang *et al.*, 2014; Zhang and Lohmann, 2010; Galbán-Malagón *et al.*, 2013a,b,c; Zhang *et al.*, 2012; and etc.] are being compiled that provide valuable information for testing and improving of multi-media modelling of POP transport and fate.

Current and previously made developments of multi-scale multi-media modelling approach, implemented in the framework of the GLEMOS modelling system, are documented in the series of MSC-E Technical reports [Shatalov *et al.*, 2013, 2014, 2015].

6. MODELLING STUDIES OF CLIMATE CHANGE INFLUENCE ON POP POLLUTION

Evaluation of interaction between the climate variability and POP pollution exhibits a challenging task due to the complexity of POP cycling in the environment. The summary of information regarding potential effects of climate variations on POP pollution was presented in the reports prepared by TF HTAP [UN ECE, 2011] and UNEP/AMAP [UNEP/AMAP, 2011]. Particularly, these reports conclude that climate change has the potential to affect all pathways of POPs in the environment and may increase vulnerability for health impacts. Main factors, directly related to climate change and influencing POP environmental fate, include increasing temperature, altering of atmospheric and oceanic transport pathways and their intensity, melting of sea and land ice, and increasing frequency and strength of extreme events. Due to the complex nature of these changes multimedia POP transport and fate models can be important tools for the evaluation of responses of future pollution levels to the climate variability. Particularly, models allow accounting for interaction of various processes governing POP fate, examining sensitivity of POP transport and levels to variations of climate parameters, and performing modelling experiments based on the climate change scenarios.

Investigation of potential effects of climate variability on POP pollution levels was performed by MSC-E at previous stages of the work [Gusev *et al.*, 2011; Shatalov *et al.*, 2012]. These studies were focused on the analysis of seasonal variations of POP air concentrations in Europe and their relationship to changes of different meteorological and environmental factors. Particularly, it is shown that such factors as temperature, precipitation amount, wind speed and direction, and vegetation cover, can in most cases explain 90% – 95% of seasonal variability of POP air concentrations indicating significant sensitivity of POP pollution levels to variations of meteorological and environmental parameters.

A number of modelling experiments aimed at evaluation of the influence of projected climate changes on POP pollution has been carried out recently. Analysis of potential effects of climate change on global distribution and bioaccumulation of POPs using multimedia POP model simulations was made by Gouin *et al.* [2013]. Evaluation of responses was carried out using chemical partitioning space approach to systematically determine the most sensitive combination of physical-chemical properties of POPs to the considered scenarios of climate changes. Based on the multimedia modelling results it is concluded that the effect of considered climate changes on POP transport, fate, and bioaccumulation in aquatic food chain is within a factor of two comparing to the baseline results.

The study of Wöhrnschimmel *et al.* [2014] applied the global multi-media model BETR to examine potential impacts of climate change on both anthropogenic emissions and global transport and fate of two typical persistent organic pollutants, namely, α -HCH and PCB-153. *Results of model simulations indicate that projected climate changes can increase POP contamination in the Arctic region by the end of 21st century up to a factor of 2 and 4 for air and water concentrations, respectively.* Substantial contribution to this increase can be made by climate-induced changes of temperature, reduction of ice cover, and changes of emission patterns, for example, enhanced usage of industrial chemicals in ice-free Arctic, and intensified agricultural activities.

POP transport to and from the Arctic region and its correlation with the Arctic and North Atlantic Oscillations (AO and NAO) under the influence of changing climate were studied by Octaviani *et al.* [2015]. Import and export of selected legacy POPs (DDT, PCBs) to and from the Arctic were quantified using multi-compartment chemistry transport model MPI-MCTM. Model simulations with present-day and future climate conditions indicate that changes of net meridional flux patterns depend on the substance behaviour and fate in the environment. Particularly, net export of PCB153 out of the Arctic

is projected to increase and net meridional flux is expected to become independent of AO and NAO. For DDT, decreasing trend of net import to the Arctic is projected to reverse to increasing trend.

The influence of projected climate changes on POP pollution of the European marginal seas was evaluated in the studies of *Kong et al.* [2014] and *O'Driscoll et al.* [2014]. The analysis of climate-induced changes in the Baltic Sea region was performed with the POPCYCLING-Baltic multimedia chemical fate model for hypothetical POP-like chemicals using wide range of physical-chemical properties. It is shown that POP concentrations in the Baltic Sea environment can change up to a factor of 3 from the baseline scenario. Among the four studied climate parameters, namely, temperature, wind speed, precipitation, and particulate organic carbon the most significant contribution was obtained for changes of temperature followed by particulate organic carbon. *O'Driscoll et al.* [2014] conducted a high-resolution modelling of potential impacts of climate variability on transport and fate of two legacy POPs in the North Sea in the 21st century. It is concluded that considered climate changes can lead to generally small influence on the simulated fate and transport of selected POPs in coastal and shelf areas of the sea. At the same time, the increased number and magnitude of storms in the 21st century can result in the events of substantial re-mobilization of POPs from sediments to seawater and to the atmosphere.

One of important issues mentioned in these studies is the level of uncertainties of the obtained projections of future POP pollution levels. Simulated responses to climatic changes comprise uncertainties of climate models and of future emission scenarios of greenhouse gases. Besides, significant uncertainties are associated with projections of future POP emissions, physical-chemical properties of POPs and their behaviour in surface media (e.g. degradation rates). Bearing this in mind the uncertainties in model predictions for the baseline scenario are likely to be substantially larger than the level of obtained climate-induced changes of future pollution levels. Thus further work is required to refine the information on POP fate in media, hosting the most part of POPs released into the environment, and estimates of future emissions of POPs.

Another important aspect related to the evaluation of future changes of pollution levels and exposure is taking into account indirect effects of climate changes. Particularly, substantial influence may also have changes of land use and vegetation cover, organic carbon cycling, species range and abundance, and anthropogenic emissions of POPs [*Gouin et al.*, 2013; *Wöhrnschimmel et al.*, 2014; *Kong et al.*, 2014]. However their effect in most of the studies of future POP pollution levels is not thoroughly evaluated and further investigations in this direction are needed.

7. COOPERATION AND EXCHANGE WITH INFORMATION

7.1. Task Force on Measurements and Modelling (TFMM)

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

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

The overview of the results of trend analysis for POP pollution for the period from 1990 to 2012 is presented in Chapter 3 of this report. These results were reported at the annual TFMM meeting in Krakow, Poland (May 2015). A number of information related to long-term HM and POP pollution changes were proposed to be highlighted in the CLRTAP Assessment Report 2016. In particular, it includes trend characteristics for modelled and observed levels at the EMEP monitoring stations, reduction rates and total reductions of HM and POP levels in the EMEP countries, analysis of factors affecting trends (changes of anthropogenic and secondary emissions, meteorological variability, boundary conditions, transboundary transport). Besides, the effect of changes of source categories of emissions and large point sources could also be overviewed in the Report.

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

7.2. Working Group on Effects (WGE)

This year MSC-E has continued co-operation with the Working Group on Effects (WGE) with regard to evaluation of POP pollution. In particular, the Centre has contributed to the WGE Assessment Report on Trends with information on temporal trends of POP contamination in the EMEP countries. This information includes analysis of long-term changes in levels of POP air concentrations in the EMEP domain from 1990 to 2012 (Section 3).

Calculated levels of POP concentrations in various environmental media and depositions on environmental ecosystems can be of use for evaluation of harmful effects for human health and environment. For a number of contaminants threshold values of concentrations in the environmental media are established by environmental-protection authorities of particular countries. For example, three threshold levels (target value of 1 ng/m³, upper assessment threshold of 0.6 ng/m³, and lower assessment threshold of 0.4 ng/m³) are established for B[a]P considered as an indicator substance for PAHs. 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). From this viewpoint, the comparison of contamination levels of POPs with corresponding thresholds can be of interest for POP risk assessment. Data on evaluated concentration/deposition levels for such comparison (see Section 4 of the report) can be provided by request.

The consideration of the information on trends that can be of use for evaluation of the harmful effects on human health and ecosystems is considered above in Section 3.5 of the Report. It includes population-based evaluation of air concentration trends and the analysis of trends of deposition of POPs to various land-use types. The latter piece of information can be important for the evaluation of harmful effects of POPs in the EMEP countries to the environment. In future, the information on deposition flux to particular environmental ecosystems can be generated for selected POPs included in the EMEP modelling activities (namely, PAHs, PCDD/Fs, HCB, and PCBs) on request.

7.3. UNEP Stockholm Convention

The recent EMEP Steering Body session has recognized the need to enhance cooperation with the UNEP Stockholm Convention (SC) in the field of assessment of environmental pollution by POPs [ECE/EB.AIR/GE.1/2014/2]. Taking up this call MSC-E performed studies of global scale transport of POPs with application of the data on emissions and measurements compiled within the Stockholm Convention. In particular, pilot modelling of PCDD/F transport and fate was carried out using experimental emission scenario based on the global emission inventory of the UNEP SC.

Model simulations took into account releases of PCDD/Fs to different media, namely, to the atmosphere and soil compartments. Predictions of the model reasonably reproduced observed levels of PCDD/F air concentrations in the European region, North America and Eastern Asia [Gusev et al., 2014b] (Fig. 7.1). Results of this study were

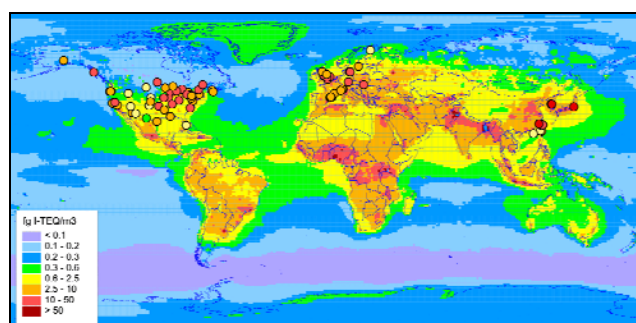


Fig. 7.1. Spatial distribution of modelled annual mean PCDD/F air concentrations (fg TEQ/m³) in comparison with measurements made in rural and remote regions (locations of measurements are illustrated as circles)

presented at the 34th International Symposium on Halogenated Persistent Organic Pollutants Conference (Madrid, September 2014) during its special session devoted to the 10th anniversary of the Stockholm Convention [Gusev *et al.*, 2014a].

Further activity with regard to cooperation and exchange of scientific knowledge with the SC was the contribution of the UN ECE LRTAP Convention to the joint Conference of Basel, Rotterdam, and Stockholm Conventions, held in Geneva in May 2015. EMEP Centres were involved in the work of the Science Fair meeting organized during the Conference, where they outlined recent EMEP activities in the field of modelling and monitoring of POP pollution levels.

In particular, MSC-E presented outcome of integrative analysis of contemporary POP pollution levels based on the modelling results and measurements collected in the SC GMP (Global Monitoring Programme) Data Warehouse. Results of model simulations permitted to estimate long-term variations of POP air concentrations in different regions of the world (Fig. 7.2). For instance, PCB-153 air concentrations declined by 70-90% during the two recent decades, which was consistent with time-series of observed air concentrations. Largest decrease of pollution levels was noted for North America, while the lowest decrease was obtained for the countries of Central and Eastern Europe. The analysis of obtained results indicated the regions with poor agreement between the model predictions and measurements (e.g. Africa), which required further refinement of information on emissions and their temporal development. In this respect further strengthening of cooperation and information exchange with international organizations, e.g. SC, AMAP, HELCOM, OSPAR, working on the assessment of POP pollution, is of importance.

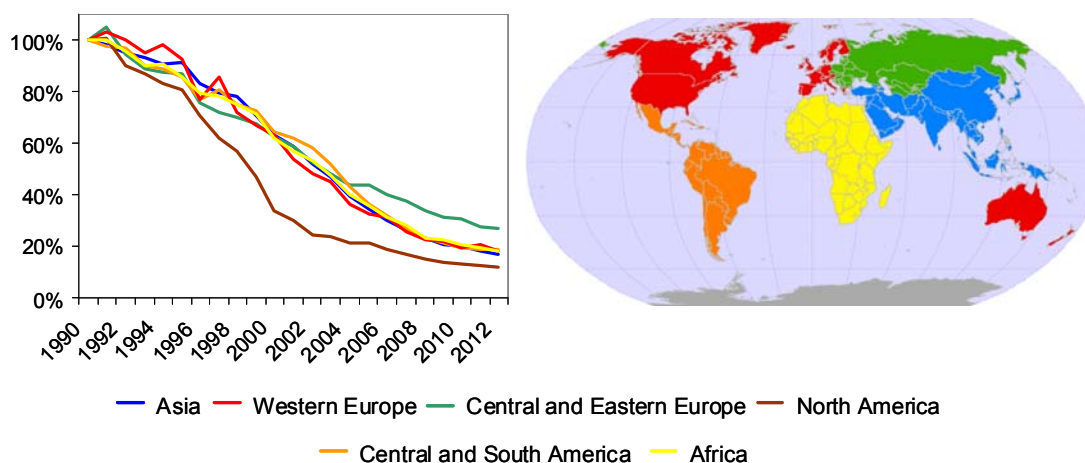


Fig. 7.2. Relative changes of annual mean PCB-153 air concentrations in different regions of the world from 1990 to 2012.

Along with these activities MSC-E was invited to take part in the preparation of the Global Monitoring Report of the Stockholm Convention. The contribution of the Centre will include model assessment of global transport and pollution levels of selected POPs and comparison of model predictions against the measurements of the GMP Data Warehouse. Brief information on these results can be seen in the Chapter 1 of this report.

7.4. Helsinki Commission

Information on airborne pollution load to 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 and is based on the long-term EMEP/HELCOM contract.

Assessment of PCDD/F pollution of the Baltic Sea comprised the information on long-term trends in deposition for the period 1990-2012 and evaluation of contributions of anthropogenic and secondary PCDD/F emissions. This year, following the requirements of the HELCOM LOAD group, the evaluation of atmospheric deposition was performed for nine sub-basins of the Baltic Sea. Results of this assessment are published in the joint annual report of the EMEP Centres [Valiyaveetil *et al.*, 2014]. 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>).

Assessment of PCDD/F contamination indicated that annual emissions in the countries surrounding the Baltic Sea dropped by 41% from 1990 to 2012 (Fig. 7.3). The largest contribution to PCDD/F emissions of the HELCOM countries in 2012 was made by Russia and Poland. Atmospheric deposition of dioxins and furans over the surface of the Baltic Sea decreased by 60% in that period. More significant changes (> 70%) took place in the western and the southern parts of the Baltic Sea, whereas decrease of deposition in the eastern part of the Sea was lower (by 30-60%).

Spatial distribution of PCDD/F deposition to the Baltic Sea in 2012 is presented in Fig. 7.4a. Relatively higher pollution levels were characteristic of the western sub-basins (the Kattegat and the Western Baltic). HELCOM countries contributed about 40% to the pollution of the Baltic Sea by PCDD/Fs. Among them the most significant contributions were made by Russia and Poland (Fig. 7.4b).

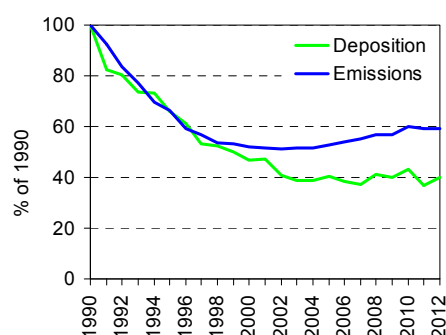


Fig. 7.3. Long-term changes of PCDD/F annual emissions of the HELCOM countries and deposition to the Baltic Sea in the period 1990-2012 (% of 1990).

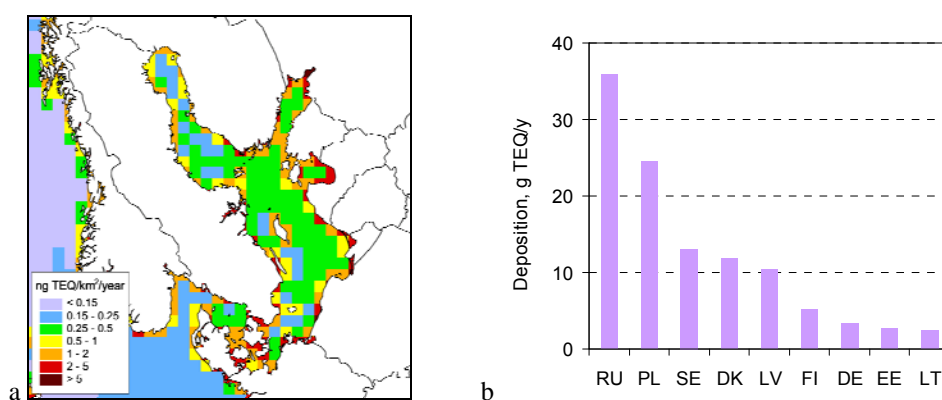


Fig. 7.4. Spatial distribution of PCDD/F deposition over the Baltic Sea, ng TEQ/km²/y (a) and contributions (in g TEQ/y) of HELCOM countries to annual total PCDD/F deposition to the Baltic Sea for 2012 (b)

8. MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

In 2016 EMEP Centres, MSC-E and CCC, will continue the work on the assessment of pollution levels of POPs in the EMEP region to support the EMEP countries with information required for the implementation of the POP Protocol. Main challenges in the assessment of POP pollution that need to be addressed in further research activities, in accordance with the Key Messages of the CLRTAP Assessment Report, are summarized below. Proposals for the EMEP workplan for the assessment of POP pollution in 2016-2017 are given in Annex B.

- Information on long-term changes of POP pollution is of importance for understanding effectiveness of environmental policy in the EMEP countries and, in particular, for the implementation of the POP Protocol. On-going evaluation of long-term trends of POP pollution during the two recent decades will be completed to support the preparation of CLRTAP Assessment Report. Provided information will comprise characteristics of pollution trends both in the EMEP region as a whole and in particular EMEP countries as well as factors affecting the long-term changes of pollution levels.
- Further development and testing of modelling approach for POPs require better knowledge of their content in relevant compartments of the environment and with global geographical coverage. In order to improve estimates of POP cycling between different ecosystem compartments parameterizations of air-water, air-soil, and air-vegetation exchange in GLEMOS will be further developed. Besides, analysis and refinement of model parameterizations of processes governing POP fate in the terrestrial and aquatic environmental compartments will be carried out to link transport and fate of POPs to the exposure and improve understanding of bioaccumulation/biomagnification in food chains.
- Completeness and consistency of national inventories of POP emissions of the EMEP countries, especially in the EECCA region, continue to be important issues with regard to the assessment of POP pollution in the EMEP region. Along with this, refinement of understanding of emissions to the atmosphere and other environmental compartments is of importance for further progress in the evaluation of POP transport and fate. Alternative methods for emission inventories based on a combination of monitoring and modelling at regional and local scales and inverse modelling techniques will be applied for the analysis of discrepancies between observed and modelled pollution levels.
- Assessment of POP pollution in the EMEP region with complementary usage of monitoring and modelling requires further improvement of spatial distribution of EMEP monitoring sites especially in South Europe and the EECCA region. Collection and analysis of data from national monitoring of POP concentrations in the atmosphere, performed by active and passive sampling, and measurements of POPs in surface media are needed to further improve estimates of POP fate in the environment. Strengthening cooperation with relevant activities of international organizations (e.g. UNEP SC, AMAP, HELCOM) to collect global scale measurements of POP pollution levels is highly appreciated.
- Cooperation with WGE with regard to evaluation of POP pollution of the EMEP countries will be continued. Ecosystem-dependent deposition of POPs in the EMEP countries will be generated to support evaluation of effects of pollution on human health and environment. Similar information will also be produced for the new EMEP grid. In addition, background pollution levels in selected cities of the EMEP countries will be estimated.

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METHODOLOGY OF TREND ANALYSIS OF AIR QUALITY DATA

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

A1. General description of the methodology

Typical measurement data on air quality for sufficiently long period (1990 – 2010) is exemplified in Fig. A.1, (upper part) where air concentrations of B[a]P at EMEP site CZ3 is shown.

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

The application of the above decomposition to the dynamics of monthly means of B[a]P air concentrations is illustrated in Fig. A.1.

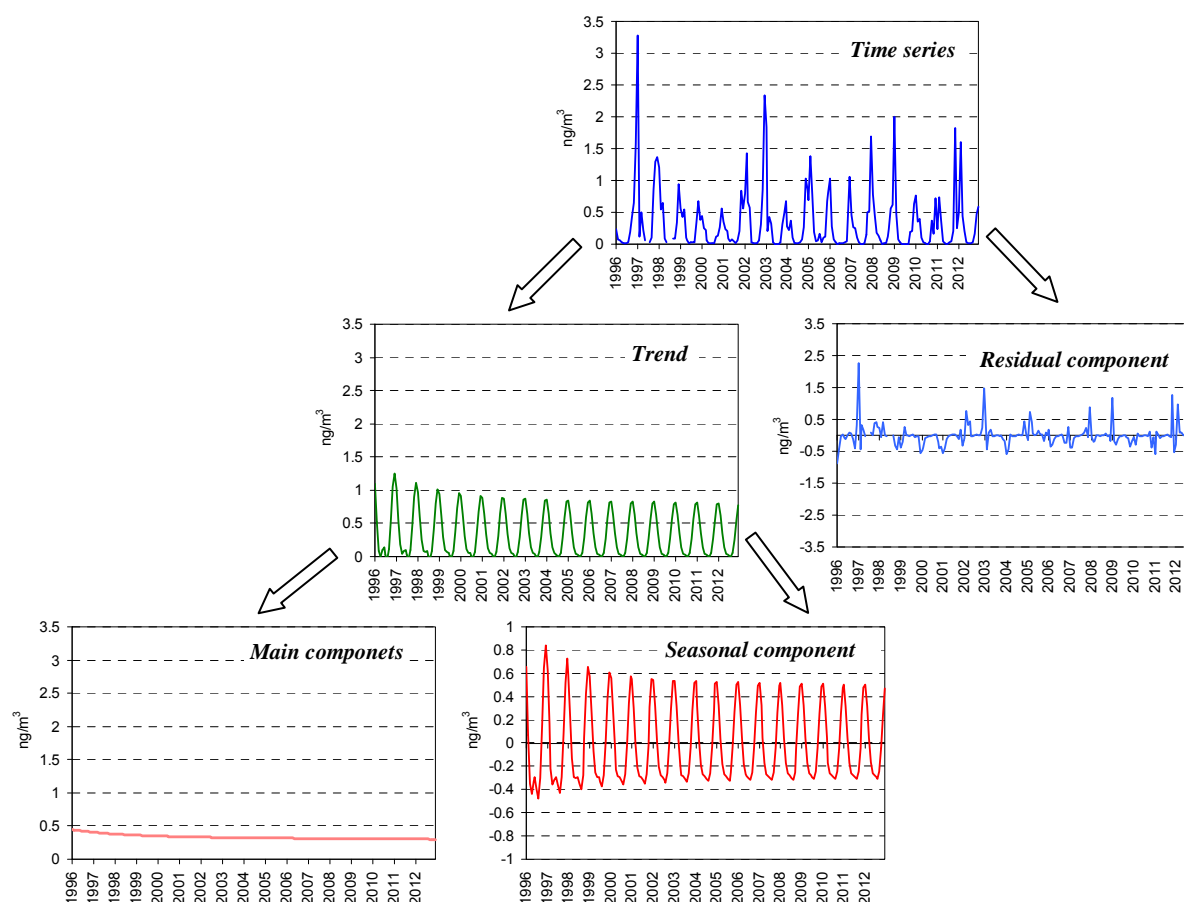


Fig. A.1. Decomposition of time series to trend and residual component exemplified by measurements of B[a]P air concentrations at EMEP site CZ3

For POPs (as well as for HMs) main component normally occurs to be non-linear (see lower left plot in Fig. A.1). Thus, for the description of the trends for these pollutants multi-exponential approach with harmonic components is applied. In fact, the approach is based on the combination of exponential trends with standard description of periodic time series (see, e.g. the monograph by T. W. Andersson, Statistical analysis of time series, Wiley-Interscience, 1994).

Analytically, the decomposition is described as follows.

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

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

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

The following trend parameters are considered for annual resolution:

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

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

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

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

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

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

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

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

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

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

The above described form of trends based on the supposition that characteristic times are constant for each individual component within the entire period of consideration. However, for some types of dependence of initial time series this supposition can be violated. For such series (see, e. g. Fig. 3.18 above), the form of trends can be modified by the assumption that time dependence parameters $\lambda_i = 1 / \tau_i$ can slowly depend on time. For the description of such trends polynomial approximation of first or second order can be used.

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

A2. Comparison of measured and calculated trends

Trend analysis can be applied for the analysis of agreement between measurement data and calculation results from the viewpoint of long-term tendencies. For this it is reasonable to compare trends rather than initial time series.

The approach is exemplified by the analysis of the agreement between measured and modelled trends of PCB-153 air concentrations at site FI96 (Pallas) (Fig. A.2).

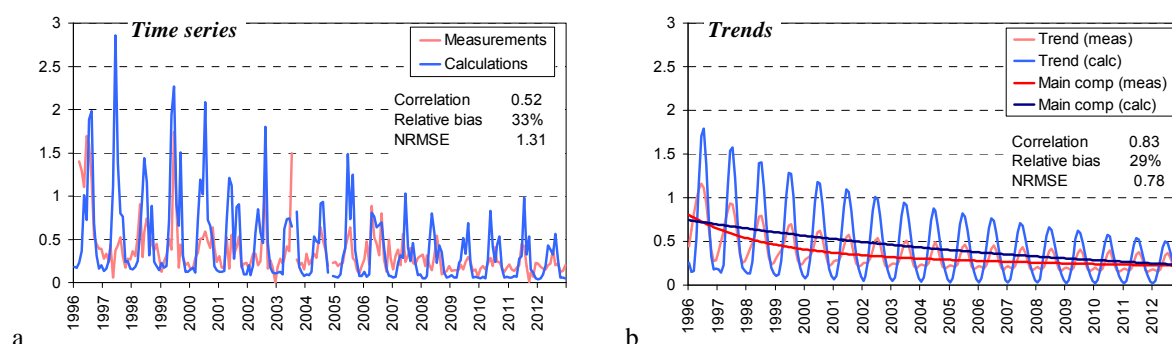


Fig. A.2. Comparison of calculated and measured values of PCB-153 air concentrations at FI96: (a) by initial values, (b) by trend.

Main trend parameters are presented in Table A.1.

Table A.1. Main trend parameters of the comparison of trends of PCB-153 air concentrations at FI96 site

	Total reduction (from 1996 to 2012)	Average annual reduction	Seasonality	Residual component
Measurements	72%	7.4%	50%	54%
Calculations	69%	6.6%	100%	57%

It is seen that the model well reproduces total reduction of contamination (and, consequently, average annual reduction) at FI96 though the dynamics of the reduction is slightly different for measurements and calculations. The relative values of residual component (normalized by the values of main component) are rather close for measurements and calculations. However, seasonal variations normalized by main component (seasonality) differ two times.

The disagreement between measured and calculation trends at particular sites can be conditioned by uncertainties in model parameterization of environmental processes for this pollutant (mainly degradation and inter-media exchange), absence of information on seasonal variations of emissions, and, possibly, by uncertainties in measurement data.

Such analysis was performed for all sites with sufficiently long measurement period (CZ3, Kosetice, FI96, Pallas, IS91, Storhofdi, NO42, Zeppelin, SE2/14, Råö, and SE12, Aspvreten). To evaluate the agreement between measured and calculated trends at these sites, the statistical parameters (correlation coefficient, relative bias and normalized root square error NRMSE) of the comparison between measured and calculated B[a]P air concentrations calculated on the basis of initial values and of trends was performed. The results of the comparison for B[a]P are presented in Fig. A.3.

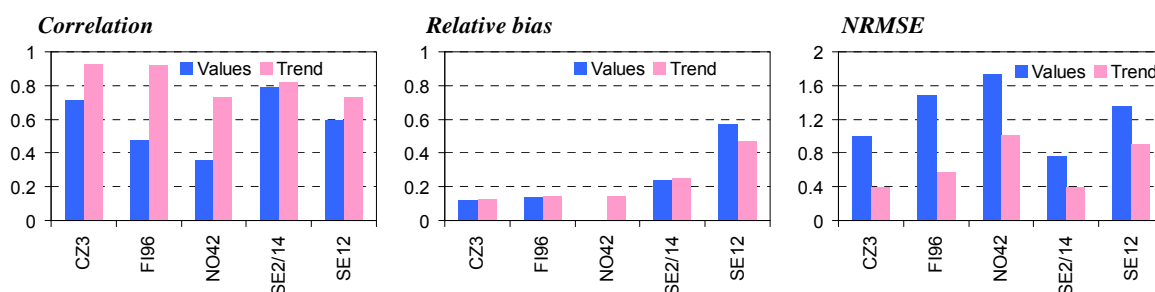


Fig. A.3. Statistical parameters of the agreement between measured and calculated values of B[a]P air concentrations calculated on the basis of initial values and of trends.

It can be seen that statistical parameters calculated on the basis of trends are always better than those calculated on the basis of initial values. This shows that **the model well enough reproduces long-term tendencies of air pollution** by B[a]P. Further, the agreement between trends of measurements and calculations is well enough at all sites except for NO42 and SE12.

Similar situation takes place for PCB-153 (Fig. A.4).

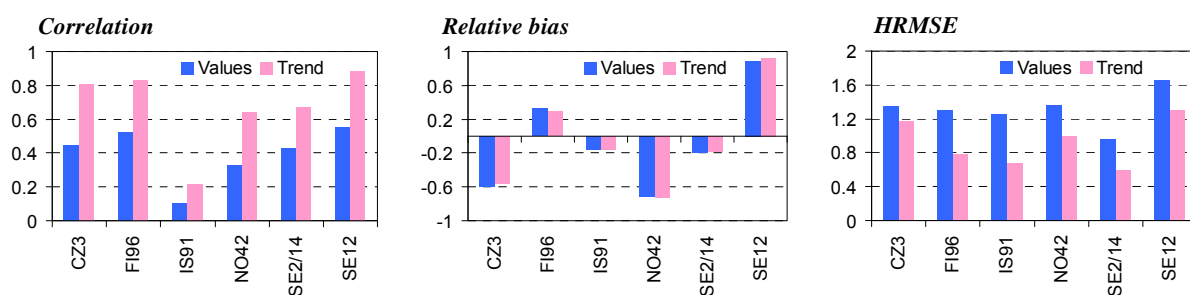


Fig. A.4. Statistical parameters of the agreement between measured and calculated values of PCB-153 air concentrations calculated on the basis of initial values and of trends.

High relative bias is found for sites NO42 and SE12. Site IS91 is characterized by low values of correlation coefficient. Further analysis of the disagreements between modelled and calculated trends by experts in modelling, emissions and monitoring is desirable.

Proposals for 2016-2017 Work-plan

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

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

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