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Persistent Organic Pollutants: Assessment of Transboundary Pollution on Regional and Global Scales

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Persistent Organic Pollutants: assessment of transboundary pollution on regional and global scales

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EXECUTIVE SUMMARY

Persistent Organic Pollutants (POPs) are semi-volatile toxic substances, characterized by significant potential to long-range transport, accumulation in the environmental media, and adverse effects on human health and wildlife. In spite of efforts to reduce their use and unintentional releases, they are still measured in the environmental compartments, posing risk to humans as well as terrestrial and aquatic ecosystems. POPs are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) since 1998, when the Protocol on Persistent Organic Pollutants came into force. Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) provides the Convention with the information on deposition and transboundary transport of selected POPs (PAHs, PCDD/Fs, PCBs and HCB) within the geographical scope of EMEP.

This report outlines recent activities of the EMEP Centres in the field of POP pollution assessment, performed in accordance with the 2016-2017 Workplan for the implementation of the Convention (ECE/EB.AIR/133/Add.1). It provides information on the pollution of the EMEP region by 4 PAHs (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)), PCDD/Fs, HCB, and PCBs in 2014, based on reported anthropogenic emissions, modelling results, and measurements. Main emphasis in the report is given to the evaluation of PAH pollution levels in the EMEP countries, including estimation of B[a]P air concentrations in urban areas. Along with this, particular attention is paid to the co-operation and exchange of information between Meteorological Synthesizing Centre East (MSC-E) and subsidiary bodies to the Convention, as well as dissemination of the outcome of pollution assessment to national and international organizations.

Total anthropogenic emissions of selected POPs within the EMEP region in 2014, reported by the Centre on Emission Inventories and Projections (CEIP), are lower than those in 2013 for PCDD/Fs, PCBs, HCB, and higher for the 4 PAHs. However, uncertainties of the reported POP emission inventories remain significant. In order to improve the quality of emission data, the Executive Body of CLRTAP has initiated an activity, aimed at refinement of POP emission inventories in the EMEP region. Following this initiative, the key parameters of POP emission, which affect model assessment results, have been analysed and ranked in terms of their priority. It has been noted that completeness of gridded emission data has the highest priority. Other important parameters include intra-annual variations of PAH and PCDD/F emissions, and information on congener profiles for PCDD/Fs and PCBs. This ranking can form a basis for further improvement of emission inventories that can be realized in cooperation with CEIP, the Task Force on Emission Inventories and Projections (TFEIP), and national experts.

Measurements of POP concentrations in air in 2014 have been performed at 33 sites of the EMEP monitoring network for POPs. Of these, 31 have reported PAH concentrations and 13 have reported data on other POPs. POP concentrations in precipitation in 2014 have been measured by 27 monitoring sites, of which 23 sites have reported PAH concentrations and 10 have reported other POPs. Eleven new sites, located in France, Spain, and Portugal, have started monitoring of POP concentrations that have improved the spatial coverage of the EMEP region by measurements of POP content in air and precipitation. Along with data of the EMEP monitoring network, substantial

amount of POP measurements is also available in the European Environmental Agency (EEA) AirBase and in the UNEP Stockholm Convention Global Monitoring Plan (SC GMP) Data Warehouse, which significantly extends the spatial coverage of the EMEP region by measurements and provides data for the global scale.

Evaluation of PAH pollution levels and transboundary transport in the EMEP countries has been carried out for the selected PAH compounds, namely, B[a]P, B[b]F, B[k]F, and IP, included into the CLRTAP Protocol on POPs. Elevated levels of annual mean PAH air concentrations (1-2 ng/m^3 and higher) in 2014 are characteristic of countries in Central and Eastern Europe (Poland, the Czech Republic, Slovakia, Slovenia, Romania, Bulgaria, Lithuania, Belarus, and Ukraine) as well as in the Iberian peninsula (Spain, Portugal). Verification of modelling results against measurements at the EMEP monitoring sites has shown reasonable agreement between modelled and observed PAH air concentrations. Results of model simulations indicate that transboundary transport of air pollution still plays an important role in the contamination of the EMEP countries. For more than 70% of the countries the contribution of external emission sources to PAH deposition in 2014 exceeds the contribution of national emission sources.

More detailed analysis of PAH pollution in the EMEP countries has been performed for B[a]P, which is used as a marker for the carcinogenic risk of exposure to PAHs. Evaluation of trends in B[a]P air concentrations in the EMEP region, carried out for two recent decades, has shown that decline of B[a]P air pollution levels slowed significantly during recent ten years and air concentrations even started to increase in some of the EMEP countries. According to measurements and modeling results, high levels of air concentrations, exceeding the EU limit values for B[a]P, are indicated in various regions of the EMEP domain, in particular, in Central and Eastern Europe. About half of monitoring stations in urban and suburban areas in Europe has measured B[a]P air concentrations in 2013 above the EU target value.

To contribute to the analysis of population exposure to elevated air pollution levels EMEP has initiated a study aiming at fine resolution modelling and quantification of B[a]P air concentrations in urban areas. First stage of this study is focused on the analysis of B[a]P air concentrations in the Czech Republic due to availability of data on emissions and measurements. The methodology of this study is based on combined evaluation of air pollution by modelling with fine spatial resolution and multiple regression analysis using measurement data of the Airbase monitoring sites.

Preliminary results show generally reasonable agreement of estimated B[a]P concentrations for urban areas in the Czech Republic with available measurements. This indicates that developing approach is capable to provide estimates of B[a]P air concentrations in urban areas consistent with the observed pollution levels. At the same time, for some of urban areas, namely, the Ostrava region, differences between modeled and measured concentrations are more significant that requires refinement of applied approach, improvement of national emission inventories for PAHs, and extension of existing monitoring network. At further stage of this work developing approach is planned to be used for other EMEP countries, provided that data on emissions and measurements are available. For this purpose close co-operation with national experts on monitoring of pollution levels and evaluation of emissions as well as with experts from TFEIP, CEIP, CCC, and EEA is of importance.

Air pollution by carcinogenic PAHs is recognized as a serious problem in areas with dense population of many EMEP countries. To consider this issue a thematic session is to be organised during the Second joint session of the Steering Body to EMEP and the Working Group on Effects. It is planned to discuss current state of monitoring and assessment of PAH pollution of the EMEP countries as well as population exposure in order to contribute to further reduction of air pollution levels.

Evaluation of contamination by PCDD/Fs, PCB-153, and HCB requires taking into account their cycling between and accumulation in different environmental compartments in order to provide information for assessment of exposure. The most important pathway for human exposure to these compounds is associated with dietary intake. Thus, their long-term accumulation in the surface media and bioaccumulation in wildlife can pose risk to human health. Model assessment of environmental pollution by PCDD/Fs, PCB-153, and HCB has been carried out using the multi-media multi-scale modelling approach, which takes into account peculiarities of POP transport and fate in the environment. Spatio-temporal variations of POP concentrations and deposition fluxes within the EMEP domain in 2014 have been analyzed on the basis of modelling results and measurement data of the EMEP monitoring network as well as measurements from the UNEP SC GMP Data Warehouse.

Analysis of long-term trends of POP pollution in the EMEP region has been continued this year with more detailed evaluation of temporal trends for PCB-153 in the EMEP countries. In particular, country-specific temporal variations of PCB pollution levels in the period 1990-2014 have been evaluated using the methodology developed by MSC-E. Estimates of trends of PCB-153 air concentrations indicate that levels of PCB-153 air pollution in the EMEP countries have declined on average by 75%. Relatively high rates of reduction (about 80%) are calculated for the EU28 countries, varying for individual countries from 60% for Estonia to almost 90% for the UK. The EECCA countries are characterized by relatively lower decrease of pollution levels (about 70%).

Long-range transport and pollution levels of PCDD/Fs, PCB-153, and HCB within the EMEP domain in 2014 have been modeled considering contributions of anthropogenic emission sources, secondary emissions, and intercontinental transport. Anthropogenic emissions of the EMEP countries substantially contribute to PCDD/F pollution of the EMEP region in 2014. Relative contribution of these sources to total PCDD/F deposition exceeds 50% in 25 countries. For PCB-153 and HCB significant contribution to the pollution levels is made by secondary emission sources. In particular, secondary emissions contribute to total deposition of HCB from 30% to 70%, and of PCB-153 from 50% to 80% depending on a country. Estimates of contribution of non-EMEP emissions to the pollution levels vary from several percents to 40% for PCDD/Fs and PCB-153, and from 20% to 60% for HCB.

Transboundary fluxes of PCDD/Fs, PCB-153, and HCB between the EMEP countries have been calculated for 2014. According to modelling results, the fraction of deposition over their territories, caused by the atmospheric transport from external emission sources, exceeds the fraction of deposition from their national emissions in almost 75% of the countries for HCB, in 60% of the countries for PCB-153, and in 50% of the countries for PCDD/Fs.

Further development of the Global EMEP Multi-media Modeling System (GLEMOS) is continued in order to improve the quality of the assessment of POP pollution levels and trends for the EMEP countries. It comprises the work on the transition of the EMEP operational modelling of POPs to the

new EMEP grid, testing and application of GLEMOS multi-scale modelling, and improvement of the GLEMOS multi-media modelling approach for POPs. Activities related to the refinement of POP model assessment include collection and analysis of information on congener composition of emissions and concentrations of PCDD/Fs and PCBs, which is important for the assessment of PCDD/F and PCB transport and fate in the environment as well as for evaluation of exposure.

Important aspect of MSC-E activities is collaboration with the subsidiary bodies to the Convention and other international organisations and programs. Progress in the MSC-E work related to model development and assessment of POP pollution in the EMEP region and selected countries was presented at the EMEP Task Force on Measurements and Modelling (TFMM). This year, MSC-E has performed analysis of trends of POP atmospheric concentrations for two recent decades and contributed obtained results to the CLRTAP scientific assessment report [*Maas and Grennfelt*, 2016], to the TFMM assessment report [*Colette et al.,* 2016], and to the WGE assessment report [*de Wit et al*., 2015]. In addition, the Centre actively participates in joint work with CEIP and TFEIP aimed at the improvement of available inventories of POP emissions.

Information exchange with other international organizations and programmes is essential for the assessment of environmental pollution of the EMEP region. In the framework of this activity MSC-E continues co-operation with the UNEP Stockholm Convention (SC), which includes the use of national inventories of PCDD/Fs emission, compiled under SC. This year refinement of the global scale scenario has been started by MSC-E on the basis of updated national inventories, submitted recently by countries to SC. The information reported by countries to SC is of importance for further studies of environmental pollution by dioxins and furans as in the EMEP region and on the global scale and further co-operation between the CLRTAP and SC in this field is highly appreciated. Along with data on emissions, significant amount of measurements of POP concentrations in the environment is collected in the UNEP SC GMP Data Warehouse. In particular, global scale measurements of POP air concentrations are available for the period of time covering approximately two recent decades. This information has been applied for the analysis of pollution levels in the EMEP countries presented in this report.

Evaluation of airborne pollution load to marginal seas within the EMEP region is of interest for various international organizations. 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 (UN ECE) and is based on the long-term EMEP/HELCOM contract.

Further activities of MSC-E will be directed to the refinement of assessment of POP pollution in the EMEP region. Specific attention will be given to PAH pollution levels in urban areas of the EMEP countries. Further development and evaluation of the Global EMEP Multi-media Modelling System (GLEMOS) will be focused on the transition of POP operational modelling to the new EMEP grid, updating open code version of GLEMOS, and refinement of parameterization of atmospheric transformation and removal processes for POPs as well as inter-media exchange. Finally, MSC-E will continue co-operation with subsidiary bodies of the Convention (TFMM, TF HTAP, TFEIP, and WGE), international organizations (AMAP, UNEP Stockholm Convention, HELCOM etc.) and national experts.

CONTENTS

INTRODUCTION

Persistent Organic Pollutants (POPs) are semi-volatile toxic substances, characterized by significant potential to long-range transport, accumulation in the environmental media, and adverse effects to human health and wildlife. In spite of efforts to reduce their use and unintentional releases, they are still measured in the environmental compartments, posing risk to humans as well as terrestrial and aquatic ecosystems. POPs are within the scope of the UNECE LRTAP Convention since 1998, when the Protocol on Persistent Organic Pollutants came into force. Along with this, POPs are included in the activities of many other international and national organizations including, for example, UNEP Rotterdam , Basel, and Stockholm Conventions, World Health Organization (WHO), the Arctic Monitoring and Assessment Program (AMAP), Helsinki Commission (HELCOM), and European Union in the framework of EU Regulation concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH).

Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe annually provides the Executive Body for the Convention with the information on deposition and transboundary transport of selected POPs (PAHs, PCDD/Fs, PCBs and HCB) within the geographical scope of EMEP. The Centre of Emission Inventories and Projections generates data on atmospheric emissions on the basis of information officially reported by the Parties to the Convention and expert estimates. Measurements of POP concentrations in air and precipitation are carried out at the EMEP monitoring network under the methodological guidance of the Chemical Coordinating Centre. The Meteorological Synthesizing Centre – East performs the model assessment of POP air concentrations and deposition fluxes for the EMEP region as well as of transboundary transport between the EMEP countries.

This report presents general description of recent activities of the EMEP Centres in the field of assessment of POP pollution levels, carried out in accordance with the 2016-2017 Workplan for the implementation of the Convention (ECE/EB.AIR/133/Add.1). It includes the outcome of model evaluation of POP pollution in the EMEP region, results of monitoring of POP concentrations, and information on POP emissions for modelling.

Specific attention in the report is paid to the evaluation of PAH pollution levels in the EMEP countries, including estimation of B[a]P air concentrations in urban areas. Air pollution by carcinogenic PAHs is recognized as a serious problem in areas with dense population of many EMEP countries. To contribute to the analysis of population exposure to elevated pollution levels, a study aimed at quantification of B[a]P air concentrations in urban areas has been initiated. First stage of this work is focused on the evaluation of B[a]P air concentrations for the Czech Republic due to availability of data on emissions and measurements. The methodology of the study is based on previously developed approaches that applied multiple regression models to describe functional relationship between measured urban concentrations, rural concentrations and secondary parameters including meteorological, geophysical data, and modeling results. At further stages this study can be continued for other EMEP countries, provided that the data on emissions and measurements are available.

Various aspects of the assessment of POP pollution of the EMEP region are considered in the report including emission data, results of monitoring and modelling activities. Gridded emission data are the most important information for model assessment of pollution. It should be noted that uncertainties of the reported POP emission inventories remain at present significant. The key emission parameters affecting quality of model estimates have been analysed and ranked in terms of their priority. Measurements of POP concentrations of the EMEP network provide important information for the analysis of spatial and temporal trends of pollution levels and evaluation of modelling results. Along with this additional measurement data on POPs, collected in the EEA AirBase and the UNEP SC GMP Data Warehouse, are used for the assessment of POP pollution of the EMEP region and for the analysis of global scale distribution of POPs. The model assessment includes evaluation of current pollution levels of PAHs, PCDD/Fs, PCB-153, and HCB in the EMEP region. For long-lived POPs multi-scale model simulations have been carried out to evaluate intercontinental transport and contributions of non-EMEP emission sources. In the framework of the assessment of POP pollution, MSC-E has performed the analysis of long-term trends of POP contamination for the period from 1990 to 2012. Results of trend analysis performed by MSC-E have been contributed to the CLRTAP Assessment Report [*Maas and Grennfelt*, 2016], the TFMM Assessment Report [*Colette et al*., 2016] and the WGE Assessment Report [*de Wit et al.*, 2016] released by the Convention this year. As a continuation of this work, the analysis of trends for PCB-153 for the period from 1990 to 2014 has been done and described in the report.

Further development of the Global EMEP Multi-media Modelling System (GLEMOS) is aimed at reduction of the model assessment uncertainties in the EMEP region. Following decisions of the Executive Body for CLRTAP [ECE/EB.AIR/113/Add.1] MSC-E continues the work on transition of the EMEP operational modelling to the new EMEP grid. In addition, this work includes testing and application of the GLEMOS multi-scale modeling, and further improvement of the GLEMOS multi-media modelling approach for POPs. To refine model assessment for PCDD/Fs and PCBs, collection and analysis of information on congener composition of their emissions and concentrations in media are carried out. This information is important for the assessment of PCDD/F and PCB transport and fate in the environment as well as for evaluation of exposure.

One of the main directions of the Convention activity is collaboration with subsidiary bodies to the Convention and other international and national organisations. All results of the research and development were presented and discussed at the EMEP Task Force on Measurements and Modelling (TFMM) and the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). Moreover, the Centre works in close co-operation with the UNEP Stockholm Convention, AMAP, HELCOM, and other organizations to broaden dissemination and facilitate exchange of the scientific and policy oriented information.

Finally, main challenges of POP pollution assessment that need to be addressed in further operational and research activities in 2017 are summarized in the report. More detailed scientific information is presented in the MSC-E Technical Report 1/2016 [*Shatalov et al*., 2016]. Detailed information on POP pollution levels in the EMEP region as well as for individual EMEP countries and marginal seas is also distributed via the Internet at the MSC-E website [www.msceast.org].

1. EMEP MONITORING NETWORK FOR POPs

Measurements of persistent organic pollutants (POPs) including polyaromatic hydrocarbons (PAHs) were included in the EMEP's monitoring program in 1999 but data are also available from the beginning of 1990s for some compounds and some stations in the EMEP database [http://ebas.nilu.no].

In 2014, a total number of 33 monitoring sites have reported data for PAHs and/or other POPs in air. Of these, 31 reported PAHs and 13 reported other POPs only. The number of sites reporting air data in 2014 are the same as in 2013 but the location of the sites are slightly different. In 2014, five new sites started reporting data (i.e. FR0023, FR0024, FR0025, PT0004, PT0006) while five sites from 2013 have not reported data. For precipitation (including total- or wet deposition), POPs and/or PAHs are available from 27 sites in 2014. Of these, 23 reported PAHs and ten reported other POPs. Benzo(a)pyrene (B[a]P) in air and precipitaion is available from 31 and 23 sites, respectively. The total number of precipitation sites in 2014 are three more than in 2013. As for air, the location of the sites differs somewhat as nine new sites started reporting data in 2014 (i.e. ES0001, ES0006, ES0007, ES0014, FR0009, FR0013, FR0023, FR0024, FR0025) and six sites from 2013 did not report data. The new sites in 2014 have increased the spatial coverage for air and precipitaion data of PAHs in south/south-west Europe.

The length of the monitoring data sets varies between sites and type of POP compound (Table 1.1). For example, B[a]P data in air are available from the 1990s at Zeppelin in Norway (1994), Rörvik/Råo in Sweden (1994), Aspvreten in Sweden (1995), Pallas in Finland (1996), and Kosetice in the Czech Republic (1999). From the 2000s, additionally data have been reported from Råö in Sweden (2002), High Muffles in the United Kingdom (2004-2013), Rucava in Latvia (2004), Niembro in Spain (2005), Westerland, Schauinsland, Schmücke, Zingst in Germany (2007), and Diabla Gora in Poland (2008) (Table 1.1). In addition, B[a]P levels in air have been reported from 18 other sites during the last years. Long-term data of B[a]P in precipitation are available from Westerland (1996) and Zingst (1999) in Germany, Kosetice in the Czech Republic (2003), Schauinsland and Schmücke in Germany (2007), and Diabla Gora in Poland (2008).

Detailed information about the sites, measurement methods and results for 2014 can be found in EMEP/CCC's data reports on heavy metals and POPs [*Aas and Nizzetto*, 2016]. All data are also available in the EMEP database [http://ebas.nilu.no]. 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]. An example is the different limit of detections from the reporting countries in 2014, which results in reduced comparability of data. For this reason, data from Portugal are not included in the further discussion. In addition, data for precipitation are reported as concentration or as deposition resulting in further complications when comparing POP data in precipitation.

Table 1.1. *Long-term data on B[a]P in air and precipitation at EMEP sites showing the starting year*

	B[a]P	
Site name (site code)	Air	Precipitation
Kosetice (CZ03)	1999	2003
Westerland (DE01)	2007	1996
Schauinsland (DE03)	2007	2007
Schmücke (DE08)	2007	2007
Zingst (DE09)	2007	1999
Niembro (ES08)	2005	
Pallas (FI36)	1996	
High Muffles (GB14)*	2004*	
Rucava (LV10)	2004	
Zeppelin (NO42)	1994	
Diabla Gora (Pl05)	2008	2008
Aspvreten (SE12)	1995	
Råö/Rörvik (SE02/14)	1994	

* Stopped measuring POPs after 2013

Measurements of Benzo[a]pyrene in air and precipitation for 2014

An overview of the concentrations reported for B[a]P in air at the individual sites in 2014 is presented in Table 1.2. This shows that the annual mean and median concentrations for B[a]P in Europe vary by two orders of magnitude with ranges from 0.004-0.602 ng/m^3 and 0.001-0.275 ng/m^3, respectively. The variability at each site is even higher with up to three orders of magnitude variance between minimun and maximum as well as the 5th and 95th percentile. All sites show a seasonal variability of the B[a]P concentrations in air with highest concentrations in winter (November-February) and lowest concentrations in summer (June-August).

A low detection frequency for B[a]P, with >50% of the samples being below detection limit, is observed at Zeppelin in Norway as well as at all sites in Spain (ES01, 06, 07, 08, 14).

The spatial pattern of the annual mean concentrations for B[a]P in air and precipitation in 2014 is presented in Fig. 1.1. In general, the highest concentrations of B[a]P in air are observed at sites in central and Eastern Europe, while the lowest are observed in the Arctic (Fig. 1.1). In precipitaion, the highest annual mean concentrations are observed in Central Western Europe. This is consistent with previous year [*Aas and Nizzetto*, 2015]. The annual mean and median concentrations in air at EMEP stations are well below (generally one to two orders of magnitude) the European Air Quality Standard (1 ng/m^3), defined by the 4th daughter directive [*EU*, 2004]. However, the 95th percentile concentrations exceed the European Air Quality Standard at Kosetice, the Czech Republic; Waldhof and Zingst, Germany; Rucava, Latvia; and Diabla Gora, Poland. In addition, two sites have maximum concentrations above the European Air Quality Standard: Birkenes, Norway; and Iskrba, Slovenia.

Table 1.2. *Concentrations in air for B[a]P (ng/m^3) at EMEP sites in 2014*

B[a]P in air, 2014								
Site	**Annual mean**	**Annual median**	**Min-Max**		**25-75%**		**5-95%**	
BE0013R	0.084	0.031	0.000	0.780	0.014	0.073	0.000	0.378
CZ0003R	0.280	0.095	0.003	1.945	0.018	0.437	0.003	1.028
DE0001R	0.188	0.098	0.010	0.578	0.015	0.428	0.010	0.578
DE0002R	0.346	0.162	0.016	1.467	0.021	0.529	0.016	1.467
DE0003R	0.023	0.020	0.004	0.046	0.011	0.039	0.004	0.046
DE0008R	0.074	0.036	0.004	0.299	0.014	0.101	0.004	0.299
DE0009R	0.309	0.145	0.014	1.675	0.034	0.389	0.014	1.675
ES0001R	*0.029*	*0.020*	*0.020*	*0.367*	*0.020*	*0.020*	*0.020*	*0.054*
ES0006R	*0.029*	*0.020*	*0.020*	*0.110*	*0.020*	*0.020*	*0.020*	*0.080*
ES0007R	*0.032*	*0.020*	*0.020*	*0.090*	*0.020*	*0.050*	*0.020*	*0.080*
ES0008R	*0.089*	*0.020*	*0.020*	*0.890*	*0.020*	*0.110*	*0.020*	*0.300*
ES0014R	*0.020*	*0.020*	*0.020*	*0.020*	*0.020*	*0.020*	*0.020*	*0.020*
FI0036R	0.013	0.005	0.000	0.073	0.001	0.016	0.000	0.073
FR0009R	0.037	0.004	0.001	0.387	0.001	0.024	0.001	0.213
FR0013R	0.017	0.004	0.001	0.148	0.001	0.017	0.001	0.083
FR0023R	0.054	0.028	0.001	0.233	0.003	0.082	0.001	0.209
FR0024R	0.046	0.009	0.001	0.352	0.001	0.062	0.001	0.234
FR0025R	0.034	0.010	0.001	0.329	0.003	0.032	0.001	0.160
GB0036R	0.054	0.029	0.002	0.234	0.020	0.073	0.002	0.234
GB0048R	0.026	0.021	0.005	0.068	0.010	0.037	0.005	0.068
LV0010R	0.317	0.202	0.003	1.470	0.024	0.423	0.003	1.316
NL0091R	0.079	0.024	0.005	0.927	0.013	0.074	0.007	0.353
NO0002R	0.056	0.006	0.002	1.450	0.003	0.014	0.002	0.377
NO0042G	*0.004*	*0.001*	*0.001*	*0.033*	*0.001*	*0.002*	*0.001*	*0.018*
PL0005R	0.602	0.275	0.012	2.957	0.036	0.900	0.014	2.374
SE0011R	0.111	0.030	0.002	0.620	0.005	0.120	0.002	0.620
SE0012R	0.057	0.031	0.005	0.240	0.007	0.073	0.005	0.240
SE0014R	0.041	0.018	0.004	0.150	0.008	0.062	0.004	0.150
SI0008R	0.141	0.085	0.009	1.248	0.009	0.195	0.009	0.475

Italic means that more than 50% of the samples are below detection limit

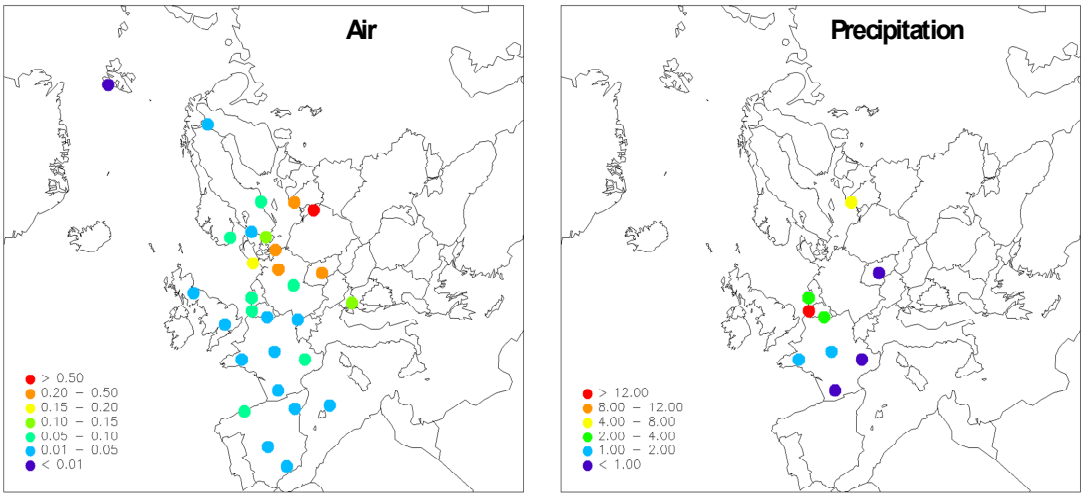


Fig. 1.1. *Annual mean concentrations (ng/m^3) in 2014 for benzo(a)pyrene (B[a]P) in air (left), and precipitation (right)*

Measurements of other selected POPs in air and precipitation for 2014

The spatial pattern, based on observed annual mean concentrations in 2014, for a selection of POPs (i.e. PCB-52, -153, α-HCH, γ-HCH, HCB and p,p'-DDT) are shown in Figures 1.2-1.4. In general, for the selected POPs the concentrations in air tend to be highest in Central Europe and decrease from south to north. An exception is HCB for which the concentrations are highest at one site in Central Europe (i.e. Kosetice, the Czech Republic) followed by the two sites in the Arctic: Zeppelin, Norway and Station nord at Greenland, Denmark which were all three-four times higher than those observed at other European sites (Fig. 1.4).

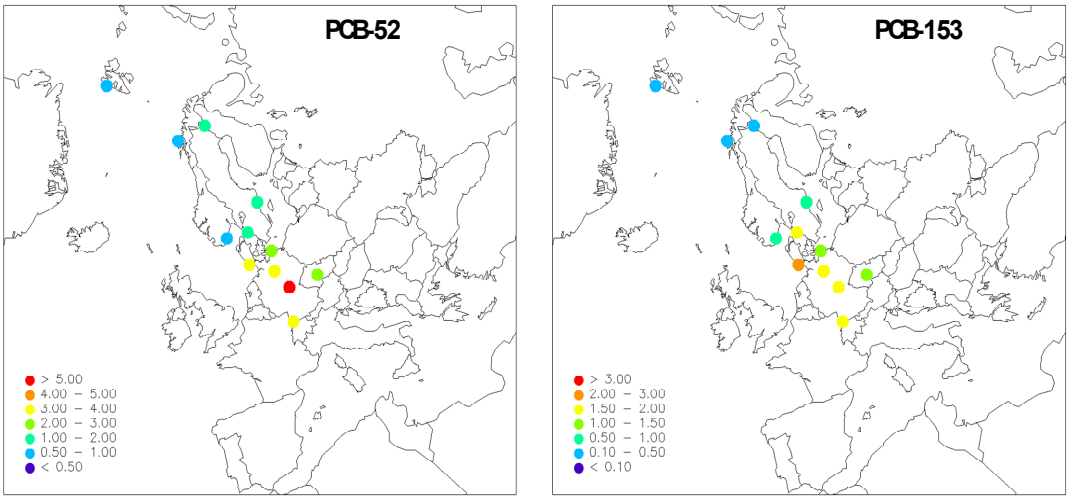


Fig. 1.2. *Annual mean concentrations (pg/m^3) in 2014 for PCB-52 (left), and PCB-153 (right) in air*

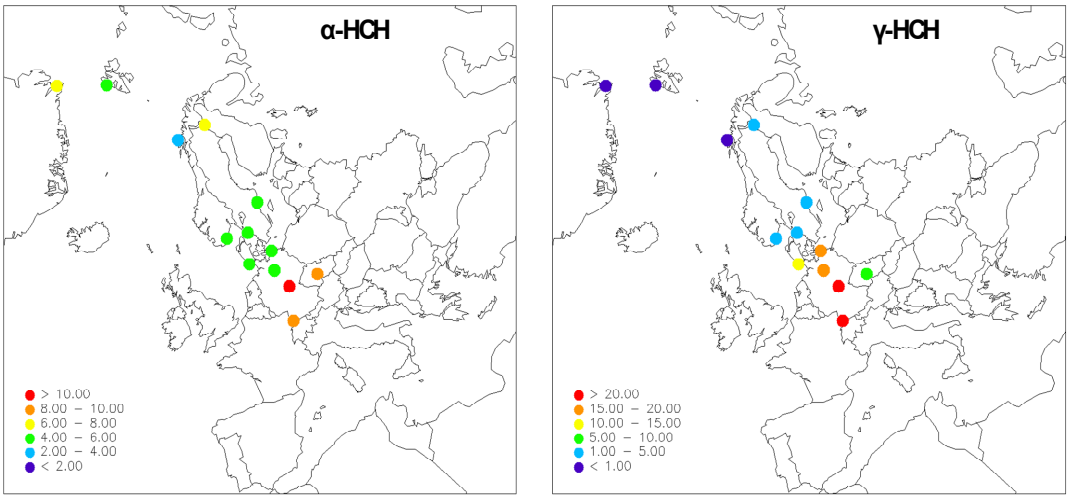


***Fig. 1.3**. Annual mean concentrations (pg/m^3) in 2014 for α-HCH (left), and γ-HCH (right) in air*

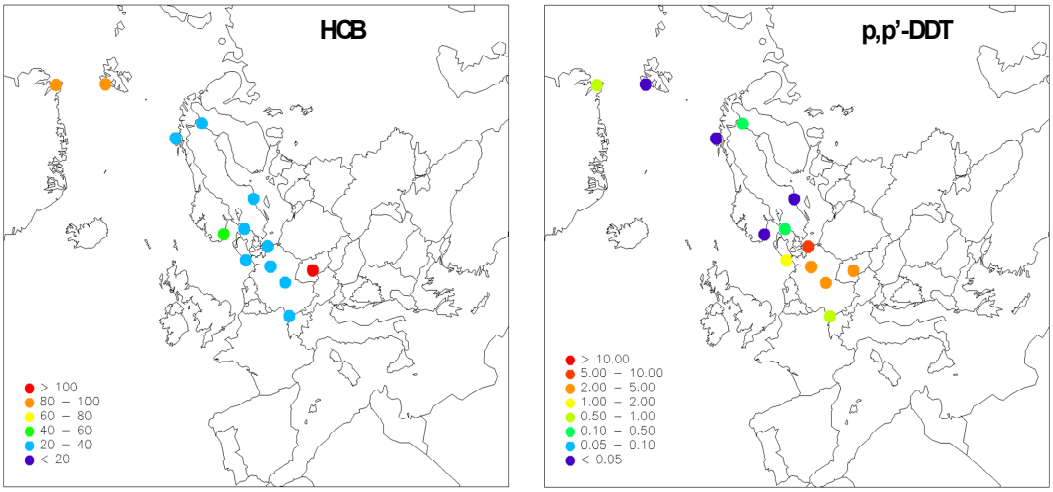


***Fig. 1.4.** Annual mean concentrations (pg/m^3) in 2014 for HCB (left), and p,p'-DDT (right) in air*

2. ASSESSMENT OF POP POLLUTION IN THE EMEP REGION

In this chapter the outcome of model assessment of long-range transport and pollution levels of PAHs (benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-cd]pyrene), PCDD/Fs, PCB-153, and HCB for 2014 is presented. Model simulations have been performed using multi-media modeling approach, which takes into account peculiarities of POP transport and fate in the environment (e.g. phase partitioning, degradation in media, multi-hop transport). To evaluate intercontinental transport and contribution of non-EMEP sources to the pollution levels in the EMEP countries multi-scale modeling has been used. Model simulations have been carried out on the basis of emission data reported by the EMEP countries and complemented by available expert estimates. Spatio-temporal variations of POP concentrations and deposition fluxes in the EMEP domain have been analyzed on the basis of modeling results and measurement data of the EMEP monitoring network as well as measurements from the EEA Airbase and UNEP SC GMP Data Warehouse. Besides, estimation of transboundary transport of pollution between the EMEP countries has been performed. Main emphasis in the assessment of this year is given to the evaluation of PAH pollution levels in the EMEP countries as well as B[a]P air concentrations in urban areas.

2.1. Polycyclic Aromatic Hydrocarbons (PAHs)

Polycyclic Aromatic Hydrocarbons (PAHs) are a group of chemicals that naturally occur in coal, crude oil, and gasoline and are contained in products made from fossil fuels. PAHs can be released to the atmosphere during the incomplete combustion of fossil fuels and biomass burning. PAHs are known as substances posing serious risk for the human health [*Theakston*, 2000].

Taking into account possible adverse effects on human health, a number of countries have introduced target values of air quality objectives for PAHs in the ambient air. Benzo[a]pyrene has been included in the list of carcinogens of category 1B by the International Agency for Research on Cancer (IARC). It is used as a marker for the carcinogenic risk of polycyclic aromatic hydrocarbons (Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004). According to this Directive, EU target value for B[a]P annual mean air concentrations is established as 1 ng/m^3. Here target value means “a concentration in the ambient air fixed with the aim of avoiding, preventing or reducing harmful effects on human health and the environment as a whole”. Besides, upper and lower assessment thresholds (0.6 and 0.4 ng/m^3 respectively) are established to assess ambient air quality.

The EU Regulation REACH includes restrictions on the manufacturing and use of chemicals, containing PAHs. In the EU legislation ((EC) No 1272/2008) the following PAHs, namely, benzo[a]pyrene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, dibenz[a,h]anthracene, chrysene and benzo[e]pyrene, are considered as carcinogens of category 1B.

Model assessment of PAH pollution levels in the EMEP domain has been 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 are

carried out for 2014 and compared with the results obtained for 2013. Assessment of PAH pollution in the EMEP countries includes analysis of spatial distribution, evaluation of transboundary transport between the EMEP countries, and comparison of obtained modeling results with available measurements.

Specific attention is given to the analysis of B[a]P air pollution levels in urban areas. To contribute to the analysis of population exposure to B[a]P concentrations, development of methodology for the evaluation of B[a]P air concentrations in urban areas is initiated. Brief outline of this activity and preliminary results are presented below in this section.

2.1.1. Emission data for model assessment

Modeling of long-range transport and pollution levels of PAHs in the EMEP domain has been carried out using gridded emission data on benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-cd]pyrene) provided by CEIP. Detailed information on PAH emissions in each country, as well as the gap-filling methods that have been used for the 2014 GNFR emission inventory (as reported in 2016) can be found in the Technical report CEIP 02/2016 [*Tista et al*., 2016].

Spatial distribution of PAH and B[a]P emissions from anthropogenic sources in the EMEP domain, used in model simulations for 2014, is shown in Figs 2.1a and b, respectively.

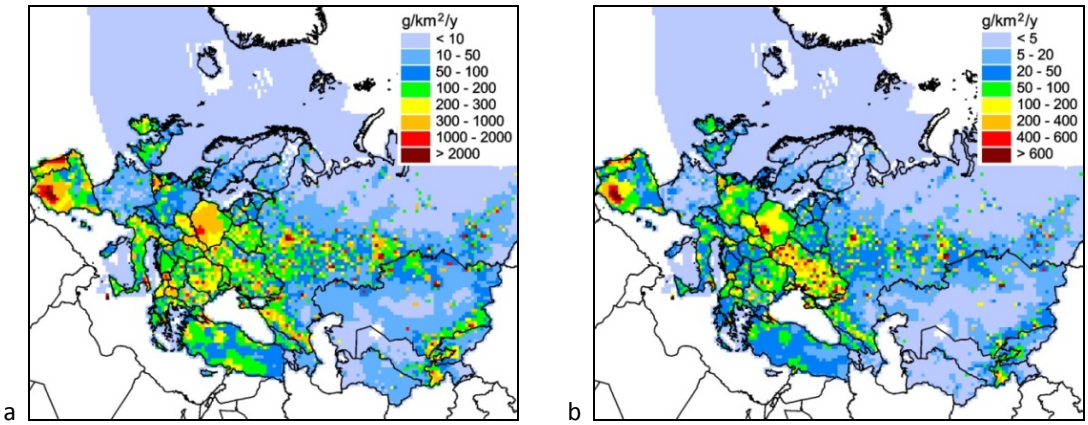


Fig. 2.1. *Spatial distribution of emissions of the sum of 4 PAHs (a) and B[a]P (b) in the EMEP countries in 2014 with resolution 50x50 km²*

Total anthropogenic emissions of 4 PAHs in the EMEP domain for 2014 are higher than the corresponding emissions for 2013, provided in previous submission of emission data [*Mareckova et al*., 2015]. *In comparison to the total amount of 4 PAHs emitted in 2013, the estimate of total emission for 2014 is larger by 55% and accounts for 2635 tonnes.*

Changes of PAH emissions for individual EMEP countries between 2014 and 2013 are shown in Fig 2.2. Values of changes are expressed as the difference of total emissions E_{2014} for 2014 and E_{2013} for

2013 (E_{2014} – E_{2013}). Positive values in the diagram indicate an increase of total 4 PAH emission in a country from 2013 to 2014, while negative values illustrate a decrease of emission.

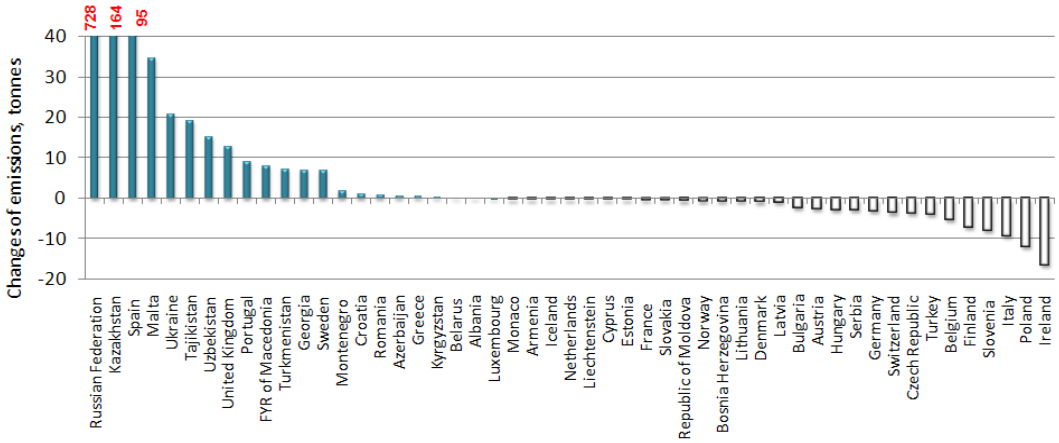


Fig. 2.2. *Changes of annual total emissions of the sum of 4 PAHs from 2013 to 2014, tonnes. Negative values denote decrease of emissions, and positive ones – increase of emissions*

Compared with 2013, PAH emissions in 2014 within the EMEP region have increased in 22 countries and decreased in 28 countries. The most significant increase of PAH emissions can be noted for the EECCA countries. Particularly, estimates of emissions for Georgia, the Russian Federation, Turkmenistan, Uzbekistan, Tajikistan, and Kazakhstan for 2014 are higher by a factor of 39, 10, 6.2, 4.4, 3.6, and 3.4 than that for 2013, respectively.

Updated evaluation of national energy balance, prepared by Georgia, has given an opportunity to improve significantly quality of inventory by estimating emissions in energy sector from much more activities than before using better methodologies and more detailed emission calculation approaches [*IIR of Georgia*, 2016]. This year PAH emission of Georgia has been calculated from almost all activities of energy sector and this has caused an increase of reported total emissions. Kazakhstan has reported data on PAH emissions for the first time, which has lead to significant change of national emissions from 2013 to 2014. To estimate total emission of the Russian Federation, Tajikistan, Turkmenistan and Uzbekistan, CEIP has used the data from the global atmospheric emission inventory of PAHs for 2004 [*Zhang and Tao*, 2009]. For subsequent years total emission of PAHs has been extrapolated by using temporal changes of gross domestic product (GDP) per capita up to the year 2014 [*Tista et al.*, 2016].

Substantial changes of PAH emissions can also be noted for Malta, the FYR of Macedonia, Sweden, the United Kingdom, and Spain. In particular, estimates of 2014 emissions for these countries are higher by a factor of 580, 3.3, 2.7, 2.2, and 1.6 in comparison to 2013, respectively. Decrease of PAH emissions from 2013 to 2014 (more than 50%) is indicated for Liechtenstein, Monaco, Slovenia, Armenia, and Switzerland. The other EMEP countries are characterized by less significant changes.

2.1.2. Pollution levels in the EMEP region

Evaluation pollution levels in the EMEP region for selected 4 PAHs has been carried out using anthropogenic emissions of the EMEP countries. The contribution of non-EMEP emissions to air pollution of the EMEP region by PAHs is comparatively low and is not currently considered in model simulations. At the same time, for countries near the boundaries of the EMEP domain these sources can notably contribute to the pollution levels. Thus, at further stages of model assessment it is planned to take into account the influence of non-EMEP emissions based on available data on global PAH emissions.

Model estimates of annual mean air concentrations and deposition fluxes of the 4 PAHs in the EMEP region for 2014 are presented in Fig. 2.3. Elevated levels of annual mean air concentrations (1-2 ng/m^3 and higher) are characteristic of countries in Central and Eastern Europe, namely, in Poland, the Czech Republic, Slovakia, Slovenia, Romania, Bulgaria, Lithuania, Belarus, and Ukraine (Fig. 2.3.a). Areas of high concentrations can also be indicated for Spain, Portugal, and northern Italy. Similar pattern of spatial distribution is obtained for total annual deposition of 4 PAHs (Fig. 2.3b). Modeling results provide information on pollution of marginal seas within the EMEP region. The largest level of PAH deposition is calculated for the Black Sea (about 44 g/km^2/y) followed by the Baltic Sea (about 33 g/km^2/y), the North Sea (about 18 g/km^2/y), the Mediterranean Sea (about 15 g/km^2/y), and the Caspian Sea (about 9 g/km^2/y).

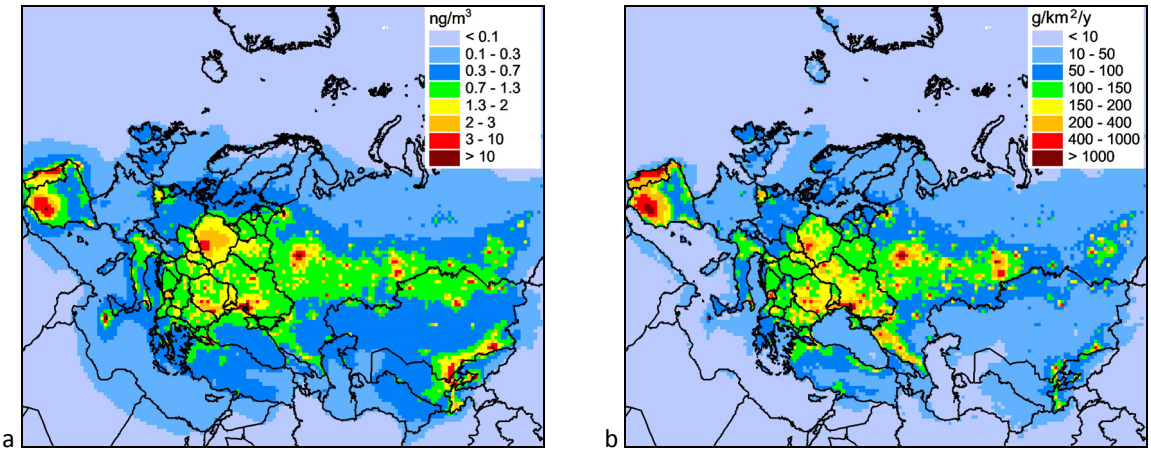


Fig. 2.3. *Spatial distribution of modeled annual mean air concentrations, ng/m^3 (a) and deposition fluxes, g/km^2/y (b) of 4 PAHs in the EMEP domain for 2014*

PAH pollution levels in the EMEP region, calculated for 2014, have been compared with the modeling results for 2013. Changes of pollution can be explained by differences in emission data used in model simulations and by inter-annual variability of meteorological conditions.

For most of the EMEP countries changes of calculated air concentrations are caused by the differences in national emissions used in model simulations for 2013 and 2014. In particular, the largest relative changes can be noted for the EECCA countries, namely, Tajikistan, Uzbekistan, Kazakhstan, the Russian Federation, Turkmenistan (more than 100%), which are explained by higher values of total PAH emissions for 2014 comparing to 2013. Substantial changes (by 50-100%) due to increased emissions can also be mentioned for Malta, Georgia, the FYR of Macedonia, Kyrgyzstan,

and Spain. In case of Iceland and Sweden higher levels of concentrations in 2014 are partly caused by changing of meteorological conditions. Lower levels of PAH concentrations (by 40-50%) in 2014 comparing to 2013 are calculated for Ireland, Slovenia, Switzerland, and Liechtenstein, which is mostly related to the effect of decreased national emissions.

To evaluate the effect of changes of meteorological conditions from 2013 to 2014 on PAH pollution levels additional model simulations for these two years have been performed with the same emission data for 2013 and varying meteorological information (for 2013 and 2014). Comparison of modeling results reflects the influence of inter-annual variability of precipitation amount, prevailing atmospheric flows, and temperature on PAH pollution levels. Relative changes of annual mean PAH air concentrations in the EMEP countries from 2013 to 2014 (C_{2013} and C_{2014}) are shown in Fig. 2.4. Model simulations indicate increase of PAH air concentrations in Northern Europe. In particular, PAH concentrations estimated for Sweden, Finland, Norway, Denmark, and Estonia are higher (by 17-50%) in 2014 comparing to 2013. The most significant decline of concentrations (by 10-20%) takes place in France, Luxembourg, Belgium, the United Kingdom, Georgia, Armenia, and Azerbaijan.

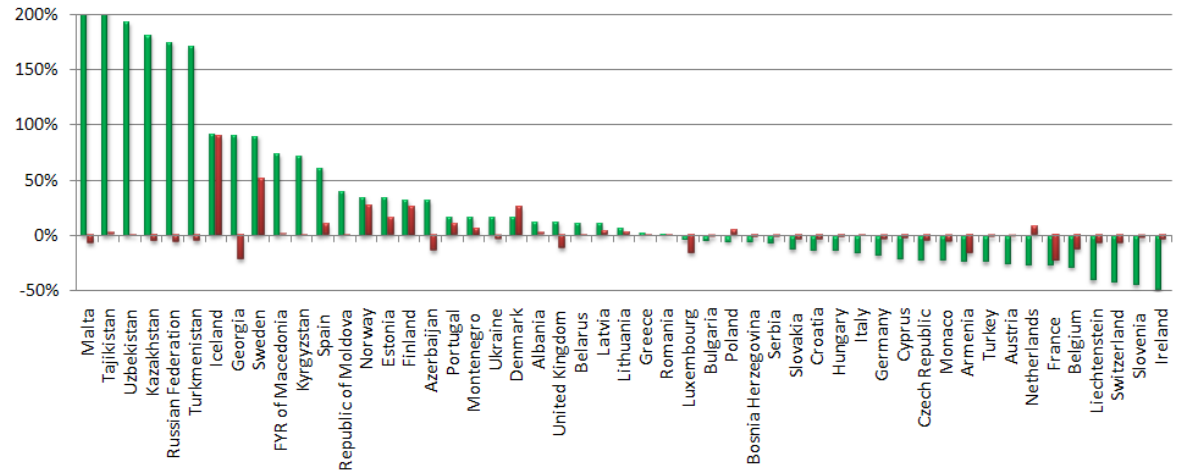


Fig. 2.4. *Changes of annual mean modelled PAH air concentrations in the EMEP countries between 2013 and 2014 (defined as $(C_{2014}-C_{2013})/C_{2013}$) due to changes of emissions and meteorological conditions (green bars), and due to changes of meteorological conditions only (red bars). Negative values denote decrease, and positive ones – increase*

In addition to the evaluation of PAH pollution levels for 2014 preliminary estimates of pollution have been made for the year 2015. In particular, "near-real time" modeling of B[a]P is carried out for 2015 and changes of pollution between 2014 and 2015 are examined. Spatial distribution of annual mean B[a]P air concentrations within the EMEP domain for 2015 as well as difference between the results for the years 2014 and 2015 (C_{2014} and C_{2015}) are illustrated in Fig. 2.5. It can be seen that *in 2015 air concentrations of B[a]P tend to decline in the countries of Northern Europe (by 10-50%). At the same time, a growth of concentrations (by 10-50%) is obtained for the countries in Western and Southern Europe. Comparison of the results for 2014 and 2015 indicates the influence of inter-annual variations of meteorological conditions on the pollution levels*, since modeling for two years has been carried out using the same set of emission data for 2014.

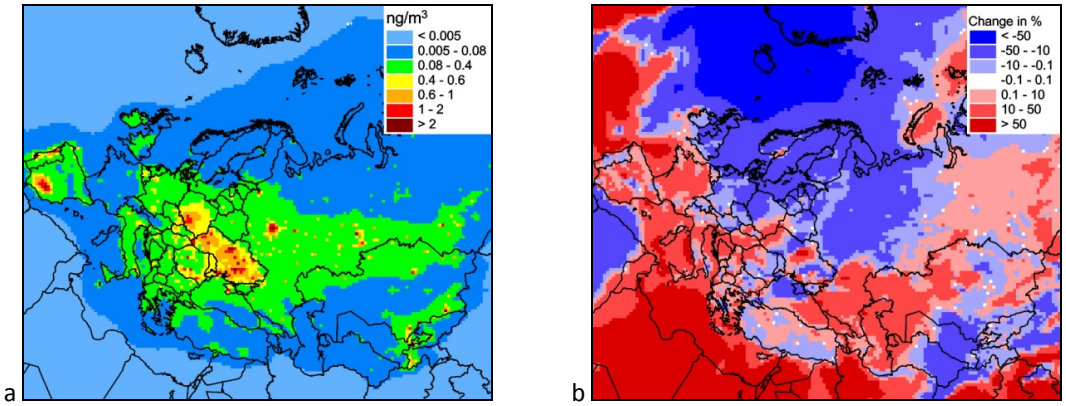


Fig. 2.5. *Spatial distribution of modeled annual mean B[a]P air concentrations for 2015, ng/m^3 (a) and relative difference between B[a]P air concentrations in 2014 and 2015, (C_{2015}-C_{2014})/C_{2014}*

2.1.3. Transboundary transport of PAHs in 2014

Model assessment of transboundary transport within the EMEP region has been evaluated for the selected 4 PAHs. Annual total deposition fluxes of the PAHs to the EMEP countries are presented in Fig. 2.6. The largest contribution to total deposition of 4 PAHs is made by B[b]F (31%). The other PAHs contribute 30% (B[a]P), 23% (B[k]F), and 16% (IP).

Elevated levels of mean PAH deposition fluxes in the EMEP countries, more than 200 g/km^2/y, are calculated for Malta, Portugal, and Montenegro. Relatively low intensity of deposition is obtained for the EECCA countries and regions located far from major European emission sources (e.g. Northern Europe).

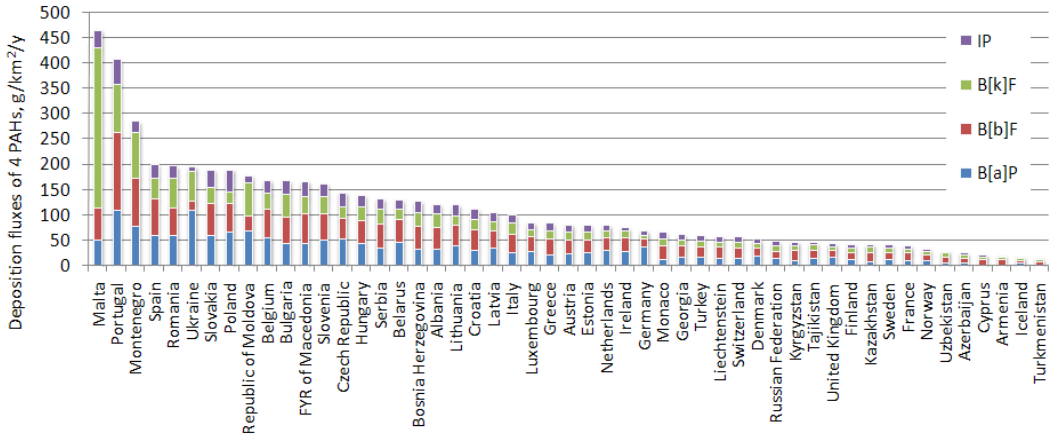


Fig. 2.6. *Annual deposition fluxes of 4 PAHs (B[a]P, B[b]F, B[k]F, and IP) in the EMEP countries calculated for 2014, g/km^2/y*

Model assessment of PAH long-range transport indicates that transboundary fluxes continued playing important role in the pollution of the EMEP countries. For the majority of countries (more than 70%) the fraction of PAH deposition over their territories in 2014, caused by the transport from external emission sources, exceeds the fraction of deposition from their national emissions (Fig. 2.7). In

particular, substantial contribution of transboundary transport to PAH deposition is estimated for countries, characterized by relatively small territory or relatively low national emissions (e.g. Monaco, Armenia, Iceland, and Lichtenstein), whereas for countries with relatively high PAH emission (e.g. Malta, Portugal, Spain, the Russian Federation) is not significant.

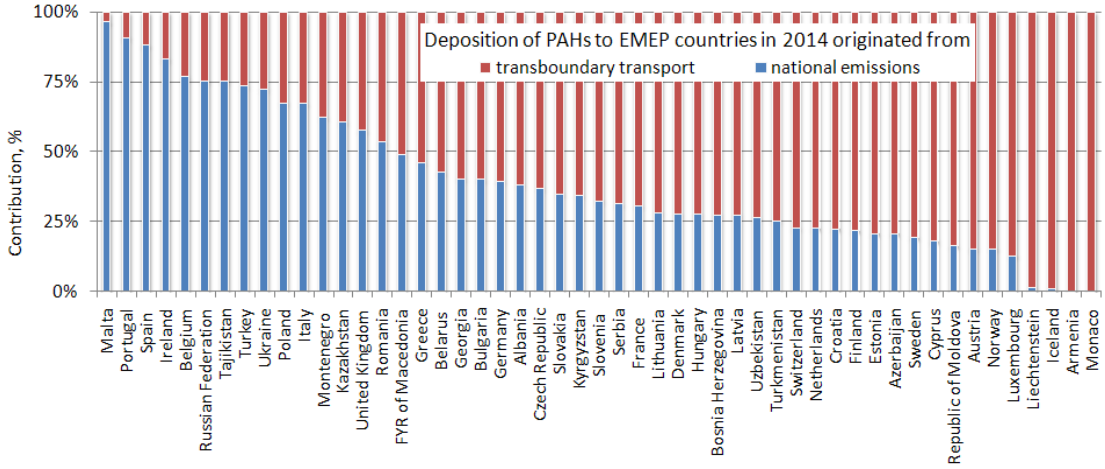


Fig. 2.7. *Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of 4 PAHs over the EMEP countries in the 2014.*

Transboundary transport of pollution from a particular country to other countries can also be characterised by the fractions of total anthropogenic deposition, originated from national emissions of a country, and fallen out within and outside its boundaries, as shown in Fig. 2.8.

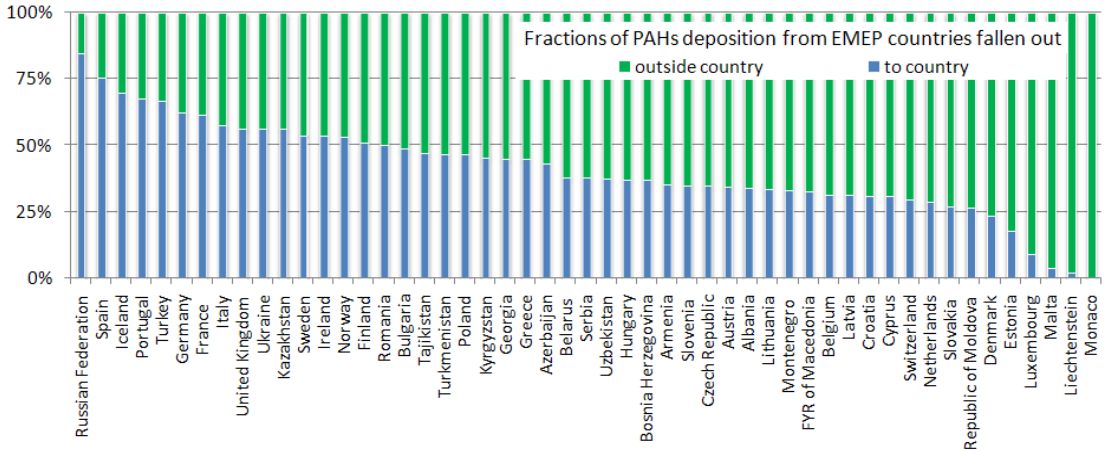


Fig. 2.8. *Fractions of PAH deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in the 2014*

For 36 countries the fraction of PAHs deposited to other EMEP countries exceeds the fraction of PAHs deposited to the country itself. About 60% of total annual deposition of 4 PAHs within the EMEP region in 2014 originated from emission sources of five countries, namely, the Russian Federation, Ukraine, Kazakhstan, Spain, and Poland.

2.1.4. Evaluation of B[a]P pollution levels in urban areas and selected cities

High levels of air pollution in urban areas and their adverse effects to population have recently received increasing attention [*Maas and Grennfelt*, 2016]. Despite the decreasing emissions to the atmosphere, air concentration levels in the EMEP countries for some of the pollutants are still significant and exceed air quality standards in areas with high population density. Benzo[a]pyrene is one of the pollutants for which monitoring stations in the EMEP countries continue measuring air concentrations near or above EU air quality guidelines [*EEA*, 2015]. Major contributions to B[a]P air pollution levels in urban areas originate from combustion of fossil fuels for domestic heating, road transport, industrial activities, and biomass burning [*Ravindra et al.*, 2008]. Combustion of wood and coal for residential heating, being important source of air pollution by fine particles and carcinogenic PAHs, is linked to serious health effects such as respiratory and cardiovascular morbidity and mortality [*Wanstall et al.*, 2015].

Analysis of temporal changes of emissions and air concentrations in the EMEP region, performed for the two recent decades, has shown that decline of B[a]P air pollution levels slowed significantly during recent ten years and air concentrations even started to increase in some of the EMEP countries [Gusev et al., 2015]. Model assessment of B[a]P pollution for 2014 indicates high levels of air concentrations exceeding the EU limit values [*EU*, 2004] in various regions of the EMEP domain (Fig. 2.9). According to these results about 5% of population in the EMEP countries is living in areas with exceeded EU limit for B[a]P air concentrations.

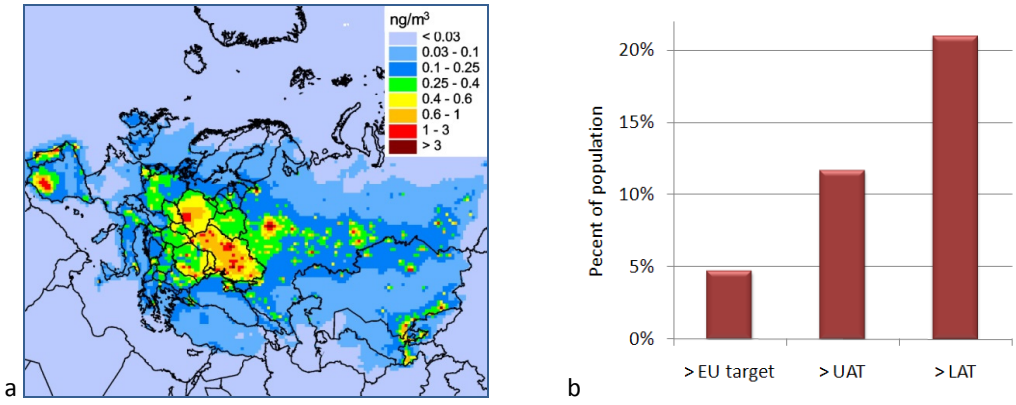


Fig. 2.9. *Modeled B[a]P annual mean air concentrations for 2014 in the EMEP region (a) and percentage of population of the EMEP countries (b) living in the areas with annual mean B[a]P air concentrations above the EU limit values: EU target value for B[a]P – 1.0 ng/m^3, UAT – 0.6 ng/m^3, LAT – 0.4 ng/m^3*

Elevated B[a]P air concentrations for 2014 near or above the EU target value can be mentioned for the countries of Central and Eastern Europe as well as for Portugal, Spain, and Italy. Modeling results satisfactory characterize regional scale transport and pollution levels in rural and background areas as it is shown in the next section. However, levels of concentrations observed in urban areas are generally underestimated by the model (Fig. 2.10).

Model assessment of POP pollution is performed using regional scale Eulerian modeling framework that provides smoothed distribution of air concentrations and does not capture significant gradients of pollution in the urban areas due applied spatial resolution. To evaluate variability of air concentrations in urban areas multi-scale modeling can be applied for limited number of areas and time periods using nested regional and urban scale models. However, for wider spatial scale and longer periods of time this approach would require significant computer resources. A number of alternative approaches have been developed that include, for example, combined use of air pollution modeling and regression analysis to estimate pollution levels for densely populated areas.

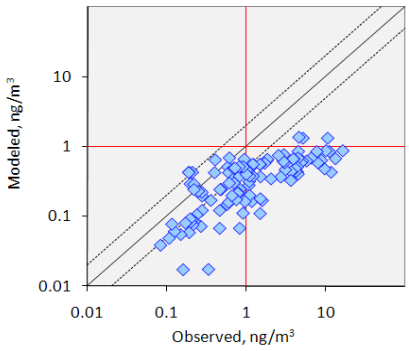


Fig. 2.10. *Modeled B[a]P air concentrations for 2013 vs. measurements of Airbase suburban/urban monitoring sites. Red lines indicate the EU target value 1 ng/m^3. Dashed lines denote the area of agreement within a factor 2*

To describe variability of B[a]P air concentrations in urban areas EMEP has initiated a study that will permit to refine assessment of pollution and to provide input information for the analysis of exposure to elevated levels of B[a]P in the EMEP countries. Initial stage of this activity is performed to estimate B[a]P air concentrations for the Czech Republic and to test capabilities of developing methodology. Analysis of pollution levels with fine spatial resolution is carried out for the year 2013 due to availability of B[a]P measurements at urban and suburban Airbase monitoring stations and emission data. The study consists of several steps, namely:

- nested modeling of B[a]P pollution levels on regional (EMEP) and national (the Czech Republic) scales;
- multiple regression analysis to determine functional relationship for selected measurements of urban concentrations;
- application of results of regression analysis to construct refined spatial distribution of B[a]P annual mean air concentrations over the Czech Republic.

Modeling of B[a]P pollution with fine spatial resolution

To evaluate regional scale transport of B[a]P and spatial variability of air concentrations with fine resolution model simulations for two nested domains are carried out. Coarse resolution domain with spatial resolution 50x50 km^2 covers the whole EMEP region. Nested inner domain for modeling with finer spatial resolution 5x5 km^2 covers the territory of the Czech Republic and parts of neighbouring countries.

Emission data for these model simulations are generated on the basis of national emission data [*Ostatnická et al.*, 2014] and official emissions for 2013 received from CEIP. The most significant

contribution to B[a]P pollution in the Czech Republic is made by the residential combustion for domestic heating followed by industrial sources and road transport [*Ostatnická et al.*, 2014]. Relative importance of these emission source categories varies across the country. In Fig. 2.11 contributions of these types of sources to B[a]P emissions are shown for urban areas of Prague and Ostrava. For the Prague urban area two most important source categories include residential combustion and road transport. At the same time, in case for Ostrava significant contribution is made by residential combustion and industrial sources.

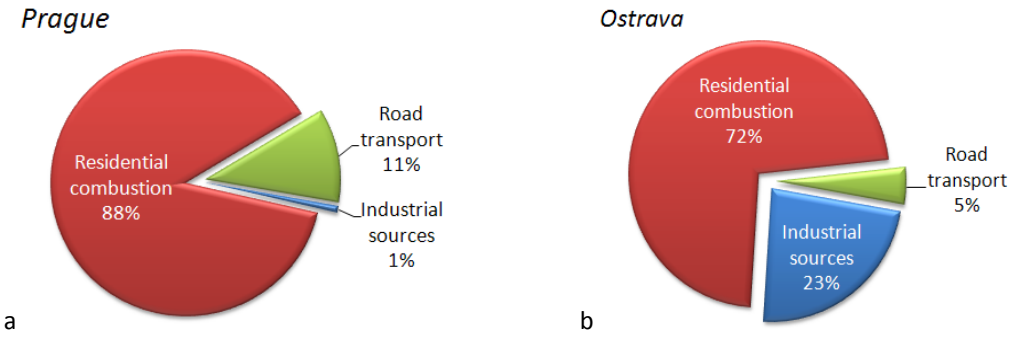


Fig. 2.11. *Relative contributions of main source categories to B[a]P emissions for urban areas of Prague (a) and Ostrava (b)*

Simulated annual mean B[a]P air concentrations for 2013 obtained from nested modeling are illustrated in the Fig. 2.12.

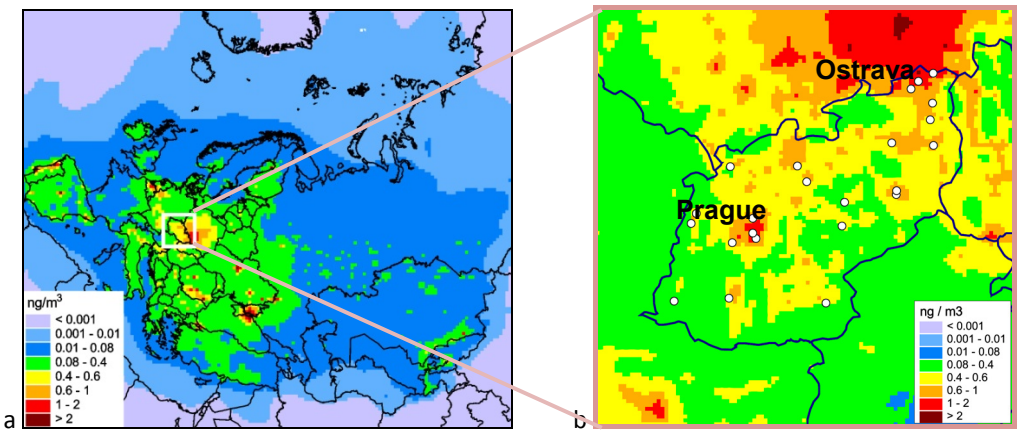


Fig. 2.12. *Spatial distribution of annual mean modeled B[a]P air concentrations for2013 calculated for two domains: EMEP domain with spatial resolution 50x50 km^2 (a) and national scale domain with resolution 5x5 km^2 (b). Circles denote locations of monitoring stations*

Comparison of modeling results with fine spatial resolution and measured B[a]P concentrations for urban and suburban sites in the Czech Republic is shown in Fig. 2.13. Level of agreement between the model predictions and measurements varies in different parts of the Czech Republic. In particular, B[a]P concentrations measured at the stations in Prague are satisfactory reproduced by

the model. At the same time, concentrations observed at the stations in Ostrava are underestimated by the model about a factor of 3. In general, the model tends to underpredict measured of B[a]P concentrations at urban and suburban sites. The underprediction can be explained by the effect of sub-grid variability of air concentrations that is not resolved by the modeling with applied spatial resolution.

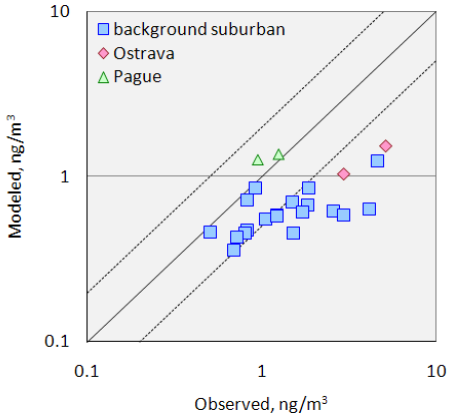


Fig. 2.13. *Comparison of annual mean modeled B[a]P air concentrations with measurements of Airbase monitoring sites in the Czech Republic for 2013. Dashed lines denote the area of agreement within a factor 2*

Multiple regression analysis to evaluate B[a]P concentrations in urban areas

The methodology of this study is based on complementary use of modeling results with fine spatial resolution, measurement data of the Airbase monitoring sites, and multiple regression analysis to determine functional relationship between measured urban concentrations and regression parameters (meteorological, geophysical data, and modeling results). A number of studies have been applied multiple regression models to evaluate levels of ozone, NO_x, PM_{10}, and $PM_{2.5}$ concentrations for urban areas using measurements as well as additional characteristics including meteorological, geophysical data, and modeling results [*Cuvelier et al*., 2005; *Amann et al*., 2007; *Moussipopolous et al*., 2011; *Ortiz and Freidrich*, 2013]. Levels of B[a]P air concentrations in the EU countries and population exposure for 2012 have been estimated in the recent study of EEA using linear regression model followed by krigging of residuals [*Guerreiro et al.*, 2014]. Experience of these studies is used for the construction of regression model for the evaluation B[a]P concentrations in urban areas.

The following characteristics are included into the regression model to describe spatial variability of concentrations in urban areas, namely, spatial variation of near surface wind speed, contributions of major emission source categories, and variance of modeled air concentrations with fine spatial resolution. This set of parameters is applied at the initial stage of the study. At subsequent stages additional characteristics of local meteorological and geophysical conditions influencing air concentrations (e.g. parameters of atmospheric stability, mixing layer height, and elevation) will be considered for the inclusion into the regression model.

The following equation is used to perform multiple regression analysis to evaluate functional relationship between observed B[a]P air concentrations in urban areas and selected set of regression parameters:

$$C_j = C_{est,j} + \omega_j = \alpha \cdot Var_j + \frac{1}{W_{av,j}}\left(\beta_{IND} \cdot e_{IND,j} + \beta_{RES} \cdot e_{RES,j} + \beta_{ROT} \cdot e_{ROT,j}\right) + \omega_j , \qquad (2.1)$$

where:

C_j is observed B[a]P air concentration in the grid cell *j*,

$C_{est,j}$ is the value of B[a]P concentration in grid cell *j* estimated by regression model,

$W_{av,j}$ is average near surface wind speed in the grid cell *j*,

$e_{IND,j}$, $e_{RES,j}$ and $e_{ROT,j}$ are emission densities of selected major B[a]P emission source groups (INDustry, RESidential heating and ROad Transport) in the grid cell *j*,

α, β_{IND}, β_{RES} and β_{ROT} are regression coefficients, and

ω_j is the value of "random component" in the grid cell *j*, which denotes the influence of factors not taken into account by the regression model.

Relative variations of modeled B[a]P air concentrations near the corresponding grid cell *j* Var_j is evaluated using 9-gridcell moving matrix: $Var_j = C_{max} / C_{min}$.

Analysis of relationship between observed B[a]P concentrations and selected parameters is performed using measurements of monitoring sites in the Czech Republic, which data have sufficient temporal coverage (more than 75%) of the year 2013. Locations of the sites are shown in Fig. 2.14. In particular, 23 background urban and suburban monitoring sites are selected for regression analysis. Additional 7 sites of industrial and traffic types are used as measurement data set for the verification of the regression model.

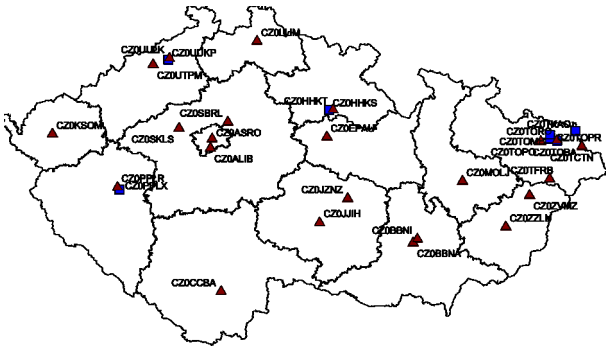


Fig. 2.14. *Locations of monitoring sites, selected for the regression analysis (triangles) and for the verification of regression model results (squares)*

Results of multiple regression analysis for the selected set of measurements made at background urban and suburban sites are shown in Fig. 2.15. In general, constructed regression model provides reasonable level of agreement with measurements of selected monitoring sites.

Fig. 2.15. *Observed annual mean B[a]P air concentrations at background urban and suburban sites vs. results of fine resolution modeling and of regression model*

To evaluate performance of the regression model, statistical parameters, describing the comparison of estimated B[a]P air concentrations with measurements and fine resolution modeling results, are calculated (Table 2.1).

Table 2.1. *Statistical parameters describing the results of the comparison of fine resolution modeling and regression model results on annual mean B[a]P air concentrations with measurements of selected background urban and suburban sites*

	Modeled concentrations	Regression model estimates
Relative bias	60%	0.01%
Normalized root mean square error (NRMSE)	85%	41%
Fractions of sites with agreement within a factor of 2	39%	96%
Correlation coefficient	0.52	0.79

It can be seen that application of the regression model leads to substantial decrease of bias between the modeled and observed concentrations and of normalized mean square error. Almost all values of concentrations, calculated by the regression model, are within a factor of 2 with observed concentrations. Besides, better correlation is obtained between regression model estimates and measurements in comparison with estimates of dispersion modeling. At the same time, for some of the monitoring sites estimated values of B[a]P concentrations differ from measurements, which will be discussed below in this section.

For the evaluation of regression model predictions, coefficients of obtained functional dependency (α, β_{IND}, β_{RES} and β_{ROT}) calculated for selected background urban and suburban monitoring sites are applied to the additional set of measurements at industrial and traffic sites in the Czech Republic. Results of testing of obtained functional dependence for these sites are shown in Fig. 2.16.

Preliminary analysis of obtained results indicates that developing approach is capable to provide reasonable estimates of B[a]P air concentrations in urban areas, which are consistent with the observed pollution levels. As it can be seen from the diagram in Fig. 2.15, estimated values of concentrations in general follow spatial variations of observed values. At the same time, for some of the urban areas, namely, the Ostrava region, differences between modeled and measured concentrations are more significant. Discrepancies between regression modeling results and measurements can be attributed to (1) uncertainties due to applied selection of parameters in the regression model, and (2) uncertainties of modeled concentrations generated by fine resolution modeling for this region. In particular, at current stage of developing approach for the regression analysis takes into account temporal and spatial variability of surface wind to evaluate the influence of meteorological conditions. At the same time, additional parameters

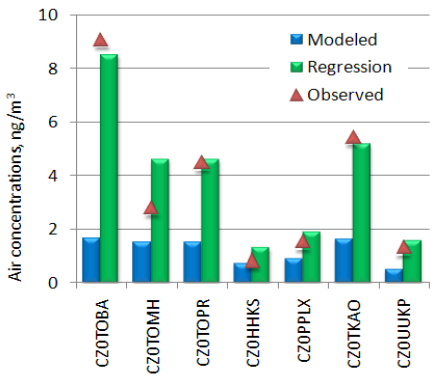


Fig. 2.16. *Observed B[a]P air concentrations at industrial and traffic sites vs. results of fine resolution modeling and of regression model*

of local variations of meteorological conditions (e.g. atmospheric stability, mixing layer height) significantly influence air concentrations and need to be considered in the regression model.

Uncertainties of estimated concentrations for the Ostrava region can be connected with underprediction of observed B[a]P concentrations by the fine resolution modeling. This difference is likely caused by the underestimation of long-range transport from emission sources of neighboring countries. The Ostrava area is a part of large agglomeration that extends to the territories of Poland and Slovakia. Thus influence of B[a]P emissions originated from these territories can be underestimated. In particular, comparison of fine resolution modeling results with measurements of B[a]P concentrations at monitoring sites in Poland and the Czech Republic shows that difference between modeled and observed concentrations for Poland is much larger than that for the Czech Republic (Fig. 2.17). This suggests that the transboundary transport from emission sources in the southern part of Poland is likely underestimated by the model.

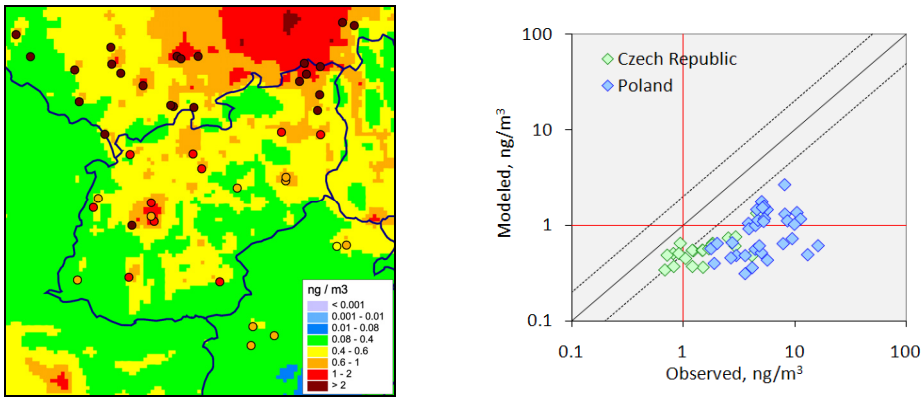


Fig.2.17. *Comparison of modeled B[a]P annual mean air concentrations with measurements of Airbase monitoring sites in Poland and the Czech Republic for 2013. Dashed lines denote the area of agreement within a factor 2*

In addition, seasonal variations of modeled and observed B[a]P pollution in the Ostrava area can be considered for the analysis of uncertainties of modeling results. Comparison of seasonal changes of B[a]P concentrations for monitoring sites in Ostrava and Prague shows that fine resolution modeling provides reasonable agreement of modeled and observed values for Prague, while for Ostrava significant differences is noted for the cold period of the year (Fig. 2.18). Similar pattern of seasonal variability and underprediction of air concentrations for the cold period is also seen for the EMEP site PL0005R in Poland (Fig. 2.23). This can indicate that B[a]P emissions in cold period of the year from the emission sources in Poland are likely underestimated and further refinement of the emission data applied for modeling is required. At further stages of this study more detailed evaluation of B[a]P pollution levels for this urban area will be made with application of trajectory analysis of pollution transport and source apportionment on the basis of measurements performed for different PAHs and specific tracers.

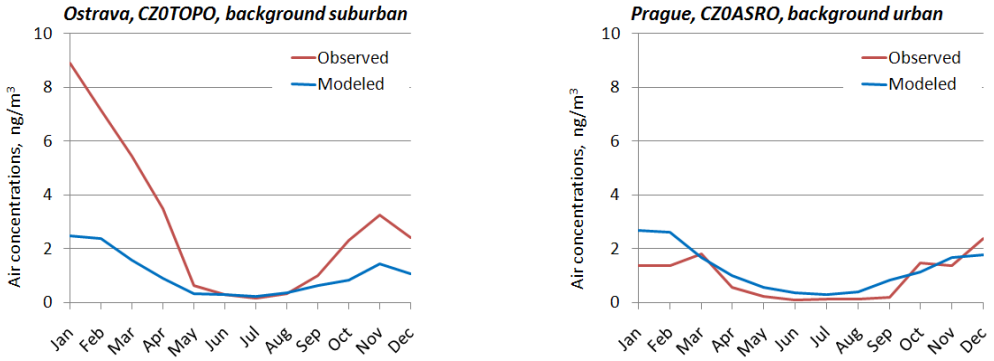


Fig. 2.18. *Comparison of monthly mean modeled B[a]P air concentrations with measurements of EEA AirBase background urban monitoring sites in Ostrava and Prague*

Evaluation of spatial distribution of B[a]P air concentrations in urban areas of the Czech Republic

Multiple regression analysis, performed for selected background urban and suburban monitoring sites, permits to obtain functional relationship between observed urban concentrations and additional parameters (see the equation 2.1). This relationship can be used then to estimate B[a]P air concentrations for urban areas of the Czech Republic where measurements are absent. In Fig. 2.19a spatial distribution of air concentrations in urban areas, calculated using regression equation (2.1) and values of coefficients α, β_{IND}, β_{RES} and β_{ROT}, derived from the regression analysis, is presented. Resulted distribution of B[a]P air concentrations is subsequently merged with the output of fine resolution model simulation using the gridded information on the fractions of urban and rural areas. Data on gridded distribution of urban and rural areas have been adapted from the outcome of the project GRUMP1 (http://sedac.ciesin.columbia.edu/). Air concentration in each cell of the model grid is evaluated as weighted average of concentration values for rural and urban areas using the following equation:

$$C_j = f_{urb,j} \cdot C_{urb} + (1 - f_{urb,j}) \cdot C_{\mathrm{mod}}$$

where $f_{urb,j}$ is the fraction of urban area in the grid cell *j*.

Combined spatial distribution of B[a]P annual mean air concentrations in the Czech Republic for 2013 is shown in Fig. 2.19b.

Concluding, it can be mentioned that presented approach in general is capable to reasonably describe spatial variations of B[a]P air concentrations in urban areas and to provide input information for exposure studies.

Discrepancies between estimated and observed B[a]P concentrations, obtained at this stage, can be related to the uncertainties of regression model estimates and uncertainties of emission data, used to perform modeling of B[a]P air pollution on regional and national scales.

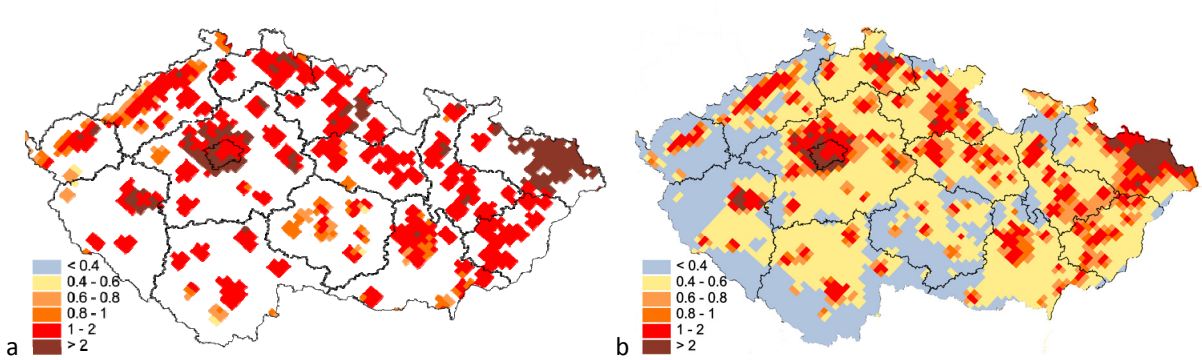


Fig. 2.19. *Spatial distribution of B[a]P air concentrations in urban areas, calculated using multiple regression analysis (a) and combined spatial distribution of B[a]P annual mean air concentrations in the Czech Republic for 2013 (b)*

To refine estimates of B[a]P air concentrations for urban areas further improvement of regression model is required, namely, inclusion of additional meteorological parameters influencing urban air concentrations (e.g. atmospheric stability, mixing layer height, etc.). *At further stage of the work developing approach is planned to be applied for other EMEP countries, provided that the data on emissions and measurements are available, in close co-operation with national experts on monitoring of pollution levels and evaluation of emissions as well as with experts from TFEIP, CEIP, CCC, and EEA.*

2.1.5. Comparison of modeled and measured PAH concentrations

Verification of modeling results has been carried out using comparison of modeled PAH air concentrations with measurements of the EMEP monitoring sites. Altogether 31 monitoring sites performed measurements of PAH concentrations in 2014 (Fig. 1.1). To analyze model predictions of annual mean PAH air concentrations measurements of 12 sites with sufficient temporal coverage (> 75%) of the year have been selected. In Fig. 2.21a modeled air concentrations of the sum of the 4 PAHs for 2014 are shown in comparison with corresponding observed concentrations. Seven monitoring sites among the selected sites performed measurements of all 4 PAHs in 2014. In general, there is a good agreement of modeled and observed air concentrations of the sum of 4 PAHs. For all selected monitoring sites agreement between observed and modeled concentration values is within 45% (Fig. 2.20).

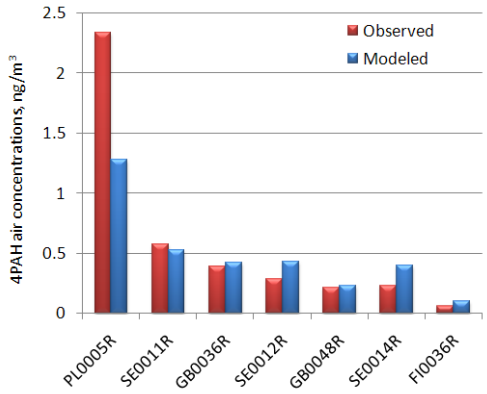


Fig. 2.20. *Comparison of modeled air concentrations of the sum of 4 PAHs with measurements of the EMEP monitoring sites for 2014*

Comparison of modeled and observed concentrations of individual 4 PAHs, namely, B[a]P, B[b]F, B[k]F, and IP, is presented in Fig. 2.21. Analysis of results indicates that modeled air concentrations

have captured the spatial distribution of observed values with correlation coefficients ranging from 0.7 to 0.8 for particular PAHs. About 70% of modeled concentrations values are within a factor of 2 in comparison with measurements of B[a]P, B[b]F, B[k]F, and IP.

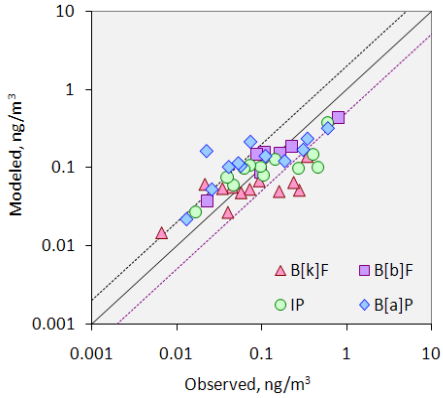


Fig. 2.21. *Comparison of modeled air concentrations of B[a]P, B[b]F, B[k]F, and IP with measurements of the EMEP monitoring sites for 2014. Dashed lines in right diagram denote the area of agreement between the modeled and measured values within a factor 2*

Comparison for individual PAHs shows that the highest differences (near or above a factor 3) between modeled and observed annual mean concentrations are obtained for monitoring sites DE0003R (for B[a]P, B[k]F), and DE0001R, DE0002R, DE0009R (for B[k]F, IP) (Fig. 2.22). Model predictions of B[a]P for the site DE0003R are substantially higher than measured concentrations, which can be partly explained by the specific location of this site. DE0003R is located in elevated place at the altitude about 1200 m. PAH concentrations measured at the site might be influenced by its local conditions, that is likely not properly captured by the model.

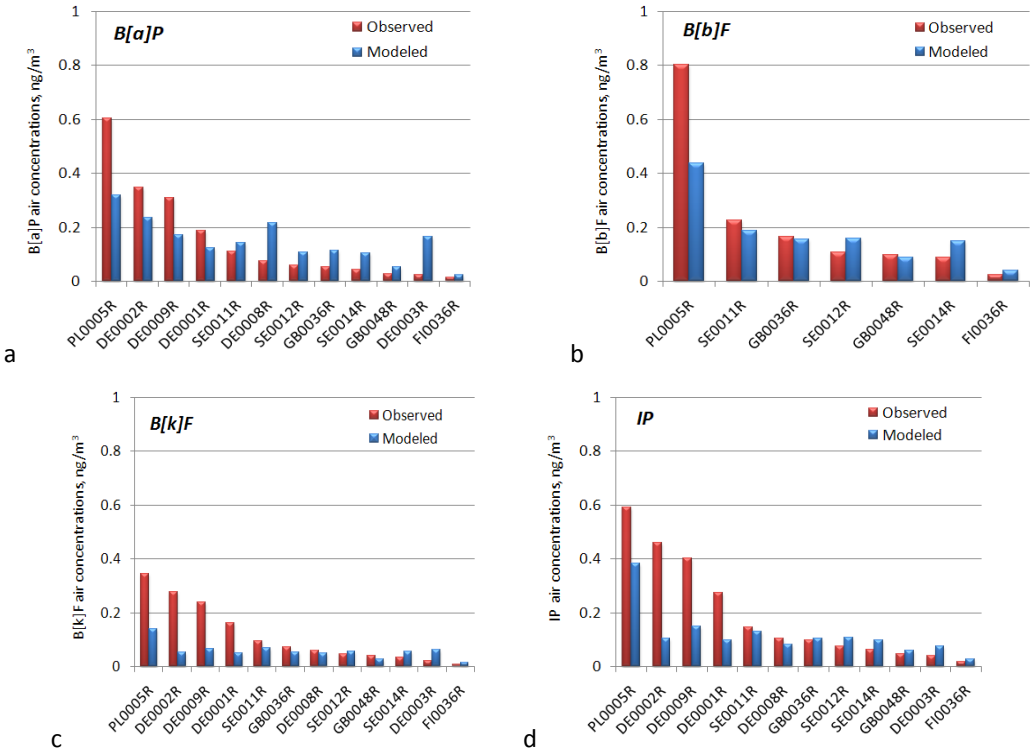


Fig. 2.22. *Comparison of modeled and observed B[a]P (a), B[b]F (b), B[k]F (c) and IP (d) air concentrations for 2014*

Significant underprediction of observed B[k]F and IP concentrations by the model is obtained for the sites DE0001R, DE0002R, and DE0009R. Among the likely reasons of this discrepancy the difference between relative contributions of individual PAH species in the emission data provided by Germany and other countries can be mentioned. In particular, B[a]P contribution to the emission of 4 PAHs, reported by Germany, exceeds 90%, while other three PAHs contribute less than 10%. This relative contributions to PAH emissions noticeably differ from averaged distribution reported by other EMEP countries, where contributions of considered PAHs vary between 30-50%.

Comparison of modeled and observed seasonal variations of B[a]P air concentrations for 2014 shows in general satisfactory agreement between model results and measurements. In Fig. 2.23 model predictions and measurements of monitoring sites BE0013R, CZ0003R, and PL005R, covering the whole year 2014, are presented. For most part of the year modeled concentrations capture variations of observed B[a]P air concentrations, being close or slightly higher than measurements. In some of the episodes the model overpredicts measured concentrations more significantly. In particular, it is seen for May and October for BE0013R, and for December for CZ0003R that requires more detailed trajectory analysis.

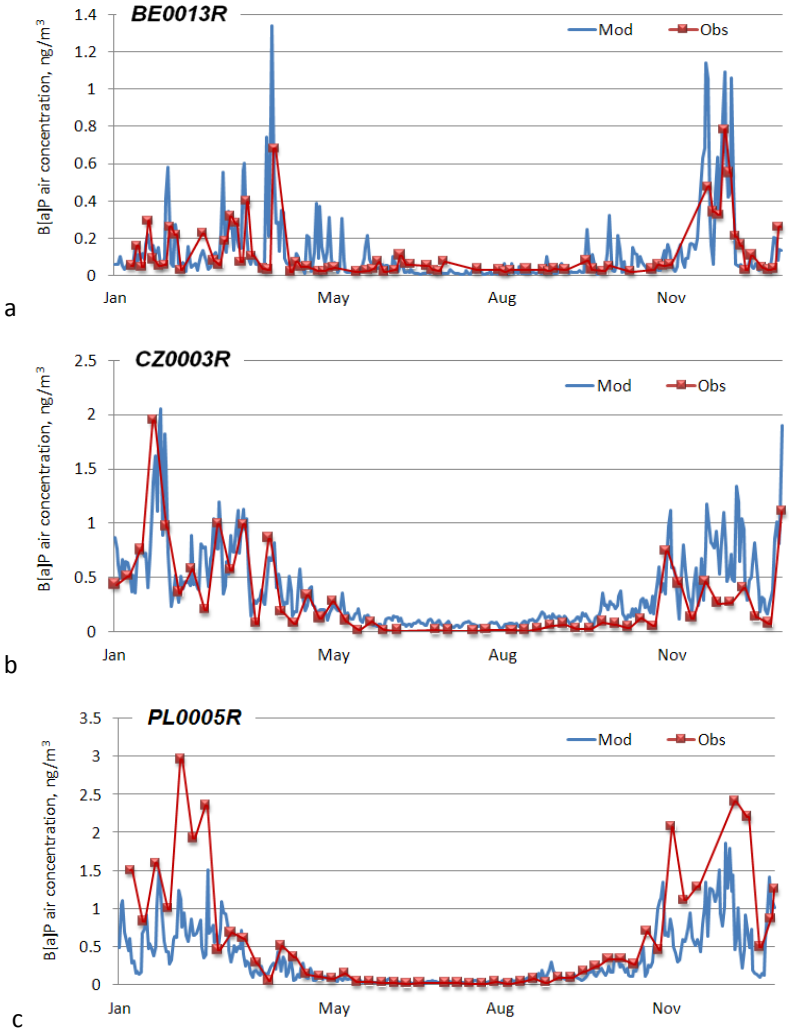


Fig. 2.23. *Comparison of modeled and observed seasonal variations of B[a]P air concentrations for 2014 for monitoring sites PL0005R, BE0013R, and CZ0003R.*

Measurements of the monitoring site PL0005R were carried out on weekly basis. Comparison of modeling results with measured B[a]P concentrations at this site shows that modeled concentrations agree well with measurements during the warm period of the year. At the same time, for colder period the model underpredicts observed concentrations, which is likely connected with underestimated B[a]P emission in cold period of the year.

Detailed information on the comparison of modeling results with measurements of the EMEP monitoring sites can be found in MSC-E Technical report [*Shatalov et al*., 2016].

2.1.6. Concluding remarks

Model assessment of PAH pollution levels and transboundary transport in the EMEP countries has been carried out for the indicator compounds, namely, B[a]P, B[b]F, B[k]F, and IP, included into the Protocol on POPs. Elevated levels of annual mean PAH air concentrations (1-2 ng/m^3 and more) in 2014 are characteristic of countries in Central and Eastern Europe (Poland, the Czech Republic, Slovakia, Slovenia, Romania, Bulgaria, Lithuania, Belarus, and Ukraine) as well as Southern Europe (Spain, Portugal). Verification of modeling results against the measurements of the EMEP monitoring sites has shown reasonable agreement between modeled and observed PAH air concentrations.

Results of model simulations indicate that transboundary transport of air pollution continues playing important role in the contamination of the EMEP countries. For the majority of countries the contribution of external emission sources to PAH deposition in 2014 exceeds the contribution of national emission sources.

More detailed analysis has been performed for the B[a]P pollution levels. Evaluation of trends in B[a]P air concentrations in the EMEP region, carried out for the period 1990-2012, has shown that decline of B[a]P air pollution levels slowed significantly during recent ten years and air concentrations even started to increase in some of the EMEP countries. According to measurements and modeling results high levels of air concentrations, exceeding the EU limit values for B[a]P, are indicated in various regions of the EMEP domain, in particular, in Central and Eastern Europe. About half of monitoring stations in urban and suburban areas in Europe measured B[a]P air concentrations above the EU target level in 2013.

To contribute to the analysis of population exposure to elevated air pollution levels EMEP has initiated a study aiming at quantification of B[a]P air concentrations in urban areas. First stage of this study is focused on the analysis of B[a]P air concentrations in the Czech Republic due to availability of data on emissions and measurements. At further stages this work can be continued for other EMEP countries in areas with exceedances of EU target levels for B[a]P.

Obtained results show in general reasonable agreement of estimated B[a]P concentrations for urban areas in the Czech Republic with available measurements. At the same time, in order to reduce the level of uncertainties in estimates of B[a]P concentrations for urban areas further development of applied approach is required. Besides, improvement of completeness of national inventories of PAH

emissions and information on seasonal variations of emissions are of importance for the evaluation of air pollution levels. To refine estimates of B[a]P concentrations in urban areas extension of existing monitoring network is needed.

Air pollution by carcinogenic PAHs is recognized as a serious problem in areas with dense population of many EMEP countries. To consider this issue a thematic session is to be organised during the Second joint session of the Steering Body to EMEP and Working Group on Effects. It is planned to discuss the current state of monitoring and assessment of PAH pollution of the EMEP countries as well as population exposure in order to contribute to further reduction of air pollution levels. Participation of experts in the preparation of emissions, monitoring of air pollution, and evaluation of health effects from national and international organizations and their contribution to this activity are highly appreciated.

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

Polychlorinated dibenzo(p)dioxins and dibenzofurans are two groups of chlorinated compounds characterized by similar chemical structures and properties, that include insolubility in water, lipophilicity, and high persistence in the environmental media (e.g. soils, sediments). PCDD/Fs are unintentionally released into the environment during various combustion processes (e.g. including industrial processes, waste incineration, and open burning of wastes and biomass). Long-term accumulation of PCDD/Fs in the surface media can pose risk to the ecosystems and human health. Monitoring and assessment of presence of dioxins and furans in the environmental media are under special attention of various national and international organizations, such as LRTAP Convention, Stockholm Convention, Helsinki Commission (HELCOM), US Environment Protection Agency (US EPA), European Union (EU), German Environmental Protection Agency (UBA), Swedish Environmental Protection Agency (SEPA), and others. A number of countries have established maximal admissible levels of dioxins/furans in the atmosphere and other environmental compartments. For example, threshold level for PCDD/F soil concentrations in Canada is set to 4 ng TEQ/kg (Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health). In Italy soil acceptable concentration limit (SACL) for PCDD/Fs in soil amounts to 10 ng TEQ/kg (D.Lgs 152/06).

Analysis of available emission data, performed in the studies of *K.Breivik* et al. [2004], *H.Fiedler* [2007], and *K.Mareckova et al.* [2012], has indicated that available national inventories of PCDD/F emissions most likely do not cover all potential sources of PCDD/Fs releases to the atmosphere and thus can not explain observed levels of pollution. In course of the evaluation of PCDD/F pollution in the EMEP countries MSC-E has carried out a number of studies in order to analyze available data on dioxins and furans emissions and to examine their uncertainties. In particular, reported PCDD/F emission data and pollution levels in the Baltic Sea region were analyzed in the framework of the BalticPOPs research program [*Shatalov et al.*, 2012a; *Wiberg et al.*, 2013]. One of the aims of this study was to trace the origin of dioxins and furans in the Baltic Sea region and to evaluate whether the observed PCDD/F air concentrations could be reproduced by the modeling using officially reported PCDD/F emissions. Combined analysis of air monitoring and modeled data showed that the use of officially reported emissions for modeling led to underestimation of measured air

concentrations and deposition fluxes. The most important source regions for the Baltic Sea area were likely the central and eastern parts of Europe, which could be characterized by underestimated emissions. Several modeling experiments with regionally adjusted emission scenarios showed that increase of emissions in these regions could substantially reduce underestimation of observed pollution levels.

Later on this work was continued with the analysis of congener specific emission of PCDD/Fs on the basis of extended set of measurements [*Shatalov et al.*, 2012b]. Application of this approach and construction of experimental scenario emissions for modeling permitted to improve agreement of modeled values with available measurements and to estimate the likely levels of PCDD/F releases to the environment. These estimates exceeded officially reported emissions about five times, which was in line with the range of uncertainties of dioxins and furans emissions in the EMEP countries indicated in studies of [*Pulles et al.,* 2005; *Pulles et al.,* 2006].

This year model assessment of PCDD/F pollution levels in the EMEP region has been continued on the basis of experimental emission scenarios for regional and global modeling. Scenarios have been prepared on the basis of emission data, officially reported by the EMEP countries, and national inventories collected under the UNEP SC. Results of the assessment include estimates of spatial distribution and transboundary transport of pollution between the EMEP countries. Specific attention has also been given to further refinement of scenarios of PCDD/F emissions. Ongoing work in this direction includes construction of scenario of congener composition of dioxins and furans emissions as well as updating scenario of global emission. Brief overview of progress and outcome of this study are presented below. Detailed information on the results of model simulations and their analysis are given in the MSC-E Technical Report [*Shatalov et al*., 2016].

2.2.1. Emission data for model assessment

Evaluation of PCDD/F pollution in the EMEP domain in 2014 has been performed using gridded emission data provided by CEIP. Detailed information on PCDD/F emissions reported by countries, as well as the gap-filling methods, used for the elaboration of GNFR emission inventory for 2014 (as reported in 2016) can be found in the Technical report CEIP 02/2016 [*Tista et al*., 2016]. Spatial distribution of dioxins and furans emissions in the EMEP domain for 2014 is shown in Fig 2.24.

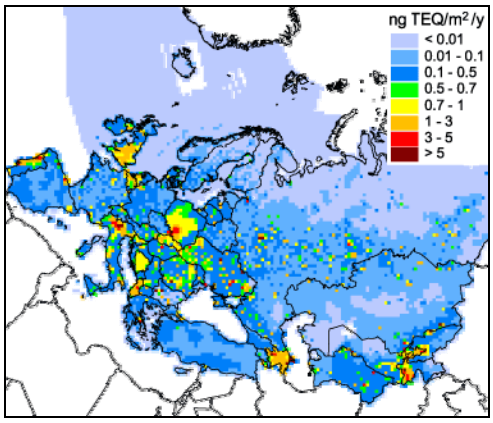


Fig. 2.24. *Spatial distribution of PCDD/F emissions in the EMEP domain in 2014 with resolution 50x50 km²*

PCDD/F emissions from anthropogenic sources of the EMEP countries in 2014 are almost 30% lower than corresponding emissions in 2013, provided in the previous submission of emission data [*Mareckova et*

al., 2015]. According to these data the value of annual PCDD/F emission for 2014 accounts for about 4.2 kg TEQ.

Changes of PCDD/F emissions from 2013 to 2014 are illustrated in Fig 2.25. In particular, differences in the reported total values for 2014 (E_{2014}) and for 2013 (E_{2013}) are displayed for each EMEP country (E_{2014}-E_{2013}). Positive values in the diagram indicate an increase of PCDD/F emission from 2013 to 2014, while negative values indicate a decrease of emission.

The most significant changes of PCDD/F emissions are noted for several EECCA countries and Turkey. Particularly, emissions of Georgia, the Republic of Moldova, Turkmenistan, and Tajikistan for 2014 are higher than emissions for 2013 by a factor of 84, 10, 4, and 3, respectively. The largest decreases of emissions are seen for Kazakhstan, Ukraine, and Turkey (by a factor of 10, 3, and 1.5, respectively).

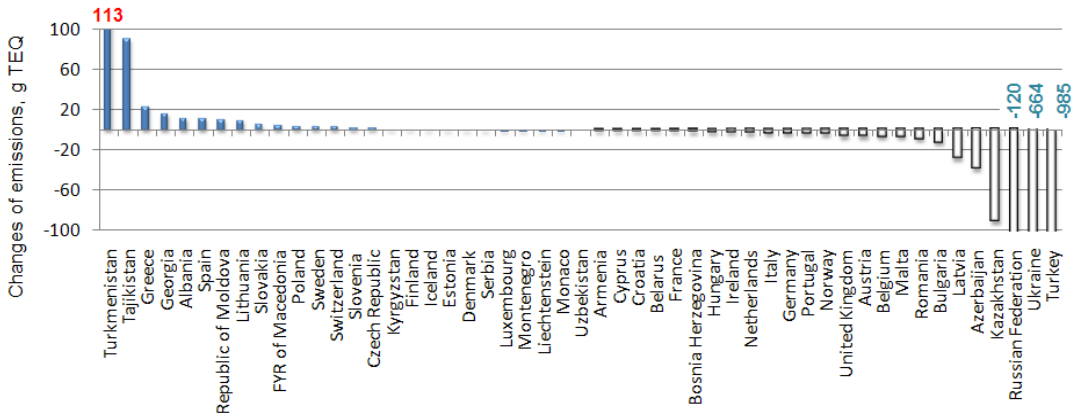


Fig. 2.25. *Changes of annual total emissions of PCDD/Fs from 2013 to 2014 (E_{2014}-E_{2013}), g TEQ. Negative values denote decrease of emissions, and positive ones – increase of emissions*

Changes of PCDD/F emissions of Georgia, similar to the PAH emissions (Section 2.1), are connected with the application of the new national energy balance, prepared by GEOSTAT of Georgia. PCDD/F emissions have covered almost all activities of energy sector, which has lead to substantially higher estimates of PCDD/F releases [*IIR of the Republic of Georgia*, 2016]. For the Republic of Moldova, recalculations of emissions have been made for all source categories using updated emission factors of the EMEP/EEA Emission Inventory Guidebook 2013 instead of factors applied before [*IIR of the Republic of Moldova*, 2016]. Significant changes of emissions for Ukraine and Turkey are caused by the use of national emissions of these countries, reported to the Stockholm Convention on POPs, which have substituted expert estimates from the TNO emission inventory. In case of Kazakhstan officially reported national data on emissions have been used, submitted by the country for the first time.

Expert estimates of PCDD/F emissions for Turkmenistan and Tajikistan, for which officially reported data are not available, have been elaborated by CEIP on the basis of literature information and data on GDP. Particularly, estimates of emission for these two countries have been extrapolated from the

data of the study [*Hodjamberdiev*, 2006] and values of GDP per capita up to the year 2014 [*Tista et al.*, 2016].

Relatively high changes of PCDD/F emissions (more than 50%) can also be noted for Greece, Liechtenstein, Iceland, Lithuania, the FYR of Macedonia, Malta, and Latvia. The other EMEP countries are characterized by less significant changes.

For the evaluation of global scale transport and fate of PCDD/Fs experimental emission scenario, constructed on the basis of the UNEP SC inventory of dioxins and furans emissions, have been used [*Gusev et al.*, 2014b; *Shatalov et al.*, 2014]. Global gridded emissions of PCDD/Fs to the atmosphere and soil were prepared using the national emission inventories of 68 countries. Spatial distribution of PCDD/F emissions to air and soil, applied in global scale model simulations for 2014, is shown in Fig.2.26.

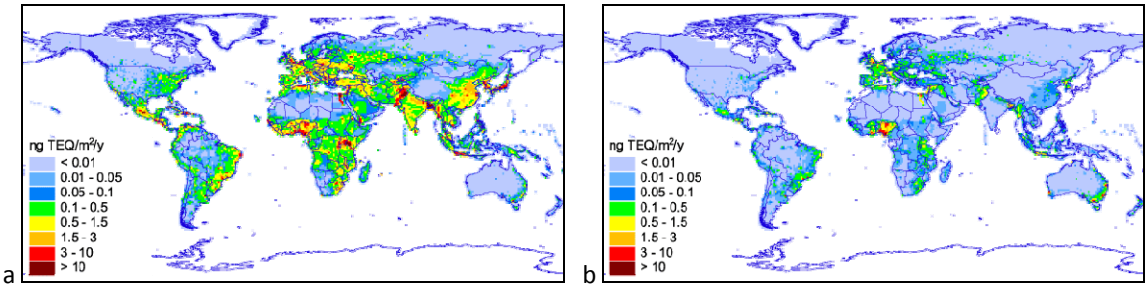


Fig. 2.26. *Spatial distribution of global annual PCDD/F emissions (ng TEQ/m^2/y) to the atmosphere (a) and to soil (b) with spatial resolution 1°x1°, applied in model simulations for 2014*

This year refinement of the mentioned above scenario has been started by MSC-E on the basis of updated national inventories of PCDD/F emissions, submitted recently by countries to the UNEP Stockholm Convention. It is planned to use updated scenario of global PCDD/F emissions next year in model simulations for 2015.

According to the national reports, published at the UNEP SC web site in the beginning of 2016, inventories of dioxins and furans emissions were available for more than 140 countries [http://chm.pops.int/Implementation/NIPs/NIPTransmission/tabid/253/Default.aspx]. The reference years of the emission data cover the period from about 1998 up to 2014, at the same time most of the inventories have been made for years around 2004. Significant number of countries has provided estimates of PCDD/F releases for several years that can be used for the evaluation of temporal changes of emissions.

Geographical distribution of latest available national PCDD/F emissions to the atmosphere reported by countries to the UNEP SC is shown in Fig.2.27. In accordance with submitted data the largest contributions to the global emission have been made by Africa (36%) and South and East Asia (34%). The EMEP countries have contributed 15% and contribution of North and South Americas is accounted for about 13%. It is seen that for some of the countries data on emissions are missing, therefore actual contributions of particular regions might be somewhat different.

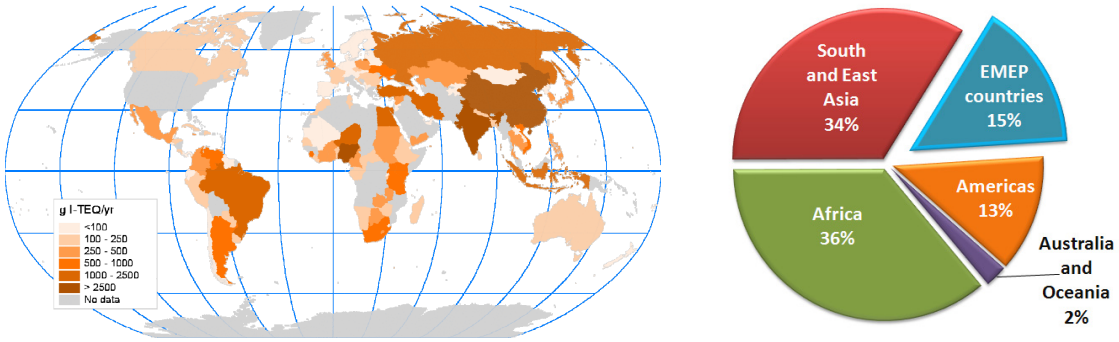


Fig. 2.27. *Total releases of PCDD/Fs to the atmosphere from anthropogenic emission sources in different countries (g TEQ/y) and contributions of major source regions to global PCDD/F emissions*

Inventories of PCDD/F releases, collected under the UNEP SC, provide important information on sector distribution of emission and emissions to media other than the atmosphere. In accordance with these data the largest contribution to global emission is made by uncontrolled open burning processes (42%) followed by metal production (19%) and power generation and heating (16%) (Fig. 2.28). At the same time, data on emissions for the EMEP region demonstrate that major contribution is made by ferrous and non-ferrous metallurgy (37%) as well as power generation and heating (21%), while open burning processes contribute only 19%. These three source categories of releases comprise the largest contribution (more than 75%) to overall PCDD/F emissions to the atmosphere.

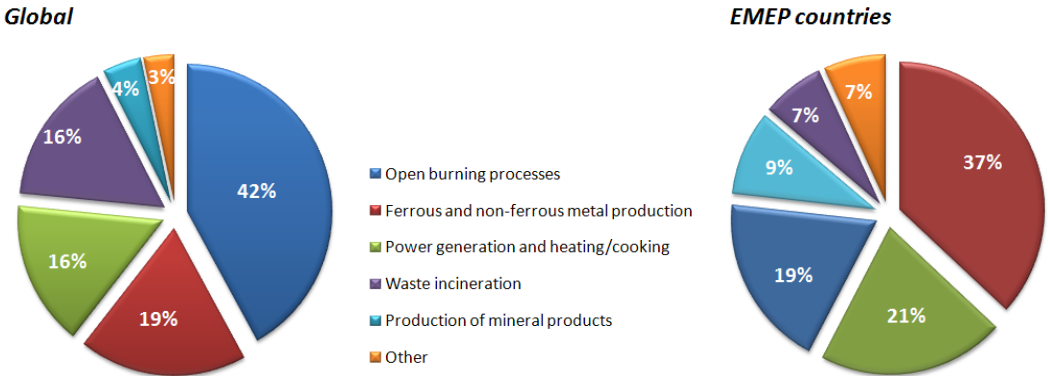


Fig. 2.28. *Major source categories of PCDD/F emissions to the atmosphere for the global scale emissions and for the emissions from the EMEP countries*

National PCDD/F emission inventories, reported by countries to the Stockholm Convention, are being updated in course of the refinement of the UNEP Standardized Toolkit and recalculations of national emission data [*Black et al.*, 2012; *Fiedler*, 2015]. In particular, according to available requirements inventories of national releases need to be revised every five years. Thus, *the information reported by countries to the Stockholm Convention is of importance for further studies of environmental pollution by dioxins and furans as in the EMEP region and on the global scale and further co-operation between the LRTAP and Stockholm Conventions in this field is highly appreciable.*

2.2.2. Congener composition of PCDD/F emissions

Congener specific information on the emissions and concentrations in the environment is of importance for the analysis of PCDD/F transport and fate. Dioxins and furans are released into the environment as complex mixtures of different congeners that have their own physical-chemical properties that differ substantially and depend on environmental characteristics (e.g. temperature). In particular, lower chlorinated congeners are semi-volatile, partitioning in the atmosphere between gaseous and particulate phase, and exchange efficiently with terrestrial and aquatic surfaces, while more chlorinated congeners are less volatile and tend to associate with atmospheric aerosol particles that control their transport. Differences in the properties lead to differential removal and fractionation of particular congeners in course of their long-range transport.

The most part of available inventories of PCDD/F releases to the environment provides emissions of the mixture of seventeen 2,3,7,8-substituted PCDD/Fs in the toxicity units without splitting on particular congeners [*Breivik et al.*, 2004]. Particularly, emission data, reported by the EMEP countries and national inventories of dioxins and furans of the UNEP SC, are prepared using the toxic equivalency approach. At the same time, to evaluate levels of pollution and toxicity of dioxins and furans, model assessment requires taking into account congener composition of PCDD/F emission to predict transport and fate of individual congeners.

In course of model assessment of PCDD/F pollution levels in the EMEP countries MSC-E performed analysis of available data on the emissions of dioxins and furans and their congener composition. In particular, collection of information on emissions of the seventeen 2,3,7,8-substituted PCDD/Fs in the EMEP countries and their congener profiles was carried out at the previous stages of the work [*Vulykh and Shatalov*, 2001]. It was indicated that major source categories of PCDD/F emissions included combustion of fossil fuels, inciniration of wastes, and secondary processing of non-ferrous metals. Individual congeners provide different relative contributions to overall emission of PCDD/Fs. According to the data collected from literature three homologue groups of PCDD/F congeners, in particular, PeCDFs, HxCDFs, and PeCDDs, contributed more than 70% to the emitted dioxins and furans mixture toxicity.

This year analysis of information on congener-specific PCDD/F emissions from various source categories has been continued in order to elaborate experimental scenario of congener patterns of reported emission and to apply it for the evaluation of environmental pollution by dioxins and furans. This study includes collection of additional information on the PCDD/F emission profiles for various types of emission sources available in literature, and statistical analysis of collected data to determine typical congener patterns for major source categories of PCDD/F emissions.

According to national inventories of PCDD/F emissions, officially reported by the EMEP countries, the most important emission source categories, contributing to the PCDD/F toxicity, are public power, stationary combustion, industry, waste incineration, road transport, and agriculture. The information on congener profiles of PCDD/F emissions related to these source categories is collected and preliminary processed using statistical methods. To analyze congener patterns of dioxins and furans emissions the hierarchical cluster analysis is planned to be used similar to a number of studies

examining congener specific emissions and concentrations of PCDD/Fs [*Fiedler et al.*, 2000; *Towey et al.*, 2010; *Quass et al.*, 2016].

Cluster analysis permits to group individual congener profiles with similar properties. Selected set of profiles can be further used to construct typical profiles for particular emission source groups and apply them for modeling purposes. Preliminary results of the analysis for two source categories, namely, combustion of fossil fuels (public power) and municipal waste incineration, are illustrated in the Fig. 2.29 and Fig. 2.30.Though this approach is subject of considerable uncertainties due to aggregation of information for different types of sources it can be used for scenario modeling of PCDD/F long-range transport and analysis of pollution levels.

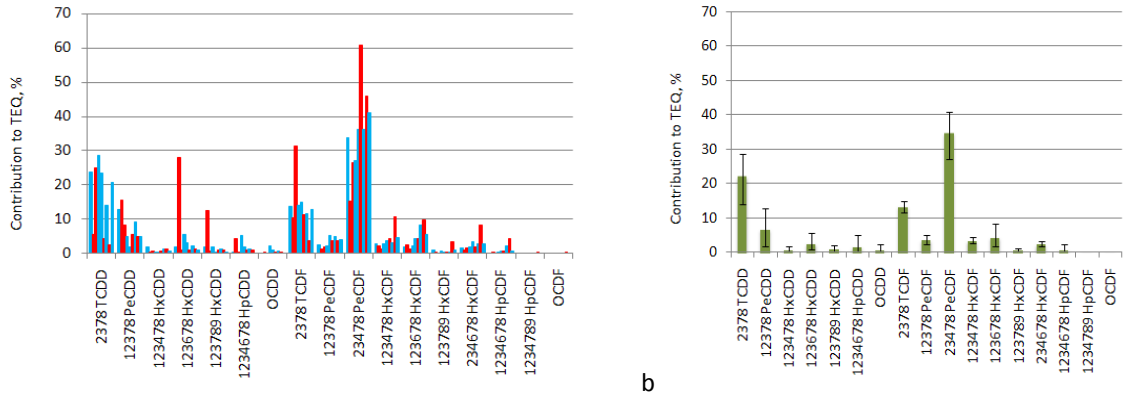


Fig. 2.29. *Congener profiles of 17 PCDD/Fs emitted from combustion of fossil fuels (public power), determined in different studies [US EPA, 2006; Nielsen et al., 2010; Oehme and Mueller, 1995; Thoma, 1988; Bacher et al., 1992] (a). Blue bars denote profiles included in the cluster, red bars denote profiles not included into the cluster. Average congener pattern of PCDD/F emission based on selected profiles for the cluster (b)*

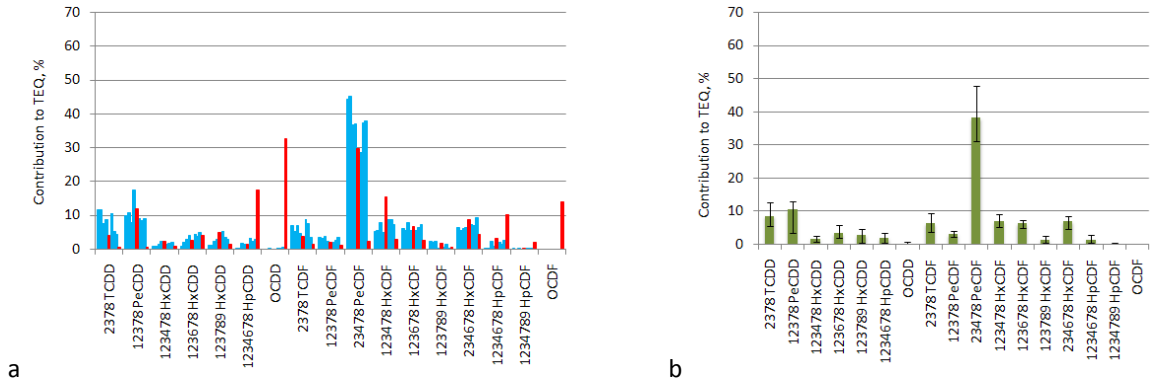


Fig. 2.30. *Congener profiles of 17 PCDD/Fs emitted from municipal waste incineration, determined in different studies [Spinnel et al., 2008; Ni et al., 2009; Kao et al., 2006; Cleverly et al., 1997; EPA, 2006; Lee et al., 2003] (a). Blue bars denote profiles included in the cluster, red bars denote other profiles not included into the cluster. Average congener pattern of PCDD/F emission based on selected profiles for the cluster (b)*

Refinement of estimates of congener composition of PCDD/F emissions is of importance for further progress in the assessment of pollution levels. Further work in this direction can be performed in course of updating of available expert estimates of congener-specific emissions for the EMEP region and for the global scale, including also estimation of emissions from particular source categories. For this purpose further co-operation with CEIP and TFEIP as well as other experts on preparation of emissions inventories is important.

2.2.3. PCDD/F pollution levels in the EMEP region and on global scale

Model assessment of PCDD/F pollution levels in the EMEP domain has been carried out using combined regional and global scale modeling. In order to set up boundary conditions for regional model simulations global model runs for 2014 have been used. Model simulations of global transport and fate of dioxins and furans have been performed using the experimental emission scenario based on the emission inventory of the UNEP SC described in [*Gusev et al.*, 2014b; *Shatalov et al.*, 2014]. Initial conditions to evaluate pollution levels in 2014 have been obtained using long-term spin-up model run for the period from 1930s to present time. Spatial distribution of global scale annual mean PCDD/F air concentrations generated for 2014 is presented in Fig. 2.31.

According to obtained results relatively high levels of PCDD/F air concentrations (10-50 fg TEQ/m^3) are noted for Africa and South Asia. Lower levels of pollution (1-25 fg TEQ/m^3) are characteristic of Europe, North and South America, and Australia.

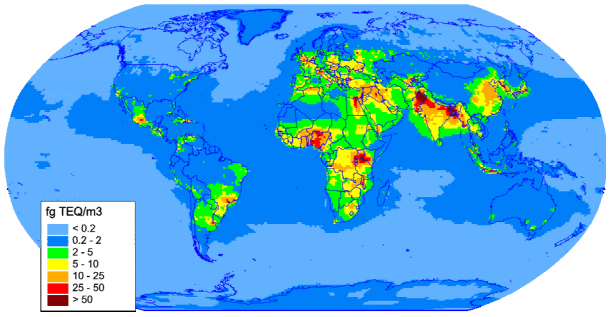


Fig. 2.31. *Spatial distribution of modeled annual mean PCDD/F concentrations in air (fg TEQ/m^3) obtained for 2014*

Comparison of model predictions with measurements, performed at the previous stages of model assessment of PCDD/F pollution levels, has shown reasonable agreement between modeled and observed air concentrations [*Gusev et al.*, 2014a; *Schuster et al.*, 2015].

In general, the model tends to slightly underestimate PCDD/F concentrations measured at rural and background sites in Europe, South America, and East Asia in the period from 2002 to 2012. At the same time, more significant underprediction of observed concentrations has been obtained for North America. To improve evaluation of PCDD/F global scale transport and fate further co-operation with the activities of the UNEP SC related to monitoring of dioxins and furans in the atmosphere and other environmental compartments is of importance.

Model estimates of annual mean PCDD/F air concentrations and deposition fluxes in the EMEP region for 2014 are shown in Fig. 2.32. *Elevated levels of annual mean air concentrations (10-50 fg TEQ/m^3 and higher) are characteristic of Poland, northern Italy, Portugal, Slovakia, Romania, Albania, and Azerbaijan (Fig. 2.32a). Areas of relatively high air concentrations can also be noted for Ukraine, the*

Russian Federation, Tajikistan, and Turkmenistan. Spatial distribution of annual deposition fluxes of PCDD/Fs has in general similar pattern to the distribution of air concentrations (Fig. 2.32b).

Results of regional model simulations provide information on the pollution of marginal seas within the EMEP region. The largest level of annual mean PCDD/F deposition fluxes is calculated for the Baltic Sea (0.5 ng TEQ/m^2/y) followed by the Black Sea (0.46 ng TEQ/m^2/y), the Caspian Sea (0.4 ng TEQ/m^2/y), the North Sea (0.35 ng TEQ/m^2/y), and the Mediterranean Sea (0.3 ng TEQ/m^2/y).

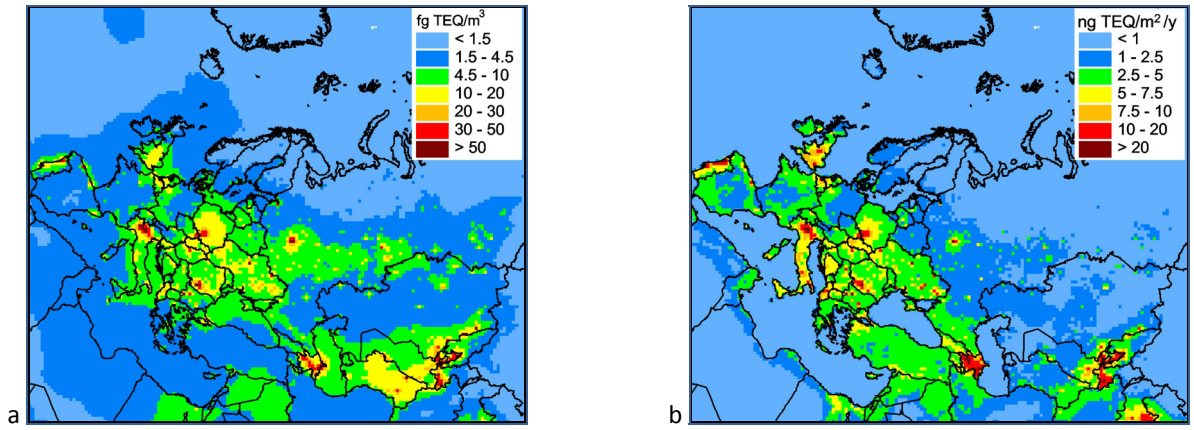


Fig. 2.32. *Spatial distribution of modeled annual mean air concentrations (fg TEQ/m^3) in the EMEP domain (a) and annual deposition fluxes (ng TEQ/m^2/y) (b) of PCDD/Fs for 2014*

To characterize inter-annual variability of PCDD/F pollution levels in the EMEP region, results of model assessment for 2014 have been compared with estimates of pollution levels, obtained for the year 2013. Changes in pollution levels between these two years are shown in Fig. 2.33 as a difference between annual mean deposition fluxes, calculated for 2014 and 2013 for each EMEP country. Variations of pollution levels take place due to changes of emission data, used in model simulations, and changes of meteorological conditions.

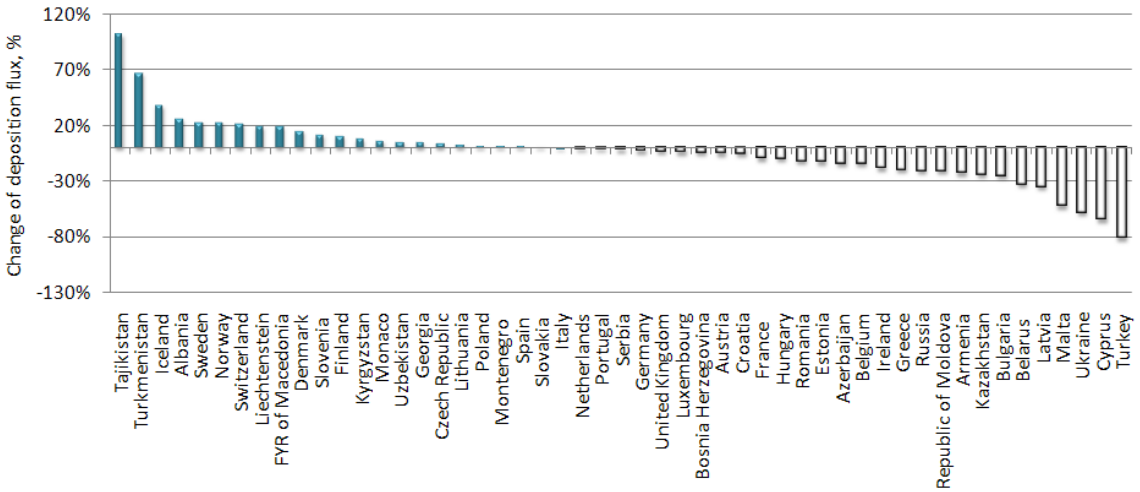


Fig. 2.33. *Changes of annual mean PCDD/F deposition fluxes in the EMEP countries between 2013 and 2014, (C_{2014}-C_{2013})/C_{2013}. Negative values denote decrease, and positive ones – increase*

Levels of annual mean PCDD/F deposition fluxes in the EMEP countries varied from 2013 to 2014 from about 80% decrease to about 100% increase. The largest decline of deposition (more than 50%) takes place for Turkey, Cyprus, Ukraine, and Malta, which is mostly caused by decreased national emissions. The largest increase of deposition (more than 50%) in 2014 is calculated for Tajikistan and Turkmenistan due higher total emissions for 2014 comparing to 2013. The other countries are characterized by lower changes of annual mean PCDD/F deposition fluxes varying in the range from about -30% to 30%.

2.2.4. Transboundary transport of PCDD/Fs in 2014

Transboundary transport of dioxins and furans within the EMEP region has been evaluated for 2014 taking into account contributions of anthropogenic and secondary emission sources. The influence of non-EMEP emission sources has been estimated using global scale model simulations with application of scenario emissions, described above. According to modeling results, *relative contribution of the EMEP anthropogenic emission sources to deposition of PCDD/Fs in the EMEP countries is accounted for about 45%, secondary emission sources contribute about 50%, and non-EMEP emission sources contribute about 5%.*

Annual total deposition fluxes of PCDD/Fs to the EMEP countries are illustrated in Fig. 2.34. Relatively high levels of mean PCDD/F deposition fluxes in the EMEP countries (more than 8 ng TEQ/m^2/y), are calculated for Azerbaijan, Albania, and Portugal. Elevated deposition fluxes (2 – 6 ng TEQ/m^2/y) are obtained for the countries of Central and Eastern Europe as well as for the EECCA countries. Relatively low levels of deposition are characteristic of the Scandinavian countries and remote regions of the Russian Federation and Kazakhstan.

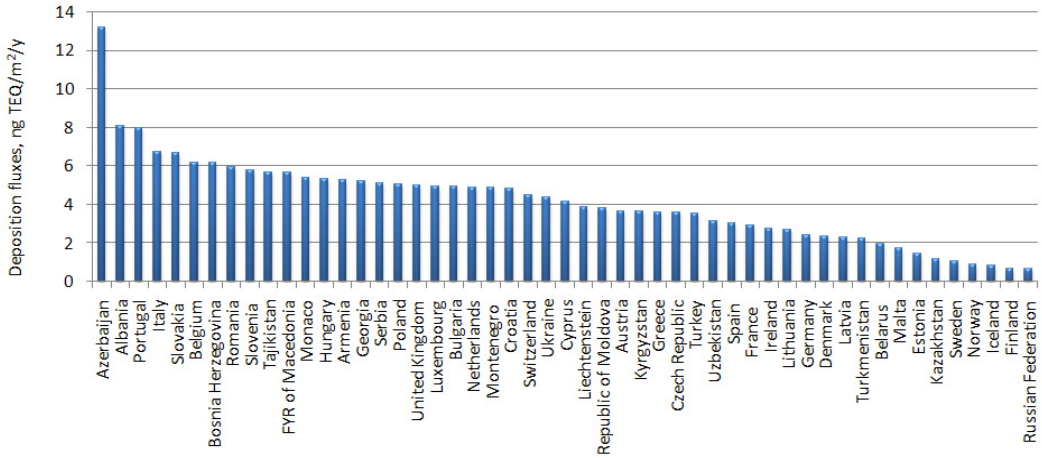


Fig. 2.34. *Annual deposition fluxes of PCDD/Fs in the EMEP countries calculated for 2014, ng TEQ/m^2/y*

Transboundary fluxes of dioxins and furans significantly contribute to the pollution of the EMEP countries. Model estimates of contributions of national emission sources and transboundary transport to PCDD/F anthropogenic deposition in the EMEP countries in 2014 are shown in the Fig. 2.35. According to modeling results, *in half of the EMEP countries the fraction of PCDD/F deposition*

over their territories in 2014, caused by the transport from external emission sources, exceeds the fraction of deposition from their national emissions. This in particular is seen for countries with relatively small territory or relatively low national emissions (e.g. Lichtenstein, Iceland, Luxembourg, and Cyprus). At the same time, countries with relatively high PCDD/F emission or located far from major emission sources (e.g. Azerbaijan, Portugal, Italy) are characterized by low contribution of transboundary transport.

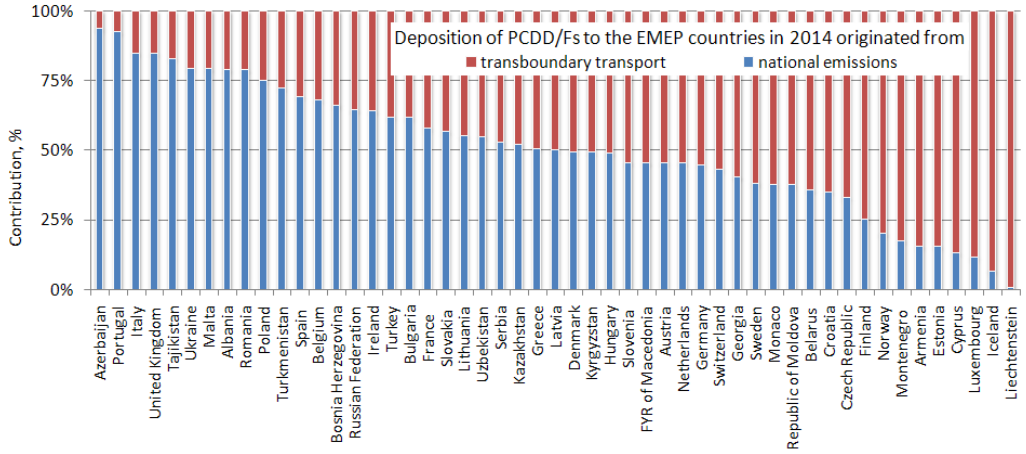


Fig. 2.35. *Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of PCDD/Fs over the EMEP countries in the 2014*

Influence of national emission sources of a particular country on the pollution levels in other countries can also be characterised by the fractions of total anthropogenic deposition, originated from national emission sources of a country, and fallen out within and outside its boundaries (Fig. 2.36). *For 16 countries the fraction of PCDD/Fs deposited outside their territories in 2014 is higher than the fraction of PCDD/Fs deposited over the country itself.* Model calculations for 2014 indicate that about 50% of annual PCDD/F deposition from anthropogenic emission sources within the EMEP region is provided by the emissions of six countries, namely, the Russian Federation, Ukraine, Poland, Italy, Romania, and Kazakhstan.

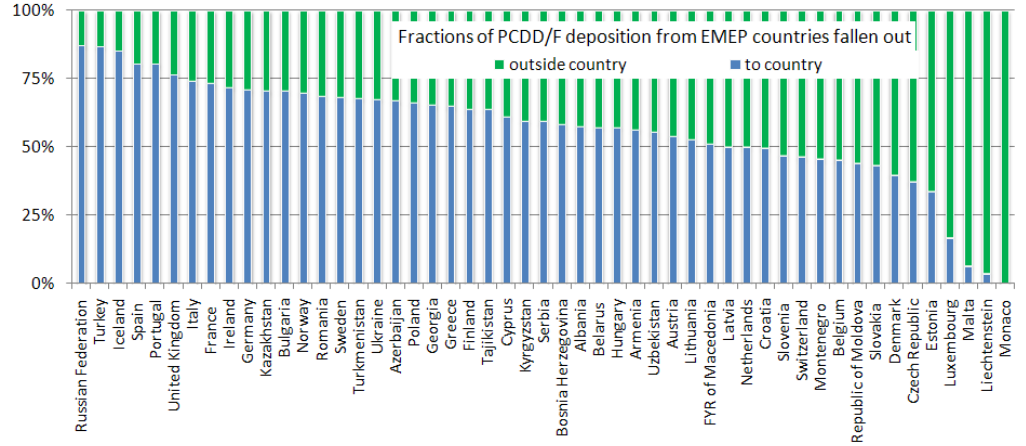


Fig. 2.36. *Fractions of PCDD/F deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in the 2014*

2.3. Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls represent a group of chlorinated toxic compounds intentionally produced for various technical applications as components in electrical and hydraulic equipment and lubricants. Similar to dioxins and furans, PCBs are hydrophobic, lipophilic, and highly resistant to degradation in the environment that leads to their accumulation in soil and sediment compartments as well as bioaccumulation along the food-chains. The production and use of PCBs has been banned in many countries of the world. Nevertheless they are still measured in various regions and continue posing risks to human health and ecosystems. The most important human exposure pathway for PCBs is dietary intake followed by additional pathways like drinking water intake and inhalation of air. PCBs have been shown to be probable human carcinogens and to cause variety of adverse health effects in humans and animals (e.g. reproductive effects, teratogenicity, and immunotoxicity).

Monitoring of use, disposal, and dispersion of PCBs in the environment is included in the activities of many international organizations, like LRTAP Convention, Stockholm Convention on POPs, Rotterdam and Basel Conventions, Helsinki Commission (HELCOM), Arctic Monitoring and Assessment Program (AMAP), as well as national organizations, such as US Environment Protection Agency (US EPA), German Environmental Protection Agency (UBA), Swedish Environmental Protection Agency (SEPA), and others.

PCBs are considered as PBT (persistent, bioaccumulation and toxic) and vPvB (very persistent and very accumulated substances) by the EU Regulation REACH following the criteria, listed in the Annex XIII of the Regulation (EC) No 1907/2006. In particular, they are subject of registration and restrictions according to the Annex III of REACH as the carcinogen, mutagen and toxic substances for reproductive organs (CMR). Further, the EU regulation for classification, labeling, and packaging ((EC) No 1272/2008) classifies PCBs as very toxic to aquatic life and establishes concentrations limits for their content in products.

Evaluation of PCB pollution levels in the EMEP region has been carried out using global and nested regional scale model simulations. To characterize PCB transport and fate, modeling has been made for the indicator congener PCB-153. The outcome of the assessment includes estimates of spatial distribution and temporal changes of PCB-153 concentrations and deposition fluxes as well as estimates of transboundary transport of pollution between the EMEP countries. Modeling results have been compared with concentrations measured at the EMEP monitoring sites and measurement data of the UNEP SC GMP Data Warehouse.

2.3.1. Emission data for model assessment

Emission data for modeling of PCB long-range transport and deposition in the EMEP region have been prepared on the basis of emission inventories, officially submitted by the EMEP countries, and available expert estimates. It should be noted that officially reported PCB emission data provide the total releases of PCB mixture to the atmosphere, without information on contribution of particular congeners. At the same time, model assessment of PCB pollution requires congener specific emission

data to evaluate the transport and fate of PCB mixture of congeners or of individual indicator PCB congeners. Taking this into account, expert estimates of global PCB congener specific emissions of *Breivik et al.* [2007] have been used for the preparation of emission data for PCB modeling. These expert estimates provide the consistent set of historical and future emissions of 22 individual PCB congeners from 1930 up to 2100. The indicator congener PCB-153 has been selected to characterize transboundary transport and pollution levels of PCBs. According to expert estimates of *Breivik et al.* [2007] as well as officially reported data on PCB releases, annual anthropogenic emissions in the EMEP countries in 2014 are on average about 5% lower comparing to the emissions in 2013.

Gridded emissions of PCB-153 for the EMEP domain are generated using officially submitted spatial distribution of PCB emissions of 19 EMEP countries as well as expert estimates, worked out by TNO [*Denier van der Gon et al.*, 2005]. The spatial distribution of PCB-153 emissions within the EMEP region, applied in model simulation for 2014, is presented in Fig. 2.37.

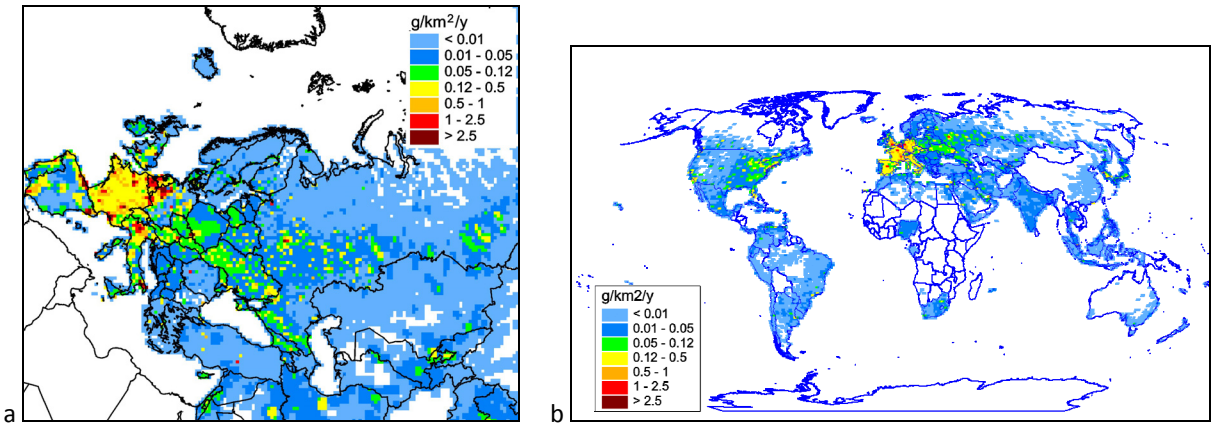


Fig. 2.37. *Spatial distribution of PCB-153 emissions in the EMEP domain with resolution 50x50 km^2 (a) and over the global domain (b) with resolution 1^{o}x1^o used in model simulations for 2014*

2.3.2. PCB pollution levels in the EMEP region and on global scale

Evaluation of PCB-153 pollution levels in the EMEP domain has been performed using global scale modeling and nested regional modeling. Spatial distribution of global scale annual mean PCB-153 air concentrations, obtained for 2014, is illustrated in Fig. 2.38. Elevated levels of air concentrations are noted for the European region (1-6 pg/m^3), while air concentrations in the other regions are lower, namely, 0.08-3 pg/m^3 in North America, 0.08-0.2 pg/m^3 in Eastern and Southern Asia, 0.02-0.2 pg/m^3 in South America, Africa, and Australia. This pattern of air concentrations reflects the spatial distribution of anthropogenic emissions. In general, model predictions of PCB-153 pollution levels for recent

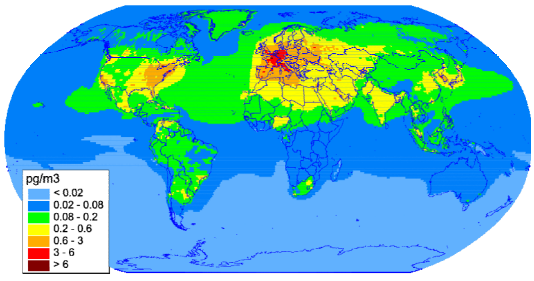


Fig. 2.38. *Spatial distribution of modeled annual mean PCB-153 concentrations in air (pg/m^3) obtained for 2014*

decade agree with available measurements, that has been shown in the comparison of modeling results with measurements of air concentrations, performed for the period from 1995 to 2013 presented at previous stage of work [*Shatalov et al*., 2015].

Model estimates of annual mean PCB-153 air concentrations and deposition fluxes in the EMEP region, generated for 2014, are presented in Fig. 2.39. Boundary conditions for regional model simulations have been set up on the basis of global modeling results. Initial conditions for the evaluation of pollution levels in 2014 have been obtained using long-term spin-up model simulations for the period from 1930s to present time.

Relatively high annual mean air concentrations (1-7 pg/m^3 and higher) can be noted for countries in Western and Central Europe, while lower levels of concentrations are obtained for Northern Europe and the EECCA countries (Fig. 2.39a). Similar spatial distribution can be seen for annual deposition fluxes of PCB-153 (Fig. 2.39b). The largest level of annual mean PCB-153 deposition fluxes to the marginal seas in the EMEP domain is calculated for the Baltic Sea (0.06 g/km^2/y) followed by the North Sea (0.05 g/km^2/y), the Mediterranean Sea (0.03 g/km^2/y), the Black Sea (0.025 g/km^2/y), and the Caspian Sea (0.016 g/km^2/y).

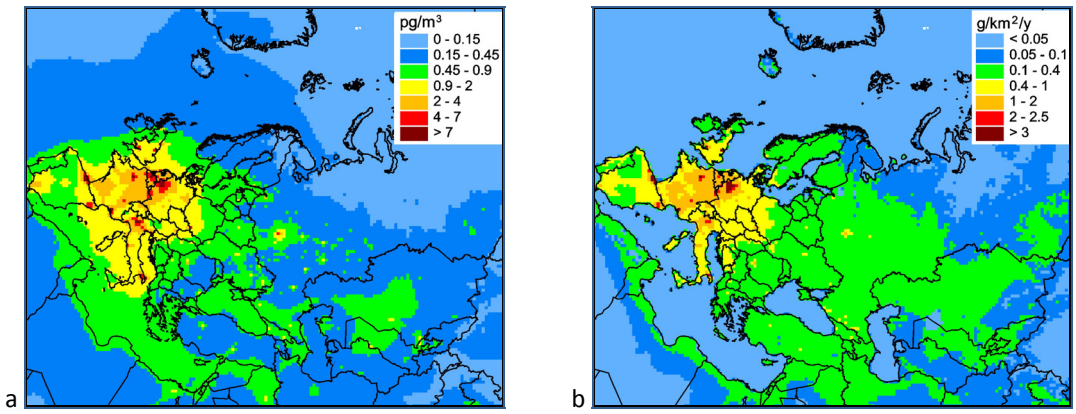


Fig. 2.39. *Spatial distribution of modeled annual mean air concentrations (pg/m^3) in the EMEP domain (a) and annual deposition fluxes (g/km$^{2/}$y) (b) of PCB-153 for 2014*

Similar to other contaminants, considered in this report, results of PCB-153 model simulations for 2014 have been compared with the results for the year 2013. Changes in pollution levels between these two years for each EMEP country are presented in Fig. 2.40 as a difference between annual mean deposition fluxes, calculated for 2014 and 2013. Levels of annual mean PCDD/F deposition fluxes in the EMEP countries between these two years vary from about 10% decrease to about 30% increase.

Changes of pollution levels are mainly caused by the changes of meteorological conditions since changes of emission data applied in model simulations are relatively low (about 5%). The largest decline of deposition (about 10%) is noted for Armenia, Kyrgyzstan, and Germany. The largest increase of deposition (about 20-30%) is calculated for Malta, Iceland, Cyprus, and Norway. The other countries are characterized by lower changes of annual mean PCB-153 deposition fluxes varying in the range from about -5% to 10%.

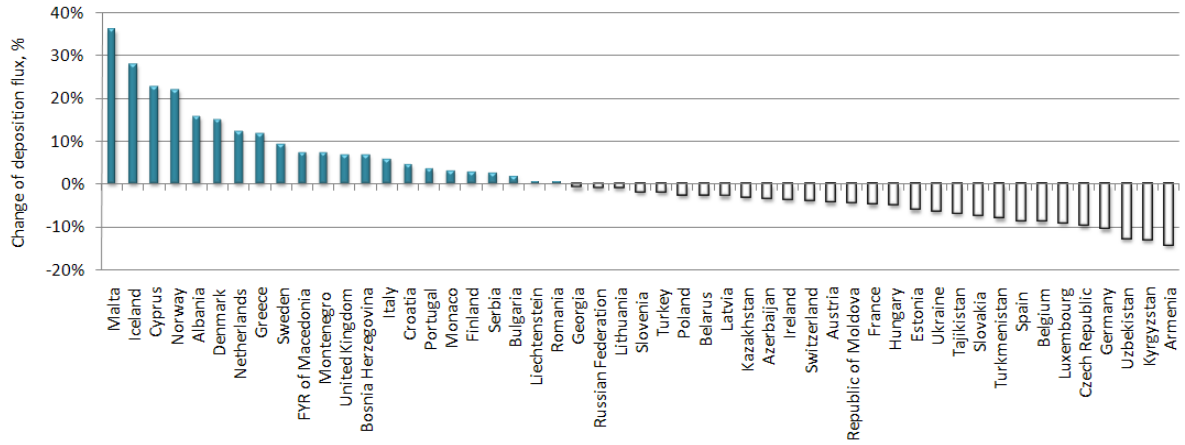


Fig. 2.40. *Changes of annual mean PCB-153 deposition fluxes in the EMEP countries between 2013 and 2014, $(C_{2014}-C_{2013})/C_{2013}$. Negative values denote decrease, and positive ones – increase*

Comparison of PCB modeling results with measurements

Model predictions of PCB pollution levels in the EMEP region for 2014 have been evaluated against measurements of the EMEP monitoring sites. Monitoring of PCB-153 air concentrations in 2014 has been performed at 12 EMEP sites. In addition to this PCB modeling results have been compared with passive sampling measurements of PCB air concentrations, collected in the UNEP SC Global Monitoring Programme (GMP) Data Warehouse. Locations of monitoring sites, performed PCB measurements, are shown in Fig. 2.41. It is seen that measurements of the GMP Data Warehouse notably extend the coverage of the EMEP region by the inclusion of measurements made in Central and Eastern Europe.

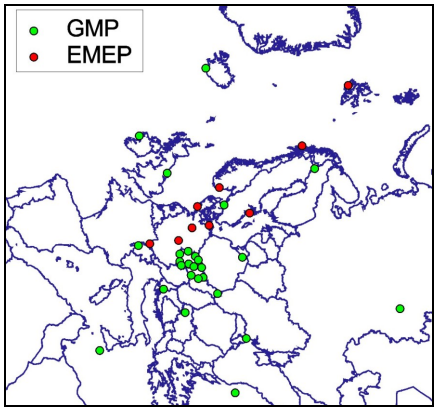


Fig. 2.41. *Locations of monitoring sites performed measurements of PCB-153 air concentrations: red circles – the EMEP monitoring sites, green circles – passive sampling measurements from the GMP Data Warehouse*

Comparison of model predictions of annual mean PCB-153 air concentrations and measurements of the EMEP monitoring sites is displayed in Fig. 2.42. For 85% of monitoring sites the agreement between the model results and observations is within a factor of 2. *Modeled concentrations capture spatial variations of observed concentrations with correlation coefficient accounting for 0.7. The relative bias of modeled concentrations in comparison with measurements is as about 0.4%, which indicates that on the average modeling results for PCB-153 air concentrations agree well*

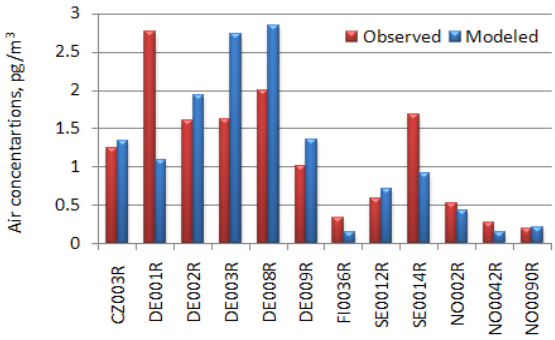


Fig. 2.42. *Comparison of annual mean modeled PCB-153 air concentrations with measurements of the EMEP monitoring sites for 2014, pg/m^3*

with measured concentrations. For some of the EMEP monitoring sites deviations between modeled and observed values exceed a factor of 2. For example, the largest discrepancies have been obtained for the monitoring sites DE0001R and SE0014R, which can be attributed to the uncertainties of emission data used in modeling.

Seasonal variations of modeled PCB-153 air concentrations for 2014 are in reasonable agreement with measurements. For the majority of monitoring sites temporal correlation between modeled and observed values is in the rage from about 0.5 to 0.8 (Fig. 2.43).

Comparison of modeled and observed monthly mean PCB-153 air concentrations for selected monitoring sites is shown in Fig. 2.44. In particular, for most part of the year modeled concentrations for CZ0003R and DE0002R reasonably reproduce variations of observed PCB-153 air concentrations. The most significant disagreement is found for the monitoring sites DE0001R and SE0014R. The model underpredicts concentrations, measured at the site SE0014R in summer months, and concentrations, measured at DE0001R for most part of the year. As it has been mentioned above, the underprediction can be explained by the uncertainty of applied emission data, namely, spatial distribution of emissions and their seasonal variability that requires more detailed analysis.

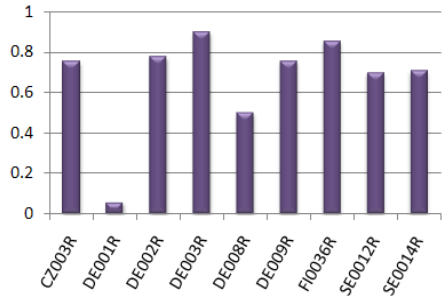


Fig. 2.43. *Correlation coefficients between observed and modeled monthly mean PCB-153 air concentrations for 2014*

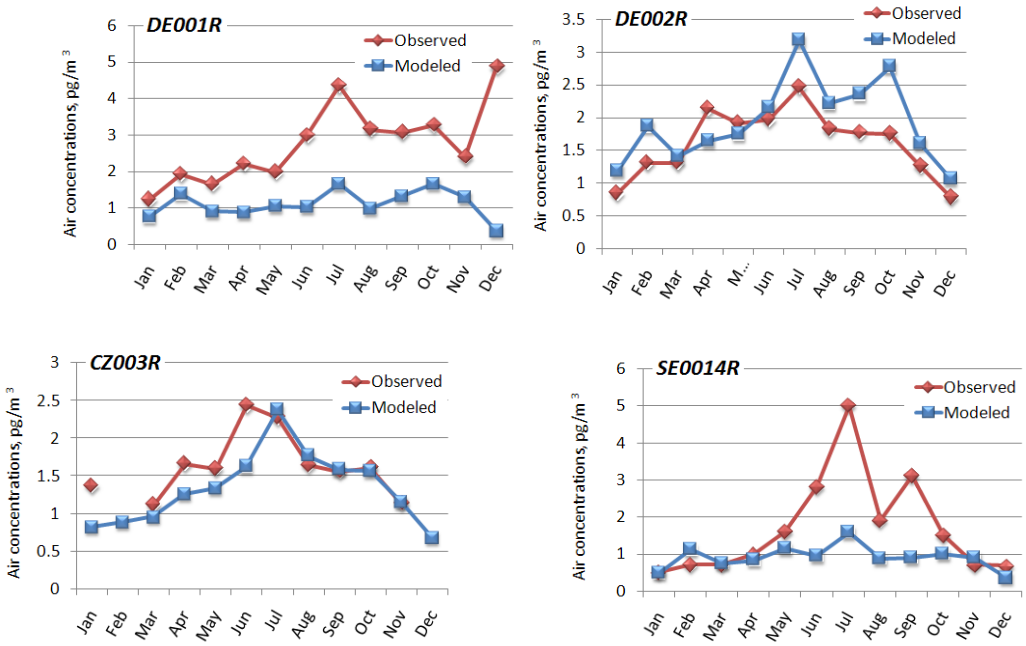


Fig. 2.44. *Comparison of monthly mean modeled PCB-153 air concentrations with measurements of selected EMEP monitoring sites for 2014, pg/m^3*

In addition to the verification of modeling results against the measurements of the EMEP monitoring sites modeled concentrations have also been compared with passive sampling measurements collected in the GMP Data Warehouse. To extend the base of the comparison model predictions and measurements for two years 2013 and 2014 are used. The scatter plot of modeled and observed concentrations of PCB-153 is shown in Fig. 2.45.

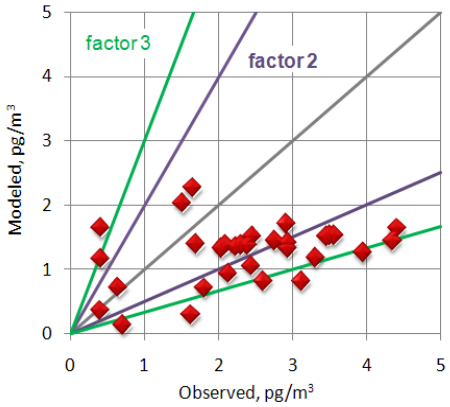


Fig. 2.45. *Comparison of modeled annual mean PCB-153 air concentrations with passive sampling measurements collected in the UNEP SC GMP Data Warehouse for 2013 and 2014*

It is seen that modeling results agree with passive sampling measurements within a factor of 2 – 3. In particular, about 88% of measurements agree with model results within a factor of 3, which can be regarded as reasonable level of agreement taking into account the uncertainties of passive sampling measurements, indicated in the number of studies applying passive sampling for POP monitoring [*Harner et al.,* 2006, *Gouin et al.,* 2008, *Wang et al.,* 2016, *Hayward et al.,* 2010, *Zhang et al.,* 2013].

2.3.3. Analysis of trends of PCB pollution in the EMEP countries

Analysis of long-term changes of POP pollution in the EMEP region has been continued this year with more detailed evaluation of temporal trends for PCB-153 in the EMEP countries. In particular, country-specific temporal variations of PCB pollution levels in the period 1990-2014 have been evaluated using the methodology developed by MSC-E [*Shatalov et al.,* 2015]. Elaborated methodology allows to decompose time-series of pollution levels into three components, namely, main component (general tendency), seasonal component, and irregular component (residue), and to determine the trend as a sum of two regular components of variations (main and seasonal).

To analyze temporal changes of the contamination of the EMEP countries, consistent set of modeling results on PCB-153 for the EMEP region, generated for the period 1990-2014, has been used. Model simulations have been carried out using official emission data complemented by expert estimates by *Breivik et al.* [2007].

Results of trend analysis of long-term variations of PCB-153 air concentrations, averaged over the territory of all EMEP countries and over the EECCA countries, are shown in Fig. 2.46.

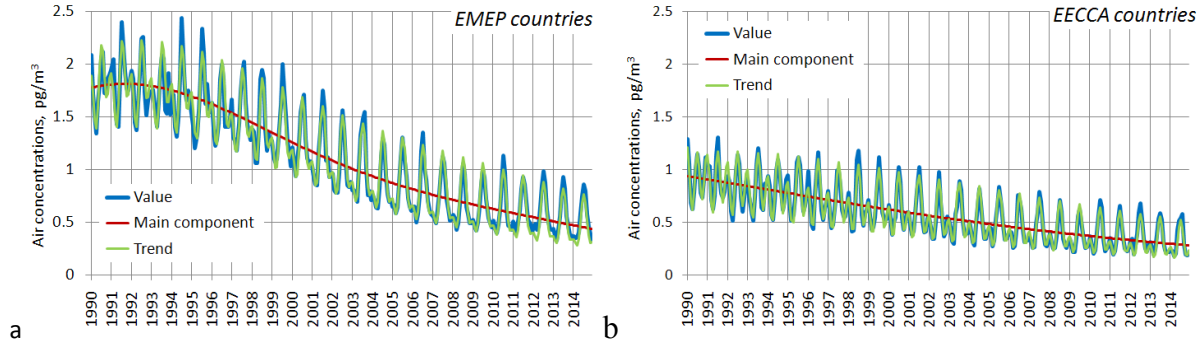


Fig. 2.46. *Trends in monthly mean air concentrations of PCB-153 for the area of the EMEP countries on the whole (a) and for the EECCA countries (b), estimated on the basis of modeling results for the period from 1990 to 2014. Red line denotes main component of trend, green line – trend of concentrations, and blue line – monthly mean PCB-153 air concentrations (pg/m^3)*

The analysis indicates that levels of PCB-153 air pollution in the EMEP countries have declined on average by 75% during the two recent decades (Fig. 2.47a). Relatively higher rates of reduction (about 80%) are estimated for the EU28 countries, varying for individual countries from 60% for Estonia to almost 90% for the United Kingdom. The EECCA countries are characterized by relatively lower decrease of pollution levels (about 70%).

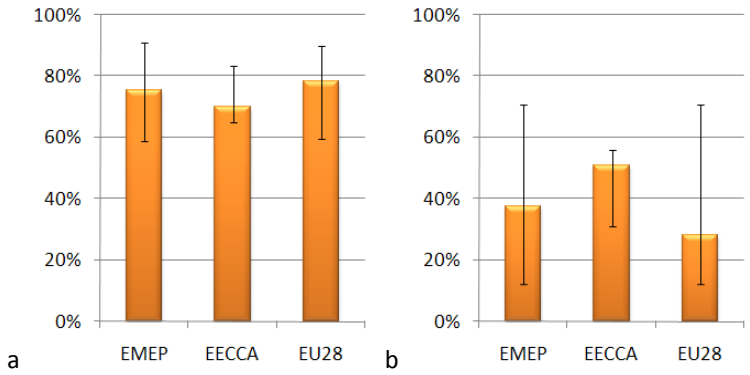
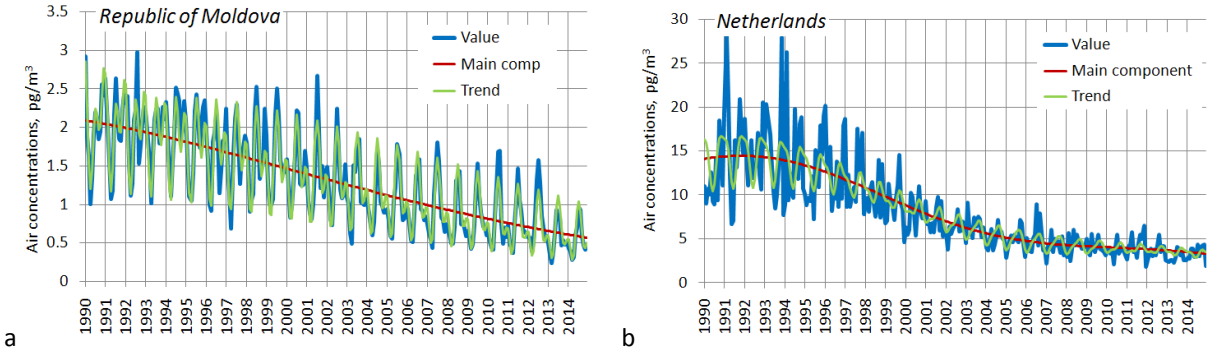


Fig. 2.47. *Total reduction of PCB-153 contamination in the EMEP countries, EECCA region and EU28 countries the period from 1990 to 2014 (a) and seasonal variation contributions (b)*

Air concentrations of PCB-153 are subject of pronounced seasonal variations with maximum concentrations in warmer months and minimum concentrations in the colder months during the year. Seasonal variability of air concentrations, averaged over the area of all EMEP countries, is about 40% of the main component. Results of the analysis indicate that seasonal variations of air concentrations, evaluated for the EECCA countries, are larger (about 50%) in comparison to the estimates of variations for the EU28 countries (about 30%) (Fig. 2.47b). This difference can be explained by the influence of geographical location of the countries. In particular, higher estimates of seasonal variations are obtained for continental countries comparing to the countries located closer to coastal areas.

Analysis of trends has been carried out also for each particular EMEP country. Examples of calculated trends for the two selected EMEP countries, namely, the Republic of Moldova and the Netherlands, are presented in Fig. 2.48.



a b

Fig. 2.48. *Trends in monthly mean air concentrations of PCB-153 for the Republic of Moldova (a) and the Netherlands (b), estimated on the basis of modeling results for the period from 1990 to 2014. Red line denotes main component of trend, green line – trend of concentrations, and blue line – monthly mean PCB-153 air concentrations (pg/m^3)*

Comparison of trends for individual EMEP countries leads to the conclusion that seasonal variations are more significant for continental countries and less significant for coastal countries. Moreover, it is found that for coastal countries the influence of non-regular meteorological perturbations is much higher than for continental ones. For example, for the Netherlands the contribution of non-regular perturbations amounts to about 25% of main component, whereas for Moldova the influence of non-regular component is evaluated as 14% only. Detailed information on the analysis of trends of PCB pollution levels in the EMEP countries can be found in the MSC-E Technical report [*Shatalov et al.*, 2016].

2.3.4. Transboundary transport of PCB-153 in 2014

To evaluate transboundary transport of PCB-153 within the EMEP region for 2014 the contributions of anthropogenic emission sources of the EMEP countries as well as secondary and non-EMEP emissions have been taken into account. Influence of non-EMEP emissions have been estimated using global scale model simulations. Relative contributions of the EMEP anthropogenic emission sources to deposition of PCB-153 in the EMEP countries vary from about 5% to 50%, while secondary emissions contribute about 50-70% and non-EMEP emission sources about 1-30% depending on a particular country.

Annual total deposition fluxes of PCB-153 to the EMEP countries are illustrated in Fig. 2.49. Relatively high levels of mean PCB-153 deposition fluxes in the EMEP countries (1-2 g/km^2/y), are calculated for Luxembourg, Belgium, France, the Netherlands, and Germany. The other countries are characterized by lower levels of deposition fluxes.

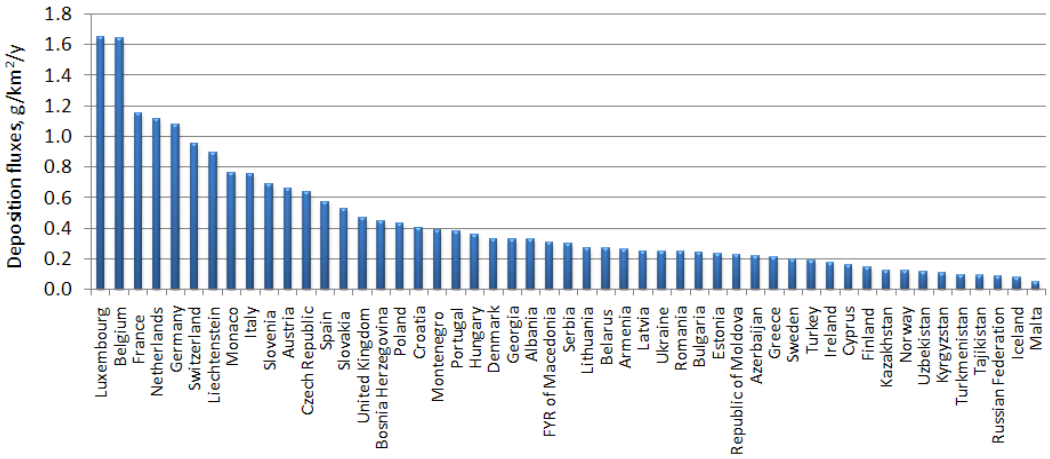


Fig. 2.49. *Annual deposition fluxes of PCB-153 in the EMEP countries calculated for 2014, g/km^2/y*

According to modeling results, transboundary transport of PCB-153 provides considerable influence on the pollution of the EMEP countries. Model estimates of contributions of national emission sources and transboundary transport to anthropogenic PCB-153 deposition in the EMEP countries in 2014 are shown in the Fig. 2.50. According to modeling results, *in more than 60% of the EMEP countries the fraction of PCB-153 deposition over their territories in 2014, caused by the transport from external emission sources, is higher than the fraction of deposition from their national emissions.* Substantial contribution of transboundary transport is obtained for the countries with relatively small territory or relatively low national emissions (e.g. Monaco, Lichtenstein, Iceland, and Montenegro), while for countries with relatively high PCB-153 emission or countries located far from major emission sources low contribution of transboundary transport is estimated (e.g. Spain, Italy, France, and the United Kingdom).

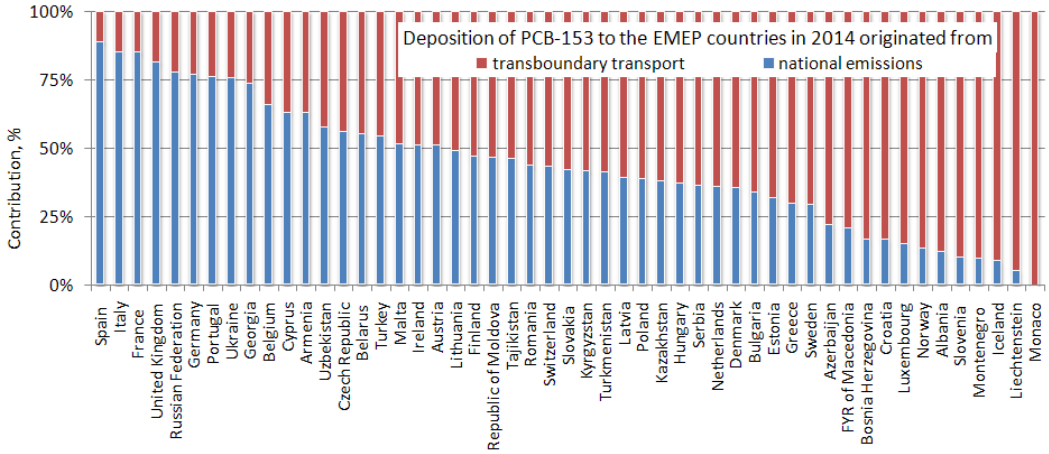


Fig. 2.50. *Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of PCB-153 over the EMEP countries in the 2014*

The influence of national emission sources of a particular country on the pollution of other countries is shown in Fig. 2.51 using the fractions of total anthropogenic deposition, originated from national

emissions of a country, and fallen out within and outside its boundaries. *For 16 countries the fraction of PCB-153 deposited outside their territories in 2014 is higher than the fraction of PCB-153 deposited over the country itself.* Model calculations for 2014 indicate that about 50% of annual deposition of PCB-153 from anthropogenic emission sources within the EMEP region is provided by the emissions of four countries, namely, the Russian Federation, France, Germany, and Kazakhstan.

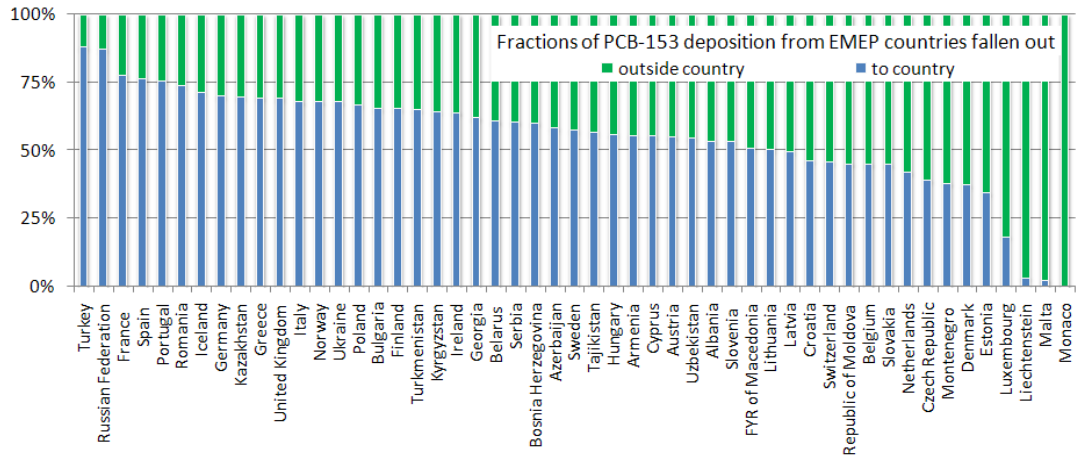


Fig. 2.51. *Fractions of PCB-153 deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in the 2014*

2.4. Hexachlorobenzene (HCB)

Hexachlorobenzene is highly persistent toxic chemical that does not occur naturally and is produced for various applications or released as unintentional by-product in course of manufacturing of chlorinated compounds. It was widely used for agricultural purposes from the beginning of 1950s until 1970s as a pesticide and fungicide. After restrictions and banning of HCB usage as a fungicide/pesticide starting from 1970s its production and use have been substantially declined leading to decrease of its content in the environment. Contemporary levels of HCB pollution are likely supported by industrial and combustion emission sources and to significant extent by the secondary releases from the environmental media previously polluted due to agricultural or industrial activities.

HCB is known as a pollutant of global concern due to significant potential to long-range transport and persistence in the environment. HCB is characterized by ability to bioaccumulation and harmful effects both for human health and for the ecosystems. In particular, it is classified under the EU Regulation CLP ((EC) No 1272/2008) as a carcinogen of the group 1B, which defines it as a toxicant for specific organs under repeated exposure, and for aquatic organisms.

Assessment of HCB pollution levels in the EMEP region for 2014 has been carried out with application of combined global and regional scale modeling. Global transport and fate of HCB in the environment has been evaluated using experimental scenario of contemporary and historical HCB emissions. Transboundary transport of HCB between the EMEP countries has been assessed on the basis of

officially reported emission data using regional scale model simulations with initial and boundary conditions, obtained from the global modeling results. Modeled air concentrations of HCB have been compared with measurements of the EMEP monitoring sites.

2.4.1. Emission data for model assessment

Assessment of HCB pollution in the EMEP domain has been carried out on the basis of gridded emission data provided by CEIP. Description of reported HCB emissions and gap-filling methods, used to prepare GNFR emission inventory for 2014 (as reported in 2016), is available in the CEIP Technical report 02/2016 [*Tista et al.*, 2016]. Spatial distribution of HCB emissions in the EMEP region, generated for 2014, is presented in Figs 2.52.

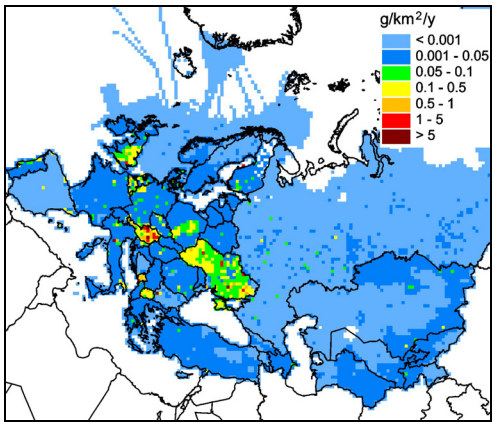


Fig. 2.52. *Spatial distribution of HCB emission fluxes in the EMEP domain in 2014 with resolution 50x50 km²*

Total annual HCB emission, reported by the EMEP countries for 2014, is accounted for about 452 kg. *Annual emissions of HCB for 2014 from anthropogenic sources of the EMEP countries are 50% lower comparing to the emission for 2013, provided in the previous submission of emission data* [*Mareckova et al.*, 2015].

Relative changes of HCB emissions in individual countries between the years 2014 and 2013 are demonstrated in Fig. 2.53 as the differences between the reported values of total emission for 2014 (E_{2014}) and for 2013 (E_{2013}). Positive values in the diagram indicate an increase of HCB emission from 2013 to 2014, while negative values indicate a decrease of emission.

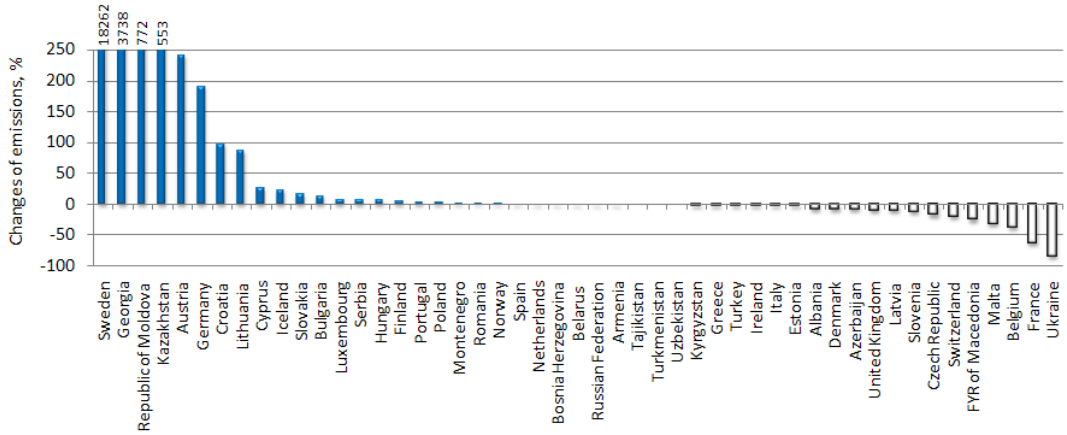


Fig. 2.53. *Relative changes of annual total emissions of HCB between 2013 and 2014, $(E_{2014}-E_{2013})/E_{2013}$. Negative values denote decrease of emissions, and positive ones – increase of emissions*

Reported HCB emissions for 2014 are lower than corresponding emissions for 2013 in 18 countries and higher in 28 countries. Significant relative increase of emissions is noted for Sweden, Georgia, the Republic of Moldova, Kazakhstan, Austria, and Germany in 183, 38, 9, 7, 3.4, and 3 times, respectively.

Changes of emissions for Sweden are caused by the use of extended coverage of NFR emission sectors in the submission of 2016 comparing to the previous submission [*IIR Sweden*, 2016]. Similar to PAHs, changes of HCB emissions of Georgia are caused by the recalculation of emissions based on the new national energy balance, prepared by GEOSTAT of Georgia [*IIR of the Republic of Georgia*, 2016]. Emissions of HCB for the Republic of Moldova have been recalculated using updated emission factors of the EMEP/EEA Emission Inventory Guidebook 2013 instead of factors applied before [*IIR of the Republic of Moldova*, 2016].

For Kazakhstan officially reported national data on HCB emissions have been used, which is higher than previously applied expert estimates. Higher value of emission from Austria is attributed to the unintentional releases of HCB from Austrian cement plant in 2014 [*Austria's IIR*, 2016]. Changes of HCB emissions for Germany are connected with the reporting of new estimates for the NFR sector 'Use of pesticides' [*German IIR*, 2016].

Notable decrease (> 50%) of HCB emissions in 2014 is reported for Ukraine and France. Annual HCB emission of Ukraine for 2014 is extrapolated based on the estimates of emissions for 1990 and 1995 made by *Pacyna et al.* [2003] [*Tista et al.*, 2016]. In France, the reduction of emissions is caused by the removal of HCB emissions of the road transport from the inventory [*CITEPA*, 2016].

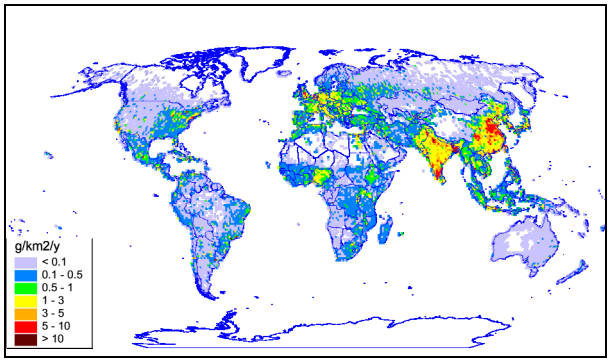


Fig. 2.54. *Spatial distribution of global scale HCB emissions with spatial resolution 1^0x1^0 applied in model simulations for 2014*

Model simulations of HCB global transport and fate have been made with experimental emission scenario describing historical HCB releases during the recent several decades [*Shatalov et al.,* 2010]. This scenario has been updated to cover the period of time up to 2014 and is used for the modeling of long-term accumulation of HCB in the environmental compartments. Spatial distributions of global HCB emission fluxes for 2014 are illustrated in Fig. 2.54.

2.4.2. HCB pollution levels in the EMEP region and on global scale

HCB pollution of the EMEP region has been evaluated using multi-scale modeling approach. Results of global scale model simulations, obtained for 2014, are shown in Fig. 2.55. Model predictions indicate relatively high levels of annual mean HCB air concentrations (70-100 pg/m^3) in Eastern and Southern Asia. Lower levels of air concentrations have been predicted for the European region (about 20-40 pg/m^3). Other regions are characterized by the concentrations about 10 pg/m^3 or below. Spatial variations of air concentrations generally correspond to the distribution of anthropogenic emissions used for model calculations. Comparison of modeled spatio-temporal variations of HCB concentrations with measurements for the period 1990-2013 have shown that modeling results reasonably agree with measurements performed in most of the regions. However, the model tends to underpredict observed air concentrations in North America and Europe [*Shatalov et al.,* 2015].

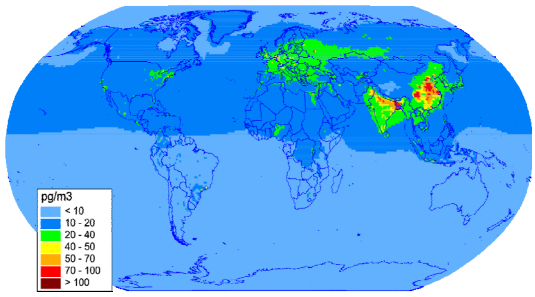


Fig. 2.55. *Spatial distribution of modeled annual mean HCB concentrations in air (pg/m^3) obtained for 2014*

Results of regional scale model simulations of HCB long-range transport in the EMEP region, generated for 2014, are presented in Fig. 2.56. In order to provide boundary conditions for modeling over the EMEP domain the outcome of global modeling has been used. HCB content in environmental media has been set up using long-term spin-up model run for the period from 1945 to the present time.

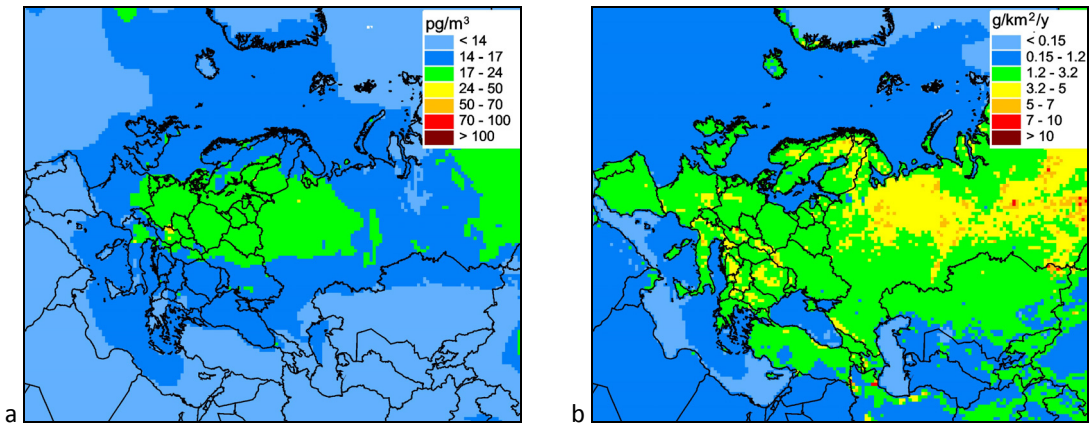


Fig. 2.56. *Spatial distribution of modeled annual mean air concentrations (pg/m^3) in the EMEP domain (a) and annual deposition fluxes (g/km^2/y) (b) of HCB for 2014*

It is seen that *elevated annual mean air concentrations of HCB (20-40 pg/m^3) are predicted for countries in Central and Eastern Europe, while lower levels of concentrations (10-20 pg/m^3) are obtained for Western and Southern Europe and the EECCA countries (Fig. 2.56a). Homogeneous distribution of HCB air concentrations can be explained by high stability of this pollutant in the atmosphere.*

Spatial distribution of HCB deposition fluxes indicates elevated levels of deposition for countries of Northern and Southern Europe as well as for the northern and eastern parts of the Russian Federation (Fig. 2.56b). In addition, levels of HCB deposition to the marginal seas within the EMEP regions are evaluated. The largest level of annual mean HCB deposition fluxes to the marginal seas in the EMEP domain is calculated for the Baltic Sea (0.52 g/km^2/y) followed by the Black Sea (0.30 g/km^2/y), the North Sea (0.28 g/km^2/y), the Mediterranean Sea (0.23 g/km^2/y), and the Caspian Sea (0.22 g/km^2/y).

Modeling results on HCB, obtained for 2014, have been compared with output of model simulations for the year 2013. Inter-annual changes of pollution levels for each EMEP country are presented in Fig. 2.57 in the form of relative difference between annual mean air concentrations for 2014 and 2013. Annual mean HCB air concentrations in the EMEP countries between these two years vary from 25% decrease to 25% increase.

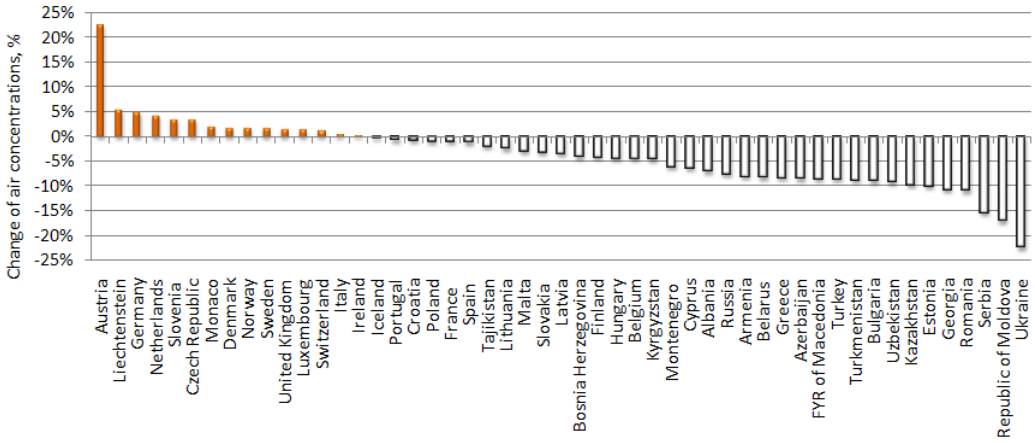


Fig. 2.57. *Changes of annual mean HCB air concentrations in the EMEP countries between 2013 and 2014, (C_{2014}-C_{2013})/C_{2013}. Negative values denote decrease, and positive ones – increase of concentrations*

Two major factors, contributed to these changes of pollution levels, include changes of meteorological conditions and changes of emission data used for modeling. The largest increase of air concentrations (about 25%) is seen for Austria, whereas decrease of air concentrations (about 20%) is characteristic of Ukraine, the Republic of Moldova, and Serbia. For other countries smaller changes of annual mean HCB air concentrations (about -10% - 5%) have been estimated.

Comparison of HCB modeling results with measurements

To compare modeling results on HCB for 2014 with observed levels of concentrations, measurements of the EMEP monitoring sites have been used. Monitoring of HCB air concentrations in 2014 has been performed at 12 EMEP sites. Results of the comparison of model predictions and measured HCB annual mean air concentrations at the background monitoring sites within the EMEP region are shown in Fig. 2.58.

It is seen that for the majority of monitoring sites (75%) the agreement between observed and modeled HCB concentrations is within a factor of 2. However, similar to the comparison results performed at the previous stages of the work, *the model exhibits the tendency to underestimate measured concentrations*. The largest deviations are noted for the monitoring sites CZ0003R, NO0002R, and NO0042R. The reasons of the disagreement can be attributed to the uncertainties of emissions, their spatial distribution, seasonal variability, as well as uncertainties of related to the measurements of HCB concentrations and deficiencies of modeling approach. Obtained discrepancies between the model predictions and measurements for HCB air concentrations require further analysis in co-operation with experts in monitoring and evaluation of HCB emissions and pollution levels.

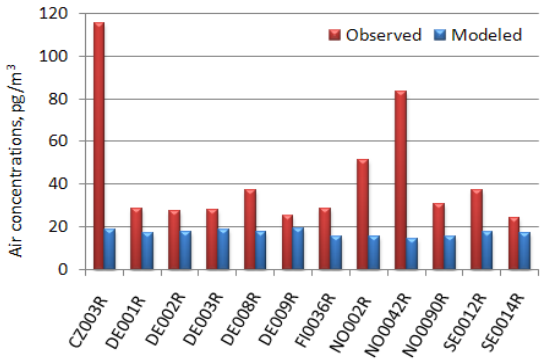


Fig. 2.58. *Comparison of annual mean modeled HCB air concentrations with measurements of the EMEP monitoring sites for 2014, pg/m^3*

2.4.3. Transboundary transport of HCB in 2014

Transboundary transport of HCB within the EMEP region has been evaluated for 2014. Model simulations take into account contributions of anthropogenic emission sources of the EMEP countries as well as influence of secondary and non-EMEP emissions. The contribution of non-EMEP emissions has been estimated using global scale modeling. According to modeling results, the share of the EMEP anthropogenic emissions in the deposition of HCB to the EMEP countries is not significant varying from less than percent to about 20%, while secondary emissions contribute about 30 to 70% and non-EMEP emission sources about 20 to 60% depending on a particular country.

Estimated annual total HCB deposition fluxes for 2014 over the EMEP countries are illustrated in Fig. 2.59. Elevated levels of HCB deposition in the EMEP countries (about 3 g/km^2/y) are calculated for Slovenia, Bosnia and Herzegovina, Montenegro, the FYR of Macedonia, Slovakia, and Austria. Relatively low deposition values are obtained for Uzbekistan, Turkmenistan, and Malta.

Model estimates of contributions of national emission sources and transboundary transport to anthropogenic HCB deposition in the EMEP region in 2014 are shown in the Fig. 2.60. In accordance with model predictions, transboundary transport of HCB has substantial contribution to deposition over the EMEP countries. *For almost 75% of the EMEP countries the fraction of HCB deposition over their territories in 2014, originated from the external emission sources, exceeds the fraction of deposition from their national emissions.* Significant contribution of transboundary transport to anthropogenic HCB deposition is estimated for countries with relatively small territory or relatively low national emissions (e.g. Monaco, Lichtenstein, Bosnia and Herzegovina, Malta, etc.). Low contribution of transboundary transport is obtained for countries with relatively high HCB emission (e.g. Austria, Ukraine, Montenegro, etc.).

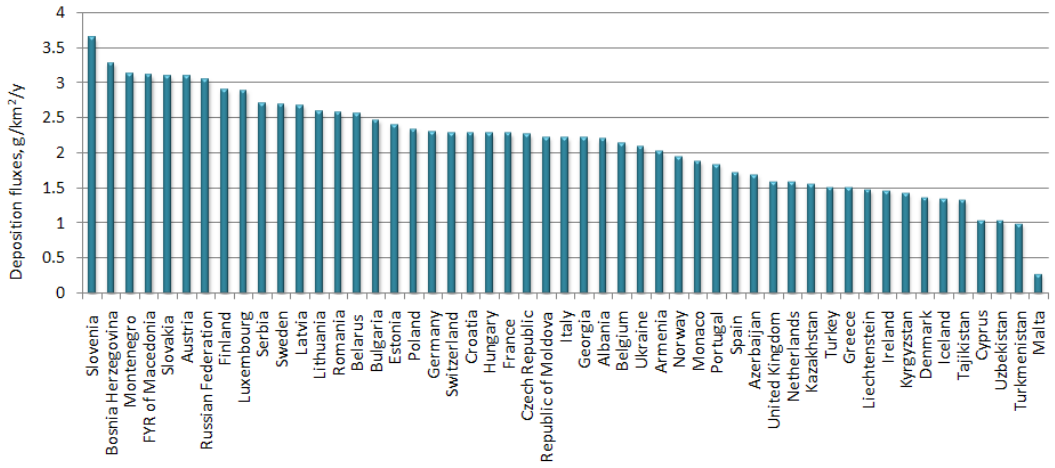


Fig. 2.59. *Annual deposition fluxes of HCB in the EMEP countries calculated for 2014, g/km^2/y*

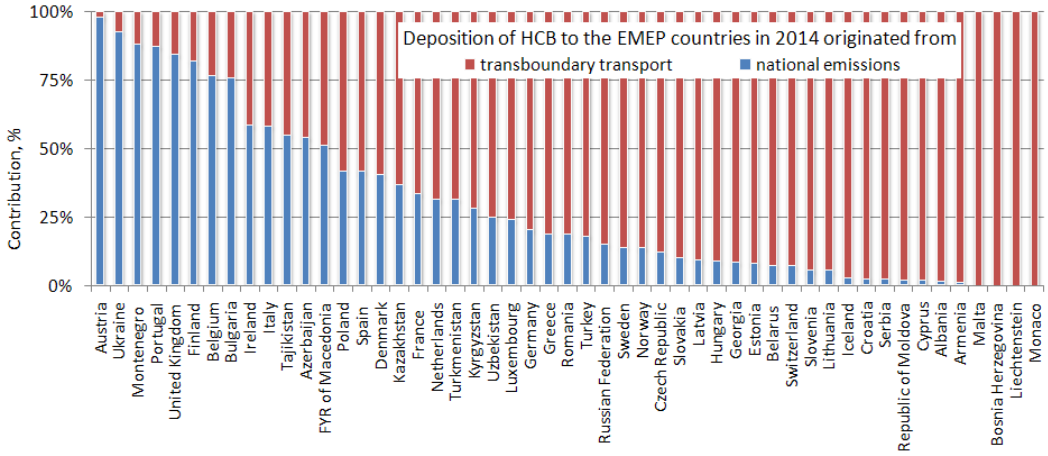


Fig. 2.60. *Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of HCB over the EMEP countries in the 2014*

Fig 2.61 illustrates the fractions of total anthropogenic deposition, originated from national emissions of a particular country, and fallen out within and outside its boundaries, contributing to the pollution of other countries. *For almost 70% of the countries the fraction of HCB deposited outside their territories in 2014 is higher than the fraction of HCB deposited over the country itself.* Model estimates show that about 70% of annual HCB deposition from anthropogenic emission sources over the EMEP countries is originated from the emissions of four countries, namely, Austria, Ukraine, Finland, and Italy.

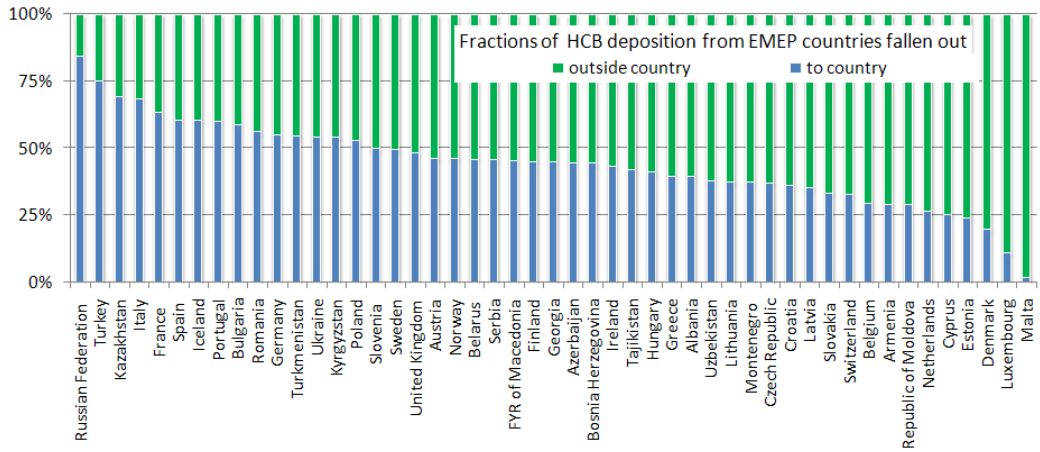


Fig. 2.61. *Fractions of HCB deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in the 2014*

3. COOPERATION AND EXCHANGE WITH INFORMATION

3.1. TRENDS OF POP POLLUTION IN THE EMEP COUNTRIES IN PERIOD 1990-2012

Assessment of long-term changes of POP pollution provides important information with respect to the effectiveness of policy measures aimed to prevent impacts of air pollution on human health and ecosystems. This year, MSC-E performed analysis of trends of POP atmospheric concentrations for two recent decades and contributed obtained results to the CLRTAP scientific assessment report [*Maas and Grennfelt,* 2016], to the TFMM assessment report [*Colette et al*., 2016], and to the WGE assessment report [*de Wit et al*., 2015]. Brief summary of the reported information on recent trends in POP pollution is given below.

The analysis of long-term trends of POP pollution of the EMEP region has been carried out for the period from 1990 (reference year) to 2012 on the basis of the methodology elaborated by MSC-E [*Shatalov et al.*, 2015]. Temporal changes of air pollution were analyzed for benzo[a]pyrene (B[a]P) (as a marker of PAHs), PCDD/Fs (17 PCDD/F toxic congeners), PCB-153 (as an indicator congener of PCBs), and HCB. Both measured and modeled values of atmospheric POP pollution levels were used in the trend analysis. In particular, for the locations of the monitoring stations, complementary analysis of modeled and measured data was performed. For other parts of the EMEP region, where there were no measurements, trends in POP pollution levels were characterized entirely by the modeling results.

Following significant reduction of anthropogenic POP emissions in the beginning of 1990s levels of POP pollution declined substantially in most of the EMEP countries. According to the analysis the largest reduction took place for HCB (about 90%), and the smallest one for B[a]P (about 30%) (Fig. 3.1). At the same time, temporal changes of pollution during the two-decade period were inhomogeneous. Rates of pollution reduction were higher in the beginning of the first decade (1990-2000), while the second decade (2001-2012) was characterized by considerably smaller changes, and levels of B[a]P pollution in the period after 2005 even started to increase.

Along with changes of POP pollution in the EMEP region as a whole the analysis of trends was performed also for the individual EMEP countries. Results of this analysis indicated that changes of pollution were different in different countries. As an example, differences in average pollution reduction rates of B[a]P air concentrations for the period 1990-2012 as well as in annual reduction rates for the year 2012 are illustrated in Fig. 3.2a for the three groups of the EMEP countries, namely, the EU28 countries, the EECCA countries, and the other EMEP countries.

It is seen that highest changes of B[a]P pollution levels took place in the EU28 countries. Besides, it is found that the growth of concentrations in the end of the two-decade period (in 2012) is characteristic for about 75% EMEP countries.

Evaluation of trends in POP pollution levels in the EMEP region was complemented by the joint analysis of modeled and observed time series of POP air concentrations for the locations of the EMEP monitoring stations. In general, the agreement between modeled and observed trends is good for most of considered POPs, with the exception of HCB measured at sites located close to boundaries of

the EMEP modeling domain that are influenced by non-EMEP emission sources. As an example, comparison of average annual reduction rates obtained from the analysis of modeling results and measurements is presented in Fig. 3.2b. As follows from the figure, the highest mean reduction of modeled B[a]P air concentrations in the EMEP region for 1990-2012 took place in the central, western and north-western parts of Europe. In a number of countries, located mainly in eastern, south-eastern and northern parts of Europe and in Central Asia an increase of B[a]P air concentrations occurred.

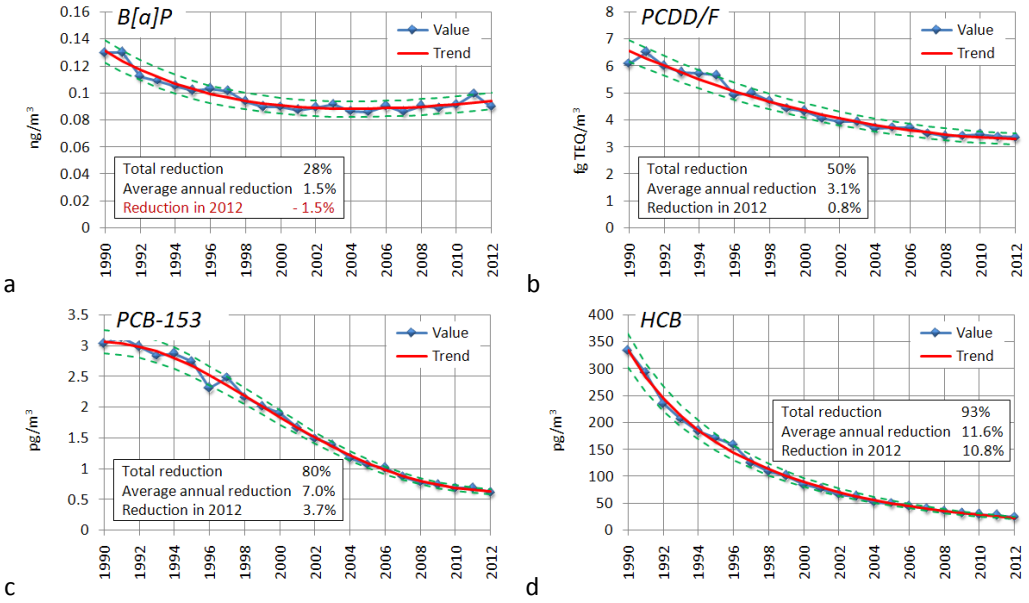


Fig. 3.1. *Trends in modeled annual mean air concentrations of (a) B[a]P, (b) PCDD/Fs, (c) PCB-153 and (d) HCB in the EMEP region for the period from 1990 to 2012. Negative reduction denotes increase of air concentrations. Dashed lines indicate 95% confidence interval*

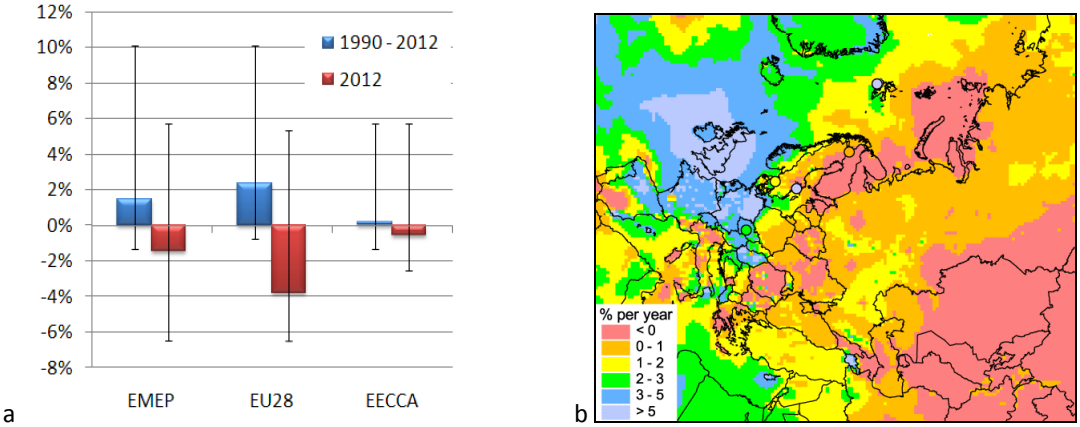


Fig. 3.2. *Average annual reduction rates of modeled B[a]P air concentrations for the period from 1990 to 2012 as well as reduction rates in 2012 for all EMEP countries, for the EU28, and for the EECCA countries. Negative values denote growth of pollution. Whiskers indicate variations between countries (a). Average reduction rates of modeled (field) and observed (circles) B[a]P air concentrations for the period from 1990 to 2012 (b)*

3.2. Task Force on Measurements and Modeling (TFMM)

In framework of co-operation with TFMM MSC-E took part in the 17th meeting of the Task Force held in May 2016 in Utrecht (the Netherlands). Participants of the meeting were provided with the information on the progress in the MSC-E activities related to the assessment of POP pollution in the EMEP region and its further development.

Ongoing work of MSC-E on the refinement of the GLEMOS modeling system comprises implementation of the new EMEP grid for the routine model calculations, updating of open code version of GLEMOS, and improvements of modeling approach for POPs. It includes collection and analysis of information on congener contributions for POP mixtures (e.g. PCDD/Fs), refinement of parameterization of atmospheric transformation and removal processes as well as inter-media exchange, further development of methodology for the evaluation of source-receptor relationships accounting for multi-media transport of POPs and evaluation of intercontinental transport to and from the EMEP region.

Research activities in these directions are performed with the exchange of information and co-operation with the UNEP Stockholm Convention on POPs including also collaboration with national experts in POP modeling. Specific attention has been given to the issues related to the preparation of emission data for model assessment.

Particular attention has been paid recently to air pollution in urban areas of the EMEP countries and exceedances of air quality limits. To contribute to the analysis of population exposure to elevated pollution levels EMEP initiated a study aimed at quantification of B[a]P air concentrations in urban areas. Outline of this study and preliminary results were presented during the meeting.

First stage of the work is focused on the evaluation of B[a]P air concentrations for the Czech Republic due to availability of data on emissions and measurements. The methodology of the study is based on previously developed approaches that applied multiple regression models to describe functional relationship between measured urban concentrations, rural concentrations and secondary parameters including meteorological, geophysical data, and modeling results. Obtained results show in general reasonable agreement of estimated B[a]P concentrations for urban areas in the Czech Republic with measurements. At the same time, some discrepancies require further development of applied approach in order to reduce the level of uncertainties in estimates of B[a]P concentrations in urban areas. Preliminary results on the evaluation of B[a]P pollution of urban areas in the Czech Republic are briefly described in this report. Detailed information on this study can be found in the Technical Report [*Shatalov et al.*, 2016].

Further work in this direction will include refinement of developing methodology and its application to the analysis of pollution levels in other EMEP countries. It is planned to discuss the issue of B[a]P pollution in urban areas at a thematic session of the forthcoming 2nd joint session of the Steering Body to EMEP and the Working Group on Effects in September 2016.

3.3. Cooperation with UNEP Stockholm Convention

Information exchange with other international organizations and programmes is of importance for the assessment of environmental pollution of the EMEP region by POPs. In the framework of this activity MSC-E continues co-operation with the UNEP Stockholm Convention (SC) which includes application of data on emissions and measurements of POPs, compiled under this Convention.

In particular, evaluation of PCDD/F intercontinental transport and contributions of non-EMEP sources to the pollution of the EMEP countries is performed using experimental emission scenario, constructed on the basis of national inventories of dioxins and furans releases submitted by countries to the UNEP SC [*Shatalov et al.,* 2014]. There is significant progress in the development and updating of these national inventories. According to the national reports, provided by countries to the Stockholm Convention up to the beginning of 2016, information on dioxins and furans emissions is available for more than 140 countries. The reference years of reported emission data vary from 1998 up to 2014, with the majority of emission estimates made for the years around 2004. In addition, substantial number of countries provided data on PCDD/F releases for several years that can be used for the evaluation of temporal changes of emissions. This information is of importance for updating of the scenario of global PCDD/F emission and for the assessment of environmental pollution as in the EMEP region and on the global scale.

Along with data on emissions, significant amount of measurements of POP concentrations in ambient air is compiled in the UNEP SC Global Monitoring Plan Data Warehouse (GMP DWH). Global scale measurements of PCDD/Fs, PCB-153, and HCB concentrations are available for the period approximately from 1990 to 2014. This information has been applied for the analysis of global scale modeling results and estimates of pollution levels in the EMEP countries presented in this report.

3.4. Arctic Monitoring and Arctic Monitoring and Assessment Program

EMEP has a long and successful experience of co-operation with the Arctic Monitoring and Assessment Programme (AMAP), which includes participation in joint assessments and projects. Possible directions of future cooperation between AMAP and CLRTAP have been discussed during the joint meeting held in Potsdam, Germany in February 2016. Potential for the collaboration has identified in a number of scientific and technical areas related to the pollution of the Arctic region by POPs.

The information on pollution, generated within EMEP as a part of its operational activity, is also relevant to the Arctic pollution assessment. The EMEP and AMAP domains share significant part of the European and Asian sectors of the Arctic (Fig. 3.3). In order to contribute to further collaboration between AMAP and CLRTAP evaluation of POP pollution levels and transboundary transport for the part of the Arctic region covered by the EMEP domain have been prepared. Examples of modeling results, obtained from regional scale model simulations for the EMEP domain, are shown below for B[a]P and PCDD/Fs.

Spatial distributions of B[a]P and PCDD/F annual mean air concentrations in the Arctic region, calculated for 2014, are presented in Fig. 3.4. Modeled air concentrations decline in northward direction, indicating the transport of the pollutants from major emission sources of the EMEP countries towards the Arctic region. Elevated levels of air concentrations can be seen over the Northern Atlantic region and the northern parts of Norway, Sweden, Finland, and the Russian Federation. Lower levels of concentrations have been calculated for the areas of the high Arctic.

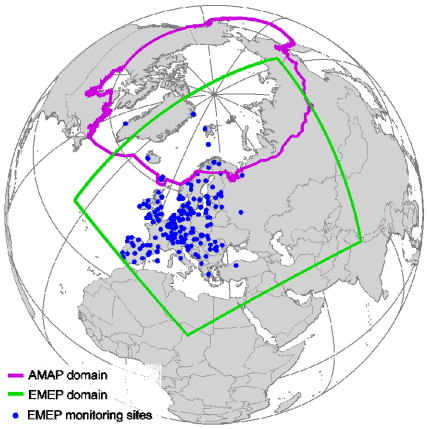


Fig. 3.3. *EMEP and AMAP domains*

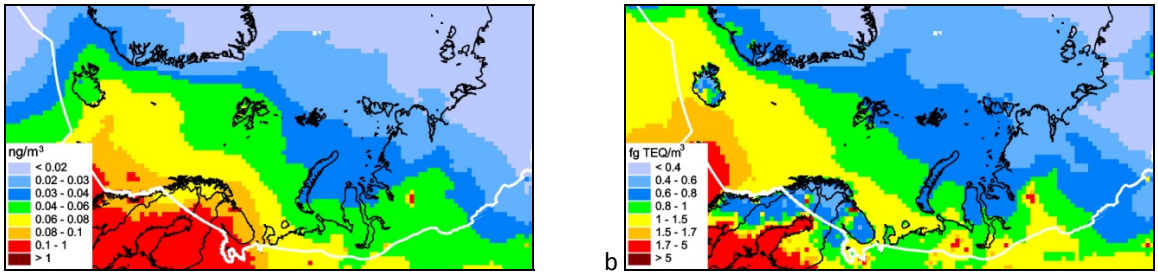


Fig. 3.4*. Spatial distribution of modeled annual mean air concentrations of B[a]P (a), and PCB-153 (b) over the Arctic region covered by the EMEP domain calculated for 2014. White line denotes the boundary of the AMAP domain*

Relative contributions to deposition over the Arctic region in 2014 from the anthropogenic emission sources of the EMEP region are presented in Fig. 3.5. Among the EMEP countries, both for B[a]P and PCDD/Fs, the largest contributions belong to the emission sources of the Russian Federation (43% and 27%, respectively). Considerable contributions to B[a]P deposition are also made by emissions of Ukraine, Poland, and Germany. In case of PCDD/Fs significant fractions of deposition are caused by the emission sources of the United Kingdom, Poland and Ukraine.

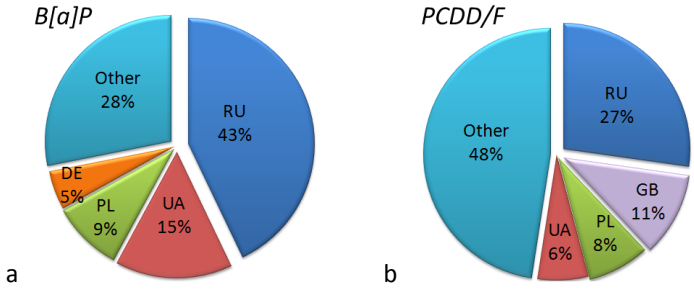


Fig. 3.5. *Relative contributions to annual deposition of B[a]P (a) and PCDD/Fs (b) in 2014 over the part of the Arctic region covered by the EMEP domain, originated from anthropogenic emissions sources of the EMEP countries*

3.5. Helsinki Commission

Evaluation of airborne pollution load to marginal seas within the EMEP region is of interest for various international marine organizations. 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 (UN ECE) and is based on the long-term EMEP/HELCOM contract.

In 2015 assessment of POP pollution load to the Baltic Sea was carried out for PCBs and included evaluation of long-term changes of deposition for the period 1990-2013, source apportionment of deposition for 2013 and verification of modeling results against measurements. Model assessment of pollution levels was carried out for indicator PCB congener PCB-153. Outcome of the assessment is presented in the indicator fact sheets published on the HELCOM website [http://www.helcom.fi] and in the EMEP Centres Joint report for HELCOM [*Semeena et al*., 2015].

According to available information, PCB-153 emissions of the HELCOM countries decreased from 1990 to 2013 by 86%. In 2013 the most significant contributions to the emissions of the HELCOM region were made by Germany (45%), Russia (40%), and Poland (5%). Temporal changes of annual emissions in the region during the two recent decades are illustrated in the Figure 3.6a.

Model assessment of pollution indicated that annual deposition of PCB-153 to the Baltic Sea decreased by 70% in the period 1990-2013 (Fig. 3.6b). Major changes (exceeding 75%) took place in the south-western part of the Baltic Sea, particularly in the Sound and the Kattegat sub-basins. Other parts of the Baltic Sea are characterized by somewhat lower decrease of deposition.

Spatial distribution of PCB-153 deposition to the Baltic Sea in 2013 is presented in Fig.3.6b. Relatively higher pollution levels were characteristic of the western sub-basins (the Sound and the Western Baltic). HELCOM countries contributed about 30% to the pollution of the Baltic Sea by PCB-153. Among these countries the largest contribution to total deposition was made by Germany (8%) and Sweden (6%).

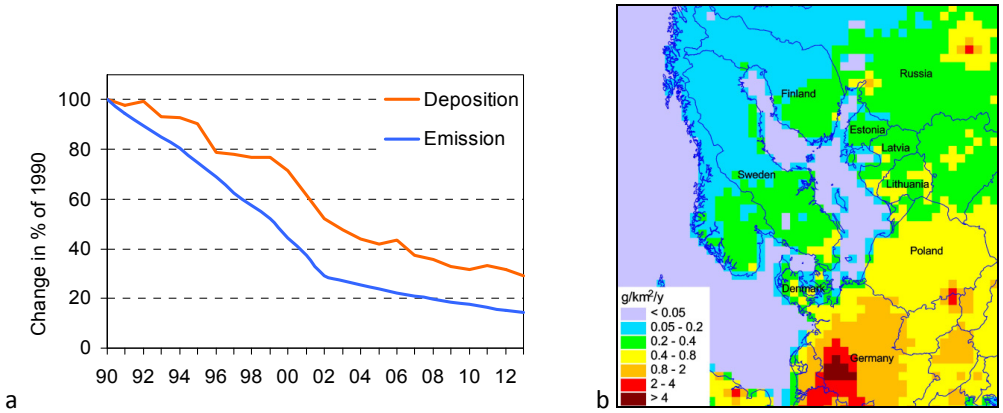


Fig. 3.6. *Changes of annual PCB-153 emissions of HELCOM countries and annual deposition to the Baltic Sea in the period 1990-2013 (a) and spatial distribution of PCB-153 deposition to the Baltic Sea region, g/km^2/y (b)*

Comparison of modeled and measured concentrations of PCB-153 generally indicated reasonable agreement between them. In particular, modeled and observed air concentrations for 2013 differed mostly within a factor of two. Besides, long-term changes of PCB-153 air concentrations observed at several monitoring sites around the Baltic Sea were satisfactory reproduced by the model (e.g. for SE12 shown in Fig. 3.7).

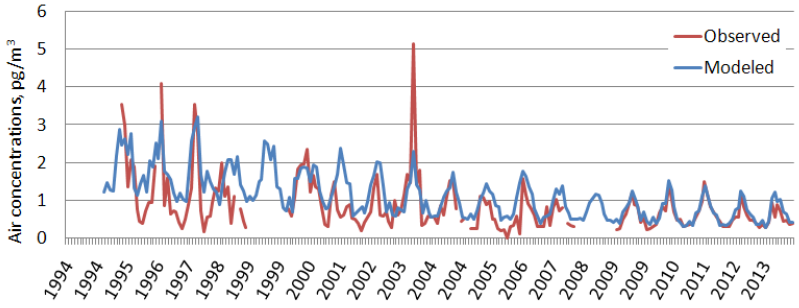


Fig. 3.7. *Comparison of monthly mean modeled PCB-153 air concentrations (pg/m^3) with long-term measurements of the EMEP monitoring site SE12*

Results of model assessment, performed by MSC-E, were discussed at the meeting of the HELCOM PRESSURE group in Copenhagen in 2015. In course of the discussion the representatives of the HELCOM countries expressed their willingness to continue co-operation and to extend the list of hazardous substances for which model assessment of the Baltic Sea pollution is performed by EMEP. In particular, this proposed list includes heavy metals (Pb, Cd, and Hg), persistent organic pollutants (PCDD/Fs, PCBs, PAHs), and new substances (PBDE, HBCDD, and PFOS), which are considered for the inclusion into the CLRTAP Protocol on POPs.

3.6. Cooperation with TFEIP/CEIP

Improvement of quality, transparency, consistency and completeness of emission inventories is an important activity of the Convention. Information on pollutant's emission to the atmosphere and other environmental media is one of the key parameters required for model assessment of pollution levels and transboundary fluxes. Uncertainties of emission data can significantly affect quality of the model estimates. At the recent session of the Executive Body [ECE/EB.AIR/133/add.1] it was stressed that scientific work to improve the quality and robustness of emission and projection data should be further maintained and reflected in the 2016-2017 work plan for the implementation of the Convention. In framework of this activity it is planned to prepare a joint CEIP/MSC-E Technical report on current state of POP emissions for the EMEP region, including description of methodological aspects with regard to gap filling, identification of the sources of errors, preparation of expert estimates, as well as proposals to upgrade current situation and practice, etc. This joint report should serve as a basis for the EMEP strategy to alleviate existing problems and guide Parties.

MSC-E contributed to the preparation of this report with information on available emission data for POP modeling and major issues regarding its quality. The key parameters of POP emission affecting

model assessment results have been analysed and ranked in terms of their priority. It has been noted that completeness of gridded emission data has the highest priority. Other important parameters include inter-annual variations of PAH emissions, and information on congener profiles for PCDD/Fs and PCBs. This ranking can form a basis for further improvement of emission inventories that can be implemented in cooperation with CEIP, the Task Force on Emission Inventories and Projections (TFEIP), and national experts. The contribution to the joint CEIP/MSC-E Technical report related to the POP emissions is available on the MSC-E web page [www.msceast.org]. It was presented at the EMEP Steering Body and WGE extended Bureau (Geneva, March 2016), the TFMM meeting (Utrecht, May 2016) and sent to TFEIP and CEIP for consideration.

MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

This report summarizes the outcome of current activities of the EMEP Centres, MSC-E and CCC, aimed at assessment of POP pollution in the EMEP region. It provides information on POP transboundary transport and pollution levels in 2014, with emphasis to PAH pollution in the EMEP countries, and identifies main directions of future improvement of POP pollution assessment and model development. In this section main challenges that need to be addressed in further research and operational work are outlined.

- Reported emission data for POPs are characterised by significant uncertainties. The key parameters of POP emission, which affect quality of model assessment, are analysed and ranked with regard to their priority. It has been noted that completeness of gridded emission data has the highest priority. Other important parameters include intra-annual variations of PAH and PCDD/F emissions, and information on congener profiles for PCDD/Fs and PCBs. In addition, development and refinement of global scale inventories as well as historical emissions for long-living POPs (PCDD/Fs, PCBs, HCB) are required. This ranking can form a basis for subsequent improvement of emission inventories that can be realized in cooperation with CEIP, TFEIP, and national experts.

- Air pollution by carcinogenic PAHs is recognized as a serious problem in areas with dense population of many EMEP countries. To improve evaluation of PAH pollution levels and to contribute to the analysis of population exposure a study aimed at quantification of B[a]P air concentrations in urban areas is initiated. Preliminary estimates of B[a]P pollution in the Czech Republic show in general reasonable agreement with measurements. At the same time, some discrepancies require further development of the applied approach in order to reduce the level of uncertainties in estimates of B[a]P concentrations. At the next stages of this work the proposed approach can be used to other EMEP countries, provided that the data on emissions and measurements are available. This activity requires close co-operation with national experts on monitoring and assessment of PAH pollution levels as well as with experts from TFEIP, CEIP, CCC, and EEA.

- Improvement of the quality of model assessment of POP pollution levels and trends is one of the important tasks under EMEP. Future development of the Global EMEP Multi-media Modeling System (GLEMOS) will include transition of the EMEP operational modelling of POPs to the new EMEP grid, updating of open code version of GLEMOS, and refinement of parameterization of atmospheric transformation and removal processes for POPs as well as inter-media exchange. Activities, related to the refinement of evaluation of POP pollution, will also comprise collection and analysis of information on congener composition of emissions and concentrations of PCDD/Fs and PCBs, which is essential for the evaluation of PCDD/F and PCB transport and fate in the environment as well as for exposure studies.

- Monitoring of POP concentrations is one of the key sources of information for the assessment of environmental contamination and temporal trends of pollution levels. At present, measurements of POP concentrations, provided by the EMEP monitoring network, are available mainly for the western, central and northern parts of Europe, whereas its southern part and the EECCA region

require improvement of spatial coverage by measurements. In addition, measurement data on POP concentrations in the atmosphere and surface media from national monitoring networks can be an additional source of information for the refinement of POP pollution assessment. Besides, further cooperation with international organizations (e.g. EU EEA, UNEP SC, AMAP, HELCOM), performing monitoring of POPs and collection of measurement data is highly appreciated. Another important issue is related to quality assurance and quality control of POP monitoring data in the EMEP region and other regions as well as comparability of measurement data of different monitoring networks.

- Cooperation between the UNEP Stockholm Convention and EMEP and exchange of information on measurements and emissions of POPs are of particular importance for the evaluation of POP pollution of the EMEP countries. National emission inventories, reported by countries to the Stockholm Convention, provide useful information on POP emissions (e.g. PCDD/Fs) for further studies of environmental pollution by POPs both in the EMEP region and on the global scale. Along with data on emissions, significant amount of global scale measurements of POP concentrations in the atmosphere and other environmental compartments is compiled in the UNEP SC GMP Data Warehouse, which can be used for the refinement of assessment of current pollution levels and their temporal trends.

WORK-PLAN FOR 2017

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

Research activity in 2017

9	Pilot simulations of long-range transport of POPs in the new EMEP grid and evaluation of modelling results: model performance and source-receptor relationships	EMEP status and technical reports	CEIP, CCC	TFMM, SB
10	Ecosystem-dependent deposition fluxes of POPs to different land use types in the new EMEP grid	Model results (Section to EMEP status report)		TFMM, WGE
11	Evaluation of POPs pollution levels in urban areas and selected cities of the EMEP countries	Section to EMEP status and MSC-E Technical reports	Selected countries	SB, TFMM
12	Explore possible use of EMEP/WGE tools, data and infrastructure to support AMAP activities	Report to the EMEP/WGE session		SB, WGE, AMAP
13	Support UNEP Stockholm Convention in relation to atmospheric observations and data management within the ECE region	Report to the EMEP/WGE session		SB
13	Continue collaboration with HELCOM related to atmospheric monitoring and modelling and data management	Report to the EMEP/WGE session		SB, HELCOM
14	Review and assess data, methodologies and competences available to deal with POPs issues in the ECE region and propose a strategy to improve emission inventories	Joint CEIP/MSC-E technical report	CEIP	TFEIP, CEIP, SB
15	Contribute to air quality assessment in the EECCA countries	Report to the EMEP/WGE session	CEIP, CCC	SB
16	Improvement of model parameterization of pollutant transformation and exchange between different compartment (air, water, soil, vegetation) based on literature data and national monitoring data on POP content in relevant compartments with global geographical coverage	Section to MSCE-E Technical report		TFMM, National experts, UNEP, AMAP
17	Facilitate the use of the EMEP model by Parties	Make annual release of the GLEMOS open source code		TFMM, SB, Parties

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