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Transboundary transport of persistent organic pollutants with emphasis on PAHs: regional and national scale assessment and transition to the new EMEP grid

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Transboundary transport of Persistent Organic Pollutants with emphasis on PAHs: regional and national scale assessment and transition to the new EMEP grid

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EXECUTIVE SUMMARY

Persistent organic pollutants (POPs) are semi-volatile toxic organic compounds, characterized by resistance to degradation, significant potential to long-range transport, and harmful effects to human health and wildlife. POPs are within the scope of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention) since coming into force the Aarhus Protocol on Persistent Organic Pollutants in 1998. Monitoring of pollution levels and compiling data on emissions as well as assessment of transboundary transport of POPs is carried out by scientific centres of Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP).

In this report the outcome of 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), is presented. The 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 is provided for 2015, based on reported anthropogenic emissions, modelling results, and measurements. Main emphasis is given to the evaluation of PAH pollution levels in the EMEP countries. Besides, the progress in the transition of operational POP modelling to the new EMEP grid is presented. Particular attention is also 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 different national and international organizations.

Assessment of POP pollution levels in the EMEP countries for 2015 was carried out using gridded emission data, prepared by the Centre on Emission Inventories and Projections (CEIP). Information on vertical distribution and intra-annual variations of emissions as well as compilation of emission scenarios for global-scale modelling were prepared by MSC-E. Emissions from anthropogenic sources within the EMEP region in 2015 were lower for PAHs, and higher for PCDD/Fs, PCBs, and HCB comparing to the level of emissions in 2014. While the completeness and consistency of national emission inventories is gradually improving, POP emissions of some countries are still subject of substantial uncertainties. Particularly important are completeness of sector-wise data on emissions and speciation of emissions for PAHs, PCDD/Fs, and PCBs.

Measurements of POP air concentrations in 2015 were carried out at 34 EMEP monitoring sites, most part of which performed monitoring of PAH concentrations. At the same time, data on PCBs and HCB were reported by less than 10 sites and no measurements were made for PCDD/Fs. Substantial amount of monitoring data on PAHs is available in the EEA AirBase and for other POPs in the UNEP Stockholm Convention Global Monitoring Plan Data Warehouse, which can be used for the evaluation of spatial and temporal variations of POP concentrations.

Available time trends of HCB at some EMEP sites show increasing air concentrations during the last five to ten years. The reasons of these changes require more research and better understanding of the relative importance of primary and secondary sources of HCB. This also highlights the critical importance of continuous and consistent long-term monitoring of regulated POPs, even after periods of decline. Comparing data from different sampling sites and laboratories is a complicating factor when interpreting POP measurements (e.g. for HCB) due to differences in sampling and analytical

methodologies. Thus, there is a need for further focus on harmonization of sampling strategies within CCC and EMEP to enhance the spatial comparability of POP monitoring data.

High levels of air concentrations of B(a)P in urban areas of the EMEP countries have been indicated as an important issue by thematic session on B(a)P during the second joint session of the Working Group on Effects and the Steering Body to EMEP. Specific attention was paid to the importance of PAH emissions from residential combustion and biomass burning as well as to spatial and temporal trends in B(a)P pollution. Analysis of B(a)P concentrations, measured by national monitoring sites in the period 2005-2015, indicates exceedances of the EU target level for B(a)P for countries in Central and Eastern Europe, with highest observed B(a)P concentrations in Poland. Monitoring sites show both decreasing and increasing pollution levels in the EMEP countries. About 65% of sites reported decreasing B(a)P concentrations during this period. Nevertheless, significant amount of sites indicates increasing concentrations, which can be noted for Poland, Czech Republic, and Italy, as well as for the UK, Ireland, Spain, Austria, Slovakia, and Cyprus.

Evaluation of PAH pollution levels and transboundary transport in the EMEP region was carried out for the selected 4 PAHs, namely, B(a)P, B(b)F, B(k)F, and IP. Relatively high annual mean PAH concentrations are estimated for countries in Central Europe, namely, in Poland, the Czech Republic, Slovakia, Slovenia, Romania, Montenegro, and Hungary. Areas of high PAH concentrations are also indicated for Spain, Portugal, Germany, and northern Italy. Assessment of PAH distribution in the EMEP domain indicates importance of the long-range transport of pollution between the countries. Source-apportionment of PAH deposition shows that for 70% of countries the contribution of emission sources, located beyond their territories, exceeds the contribution of their own national emissions.

Comparison of model predictions with measurements of B(a)P, B(b)F, B(k)F, and IP for 2015 demonstrates generally reasonable capturing of observed spatial variations of PAH air concentrations by the model. At the same time, for several monitoring sites in France, Belgium, the Netherlands, and Sweden the model tends to overestimate observed concentrations. The overestimation is most likely caused by the uncertainties in the reported PAH emissions as well as in their speciation and spatial distribution. Particularly, analysis of monitoring data and reported emissions for B(a)P shows noticeable disagreement between observed concentrations and mean emission fluxes for Poland, Germany, and Belgium, which may point out possible underestimation of PAH emissions in Poland, and overestimation of emissions in Germany and Belgium. Thus, further refinement of PAH emission data, reported by the EMEP countries, is of importance for the reduction of uncertainties of pollution assessment.

Special attention in the activities of MSC-E during this year was paid to the transition of POP operational modelling to the new EMEP grid system. The Centre continued the work on the adaptation of the Global EMEP Multi-media Modelling System (GLEMOS) for modelling using the new grid and preparation of necessary input data including meteorological and geophysical information. Particularly, the input of gridded emission data including vertical structure, intra-annual variations, and distribution by sectors was refined. A new system of preparation and input of the initial and boundary conditions for regional modelling was developed. A number of modifications were made in the parameterizations of POP deposition, gas-particle partitioning, and degradation processes applied in the model. Pilot simulations of B(a)P pollution on the new EMEP grid have shown generally better agreement with

measurements of EMEP monitoring network in comparison with the results for older EMEP grid. At the same time, for some of the stations model predictions noticeably differ from the observed pollution levels. For better performance of the GLEMOS model there is a need of further refinement of model parameterization of gas-particle partitioning, degradation, and deposition processes.

Detailed analysis of PAH pollution at a country scale has been initiated for Spain in the framework of country-specific case studies following the recommendation of the second joint session of the Working Group on Effects and the Steering Body to EMEP. To analyze spatial variations of B(a)P concentrations over the country, fine resolution modelling using detailed emissions data and measurements of B(a)P at national monitoring network was carried out. According to available measurements, spatial distribution of B(a)P pollution levels in Spain is characterized by relatively higher levels of B(a)P pollution in its northern areas and lower in other areas of the country. Comparison of model predictions with measurements indicates overestimation of observed B(a)P air concentrations for central and western parts of the country, which can be attributed to possible uncertainties in the reported national PAH emissions as well as uncertainties in modelling approach and measurements.

Particularly, national PAH emission inventory of Spain is characterized by noticeably high contribution of emissions from field burning of agricultural residues (about 70%), while contribution of residential combustion is less important. This pattern of emissions distinctly differs from the inventories of other EMEP countries, where residential combustion is dominating source category (except for Portugal and Cyprus). Experimental model simulations with several emission scenarios and analysis of observed levels of PAH pollution have indicated possible uncertainties in estimates of PAH emissions from agriculture. Thus, further refinement of emissions related to the burning of agricultural residues and analysis of their uncertainties is appreciated to improve accuracy of model predictions. In addition, analysis of major factors, influencing B(a)P pollution levels, and comparison of EMEP and national modelling results for Spain is planned to be performed.

Evaluation of pollution by PCDD/Fs, PCB-153, and HCB was based on multi-media modelling approach to take into account their long-term cycling between and accumulation in different environmental compartments. Spatial variability of air concentrations and deposition fluxes within the EMEP domain was analyzed for 2015 on the basis of nested regional and global scale model simulations. Model simulations indicated substantial contribution of anthropogenic emissions to PCDD/F pollution levels in the EMEP countries. For PCB-153 and HCB significant contribution was estimated for secondary emission sources. Model predictions for non-EMEP emissions varied from several percents to 50% for PCDD/Fs and PCB-153, and from 30% to 70% for HCB. Estimates of PCDD/Fs, PCB-153, and HCB transboundary fluxes indicated significant role of air pollution transport between the EMEP countries. According to modelling results, the fraction of POP deposition over the territories of the countries, caused by the atmospheric transport from external emission sources, exceeds the fraction of deposition from their national emissions in 75% of the countries for HCB, in 65% of the countries for PCB-153, and in 55% of the countries for PCDD/Fs.

Important aspect of MSC-E work is co-operation with subsidiary bodies to the Convention. The outcome of recent research activities and model developments, performed by the Centre, was discussed at the EMEP Task Force on Measurements and Modelling (TFMM). In particular, the progress in the transition of POP modelling to the new EMEP grid was demonstrated. Besides, preliminary results of country-

specific case study of B(a)P pollution in Spain were presented and discussed during the meeting. The representative of Spain informed participants about ongoing refinement of national inventory of PAH emissions in Spain and the need to update modelling results using refined emissions. To continue activities in framework of country scale pollution studies it was suggested to perform analysis of B(a)P pollution levels in France and Poland. As an important topic, relevant to further improvement of pollution assessment, the application of modelling and monitoring data fusion approaches can be discussed at future meetings of the Task Force.

Collaboration of MSC-E and national experts of the EMEP countries is continued. To support national study of POP pollution in Italy, regional-scale modelling for the evaluation of POP air concentrations in the Mediterranean region was carried out. Results of model simulations will be used as information on boundary concentrations in national scale modelling. Besides, the Centre collaborated with national experts from Germany providing model estimates of atmospheric deposition fluxes of PAHs to inland waters of the country.

Co-operation and exchange of information with other international organizations and programmes is essential for the assessment of pollution of the EMEP region. In the framework of this activity MSC-E continues co-operation with the other EMEP Centres, aimed to assessment of atmospheric input of various pollutants including POPs to the Baltic Sea. This work is carried out in accordance with the Memorandum of Understanding between CLRTAP and the Baltic Marine Environment Protection Commission (HELCOM). Data of national emission inventories, provided by countries to the UNEP Stockholm Convention (SC), represent important source of information for studies of environmental pollution by PCDD/Fs and other POPs as for the EMEP region and for the global scale. Thus, further co-operation between the CLRTAP and SC in this field is highly appreciated.

Further activities of MSC-E will be directed to the refinement of assessment of POP pollution in the EMEP region. Development and testing of the Global EMEP Multi-media Modelling System (GLEMOS) performance for the new EMEP grid will be continued. Besides, analysis of key factors that affect POP transport in the atmosphere and exchange surface compartments will be performed. Country-specific case studies will include analysis of PAH pollution for France and Poland. Specific attention will be paid to the 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. These directions of future research and development are outlined in the MSC-E proposals for the EMEP workplan for 2018-2019 and the updated Mandate of the Centre.

CONTENTS

INTRODUCTION

Persistent Organic Pollutants (POPs) are characterized by significant adverse effects to human health and wildlife. Due to international cooperation and measures to eliminate or reduce unintentional production and use of many POPs, their releases into environment have considerably declined. At the same time, POPs still present in the environmental compartments, posing risk to human health as well as terrestrial and aquatic ecosystems. POPs are within the scope of activities of the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention). Along with this, special attention to the reduction of POP pollution levels is paid by many other international and national organizations including 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 (EMEP) and its scientific centres annually provide the Executive Body for the Convention with the information on emissions, concentrations, deposition and transboundary transport of selected POPs (PAHs, PCDD/Fs, PCBs and HCB) within the geographical scope of EMEP. In particular, the Centre of Emission Inventories and Projections generates the data on POP emissions based on the national emission inventories of Parties to the Convention and expert estimates. Collection and analysis of POP measurements of EMEP monitoring network as well as methodological guidance of monitoring activities is made by 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. More detailed information on the EMEP structure and operational activities of the research centres is available at the EMEP website (www.emep.int).

This Status Report presents results of recent activities of the EMEP Centres in the field of POP pollution assessment, carried out in accordance with the work-plan of the Convention for 2016-2017 [ECE/EB.AIR/133/Add.1]. It includes the outcome of model assessment of POP pollution in the EMEP region, progress in the transition of POP operational modelling to the new EMEP grid, and preliminary results of country-specific case study of B(a)P pollution in Spain. In addition, measurements of POP concentrations and information on POP emissions for 2015 are presented.

Specific attention in the report is given to the carcinogenic PAHs, and, in particular, to benzo(a)pyrene (B(a)P) as the indicator PAH compound. Air pollution by B(a)P was indicated as an important issue by thematic session on B(a)P during the second joint session of the Working Group on Effects and the Steering Body to EMEP. Assessment of PAH pollution levels in the EMEP countries is based on model simulations and measurements of EMEP monitoring network. Along with EMEP monitoring data, results of national monitoring of B(a)P air concentrations, collected in the EEA/AirBase, are applied to analyze spatial and temporal trends in the pollution levels. Model assessment of pollution provides information on air concentrations, deposition fluxes, and transboundary fluxes between the EMEP countries as well as contamination of marginal seas and the Arctic area within the EMEP region. Along with this, changes of pollution levels between the two years, 2014 and 2015, are characterized. Similar information on the

pollution levels of other selected POPs, namely, PCDD/Fs, PCBs, and HCB, and their inter-annual changes is presented in the report. Model evaluation of pollution for these POPs is based on nesting of regional and global scale model simulations in order to estimate intercontinental transport of pollutants and contributions of non-EMEP emission sources.

Recent activities of MSC-E, related to the development of the Global EMEP Multi-media Modelling System (GLEMOS) for POPs, were mostly focused on the transition of operational modelling to the new EMEP grid. Following the decisions of the Executive Body for CLRTAP [ECE/EB.AIR/113/Add.1], MSC-E initiated adaptation of GLEMOS model and preparation of necessary input data. This year transition of the modelling to the new EMEP domain is continued and includes modifications of POP-specific modules of the GLEMOS model. Updated version of the model is tested using pilot simulations of B(a)P long-range transport over the new EMEP grid with finer spatial resolution. At the same time, for PCDD/Fs, PCBs, and HCB implementation of the modelling on the new grid requires further adaptation and testing of model performance.

Collaboration of MSC-E with Parties to the Convention in the field of pollution assessment is continued. Model assessment of PAH pollution with fine spatial resolution in framework of country-specific case studies is started for Spain in accordance with the recommendation of the second joint session of the Working Group on Effects and the Steering Body to EMEP. Objective of this study is to perform analysis of B(a)P pollution levels in Spain using national emission data with fine resolution, measurements of monitoring network of Spain and results of EMEP GLEMOS model and CHIMERE model. Preliminary outcome of the study is presented in the report and directions of further work are outlined. Results of this activity are also important for the ongoing transition of POP modelling to the new EMEP grid with finer spatial resolution.

Important part of the MSC-E work is co-operation with subsidiary bodies to the Convention and other international organisations. Results of recent activities of the Centre were presented and discussed at the EMEP Task Force on Measurements and Modelling (TFMM). Special attention was also paid to collaboration with other international organizations and programmes (the UNEP Stockholm Convention, the Arctic Monitoring and Assessment Programme, Helsinki Commission etc.) in order to broaden dissemination of the scientific and policy oriented information, generated by EMEP.

Detailed information on POP pollution levels in the EMEP region and in individual EMEP countries as well as transboundary transport are presented at the MSC-E website (www.msceast.org). Along with this, the information on POP pollution in the EMEP countries of Eastern Europe, Caucasus and Central Asia (EECCA) at the MSC-E website is given in Russian (www.ru.msceast.org).

1. EMEP MONITORING DATA FOR POPS IN 2015 WITH FOCUS ON HEXACHLOROBENZENE

Measurements of persistent organic pollutants (POPs) 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 2015, 34 sites have reported data for PAHs and/or other POPs in air. Of these, 27 reported PAHs only and seven reported other POPs. For precipitation in 2015, PAHs and/or other POPs have been reported from 29 sites. Of these, 25 reported PAHs and nine reported other POPs.*** In this chapter, we have selected hexachlorobenzene (HCB) to illustrate and discuss some critical issues related to comparability and consistency of EMEP data, as well as the importance of continued long-term monitoring. HCB in air and precipitation in 2015 is available from eight and five sites, respectively.

The chemical coordinating centre (CCC) supports EMEP with monitoring and analytical strategies and guidelines in order to gather quality data to assess regional-scale air pollution across Europe. A long-term goal of CCC is to collect comparable monitoring data to facilitate consistent data sets for evaluation of spatial and temporal trends. To achieve this goal, CCC attempts to harmonize sampling methodologies and chemical analytical methodologies [*EMEP/CCC*, 2014]. The EMEP Monitoring strategy for 2010-2019 recommends monitoring of six POP classes in the first step: polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), chlordanes (CHLs), α- and γ-hexachlorohexanes (HCHs), DDT/DDE and hexachlorobenzene (HCB).

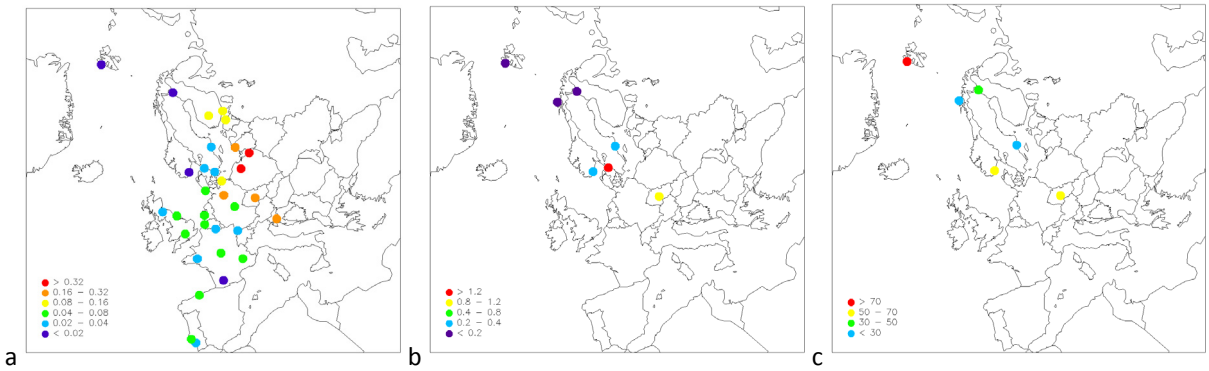


Fig. 1.1*. Annual mean concentrations of B(a)P (ng/m^3) (a), PCB-153 (pg/m^3) (b), and HCB (pg/m^3) (c) in air and aerosols observed in 2015*

Figure 1.1 shows the regional distribution of some selected POPs in air and aerosols: benzo(a)pyrene, PCB-153, and HCB. For B(a)P the annual mean concentrations in air ranges from 0.002 ng/m^3 in the Arctic to 0.6 ng/m^3 in Poland, while for PCB-153 the highest annual mean concentration (1.47 pg/m^3) was observed in Sweden and the lowest one was in the Arctic (0.11 ng/m^3). In contrast, the highest annual mean concentration of HCB (86 ng/m^3) was observed at the Arctic site in Spitsbergen.

While most of the POP classes generally tend to show decreasing time trends at monitoring sites within the EMEP domain, long-term temporal trends are rarely always consistent across sites. Nor are temporal trends of individual POP compounds, measured at individual sites, always showing continuous decline. Occasionally, there might be periods showing a decrease, followed by increasing

concentrations. It is therefore interesting to illustrate such patterns with an example. We have selected HCB as an example because (i) it shows divergent temporal trends compared to other POPs, which has triggered a debate in the scientific literature about the possible cause(s), and (ii) this chemical has a relatively high volatility, and there are well-know challenges associated with active air sampling of this compound. The latter may result in a limited comparability of data across EMEP sites, while temporal trends from individual sites may still be internally consistent.

Long-term monitoring data sets for HCB are available from 1990s to 2012/2015 from three sites for air and four sites for precipitation (Table 1.1). In detail, HCB data in air is available from Zeppelin in Norway (1993), Birkenes in Norway (1996), and Storhofdi in Iceland (1995) while HCB data in precipitation is available from Westerland and Zingst in Germany (1996 and 1999), Storhofdi in Iceland (1995) and Birkenes in Norway (1992). From the 2000s, additionally ten sites have reported HCB data in air for at least five years while no additional sites have reported HCB data in precipitation. The start date of these additional monitoring data sets varies between sites and matrix (Table 1.1).

Table 1.1. *Available long-term data on HCB in air and precipitation at EMEP sites showing the starting year*

Site name (site code)	Air	Precipitation
Kosetice (CZ03)	2006	
Westerland (DE01)	2007	1996
Schauinsland (DE03)	2007	
Schmücke (DE08)	2007	
Zingst (DE09)	2007	1999
Station Nord (DK10)	2009	
Pallas (FI36)	2002	
Storhofdi (IS91)*	1995*	1995
Birkenes (NO99/01/02)	1996	1992
Zeppelin (NO42)	1993	
Andøya (NO90)	2009	
Aspvreten (SE12)	2006	
Råö (SE14)	2006	

* Stopped measuring POPs after 2012

Detailed information about the sites, measurement methods and results for 2015 can be found in EMEP/CCC's data reports on heavy metals and POPs [*Aas and Nizzetto*, 2017]. All data are also available in the EMEP database (http://ebas.nilu.no).

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 [*Su and Hung*, 2010; *Schlabach et al*., 2011 and *Melymuk et al*., 2014]. This is a special challenge for HCB data in air as HCB, due to its volatility, has been shown to undergo breakthrough in high volume air samples [*Melymuk et al.*, 2016]. The amount of breakthrough and thereby the amount of underestimations in air samples are dependent on the total sample volume, the outdoor temperature during sampling and the sampler configuration. Experimental studies have shown

that HCB experience breakthrough losses when the sample volumes exceed 700 m^3. The sample volumes at the EMEP sites are often above this volume and breakthrough is therefore expected to occur. In addition, as both sample volumes and temperatures vary between the sites, ***there is most likely limited comparability of HCB data across sites within the EMEP network. This example illustrates the need for continued focus on harmonization of sampling strategies within CCC and EMEP to enhance the spatial comparability.*** However, for temporal trends at the individual sites, the influence of breakthrough effects is minor as long as the sampling methodologies are kept intact within the time period. Time trends for HCB at individual sites may therefore be considered consistent and informative for EMEP nevertheless.

HCB in air and precipitation, temporal trends until 2015

An overview of the annual mean concentrations reported for HCB in air at the individual sites in the time frame of POP monitoring in EMEP (1993-2015) is presented in Table 1.2. The spatial pattern for HCB in the EMEP region is hampered by the limited number of stations in combination with limited comparability across stations for reasons discussed above (Fig. 1.1). This motivates the need for continuous work to improve comparability, e.g. better coordination and harmonization within CCC and EMEP, and a better understanding of the influence of breakthrough or other sampling artefacts under variable environmental and sampling conditions across various POPs. It also illustrates that alternative sampling strategies, such as passive air samplers, may provide complementary insights into spatial patterns in European background air [e.g. *Halse et al.*, 2011].

Table 1.2. *Annual mean concentrations in air for HCB (pg/m^3) at EMEP sites 1993-2015.*

	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
CZ0003R														70	19	67	46	40	77	97	105	115	53
DE0001R															63		56	25		29	30	28	
DE0003R															60		58			31	32	28	
DE0008R															63		64			33	29	37	
DE0009R															72		55	21	21	30	28	25	
DK0010R																	49	75	78	86	107	93	
FI0036R										38							19	32			31	29	40
IS0091R			7	6	10	13	6	6	7	4	4	3	2	4	6	7	5	5	3	4	5	4	
NO0099R/ NO0001R/ NO0002R				89		93	83	55	51	57	65	64	64	64	86	63	42	51	47	53	52	52	56
NO0042R	93	116	98	92	99	82	88	56	55	56	54	65	67	71	67	73	76	79	81	83	81	83	86
NO00090R																	88	29	24	28	27	31	28
SE0012R																	18	25		22	35	37	30
SE0014R																		15	26	23	19	24	

Table 1.3. *Annual mean concentrations in precipitation for HCB (ng/L) at EMEP sites 1993-2015*

	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
DE0001R				0.09	0.12	0.12	0.02	0.20	0.15	0.10	0.09	0.24	0.06	0.02	0.03		0.05	0.04	0.05	0.05	0.05		
DE0009R							0.01	0.07	0.15	0.11	0.08	0.03	0.07	0.05	0.07	0.04	0.34	0.05	0.05	0.07	0.06		
IS0091R			0.03		0.02	0.02	0.02	0.02	0.01	0.01	0.01	0.02	0.02	0.01	0.01	0.01	0.01	0.02	0.02	0.01			
NO0099R/ NO0001R	0.40	0.57	0.81	1.41	0.90	0.68	0.55	0.49	0.33	0.34	0.43	0.12	0.12	0.13	0.18	0.09	0.06	0.09	0.08	0.10	0.08	0.07	0.09

- *50% of the samples below detection limit*

Time trends for HCB at individual sites can nevertheless be assessed when the sampling and analytical methodologies have been kept consistent. ***The available time trends within EMEP shows an initial decline of HCB concentrations in air during the 1990s. In contrast to many other POPs (e.g. HCHs) that shows continuous declining levels or apparent steady-state conditions in air also in the 2000s, HCB in air is found to increase at some EMEP sites during the last five to ten years*** (Tables 1.2, 1.3). These upward trends have been most evident at Zeppelin (NO0042), Norway, Kosetice (CZ0003), Czech Republic and Station Nord (DK0010), Denmark (Fig.1.2).

The reasons for the increasing concentrations in air are currently not known. The two main hypotheses identified are i) potentially increasing primary emissions of HCB in some parts of the world, and ii) re-volatilization from secondary sources in response to climate changes [*Barber et al.*, 2005; *Hung et al.*, 2010; *Ma et al*., 2011]. ***More research is needed to understand the reasons, as a better understanding of the relative importance of primary and secondary sources of HCB is vital to inform potential control strategies. The increasing trends for HCB also highlight the critical importance of continuous and consistent long-term monitoring of regulated POPs, even after periods of decline.***

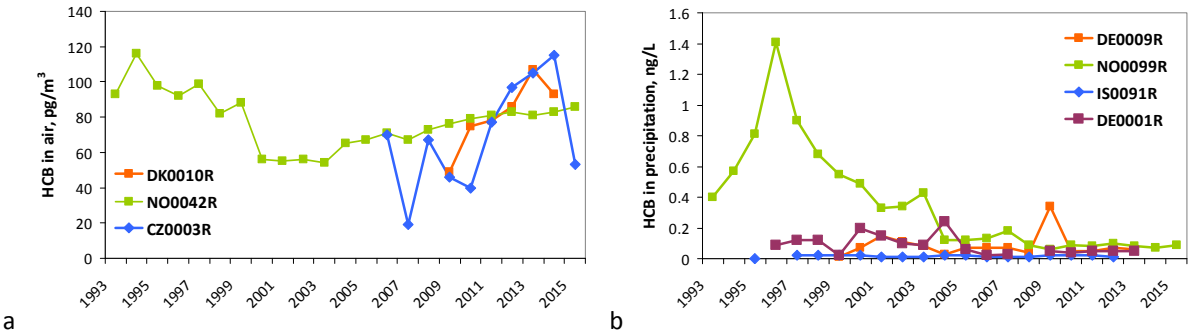


Fig. 1.2. *Long-term annual mean concentrations in air (a) and precipitation (b) for HCB*

New reporting guidelines for POPs

As illustrated above, an important measure to help support interpretation of monitoring data is to strengthen efforts to document methodologies and data quality across EMEP sites. The EBAS database (http://ebas.nilu.no/), which host data from EMEP, GAW, AMAP, CAMP, HELCOM, ACTRIS and more, has been improved to include new possibilities for metadata and information on quality assurance (QA). To discuss these new possibilities and the increasing need for documenting the data quality, EMEP/CCC arranged a workshop in October 2016, and new reporting guidelines were agreed upon.

Three important metadata points for POPs were discussed and approved at the meeting:

- *Medium*: This is the place to indicate what type of filter and/or other adsorbents that are used for collecting the air samples. For POPs this is usually either a filter or a filter/PUF/XAD combination. One combination is i.e.: Glass fiber filter+PUF+XAD+PUF.

- *Sample preparation*: This describes how the sample has been prepared after being received in the lab. Four important steps were identified (extraction+solvent+internal std+cleanup), and the combination of how this is performed needs to be specified. One example is: Soxhlet+Dichloromethane+13C+Silica.

- *QA measure*: This can be an on-site or off-site intercomparison, round-robin or an on-site audit. In previous years this information was only stored separately from the database, but this information can now be included together with the relevant data series, and thus will also be exported to those using the data. Laboratory intercomparisons of POPs are not done on an annual interval in EMEP, but there has been a few ones which are relevant, e.g. in 2010 [*Schlabach et al.*, 2011]. If the lab has participated in any relevant laboratory intercomparison, this should be included in the QA measures.

For all these metadata items, only predefined combinations and input can be included. If other options are used, the EBAS data base group should be informed about this (e.g. by e-mail ebas@ebas.no).

The reporting templates and instruction on how to submit data are found at the http://ebas-submit.nilu.no. Further, CCC has developed the submission tool (http://ebas-submit-tool.nilu.no), which the data submitters should use to check their files, and the files should be submitted via submission tool unless a special agreement is mutually agreed upon.

2. ASSESSMENT OF PAH POLLUTION IN THE EMEP REGION

Polycyclic Aromatic Hydrocarbons (PAHs) represent a group of chemicals that naturally occur in the environment in coal, crude oil, and gasoline and can be released to the atmosphere during incomplete combustion of fossil fuels and biomass burning. PAHs are considered as substances posing serious risk for the human health [*Theakston*, 2000]. Benzo(a)pyrene has been included in the list of carcinogens of category 1B by the International Agency for Research on Cancer (IARC). Taking into account possible adverse effects on human health, target values of air quality objectives for B(a)P in Europe have been established (Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004). In particular, EU target value for B(a)P annual mean air concentrations is set to 1 ng/m^3. Further, the reference level of 0.12 ng/m^3 for B(a)P was estimated by WHO as level of air concentrations corresponding to excess lifetime cancer risk level of 10^{-5} [*Theakston*, 2000].

In order to evaluate PAH pollution levels and exceedances of target values of concentrations, complementary use of regional and national scale modelling as well as monitoring of air concentrations is required. In this chapter a summary of the assessment of pollution levels and transboundary transport of selected PAHs, namely, Benzo(a)pyrene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, and Indeno(1,2,3-cd)pyrene), in the EMEP region for 2015 is presented. Model simulations of PAH transport and fate have been performed using multi-media GLEMOS modelling system on the basis of official emission data, reported by EMEP countries, and available expert estimates. More detailed analysis of pollution levels in the EMEP countries has been performed for B(a)P, which is used as a marker for the carcinogenic risk of population exposure to PAHs.

To characterize the spatial and temporal variability of PAH pollution levels within the EMEP domain, analysis of modelling results and measurements of the EMEP monitoring network as well as national measurements from EEA/Airbase has been carried out. Transboundary transport of selected PAHs between the EMEP countries has been evaluated. Results of model simulations for 2015 have been compared with measurements and previously reported modelling results for 2014.

Main emphasis in the activities of this year has been given to the transition of POP operational modelling to the new EMEP grid and finer spatial resolution. Specific attention has also been paid to the co-operation with the EMEP countries in the assessment of pollution. In particular, country-specific case study of B(a)P pollution in Spain has been initiated. The outcome of these activities is presented below.

2.1. B(a)P pollution levels observed in the EMEP countries

Measurements of B(a)P, performed by EMEP and national monitoring networks, provide detailed information on air quality in European countries that can be used for the analysis of spatial variations of pollution and long-term trends. Along with monitoring of air pollution in rural and remote areas at EMEP stations, countries, members of EU, deliver information on air quality in urban and sub-urban areas as well as areas, affected by industrial sources and traffic. To characterize temporal variations of observed B(a)P air concentrations in the EMEP region, available monitoring data in the EEA/AirBase for the period 2005-2015 were analyzed by MSC-E.

According to available national measurements for 2015, collected in the EEA/AirBase, high levels of annual mean B(a)P air concentrations, exceeding the EU target level for B(a)P, can be noted for nine countries, with highest observed levels (up to 15 ng/m^3) in Poland (Fig. 2.1a). Spatial distribution of measured B(a)P concentrations (Fig. 2.1b) indicates significant pollution levels in the countries of Central and Eastern Europe as well as in Italy and the UK. High values of B(a)P concentrations, near or above the EU target value 1 ng/m^3, were mostly observed in urban and suburban areas of these countries.

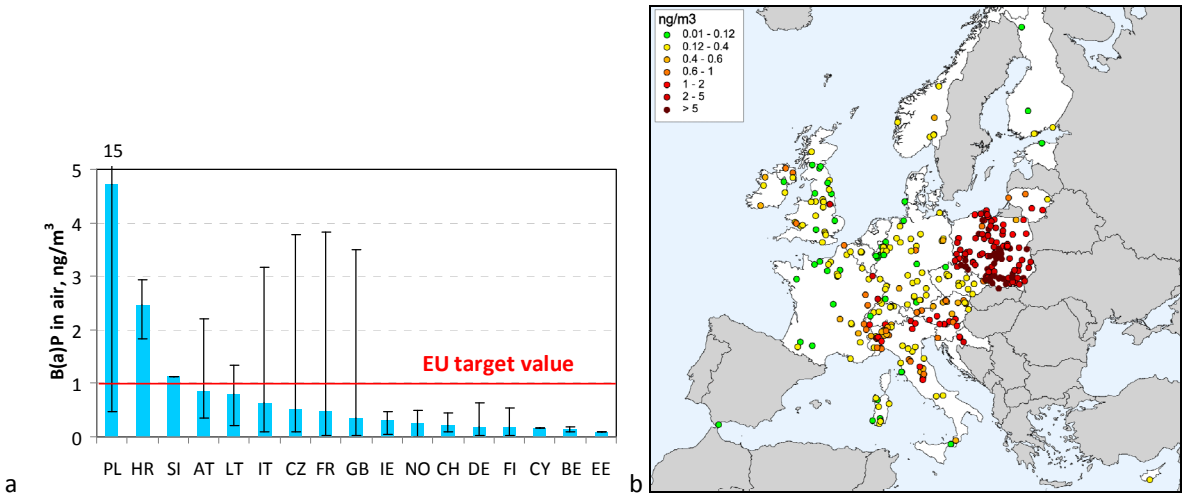


Fig. 2.1. *Annual mean B(a)P air concentrations in the EMEP countries (a) and their spatial distribution (b) observed in 2015 (EEA/AirBase). Bars represent average annual mean B(a)P concentrations in particular countries and whiskers denote the range from minimum to maximum measured concentrations*

To characterize temporal variations of observed B(a)P concentrations during recent decade, analysis of time-series of measured concentrations, reported by national monitoring sites for the period 2005-2015 was carried out. For the majority of monitoring sites time-series of measured concentrations have incomplete coverage of this period, namely, different starting and ending times as well as gaps, that complicated analysis. Therefore, to explore general tendencies in changes of B(a)P concentrations at particular monitoring sites, regression analysis of available time series was applied. For this purpose the linear regression for the time-series of each site with at least three values of annual mean measured concentrations available was calculated. In addition, to test statistical significance of B(a)P concentrations changes, 95% confidence intervals for slopes of linear regression, based on the data of each monitoring site, were evaluated. In case when all the values of regression slopes from confidence interval for a given site were positive, it was assumed that statistically significant increase of concentrations at this site took place. On the opposite, when all the values of regression slopes were negative, statistically significant decrease was assumed for the considered site.

Amount of monitoring sites indicating increase or decrease of annual mean B(a)P air concentrations in period 2005-2015 is compared in Fig. 2.2a for three types of site locations, namely, rural, suburban, and urban. More significant number of sites reported declining of B(a)P concentrations in this period. In particular, ***measurements of about 65% of sites showed decreasing levels of B(a)P concentrations. For about 30% of sites these changes can be considered as statistically significant. At the same time, almost 35% of sites showed increasing concentrations. For 10-20% of time-series statistically significant increase of pollution levels can be noted.***

Prevailing of declining B(a)P concentrations can also be seen from the results of similar comparison made for selected European countries (Fig. 2.2b). For most of these countries the number of sites, indicated decreased B(a)P concentrations, is more significant. However, for Slovakia, Italy, Poland, Estonia, and Ireland percentage of time-series with increasing pollution levels is relatively high (more than 40%).

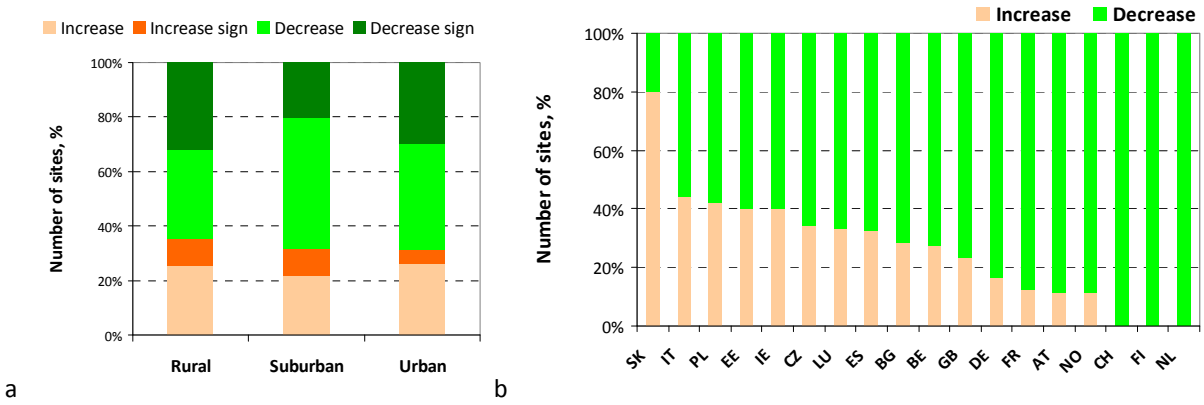


Fig. 2.2. *Number of monitoring sites (in %), indicated increase (light red) or decrease (light green) of annual mean B(a)P air concentrations in European countries in period 2005-2015 based on the data of EEA/AirBase. Amount of sites indicated statistically significant changes is shown using bright green or bright red colours*

Spatial distributions of monitoring sites with indication of increase or decrease of annual mean observed B(a)P air concentrations in period 2005-2015 are presented in Fig. 2.3a and 2.3b.

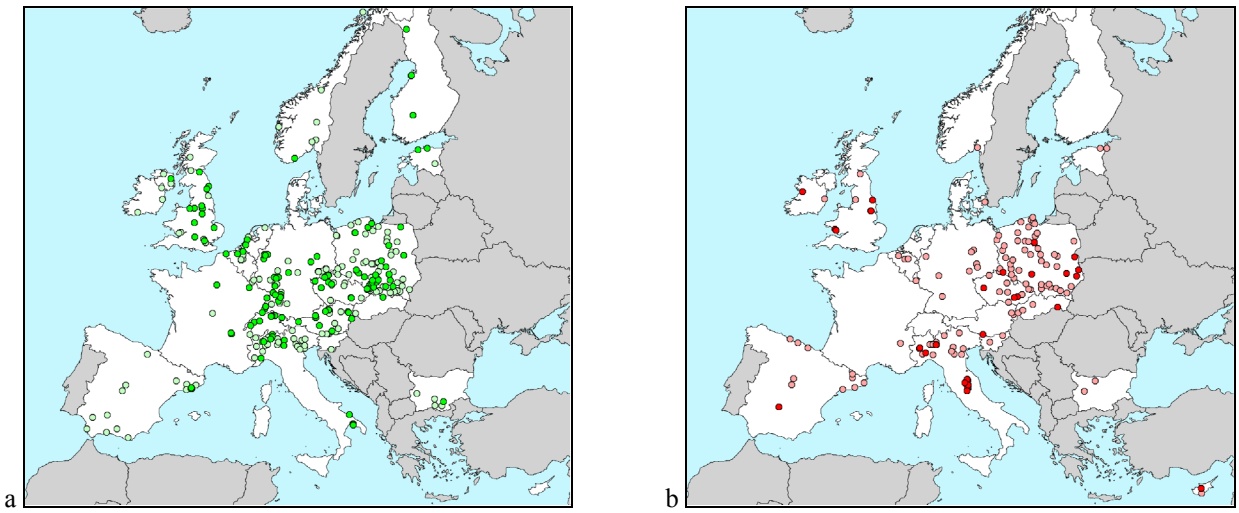


Fig. 2.3. *Locations of monitoring sites, reported decreasing (a) or increasing (b) annual mean B(a)P concentrations in period 2005-2015. Monitoring sites with statistically significant decrease or increase are shown using bright green or red colour. Other sites with increasing or decreasing concentrations are marked with light red or green, respectively*

For most of the countries time-series of measured concentrations indicate both kinds of tendencies. As it was mentioned above, more sites indicated declining concentrations. ***Increase of B(a)P pollution levels is noted for Poland, Czech Republic, and Italy. Monitoring sites, indicating statistically significant increase of B(a)P air concentrations, can be seen in the UK, Ireland, Poland, Italy, Spain, Austria, Slovakia, and Cyprus.***

2.2. Emission data for model assessment

Model assessment of PAH long-range transport and pollution levels in 2015 within the EMEP domain has been carried out for B(a)P, B(b)F, B(k)F, and IP. Gridded data on PAH emissions for 2015 were provided by CEIP. Spatial distributions of PAH and B(a)P emissions from anthropogenic sources in the EMEP domain, used in the model simulations for 2015, are presented in Figs 2.4a and 2.4b, respectively.

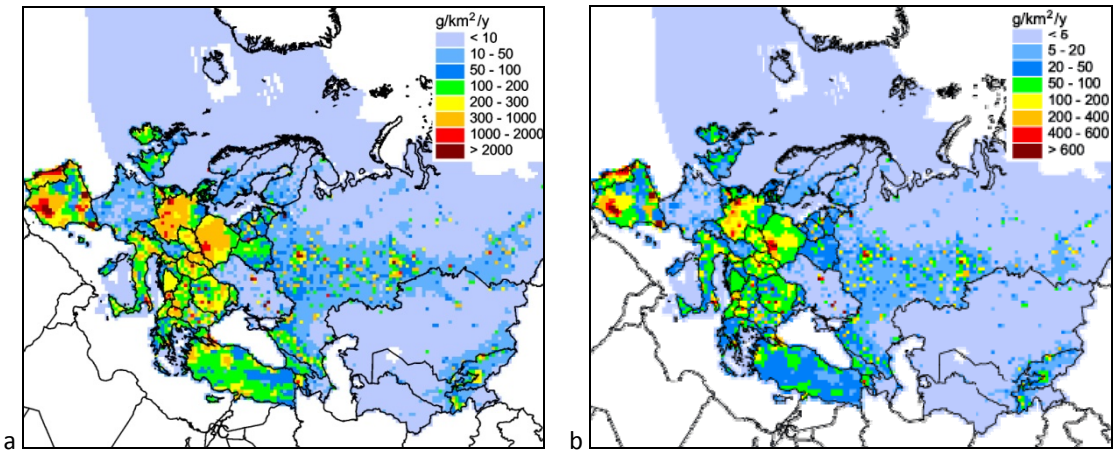


Fig. 2.4. *Spatial distribution of PAH emissions (sum of 4 PAHs) (a) and B(a)P emissions (b) in the EMEP countries in 2015 with resolution 50x50 km²*

According to the data, reported by countries in current and previous submissions of national inventories, ***anthropogenic emissions of 4 PAHs in the EMEP domain in 2015 accounted for 2042 tonnes, which is lower than level of emissions for 2014 by 23%.*** Changes of PAH emissions in the EMEP countries between 2014 and 2015 are illustrated in Fig. 2.5. Negative values denote a decrease of emissions, while positive values mean an increase of emissions between these two years.

Comparison of emission totals indicates that PAH emissions have increased in 26 countries, while 24 countries are characterized by the decrease of emissions. The largest decline of total PAH emissions in tonnes per year is noted for the Russian Federation, Kazakhstan, and Ukraine. Corresponding relative changes amount to 2.3, 8.2, and 1.3 times, respectively. In case of Malta estimates of PAH emission drop from 34.9 tonnes in 2014 down to 0.01 tonnes in 2015.

The most significant increase of total PAH emissions from 2014 to 2015 is seen for Germany, Spain, Turkey, and Hungary. Emissions of these countries in 2015 increased comparing to the previous year by 5.8, 1.2, 1.2, and 2.1 times, respectively. Comparing to 2014, significantly higher PAH emission for 2015 was estimated by CEIP for Armenia. Detailed description of reported PAH emissions in the EMEP countries, including information on the gap-filling of incomplete emission data and preparation of expert estimates, can be found in the Technical report of CEIP [*Tista et al.*, 2017].

PAH emissions in different parts of the EMEP domain are characterized by varying level of completeness and uncertainties. Officially reported information on PAH emissions is available for about 60% of the EMEP countries, which mostly comprise the EU countries. For other EMEP countries values of PAH emissions for 2015 were estimated by CEIP using previously reported national emissions or expert estimates from available regional/global emission inventories [*Tista et al.,* 2017].

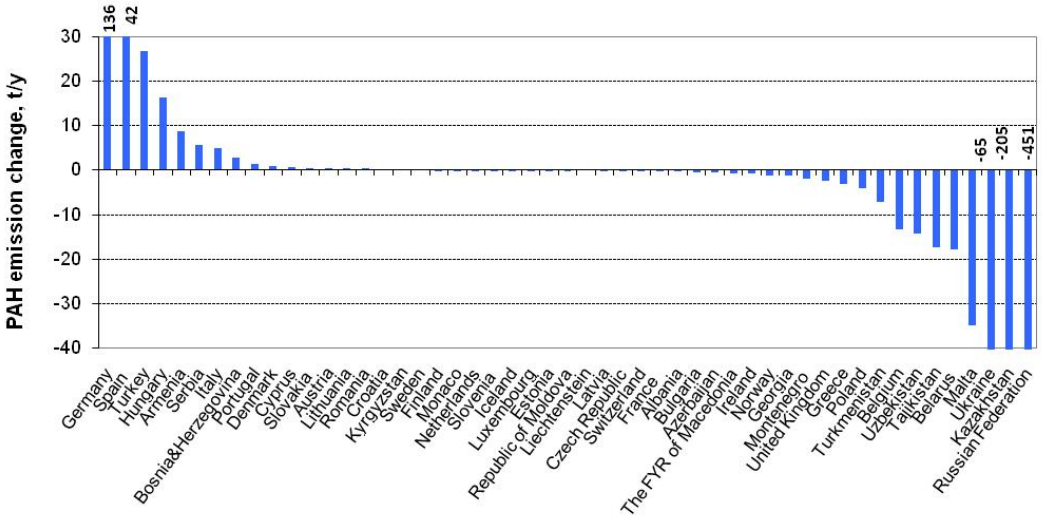


Fig. 2.5. *Changes of annual emissions of 4 PAHs in the EMEP countries from 2014 to 2015 (tonnes). Negative values denote decrease of emissions, and positive ones denote increase of emissions*

Completeness and accuracy of PAH emissions in the EMEP countries are of particular importance for model assessment of pollution in the EMEP region. Several aspects related to uncertainties of reported PAH emission data were analyzed by MSC-E and are discussed below. Model simulations requires detailed information on PAH emissions, including gridded data, speciation of emissions, and their distribution by sectors. Besides, estimates of uncertainties of national emission inventories are essential for the assessment of pollution.

For the majority of the EMEP countries data on speciated PAH emissions are presented in their national emission inventories. At the same time, data, reported by some of the countries, are characterized by inconsistencies between the total emission of 4 PAHs and emissions of individual PAHs, or do not contain speciation, which leads to considerable uncertainties in modelling results. In the inventories of Germany, Portugal, and Serbia the sum of speciated PAH emissions does not match the total national PAH emissions. Austria, Spain, Finland, and Italy provided information on total emission of 4 PAHs without speciation. For these countries expert estimates of splitting of total emissions into individual PAHs were made by CEIP [*Tista et al*., 2017]. In case of Germany reported total national emission of 4 PAHs (163 tonnes) was split into emissions of B(a)P, B(b)F, B(k)F, and IP using the average speciation of 4 PAHs in the inventories of other EMEP countries. Comparing to the level of PAH emissions, used for model simulations for the previous year 2014 [*Gusev et al*., 2016], equal to 27 tonnes, expert estimate for 2015 is significantly higher. Impact of these changes on model predictions of PAH pollution in Germany and surrounding countries will be discussed below in this chapter.

Another important source of information on PAH emissions is distribution by sectors. Sector-specific PAH emissions in the EU and non-EU countries and their temporal changes during the recent decade are shown in Fig. 2.6.

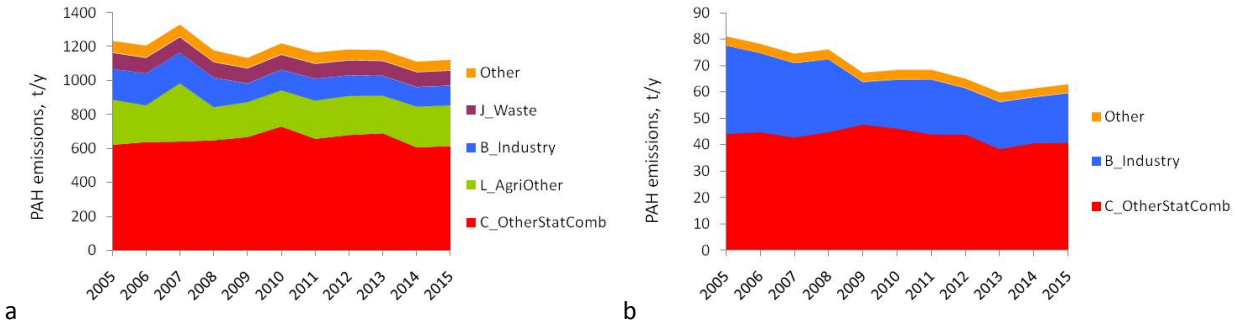


Fig. 2.6. *Long-term changes sector-specific PAH emissions in the EU28 (a) and non-EU (b) countries within the EMEP region from 2005 to 2015*

According to officially submitted emission inventories, in the majority of EMEP countries the largest contribution to PAH emissions was made by the Residential combustion sector (C_OtherStatComb). The issue of residential combustion, and in particular, wood combustion and B(a)P emissions, was discussed during the recent Steering Body meeting in 2016 at the specific session. It was emphasized that evaluation of emissions from this source category is currently subject of substantial uncertainties.

Along with residential combustion, burning of agricultural residues and wastes can also have significant contribution to PAH pollution. A number of countries, namely, Spain, Portugal, and Cyprus, reported comparatively high estimates of PAH emissions from these sectors, noticeably contributing to level of emissions in the EU countries (Fig. 2.6a). At the same time, inventories of other countries provided significantly lower emissions from biomass burning or did not estimate these emissions (Fig. 2.6b). Thus, differences in the evaluation of emissions from biomass burning or absence of their estimates may lead to additional uncertainties in the results of model simulations. ***To improve evaluation of pollution in the EMEP countries, further work in cooperation with national expert, CEIP, and TFEIP is required to refine estimates of sector-specific PAH emissions as well as their speciation.***

2.3. PAH pollution levels in the EMEP region

Model simulations of PAH long-range transport and pollution levels in the EMEP region for 2015 were performed using anthropogenic emissions of the EMEP countries. Contribution of non-EMEP emission sources outside the EMEP domain was not taken into account due to its relatively small effect. At the same time, available inventories of global PAH emissions [e.g. *Zhang and Tao*, 2009; *Shen et al.,* 2016] can be applied for the evaluation of intercontinental transport and impact of distant emission sources on the pollution of the EMEP countries and remote areas within the EMEP domain. It is planned to carry out global-scale modelling of PAHs for generating boundary concentrations for the EMEP region and evaluation of source-receptor relationships accounting for non-EMEP emissions. Additional contribution to PAH pollution, at least for particular seasons or episodes, can also be expected from the natural emission sources (e.g. forest fires), estimates for which are currently not included into the national inventories of EMEP countries.

Annual mean modelled air concentrations and deposition fluxes of the sum of 4 PAHs within the EMEP region for 2015 are presented in Fig. 2.7. ***Relatively high annual mean air concentrations (> 1 ng/m^3)***

are estimated for countries in Central Europe, namely, in Poland, Germany, the Czech Republic, Slovakia, Slovenia, Romania, Montenegro, and Hungary (Fig. 2.7a). Areas of high concentrations are also indicated for Spain, Portugal, and northern Italy. At the same time, estimates of PAH pollution levels in France, UK, and Scandinavian countries are relatively small. Spatial distribution of modelled annual deposition fluxes of 4 PAHs has mostly the same pattern (Fig. 2.7b).

The largest level of PAH deposition to the marginal seas within the EMEP domain is estimated for the Black Sea (about 50 g/km^2/y) followed by the Baltic Sea (about 29 g/km^2/y), the North Sea (about 14 g/km^2/y), the Mediterranean Sea (about 16 g/km^2/y), and the Caspian Sea (about 8 g/km^2/y).

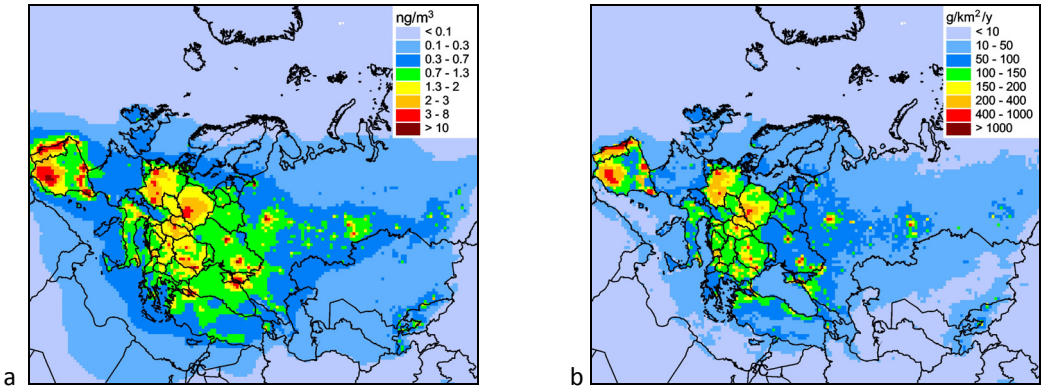


Fig. 2.7. *Spatial distribution of modelled annual mean air concentrations, ng/m^3 (a) and deposition fluxes, g/km^2/y (b) of 4 PAHs in the EMEP domain for 2015*

To evaluate inter-annual changes in PAH pollution levels from 2014 to 2015, modelling results for 2015 were compared with the results of the assessment, carried out for 2014 [*Gusev et al*., 2016]. Relative changes of annual mean PAH air concentrations in the EMEP countries from 2014 to 2015 are displayed in Fig. 2.8. Changes of pollution between these two years are explained by differences in emission data used in model simulations and by inter-annual variability of meteorological conditions. The effect of changes in meteorological conditions between 2014 to 2015 on PAH air concentrations is evaluated using the comparison of model simulations results, performed for 2014 and 2015 with corresponding meteorological input data and the same emissions for 2014. The figure illustrates the influence of inter-annual variability of precipitation amount, prevailing atmospheric flows, and temperature on PAH pollution levels.

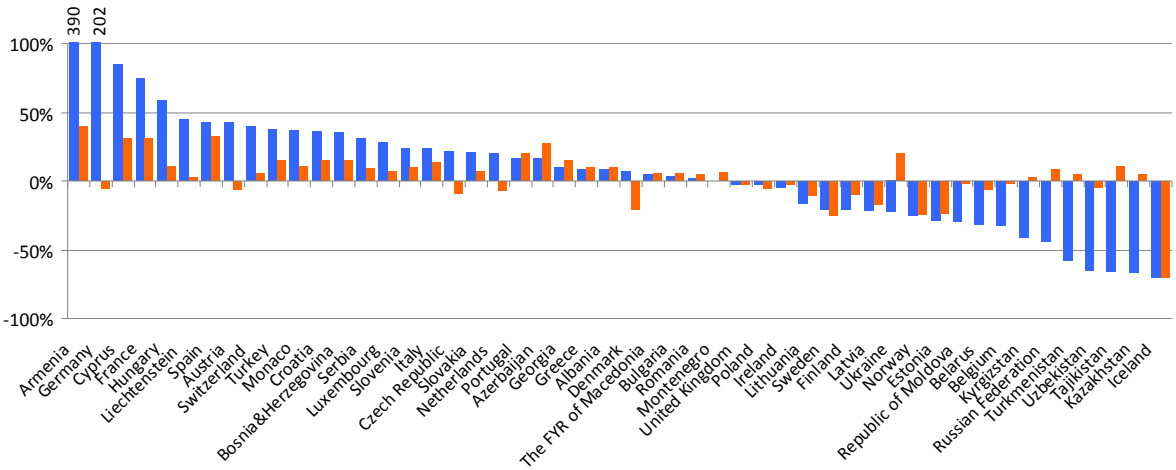


Fig. 2.8. *Changes of annual mean modelled PAH air concentrations in the EMEP countries between 2014 and 2015 (defined as (C_{2015}-C_{2014})/C_{2014}) due to changes of emissions and meteorological conditions (blue bars), and due to changes of meteorological conditions only (red bars). Negative values denote decrease, and positive ones denote increase*

In most cases changes in annual mean modelled air concentrations for 2014 and 2015 are caused by the differences between the estimates of national emissions for these two years. ***The largest relative decrease of concentrations is seen for the EECCA countries, namely, Kazakhstan, Tajikistan, Uzbekistan, Turkmenistan, and the Russian Federation (about -60%), which is explained by lower PAH emissions for 2015 comparing to 2014.*** In case of Iceland lower concentrations in 2015 are caused by changing of meteorological conditions. ***Increased PAH concentrations in 2015 comparing to 2014 are estimated for Armenia, Germany, Cyprus, and Hungary.*** This is explained by the higher values of national emissions used in model simulations for 2015. Higher level of PAH air concentrations in France, estimated for 2015, is mostly caused by higher emission of Germany comparing to that applied in model simulation for 2014.

2.4. Transboundary transport of pollution in 2015

Transboundary transport of pollution and source apportionment of annual deposition fluxes for 2015 were evaluated for the selected 4 PAHs. ***The most significant contribution to the sum of deposition of 4 PAHs is made by B[b]F (31%) followed by B(a)P (30%), B(k)F (23%), and IP (16%).*** The fraction of deposition fluxes of particular PAHs in total deposition of 4 PAHs depends on both their contributions to total emissions and difference in physical-chemical properties, governing their transport in and removal from the atmosphere. Annual total deposition fluxes over EMEP countries, estimated for 2015 with splitting into particular PAHs, are illustrated in Fig. 2.9.

Relatively high PAH deposition fluxes in the EMEP countries, exceeding 200 g/km^2/y, are estimated for Portugal and Montenegro. Significant values of deposition fluxes are noted for Spain, Slovakia, and Germany. At the same time, model estimates of PAH deposition fluxes in the EECCA countries are comparatively low.

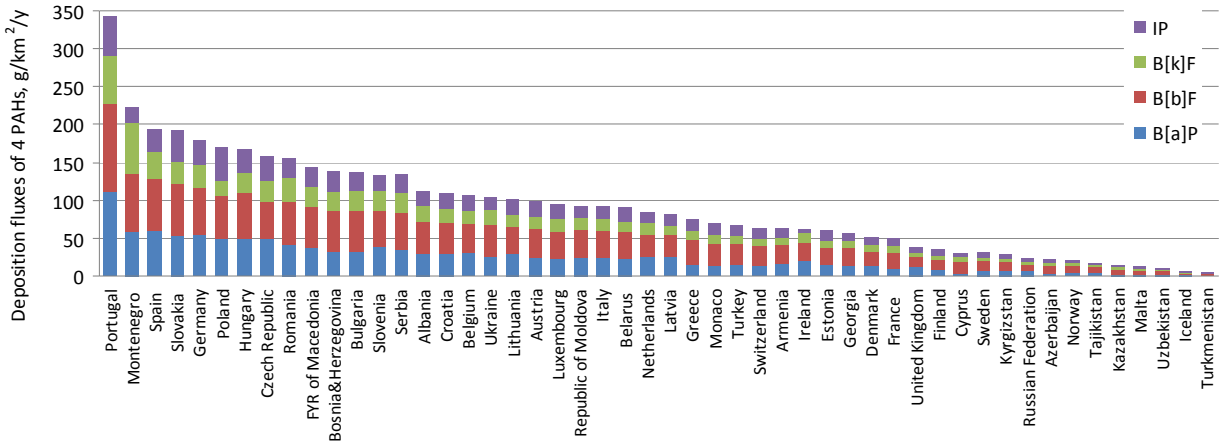


Fig. 2.9. *Annual deposition fluxes of 4 PAHs (B(a)P, B(b)F, B(k)F, and IP) in the EMEP countries calculated for 2015, g/km^2/y*

Assessment of PAH distribution in the EMEP domain indicates importance of the long-range transport of pollution between the countries. ***Source-apportionment of PAH deposition shows that for 70% of countries the contribution of emission sources, located beyond their territories, exceeds the contribution of their own national emissions*** (Fig. 2.10).

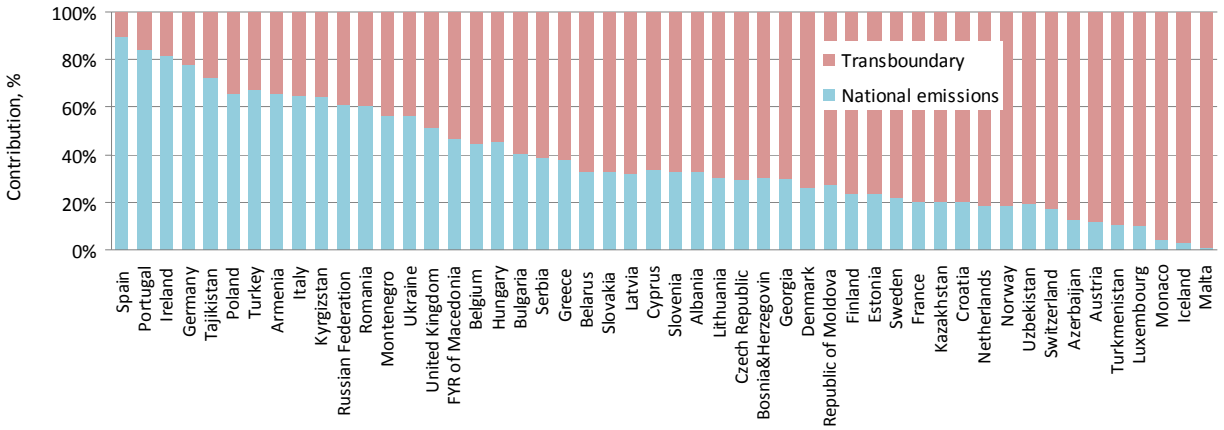


Fig. 2.10. *Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of 4 PAHs over the EMEP countries in the 2015*

Long-range transport of pollution from the emissions of a given country to other countries can also be characterised by the ratio between the value of deposition to the territory of the country and to the areas outside its boundaries, as shown in Fig. 2.11.

As follows from the Figure, ***as much as 37 countries are characterized by the exceedance of fraction of PAHs, deposited to other EMEP countries, comparing to the fraction of PAHs, deposited to the country itself.***

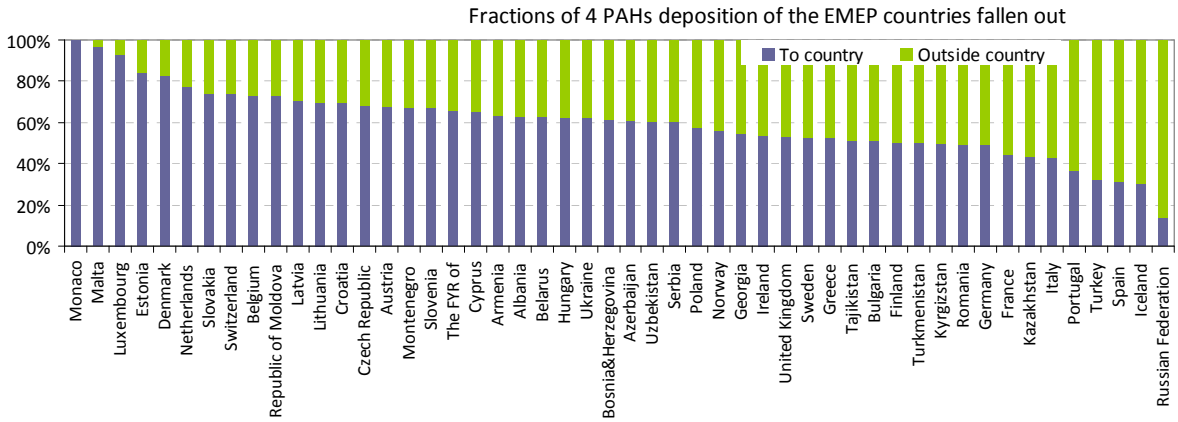


Fig. 2.11. *Fractions of PAH deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in the 2015*

According to modelling results, almost 60% of total annual deposition of 4 PAHs over EMEP countries in 2015 was originated from the emission sources of five countries, namely, the Russian Federation, Spain, Germany, Ukraine, and Poland.

2.5. Comparison of modelling results with measurements

Evaluation of model predictions of PAH pollution levels in 2015 was made using comparison of modelled air concentrations of 4 PAHs with measurements of the EMEP monitoring network and results of national monitoring, collected in the EU EEA/AirBase.

Monitoring of PAHs in 2015 was carried out at 27 EMEP monitoring sites located in Belgium, Czech Republic, Estonia, Finland, France, Latvia, Netherlands, Norway, Poland, Portugal, Slovenia, Spain, Sweden, and UK. Simultaneous measurements of 4 PAHs in 2015 were performed out at 14 monitoring sites. Comparison of modelled PAH air concentrations with measurements are presented in Fig. 2.12a. The model satisfactory described observed spatial distribution of PAH air concentrations with correlation 0.82. For 60% of selected monitoring sites the difference between measured and modelled concentrations is within a factor of 2. At the same time, for several monitoring sites, located in France, Norway, and Sweden, the model overestimates observed concentrations.

Modelled air concentrations of B(a)P, B(b)F, B(k)F, and IP against corresponding measurements of EMEP monitoring sites are shown in Fig. 2.12b and Fig. 2.13.

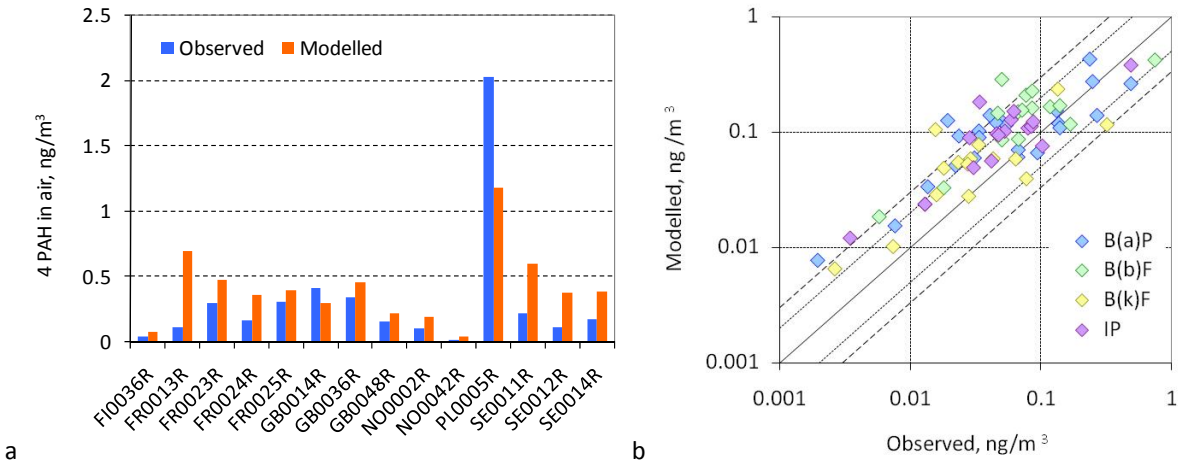


Fig. 2.12. *Comparison of modelled and observed 4 PAH air concentrations for 2015 (a) and scatter plot of modelled and observed B(a)P, B(b)F, B(k)F, and IP air concentrations for 2015. Dashed lines in right diagram denote the areas of agreement between the modelled and measured values within a factor of 2 and 3*

Comparison of model predictions with measurements of B(a)P, B(b)F, B(k)F, and IP indicates that the model is able to capture observed spatial variations of individual PAH air concentrations (correlation coefficient varies from 0.54 for B(k)F to 0.89 for IP). More than a half of model predictions are within a factor of 2 and about 80% of them are within a factor of 3 in comparison with measurements.

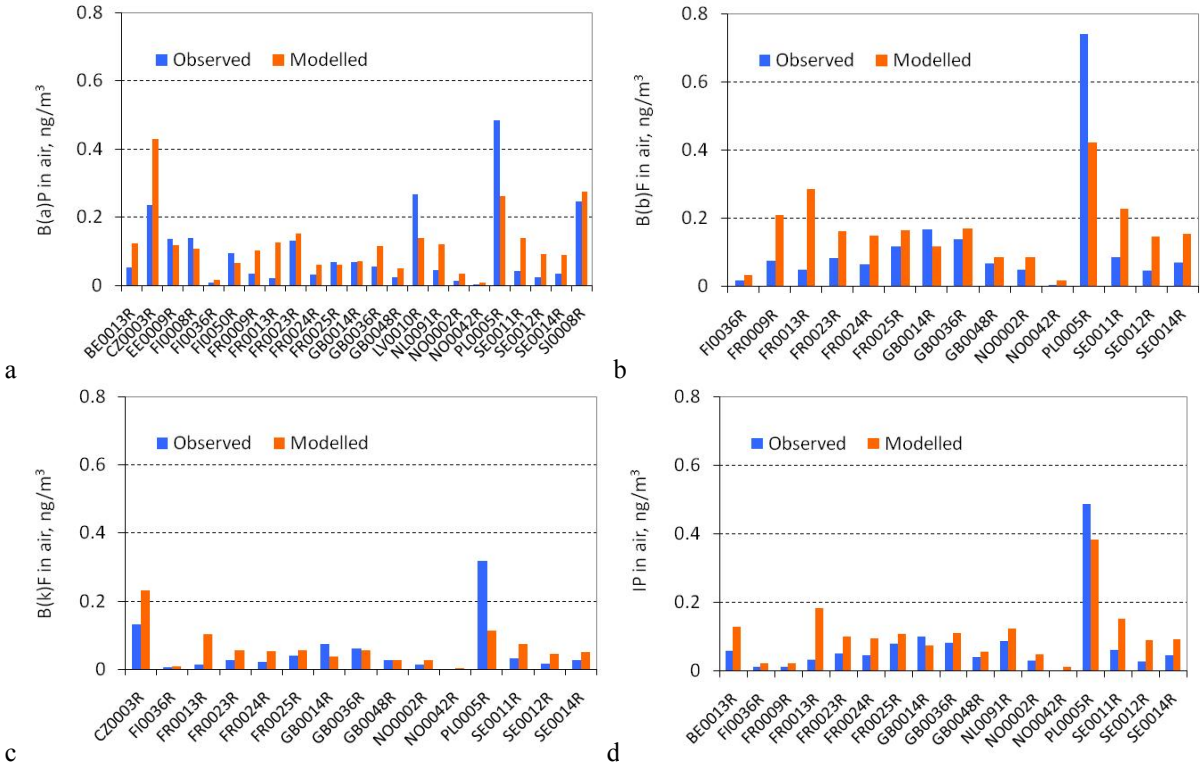


Fig. 2.13. *Comparison of modelled B(a)P (a), B(b)F (b), B(k)F (c) and IP (d) air concentrations with measurements of EMEP monitoring sites for 2015*

Similar to model estimates of the sum of 4 PAHs, modelled concentrations of B(a)P, B(b)F, B(k)F, and IP overpredict air concentrations, measured at monitoring sites FR0013R, SE0011R, SE0012R, SE0014R, BE0013R, and NL0091R.

The largest discrepancy, exceeding a factor of 3, was found for the monitoring site FR0013R, located in southern part of France. According to modelling results, air pollution levels at the location of this site are largely influenced by the emission sources of France and Spain. Model predictions for previous year (2014) fitted well with observed air concentrations of PAHs. ***Significant overestimation of concentrations, measured at FR0013R in 2015, may be explained by the changes of reported PAH emissions of Spain in new and previous national inventories for 2015 and 2014, respectively.*** The new inventory of PAH releases in Spain provided higher emissions for 2015 comparing to the level of emissions for 2014 by 16%.

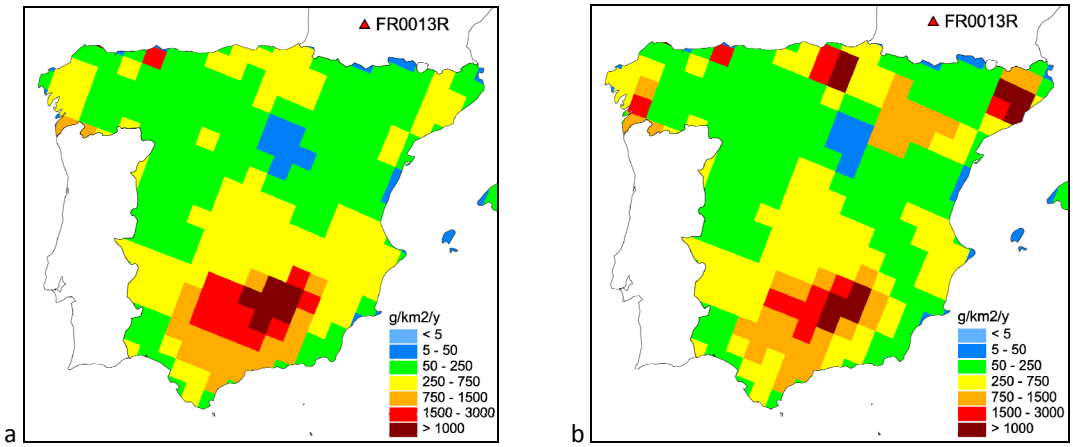


Fig. 2.14. *Spatial distribution of PAH emissions in Spain in 2014 (a) and in 2015 (b), g/km^2/y. Location of the monitoring site FR0013R is marked by the red triangle*

Besides, the changes in the distribution of emission by sectors led to allocation of higher PAH emissions in northern part of the country in 2015 comparing to the spatial distribution for 2014 (Fig. 2.14). These changes resulted in more significant contribution of Spanish emission sources to the pollution of southern part of France and thus to higher model predictions. To improve accuracy of model predictions for this part of EMEP region, refinement of emission as well as fine resolution modelling and more detailed analysis of PAH pollution levels is of importance. This can be achieved in the framework of ongoing case studies of PAH pollution in Spain and France.

Measurements of PAH concentrations, performed at monitoring sites SE0011R, SE0012R, and SE0014R in 2015, were overestimated by the model about a factor of 3 on average. Similar level of overestimation was obtained also for the measurements of B(a)P air concentrations of BE0013R and NL0091R. For all these sites measured pollution levels decreased from 2014 to 2015 by 20-60%. At the same time, model simulations did not reproduce this decline, providing less significant changes, which led to higher difference of modelled and measured values comparing to the results for 2014 [*Gusev et al.,* 2016].

Increased deviation of model predictions from the concentrations observed in 2015 can be attributed to several reasons including uncertainties of total emissions, estimated for these two years, and their spatial distribution. Substantial contribution to this can be made by the difference in Germany emissions used in model simulation for 2014 and 2015. Officially reported data on PAH emissions in Germany are subject of noticeable uncertainties. As it was mentioned in previous EMEP Status report

[*Gusev et al.,* 2016], the information on emissions of B(b)F, B(k)F, and IP in the inventory of Germany was likely underestimated. Similar situation takes place in the inventory for 2015. Emissions of B(b)F, B(k)F, and IP from some of the source categories (e.g. Public power, Industry, Residential combustion) are not estimated. Besides, the sum of the emissions of 4 individual PAHs (29.4 tonnes) is considerably lower than the total reported emissions of 4 PAHs (163 tonnes).

Taking this into account expert estimate of B(a)P, B(b)F, B(k)F, and IP emissions in Germany for 2015 was made by CEIP [*Tista et al.*, 2017]. In particular, total reported PAH emission for 2015 (163 tonnes) was split into individual PAHs (50, 56, 25, and 33 tonnes, respectively) following their average distribution in the emissions of other EMEP countries. This resulted in significant difference between emissions of Germany, used for the model simulation of pollution for 2014 and 2015, for instance, B(a)P emission changed from 25 tonnes in 2014 to 50 tonnes in 2015, and total emission of 4 PAHs changed from 28 tonnes to 163 tonnes. Available uncertainties in estimates of PAH emissions are also discussed in recent inventory information report of Germany [*Strogies et al.*, 2017]. In particular, it is mentioned that estimated total national emission of 4 PAHs of Germany is likely overestimated.

Higher levels of national PAH emissions for 2015 caused more significant contribution of Germany in model estimates of pollution levels for surrounding countries. These changes are exemplified by source attribution of the model estimates of B(a)P air concentrations for the monitoring sites NL0091R, BE0013R, and SE0014R, obtained in model simulations for 2014, for 2015 with emissions of 2014, and for 2015 with emissions of 2015, respectively (Fig. 2.15). Model simulations for 2015 with the emissions of 2014 show somewhat lower contributions of pollution transport from Germany to the locations of these sites, comparing to modelling results for 2014. Decrease of contributions can be attributed to inter-annual changes of meteorological conditions. In case of model simulations with estimates of B(a)P emissions for 2015 these contributions are increased almost twice.

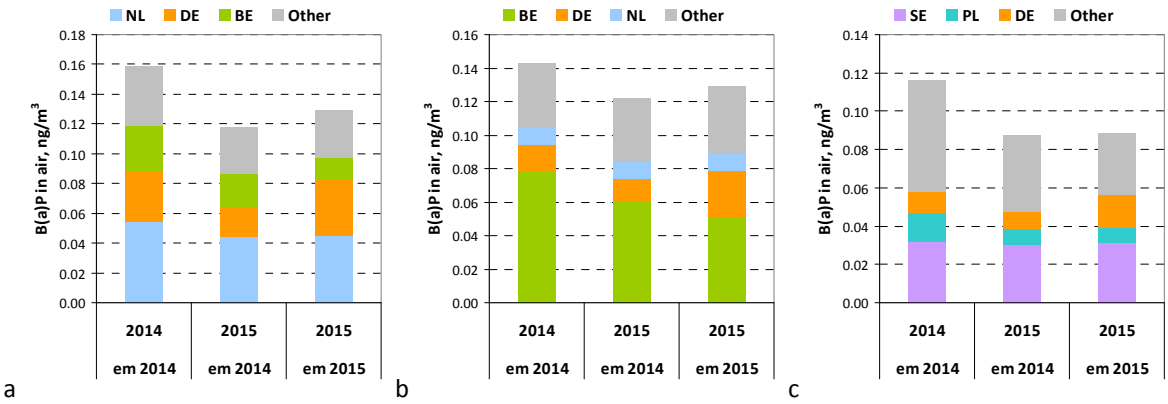


Fig. 2.15. *Source attribution of model predictions of B(a)P air concentrations for monitoring sites NL0091R (a), BE0013R (b), and SE0014R (c) obtained in the model simulations for 2014 with emissions of 2014, the model simulations for 2015 with emissions of 2014, and the model simulations for 2015 with emissions of 2015*

Important factor affecting quality of modelling results is the accuracy of spatial distribution of emissions. According to officially reported gridded data, significant B(a)P emissions in Germany took place in the western and southern parts of the country (Fig. 2.16a).

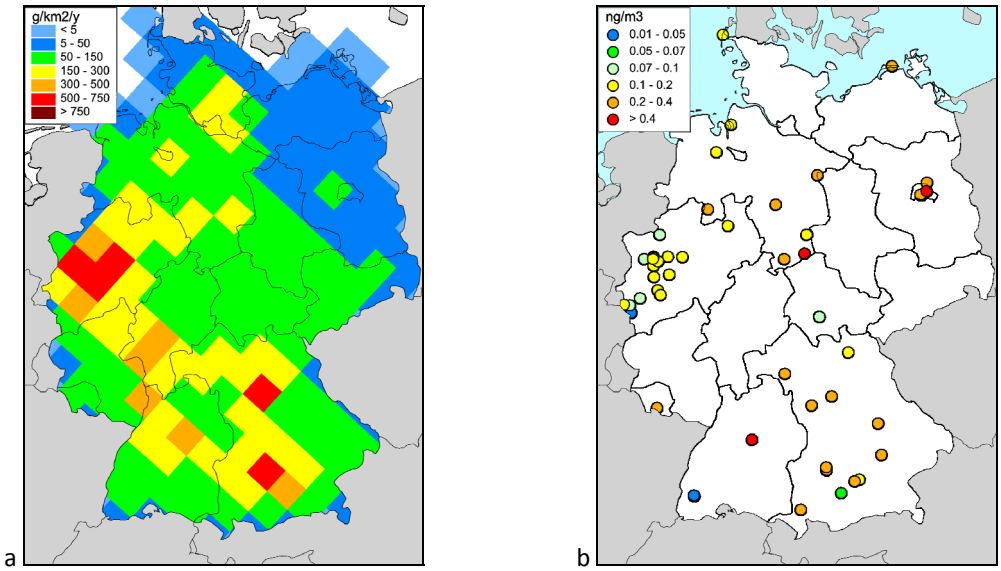


Fig. 2.16. *Spatial distribution of B(a)P emissions for 2015 (a) and annual mean observed B(a)P air concentrations at national monitoring sites of Germany in 2015 (EEA/AirBase)*

At the same time spatial distribution of observed B(a)P air concentrations for 2015, available in the EEA/AirBase, differs from that of emissions. For instance, western part of the country is characterized by relatively low B(a)P concentrations, whereas in the central and eastern parts observed B(a)P concentrations are relatively higher (Fig. 2.16b). Thus spatial distribution of emissions may also contribute to the overestimation of PAH concentrations obtained in Belgium and Netherlands.

Level of uncertainties in the officially reported emission data can also be analyzed using national data on observed PAH concentrations (EEA/AirBase). Comparative analysis of monitoring data and emissions can be used for the indication of potential uncertainties in the reported national inventories leading to underestimation or overestimation of PAH releases in EMEP countries and thus may help to further improve quality of pollution assessment.

Averaged annual mean B(a)P air concentrations and annual mean B(a)P emission fluxes in the selected EMEP countries for 2015 are shown in Figs. 2.17a and 2.17b. ***The most noticeable contradictions between the mean emission fluxes and observed B(a)P concentrations can be noted for Poland, Germany, and Belgium.*** Particularly, average level of observed B(a)P concentrations in Poland is substantially higher comparing to other countries, performing monitoring of B(a)P concentrations. At the same time, average B(a)P emission flux does not differ much from the emissions of neighbouring countries indicating possible underestimation of emissions. On the opposite, significant B(a)P emission of Germany is not consistent with monitoring data that indicate relatively low level of B(a)P air concentrations in the country. Similar situation can also be noted for B(a)P air concentrations and emissions of Belgium.

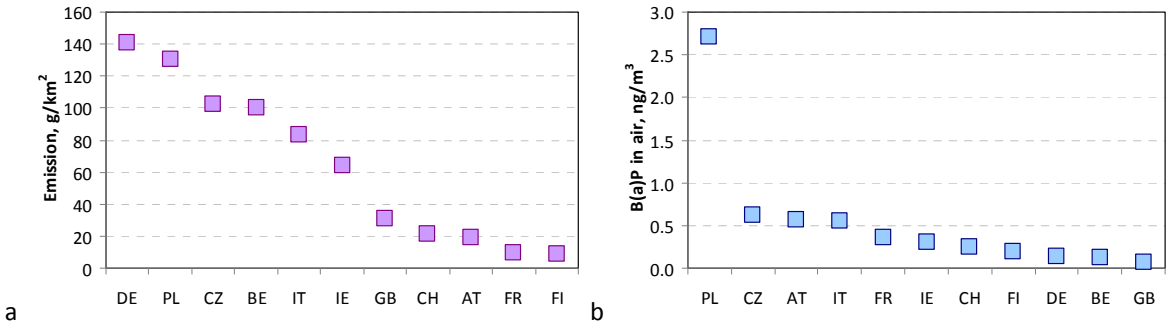


Fig. 2.17. *Mean annual emission fluxes (a) and observed air concentrations (b) of B(a)P in the selected EMEP countries for 2015*

Thus further refinement of PAH emission data in the EMEP countries is important for the reduction of uncertainties of pollution assessment.

3. TRANSITION OF GLEMOS MODELLING SYSTEM TO THE NEW EMEP GRID

Global EMEP Multi-Media Modelling System (GLEMOS) is an up-to-date chemistry transport model used by MSC-E for the operational modelling of transboundary pollution of the EMEP countries by heavy metals and POPs. In accordance with decisions of the Executive Body for CLRTAP [ECE/EB.AIR/113/Add.1] MSC-E has initiated the work on the transition of operational modelling of HMs and POPs to the new EMEP grid. The new grid system is defined in regular geographic (latitude-longitude) projection of the Earth surface and has finer spatial resolution comparing to the former EMEP grid in polar stereographic projection. Results of preparatory work for the transition to the new grid and pilot model simulations for mercury were discussed in previous reports of the Centre [*Ilyin et al.*, 2014; 2016].

This year MSC-E has continued this activity aiming to complete the transition process. The new developments include extension of the list of pollutants, simulated on the new grid, preparation of additional input data for these pollutants, and thorough testing of updated modelling system. Besides, the modelling system underwent considerable structural revision, aimed to prepare it for public distribution as open-source software. Current progress in updating and further development of GLEMOS modelling system for POPs is discussed below. The outcome of activities, related to the general aspects of further GLEMOS model development, as well as to the refinement of modelling approach for heavy metals, is described in the EMEP Status Report for heavy metals [*Ilyin et al.*, 2017].

3.1. Model update and further development

Adaptation of GLEMOS modules for simulations on the new EMEP grid

The GLEMOS modelling system consists of a number of functional modules controlling the data flows and describing various physical and chemical processes important for HMs and POPs. They include the input and output systems, general modules, describing dispersion processes in the environmental media (atmosphere, ocean, soil, vegetation), and modules, representing behaviour and properties of particular pollutants. The transition of the model to the new EMEP grid implies the adaptation of the modules, designed for general purposes, as well as modules specific for particular pollutants. Besides it includes preparation of required input data for the new modelling domain. The work in this direction was started with generation of various input data (meteorological fields, geophysical information, atmospheric concentrations of chemical reactants etc.) for the new EMEP grid [*Ilyin et al.*, 2014]. General modules of the model, describing atmospheric transport and removal processes, were adapted to the new grid and tested using pilot simulation of mercury pollution [*Ilyin et al.*, 2016].

This year input and output system modules as well as various pollutant specific modules have been updated and tested in application to the modelling over the new EMEP grid. In particular, the input of gridded emission data including information on vertical structure, intra-annual variations, and distribution by sectors was refined. A new system of preparation and input of the initial and boundary conditions for regional modelling has been developed based on the one-way nesting of the global-scale simulations. The output system of the GLEMOS model was further updated to provide flexibility in the selection of various output model parameters, their temporal resolution, and output formats (e.g. text, NetCDF).

With respect to the modules related to the processes, governing POP fate in the atmosphere, unification of parameterizations of deposition processes with those, applied for HMs, was carried out. A number of modifications were introduced to the parameterizations of POP gas-particle partitioning and degradation processes following the results of recent investigations available in literature.

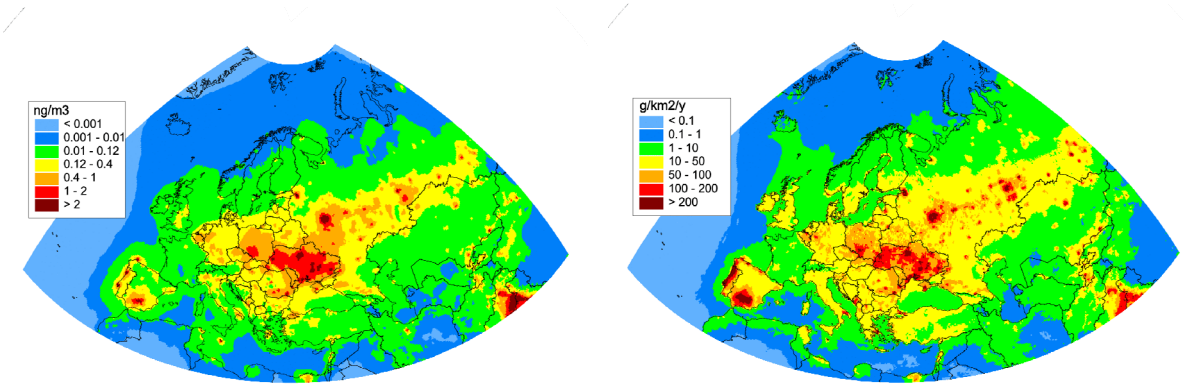
Implementation of POP modelling on the new grid requires further adaptation of the modules describing pollutants behaviour in environmental media other than the atmosphere (ocean, soil and vegetation) and air-surface pollutants exchange. The work on the updating of these pollutant specific modules is initiated and will be continued on subsequent stages of GLEMOS model development.

Information of POP deposition to various types of land cover is of importance for the evaluation of harmful effects to human health and wildlife. Model performance to evaluate ecosystem-specific deposition using the new EMEP grid was tested this year for HMs [*Ilyin et al.*, 2017]. Similar analysis for selected POPs will be performed in course of further stage of GLEMOS model development.

3.2. Pilot simulations of B(a)P on the new EMEP grid

Transition of regional-scale modelling of POPs to the new EMEP grid system involves both modifications of the GLEMOS model and preparation of necessary input data including meteorological and geophysical information. Emission data for the new grid were constructed on the basis of official B(a)P emissions provided by CEIP. It should be noted that official emissions in the new grid for 2014 were reported only by limited number of countries. Therefore, gridded B(a)P emissions for the new domain were prepared using interpolation of emissions, defined in the old 50×50 km^2 grid, taking into account mass conservation. Gridded emissions for non-EMEP sources within the new and old EMEP domains were constructed using available expert estimates of global B(a)P emissions [*Zhang and Tao*, 2009].

To evaluate performance of the GLEMOS model on the new grid with increased spatial resolution, pilot simulations of B(a)P atmospheric transport were performed for 2014 with spatial resolution 0.2°×0.2°. This level of resolution was selected for testing purposes as intermediate one between the resolutions of old and new EMEP grid systems. Spatial distribution of modelled annual mean B(a)P air concentrations and deposition fluxes for 2014 is shown in Fig. 3.1a,b, respectively.



a b

Fig. 3.1. *Annual mean air concentration (a) and deposition fluxes (b) of B(a)P in 2014 simulated over the new EMEP grid with spatial resolution 0.2°x0.2°*

3.3. Evaluation of modelling results against measurements

To analyze the effect of transition to the new grid, modelling results, generated by the updated model for the new EMEP domain, were compared with the results of previous model version for the old EMEP domain (in polar stereographic projection). Model simulations were carried out using similar levels of B(a)P emissions. The Figs. 3.2a and b present spatial distributions of modelled annual mean B(a)P air concentrations in 2014, calculated for the new and old EMEP domains, respectively.

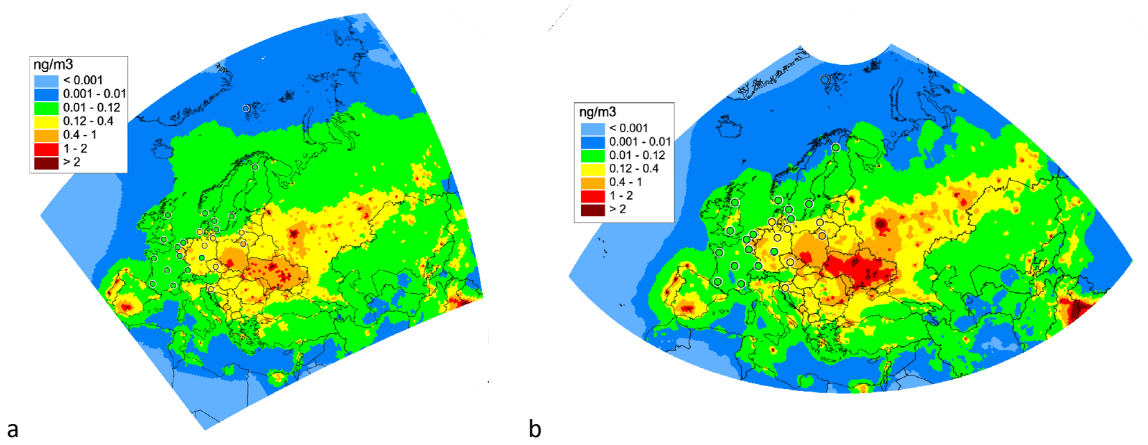


Fig. 3.2. *Annual mean air concentration of B(a)P in 2014 simulated over the old (a) and new (b) EMEP domains. Coloured circles show annual mean B(a)P air concentrations measured at the EMEP monitoring network*

In general, model simulations for both domains provided similar patterns of spatial distribution of B(a)P air pollution levels. In particular, elevated air concentrations, above 0.12 ng/m^3 and reaching 1 ng/m^3, were estimated for the countries in Central and Eastern Europe. Lower levels of concentrations, about 0.01 – 0.1 ng/m^3, were obtained for the countries in Western and Northern Europe.

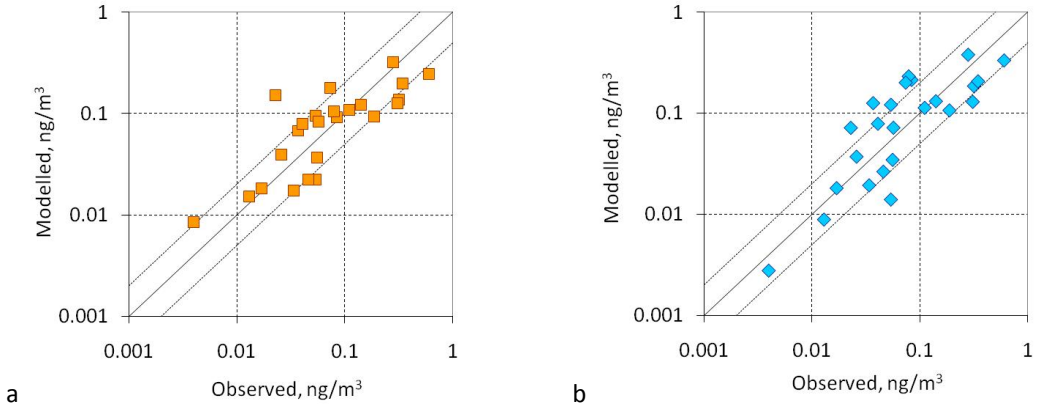


Fig. 3.3. *Comparison of annual mean B(a)P air concentrations simulated over old (a) and new (b) EMEP grid with EMEP measurements for 2014*

Both sets of modelling results were compared with annual mean B(a)P air concentrations, measured in 2014 at 24 EMEP monitoring sites in Belgium, the Czech Republic, Finland, France, the UK, Latvia, the Netherlands, Poland, Sweden, Slovenia and Germany (Figs. 3.3a,b). Statistical parameters of model-to-measurements comparison are presented in Table 3.1.

Table 3.1. *Statistics of the model-to-measurements comparison for B(a)P simulations on the new and old EMEP grids*

	Modelled B(a)P air concentrations	
	New EMEP grid	Old EMEP grid
Correlation with measurements	0.73	0.73
Relative Bias	-5%	-20%
Factor 2	63%	67%

It can be seen that the ***two model runs showed reasonable reproduction of observed B(a)P pollution levels. In particular, there is significant spatial correlation, about 0.7, between the modelled and observed concentrations. For the substantial part of model predictions the difference between the modelled and measured values is within a factor of two.*** At the same time, in both cases the model tended to underestimate measurements of B(a)P content in air. The value of relative bias is smaller for the new EMEP grid (about -5%) comparing to the results for the old EMEP grid (about -20%).

To analyze the differences between the modelled data generated by the updated and previous versions of the model, more detailed statistical analysis of obtained results was carried out. For this purpose modelled and measured B(a)P concentrations were converted to natural logarithms, taking into account log-normal distribution of air concentration data [e.g. *Venier et al.*, 2012]. Following this transformation the level of agreement between measured and calculated data was evaluated with the help of regression analysis. The scatter plot in Fig. 3.4a demonstrates the regression relationships for the two sets of model estimates, obtained by the use of updated and previous versions of the model, applied for the new and old EMEP domains, respectively. The Fig. 3.4b illustrates the ratios between the modelled and observed concentrations in accordance with regression relationships, shown in Fig. 3.4a. *It can be seen that* ***updated version of the model demonstrates better fit to measured B(a)P air concentrations*** (Fig. 3.4b).

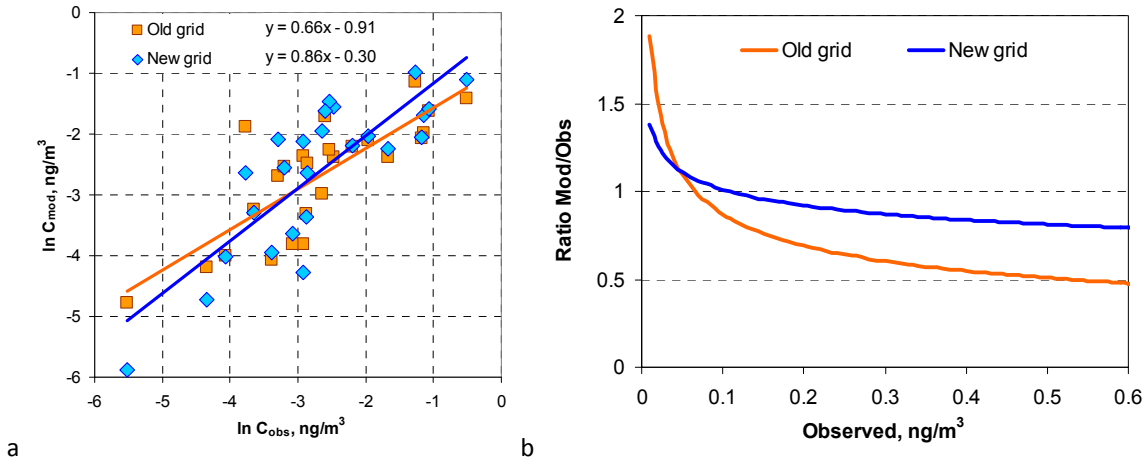


Fig. 3.4. *Scatter plot of the comparison of annual mean B(a)P air concentrations, simulated for the old grid by the previous model version and for the new grid by the updated model version, with EMEP measurements for 2014 (a), and ratios between the modelled and observed concentrations based on regression relationships, shown on the left plot (b)*

In particular, it tends to provide lower estimates for the locations with low measured concentrations (less than, approximately, 0.06 ng/m^3) and higher estimates for the locations with elevated pollution levels, comparing to the previous model version. This results in lower values of ratio between modelled and observed air concentrations and thus in better agreement with measurements. The reason of the difference between the modelling results for the new and old EMEP grids can be explained both by the updates in model parameterizations of partitioning and removal processes in the model as well as by the change of the grid system and increase of spatial resolution.

3.4. Analysis of factors affecting B(a)P pollution levels

In order to improve the quality of pollution assessment and agreement of modelling results with measurements, MSC-E continued the work on the refinement of modelling approach for POPs. In current year this activity included analysis of factors, influencing pollution transport in the atmosphere, and further updating of pollutant specific model parameterizations as well as input data for modelling. Particular attention was paid to the most important processes, controlling the long-range transport and fate in the atmosphere on the example of B(a)P, namely, interaction of B(a)P with aerosol particles, degradation, and dry and wet deposition.

Gas-particle partitioning of B(a)P in the atmosphere

According to present understanding, available in scientific literature, partitioning of B(a)P between its gaseous and particle bound phases in the atmosphere comprises a combination of absorption into organic matter and adsorption to soot in the aerosol particles [*Keyte et al*., 2013]. The process of partitioning depends on several factors, e.g. meteorological parameters, chemical composition of aerosol particles, and content of atmospheric reactants, as well as their spatial and temporal variations. To explore relationship between temporal variability of observed concentrations of B(a)P and concentrations of elemental carbon (EC), organic carbon (OC), and ozone, multiple regression analysis of measurements, reported by EMEP monitoring stations, was carried out. Specifically, monthly mean concentrations of OC, EC, ozone, and B(a)P, measured in parallel in 2013 and 2014 at the sites in Germany (DE2, DE3, DE8), France (FR9, FR23, FR25), and Poland (PL5), were used.

Results of the analysis indicates that observed seasonal variations of B(a)P are to significant extent connected with temporal variations of OC, EC, and ozone concentrations. For the majority of sites the coefficient of determination (R^2) is about 0.6 and higher (Fig. 3.5). This shows that for these sites temporal variations of B(a)P concentrations can be explained by the variability of OC, EC, and ozone, influencing partitioning and degradation of B(a)P in the atmosphere. Examples of multiple regression results versus measured B(a)P in air for particular monitoring sites are displayed in Fig. 3.6.

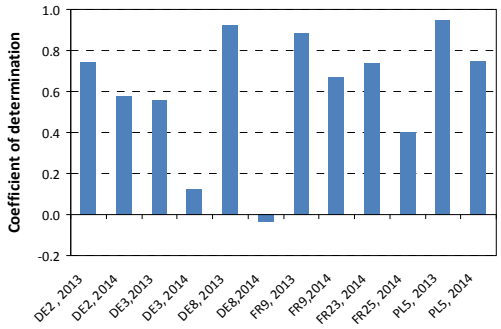


Fig. 3.5. *Values of coefficient of determination (R^2) obtained from multiple regression of measured B(a)P concentrations versus measured OC, EC, and ozone concentrations reported by EMEP monitoring sites in 2013 and 2014*

At the same time, in three cases, namely, for measurements of DE3 in 2014, DE8 in 2014, and FR25 in 2014, low R^2 values were obtained. For example, variability of measured B(a)P at DE3 and DE8 for the second part of the year 2014 was poorly captured by the multiple regression.

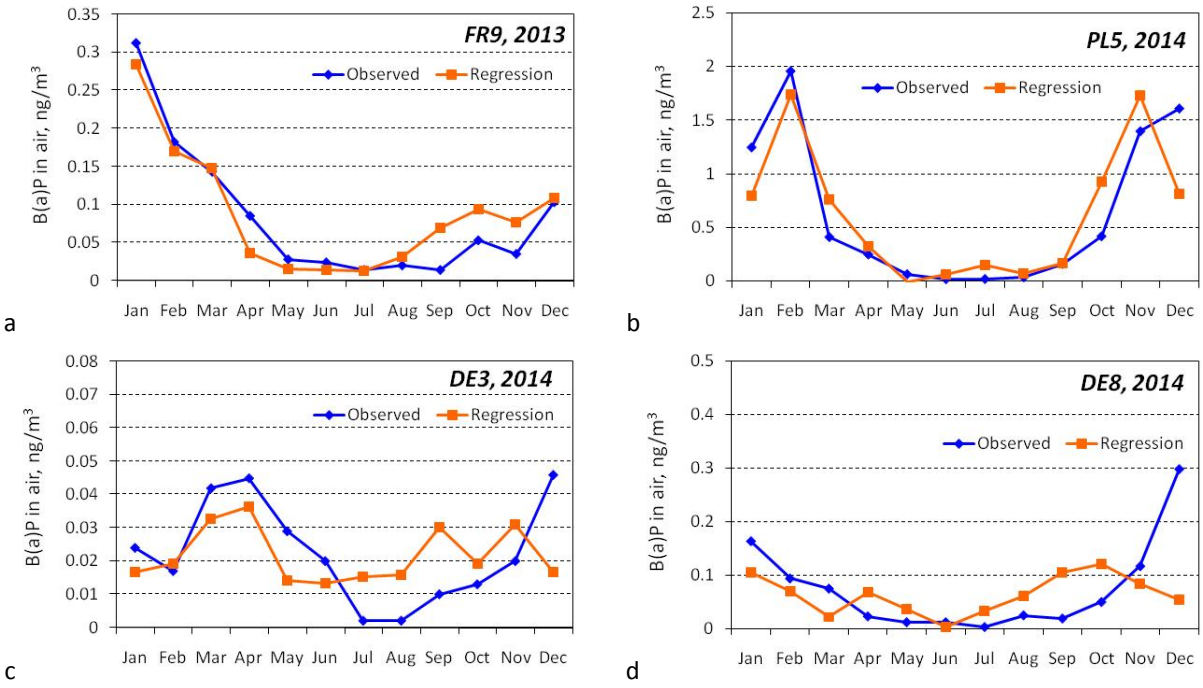


Fig. 3.6. *Monthly mean observed B(a)P concentrations (blue lines), reported by EMEP monitoring sites FR9, PL5, DE3, and DE8 in 2013 and 2014, and results of multiple regression (red lines) of B(a)P concentrations versus observed OC, EC, and ozone*

The reason of this disagreement can be explained by the effect of additional parameters, not taken into account in the regression model (e.g. meteorological parameters). Thus further analysis of factors, influencing variability of measured concentrations at these sites, needs to be carried out in co-operation with national experts.

PAHs and particulate matter are co-emitted to the atmosphere as a result of various combustion processes. Following the emissions, PAHs undergo partitioning to the aerosol particles in the atmosphere. Thus their atmospheric transport can be closely connected with the dispersion of particulate matter. Relationship between the spatial distributions of B(a)P and PM10 is analyzed using annual mean observed air concentrations of these pollutants, collected in the EEA/AirBase. The scatter plot of B(a)P versus PM10, transformed to natural logarithms, is shown in Fig. 3.7, illustrating interrelation of their concentrations.

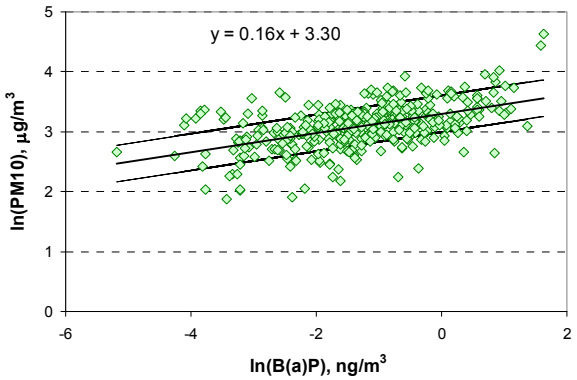
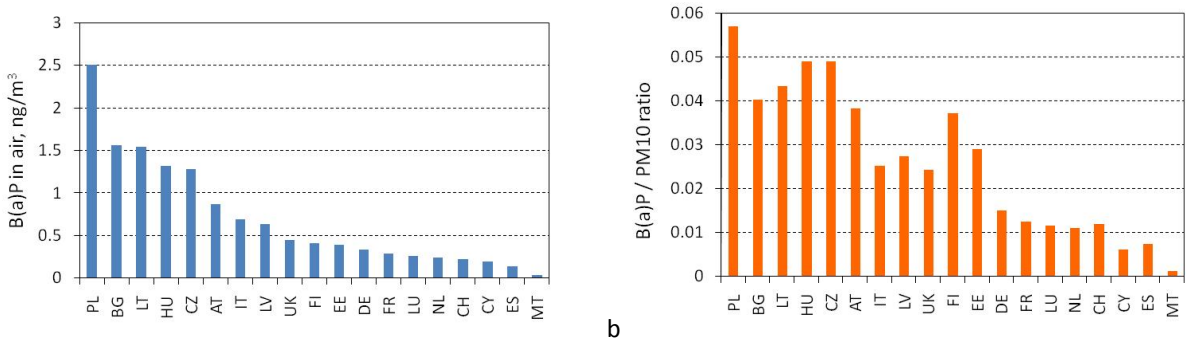


Fig. 3.7. *Scatter plot of annual mean B(a)P versus PM10 air concentrations measured at monitoring sites of European countries in 2013 (EEA/AirBase). Solid lines indicate linear regression and confidence interval*

Relationship between observed B(a)P and PM10 air concentrations can be further explored using averaged national measurements of European countries (Fig. 3.8). The figure shows higher ratios of B(a)P to PM10 concentrations for the countries with high averaged annual mean B(a)P concentrations, namely, Poland, Bulgaria, Lithuania, Hungary, and the Czech Republic. In contrast, low values of observed B(a)P air concentrations and low B(a)P/PM10 ratios can be seen, for example, in Switzerland, Cyprus, Spain, and Malta. Spatial variations of fraction of B(a)P in the particulate matter can be connected with variability of relative contribution of different source categories to the emissions of B(a)P and PM.



a b

Fig. 3.8. *Averaged annual mean B(a)P air concentrations (a) and ratio of B(a)P and PM10 concentrations (b) measured in the EMEP countries in 2013*

Similar analysis of measured B(a)P and PM air concentrations was carried in the study of *Lobscheid et al.* [2007], indicating relatively strong correlation between these two pollutants. Analysis of interactions between B(a)P and PM pollution levels is of importance for further refinement of modelling approach applied for the description of gas-particle partitioning of PAHs in the atmosphere. At subsequent stages of the work the study of relationships between measured B(a)P and PM10 concentrations will be continued to verify model predictions against measurements and improve their accuracy.

Degradation of B(a)P in the atmosphere

Degradation of B(a)P, sorbed on aerosol particles of various chemical composition, can substantially affect its long-range transport and levels of concentrations [*Keyte et al.*, 2013]. The work on the elaboration of model parameterization of this process, initiated at previous stages of the work [*Shatalov et al*., 2013, 2014], has been continued this year. In particular, estimates of B(a)P degradation rate constants, reported in scientific literature for different constituents of aerosol particles, namely OC, EC, and mineral fraction, were summarized for the refinement of model parameterization. Updated values of degradation rate constants for heterogenic reactions of B(a)P with ozone were applied in model simulations on the new EMEP grid. In addition, it is supposed to further update parameterization of degradation process with the inclusion of the dependence of degradation on the air humidity. To explore the sensitivity of modelling results to different estimates of degradation rate of particle-bound B(a)P, experimental modelling and analysis of the results is planned to be carried out.

Dry and wet deposition

B(a)P is present in the atmosphere mostly in the particulate phase. Following the release to the atmosphere it is partitioning to the aerosol particles mostly of ultrafine and accumulation modes (<1 μm) [*Keyte et al.*, 2013]. The rate of removal of particle-associated B(a)P from the atmosphere due to dry and wet deposition is dependent on the particle size. According to various studies of B(a)P particle size distribution in the atmosphere, the mass median diameter of particle-bound fraction of B(a)P can be in the range 0.1-0.8 μm (Table 3.2).

Table 3.2. *Values of mass median diameter for particle-bound B(a)P, reported in literature*

Location of the study	MMD (μm)	Reference
Boston, USA, June, 1994	0.41	*Allen et al.,* 1997
Guangzhou, China, January 2002, urban	0.35	*Bi et al*., 2005
Guangzhou, China, January 2002, rural	0.67	*Bi et al*., 2005
Guangzhou, China, January 2002	0.40 – 0.47	*Tang et al*., 2006
Pico Rivera, Los Angeles, USA, winter 1989	0.1	*Venkataraman and Friedlander*, 1994
Pico Rivera, Los Angeles, USA, summer 1989	0.46	*Venkataraman and Friedlander*, 1994
Mumbai, India, 1996	0.76	*Venkataraman et al*., 1999
USA, Combustion of wood fuel	0.16 – 0.31	*Hays et al.*, 2003
Thailand, March, 2009	0.2	*Phoothiwut and Junyapoon,* 2013

Model parameterization of dry deposition of particle associated PAHs in the GLEMOS model was updated to follow the parameterization of dry deposition, applied for heavy metals, which includes dependence of deposition rate on the particle size using the mass median diameter [*Travnikov and Ilyin*, 2005].

Experimental modelling of B(a)P pollution using the GLEMOS model and updated parameterization of dry deposition were performed for different values of mass median diameters from 0.1 to 0.8 μm. Model simulations showed considerable sensitivity of calculated B[a]P air concentrations to the

definition of particle size distribution. In particular, in Fig. 3.9 mean relative biases of model estimates and measurements are displayed. Comparison of modelling results with measurements of EMEP monitoring sites indicates decrease of estimates of B(a)P air concentrations with the increase of mass median diameter due to increasing of dry deposition intensity. Better agreement between results of model simulations and observed B(a)P concentrations is obtained in case of lower values of mass median diameter within the defined range.

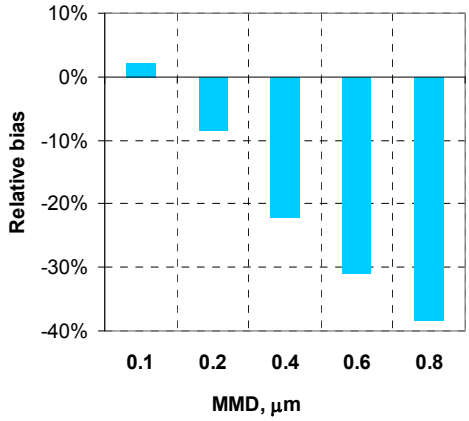


Fig. 3.9. *Relative biases between the measured and modelled B(a)P air concentrations, obtained in model simulations with different values of mass median diameter in the parameterization of dry deposition*

3.5. Concluding remarks and further activities

Pilot simulations of B(a)P pollution on the new EMEP grid, performed with updated GLEMOS model, provide comparable results with previous version of the model, showing generally better agreement of these results with measurements of EMEP monitoring network in comparison with previously used model version and older EMEP grid. At the same time, for some of the stations model predictions noticeably differ from the observed pollution levels.

To further improve the agreement between model estimates and measurements, analysis of factors, influencing the accuracy of modelling results for the new EMEP grid, is initiated. Preliminary results of this activity indicate the importance of relationships between B(a)P pollution levels and the content of particulate matter as well as atmospheric reactants in the atmosphere. For better performance of the GLEMOS model there is a need of further refinement of model parameterization of gas-particle partitioning, degradation, and deposition processes.

4. CASE STUDY OF B(a)P POLLUTION IN SPAIN

MSC-E and the Task Force on Measurements and Modelling (TFMM) of EMEP have organized a number of country–specific case studies on heavy metal pollution assessment in several EMEP countries (e.g. the Czech Republic, Croatia, the Netherlands, Belarus, the UK, and Poland). This year, following the second joint session of the Working Group on Effects and the Steering Body to EMEP, these country-specific studies are continued with the evaluation of B(a)P pollution levels in Spain. Objective of the study is to perform analysis of B(a)P pollution levels in Spain using national emission data with fine resolution, national monitoring of B(a)P concentrations and modelling of B(a)P dispersion over Spain on the basis of EMEP GLEMOS model and CHIMERE model. The study will include the following activities:

- Analysis of information on major sources of B(a)P pollution in Spain (i.e. industrial sources, residential combustion, road traffic, field burning in agriculture, natural sources and forest fires)
- Preparation of input data for model assessment of pollution levels.
- Modelling of B(a)P pollution over EMEP domain to prepare boundary conditions for the fine resolution model simulations.
- Model simulations of B(a)P pollution using different scenarios of B(a)P emissions in Spain.
- Modelling of B(a)P using sector specific B(a)P emission data to evaluate pollution levels and contributions of particular emission source categories.
- Source-receptor modelling for the evaluation of B(a)P long-range transport between administrative regions of Spain.
- Evaluation of modelling results against measurements of EMEP network and national monitoring stations.
- Analysis of pollution levels using EMEP GLEMOS and CHIMERE modelling results.

Major attention at the current stage of the work has been paid to the collection of available information on the sources and levels of B(a)P pollution in Spain from various studies and experimental model simulations. Data on PAH emissions are taken from the officially submitted inventories of Spain. National inventories cover the period from 1990 to present time and provide information on spatial distribution of emissions and key source categories. For the analysis of B(a)P pollution levels measurements of national monitoring network are used. Available inventories on PAH emissions in Spain were used to prepare scenarios of B(a)P emission for modelling purposes. Preliminary model simulations has been carried out with total B(a)P emissions and with emissions from specific source categories. Results of model simulations were compared with measurements of national monitoring sites in Spain.

4.1. Available data on PAH emissions in Spain

In order to prepare necessary input data for model assessment of B(a)P pollution three recent inventories of PAH emissions, provided by Spain, were considered, namely, inventories submitted in 2015, 2016, and 2017. According to these data PAH emission of Spain is one of the most significant among the EMEP countries.

PAH emissions from major source categories and their changes in three recent inventories of Spain are shown in Fig. 4.1. The most significant contribution is reported for the agricultural sources (L_AgriOther), followed by industrial sources (B_Industry), residential combustion (C_OtherStatComb), and road transport (F_RoadTransport). Additional contribution to B(a)P pollution levels can also be made by natural emissions (e.g. forest fires) occurred episodically (Informative inventory report of Spain, [*Perea*, 2016]).

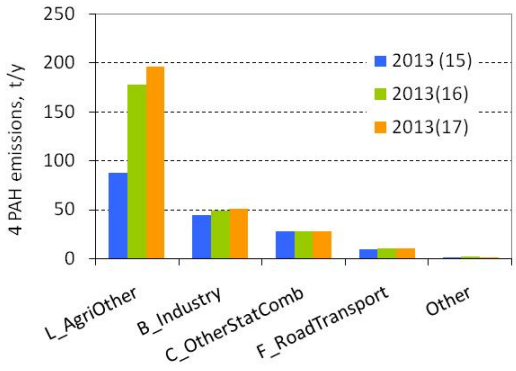


Fig. 4.1*. Sector distribution of PAH emissions for the year 2013 in Spain according to the inventories submitted in 2015, 2016, and 2017. It should be noted that in case of the inventory of 2017 the value of L_AgriOther represents the sum of PAH emissions from NFR sectors 3F and 5C2*

Important feature of these inventories is dominating contribution of the sector L_AgriOther (about 70%), related to field burning of agricultural residues. Similar structure of sectors can be seen in the inventories of Portugal and Cyprus, while most of other EMEP countries reported significantly lower values of PAH emissions from agriculture and considerably higher emissions from residential combustion.

It can be seen that values of PAH emissions from agriculture substantially differ from one inventory to another. Significant changes in estimates of agricultural emissions can also be mentioned for the time-series of annual emissions shown in Fig. 4.2. Considering the spatial distribution of PAH emissions from agriculture it can be noticed that the most significant emissions takes place in the southern areas of Spain associated with agricultural activities (Fig. 4.3). At the same time B(a)P concentrations, observed on monitoring stations in this area, do not reflect these high emissions. Besides, spatial distribution of observed concentrations better correlates with spatial distribution of industrial emissions.

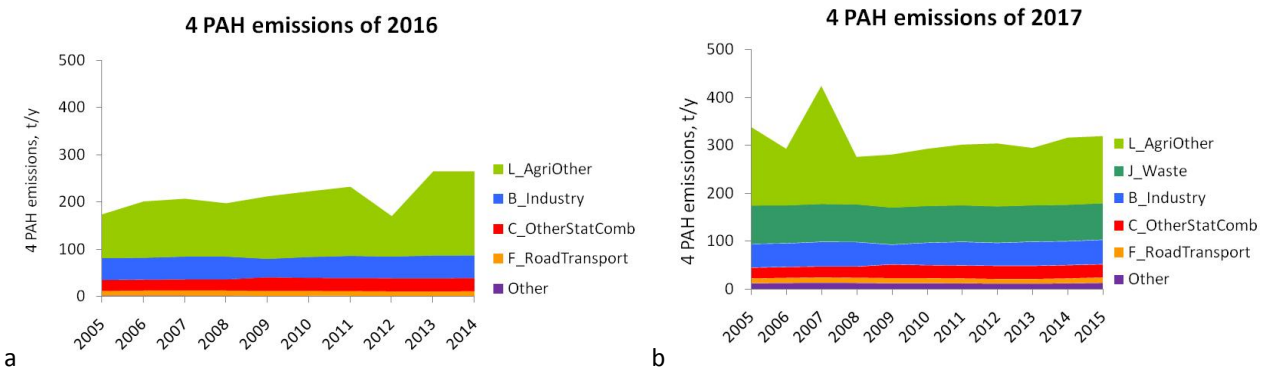


***Fig. 4.2**. Time-series of PAH emissions of Spain splitted by sectors for the recent decade according to the inventories submitted in 2016 (a) and 2017 (b)*

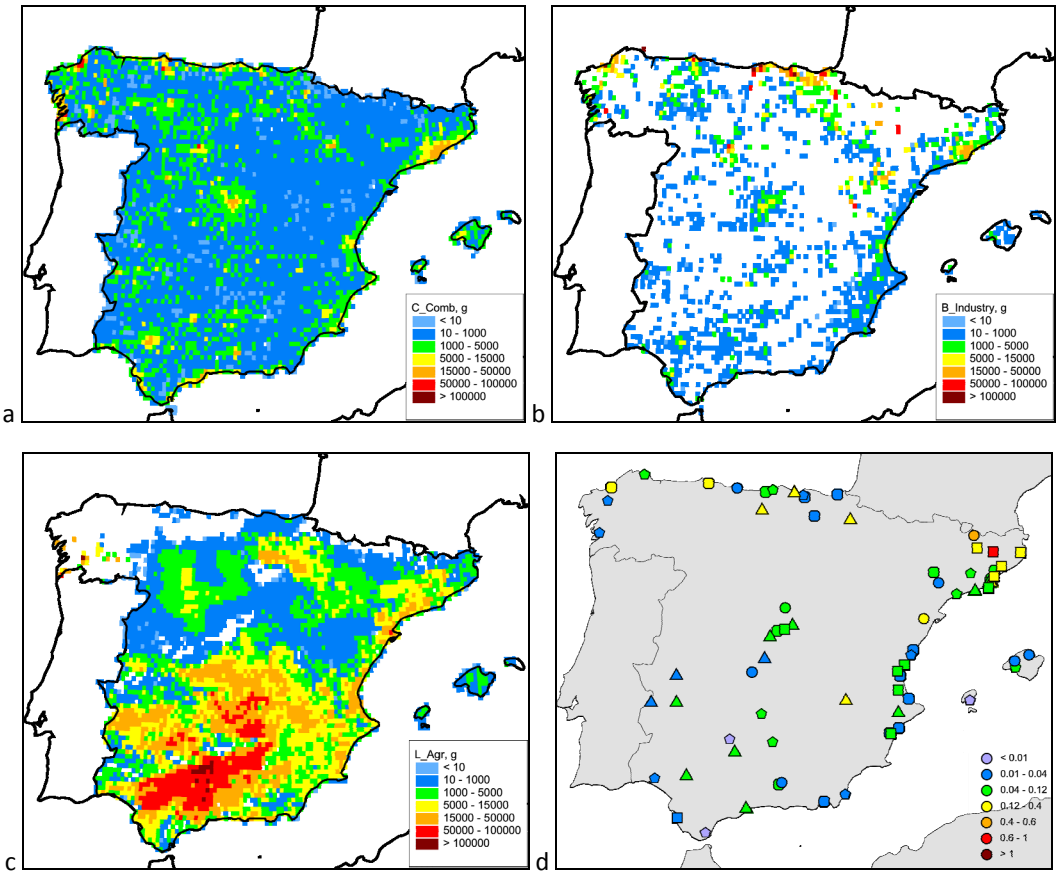


***Fig. 4.3**. Spatial distribution of PAH emissions from major source categories, namely, residential combustion (a), industrial sources (b), agricultural sources (c), in comparison with measurements of B(a)P air concentrations of national monitoring network in 2014, ng/m^3 (d)*

Taking into account possible uncertainties in the estimates of PAH emissions from agricultural sources several scenarios of PAH emissions were constructed for modelling, assuming different contributions of this sector to the total emission. Description of these scenarios is given below.

4.2. Gridded data on emissions and scenarios

Gridded dataset of B(a)P emissions has been prepared for two modelling domains, namely, for the EMEP domain and for the nested model domain over Spain. Modelling of long-range transport and pollution levels of B(a)P in the EMEP domain has been carried out using emission data for 2014 provided by CEIP. Detailed information on PAH emissions in the EMEP countries, as reported in 2016, can be found in the Technical report of CEIP 02/2016 [*Tista et al.*, 2016]. Gridded emission of B(a)P within the new EMEP region for 2014 with spatial resolution 0.4°x0.4° is generated on the basis of emissions with 50x50 km resolution applying interpolation with conservation of emitted mass of a pollutant.

To prepare emission input data for the nested domain over Spain, available inventories of PAH emissions, submitted by Spain, were used. Spatial distribution of annual total B(a)P emissions within the EMEP region and in Spain is shown in Fig. 4.4.

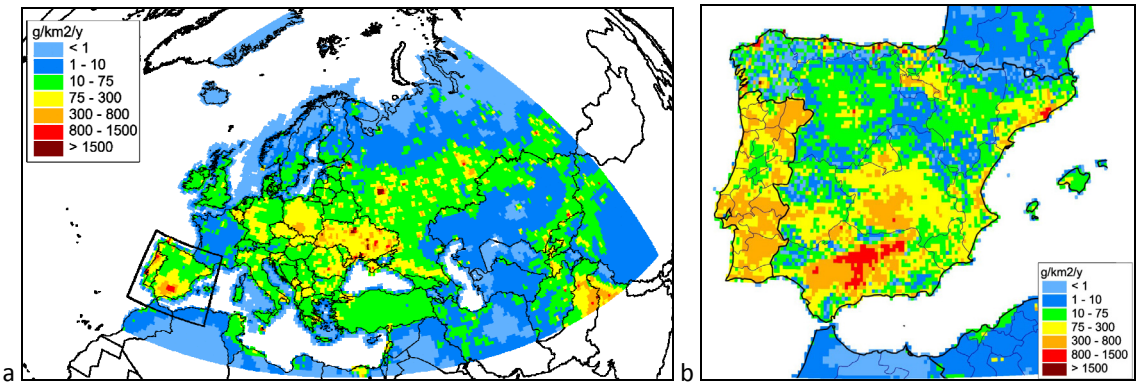


***Fig. 4.4**. Spatial distribution of annual B(a)P emission fluxes in the EMEP domain with spatial resolution 0.4x0.4 degrees (a) and in nested domain over Spain with spatial resolution 0.1x0.1 degrees (b) for 2014, g/km^2/y*

National inventory of PAH emissions is provided as the sum of 4 PAHs emissions without speciation for particular PAH compounds. In order to prepare dataset of B(a)P emissions for modelling, average content of B(a)P in the emissions of 4 PAHs, based on PAH emission data of other EMEP countries, is used. According to their inventories of PAH emissions the fraction of B(a)P can be about 28%. The fraction B(a)P/4PAHs can be different for different sectors that might have effect on total emissions of B(a)P and emissions from specific sectors. In particular, according to the information from national experts on emissions, with reference to the EMEP/EEA Guidebook 2016, the fraction of B(a)P in 4PAHs could be 14% for the category 5C2. Assumption on the average fraction for all sectors can lead to some uncertainties in evaluation of B(a)P emissions that will be evaluated at further stages of the study.

For modelling purposes three datasets of B(a)P emission in Spain have been prepared. The first dataset is based on sector distribution and spatial allocation, defined in the official emission data (base case scenario). Additional two scenarios (scenario 1 and scenario 2) are constructed to explore the effect of possible uncertainties related to the level of emissions from agricultural sources in the officially submitted data (Fig. 4.5). In particular, the first scenario assumes two-fold lower emission from the agricultural sector, which is defined following the difference between the lower and default values of

emission factor used for calculation of emissions [*EMEP/EEA Guidebook*, 2013]. The second scenario assumes the twenty-fold lower emissions from agricultural sources. In this case relative contribution of this sector to the total emissions is become comparable to the emission data of several other countries (e.g. Lithuania, France, Denmark), which provided estimates of emissions from field burning of agricultural residues (Table 4.1).

Table 4.1. *PAH emissions from selected sectors and their contributions to total emissions based on the officially submitted inventories in 2017*

	ES	PT	FR	LT	CY	DK	AT	LV	FI
Industry	51.21	1.30	0.51	0.23	0.04	0.11	0.38	0.57	0.32
Industry, %	*16.6*	*1.1*	*2.7*	*2.1*	*3.0*	*1.5*	*7.8*	*5.9*	*3.4*
ResComb	27.35	11.14	11.40	8.66	0.03	6.02	3.85	7.76	7.74
ResComb, %	*8.9*	*9.9*	*59.6*	*80.2*	*2.5*	*82.7*	*78.7*	*80.4*	*82.3*
Waste	76.30	0.00	0.98	0.09	0.01	0.24	0.00	0.00	0.15
Waste, %	*24.8*	*0.0*	*5.1*	*0.8*	*0.7*	*3.2*	*0.0*	*0.0*	*1.6*
AgriOther	139.83	99.62	1.27	0.96	0.79	0.34	0.09	0.08	0.00
AgriOther, %	*45.5*	*88.0*	*6.7*	*8.9*	*64.9*	*4.6*	*1.9*	*0.9*	*0.0*
Total	307.62	113.14	19.11	10.81	1.22	7.28	4.89	9.66	9.41

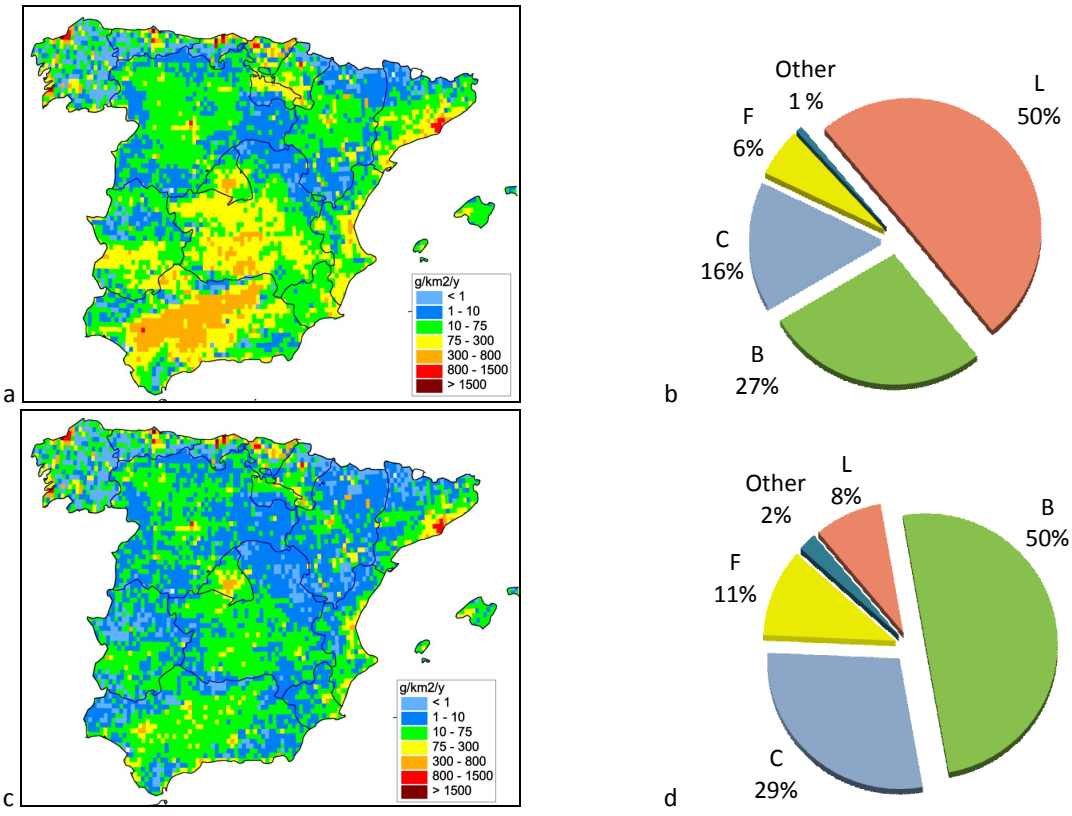


Fig. 4.5*. Spatial distribution of annual B(a)P emission fluxes in Spain (g/km^2/y) and contributions of key source categories to total emissions according to scenarios with the decreased emissions from agricultural sources: Scenario 1 with two-fold decrease (a and b) and Scenario 2 with twenty-fold decrease (c and d). Definition of sectors: B – industrial sources, C – residential combustion, F – road transport, L – agricultural sources*

4.3. Preliminary modelling results and their analysis

Setup of model simulations

Model assessment of B(a)P pollution levels in Spain is carried out using nested model simulations over two domains, namely, coarse domain over the whole EMEP region and nested domain over Spain. Regional-scale EMEP domain is used for generation of boundary conditions required for nested modelling. Model simulations for this domain are made for the new EMEP grid in longitude-latitude projection with spatial resolution 0.4°x0.4°. For national scale domain over Spain model simulations are carried out with finer spatial resolution 0.1°x0.1°.

Model assessment is performed using the recent version of EMEP multi-scale multi-media chemistry-transport model GLEMOS developed for the evaluation of pollution by HMs and POPs. For the analysis of B(a)P pollution in Spain the year 2014 is selected due to availability of gridded emission data and measurements of air concentrations. To perform model simulations necessary meteorological input data were generated for the year 2014 using numerical Weather Research and Forecasting (WRF) model on the basis of meteorological analyses of ECMWF as the driving input.

Results of model simulations with the base case scenario

Initial model simulations of B(a)P pollution for the EMEP region and over Spain were performed for 2014 with the officially reported emissions according to the base case scenario. Spatial distribution of annual mean modelled B(a)P air concentrations in the EMEP countries and in the Spain with finer spatial resolution (0.1°x0.1°) is shown in Fig. 4.6.

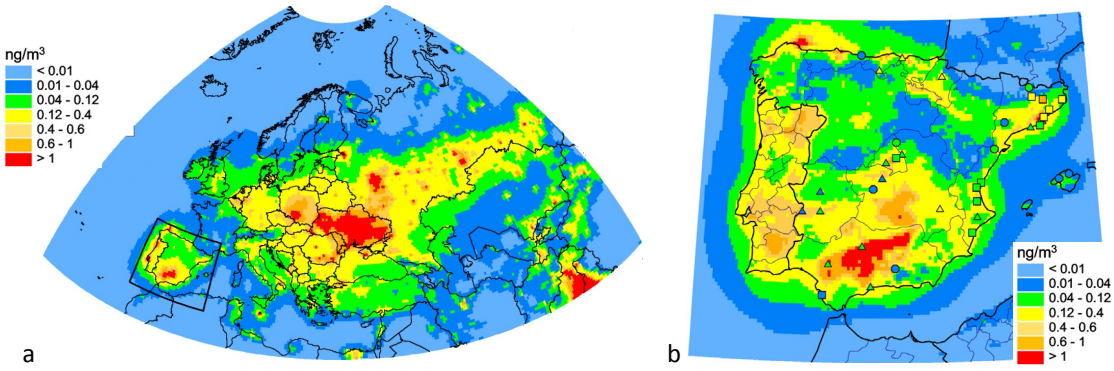


Fig. 4.6*. Results of base case model simulations. Spatial distribution of modelled B(a)P air concentrations in the EMEP domain with spatial resolution 0.4x0.4 degrees (a) and in nested domain over Spain with spatial resolution 0.1x0.1 degrees (b) for 2014, ng /m^3*

Analysis of modelling results has been performed for both model domains. Modelled B(a)P air concentrations were compared with measurements of EMEP background monitoring stations performed in 2014. In general, modelled air concentrations correspond to the concentrations observed at background and remote stations in the EMEP countries (Fig. 4.7a). The correlation between the annual mean modelled and measured concentrations amounts to about 0.7 and the bias is about 2%, which shows that the model is capable to satisfactory describe distribution of annual mean levels of air concentrations.

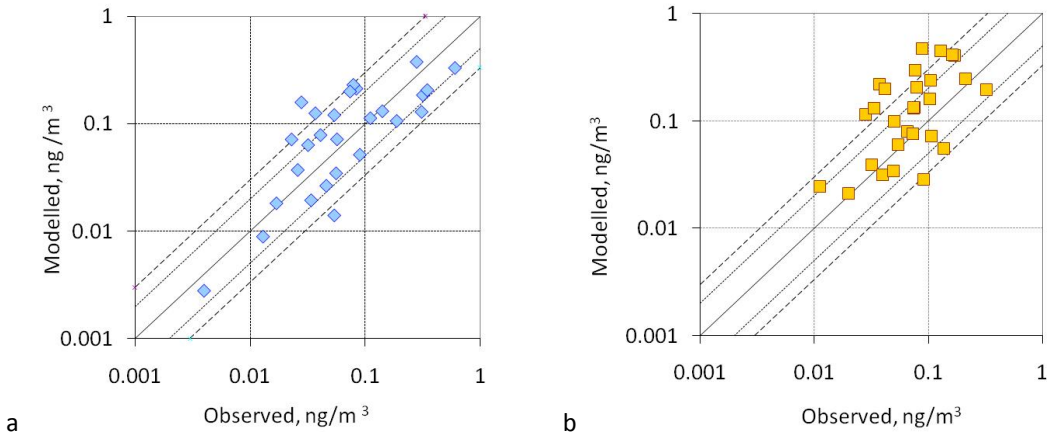


***Fig. 4.7**. Comparison of modelled B(a)P air concentrations for the EMEP domain with measurements of EMEP monitoring sites (a) and for the nested domain over Spain with measurements of Spanish monitoring sites (b) carried out in 2014, ng/m^3. Dashed lines indicate the areas of agreement between modelled and observed values within factors of 2 and 3*

At the same time, ***comparison of modelled concentrations*** for the nested domain (Fig. 4.7b) ***with measurements of background sites in Spain indicates substantial overestimation of observed concentrations for part of the sites.*** In this case the bias between modelled and observed concentrations is about 90%.

Comparing levels of emissions in the EMEP countries with measured concentrations it can be seen that B(a)P concentrations in Spain are relatively low, at the same time the average emission flux is comparatively high (Fig. 4.8).

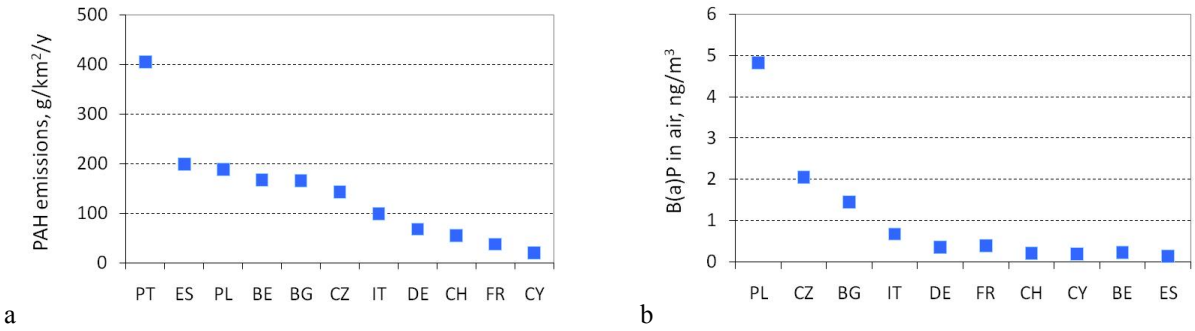


***Fig. 4.8**. Average annual emission fluxes of 4PAHs in the selected EMEP countries (a) versus annual mean observed B(a)P air concentrations in 2013 (b). Measurements of B(a)P air concentrations were taken from the EEA AirBase*

Taking this into account ***it was supposed that one of the reasons of the overestimation may be connected with high values of estimated national emissions in Spain, in particular, from the sector related to the field burning of agricultural sources. Additional contribution to this overestimation can also be due to assumption of the fraction B(a)P/4PAHs as well as the uncertainties in the parameters of model parameterization describing degradation and deposition processes.*** At this stage analysis of modelling results is focused on the effect of possible uncertainties in the emissions data. Other factors affecting modelling results will be analysed at further stages of this work.

Results of model simulations with scenario 1 and scenario 2

Model simulations with the scenarios, reflecting possible range of uncertainties in the emissions from agricultural sources, provided generally better agreement with most part of measurements of background stations (Fig. 4.8b and c). Results of the comparison of modelled concentrations with measurements are illustrated in Fig. 4.9.

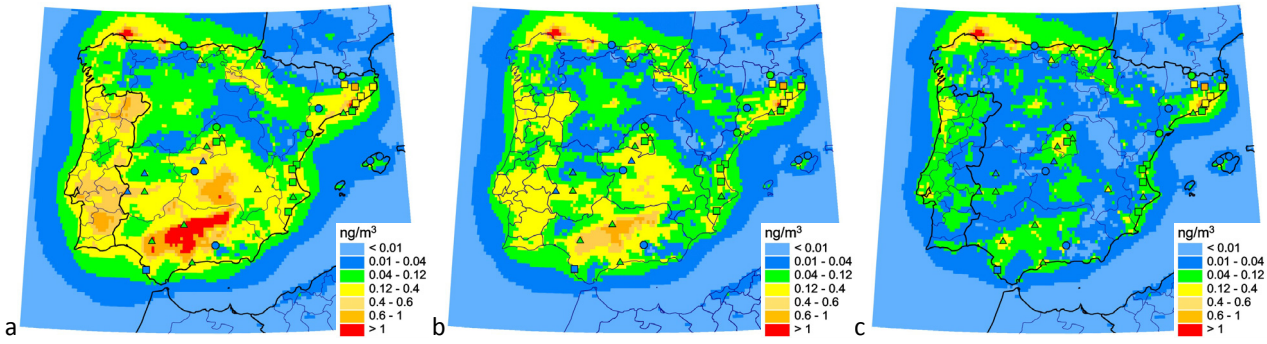


***Fig. 4.8**. Modelled B(a)P air concentrations obtained with application of emissions according to the base case scenario (a), Scenario 1 (b) and Scenario 2 (c), ng/m^3*

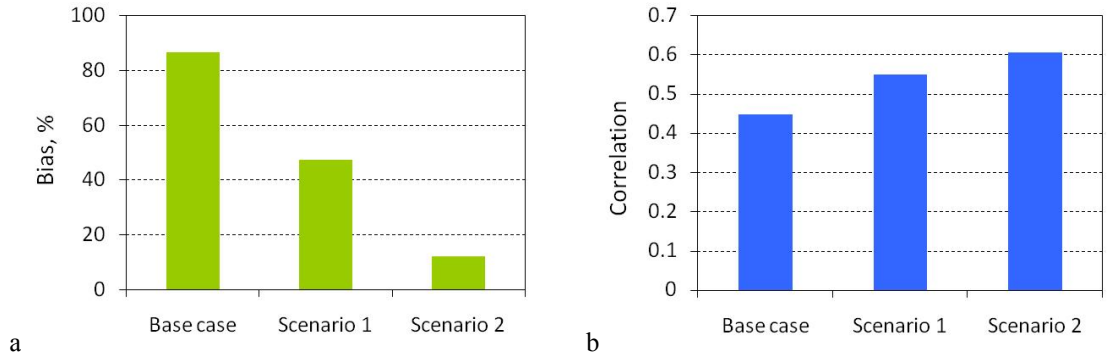


***Fig. 4.9**. Relative bias (a) and correlation (b) between observed and modelled B(a)P air concentrations obtained in model simulations with three datasets of emissions: base case, scenario 1, and Scenario 2*

In general, the application of scenarios 1 and 2 resulted in decreased bias and improved correlation between observed and modelled B(a)P air concentrations for most part of monitoring sites in Spain. These results may be regarded as the indication of available uncertainties of emission estimates, in particular, emissions from the agricultural sources. Thus further refinement of emissions reflecting releases of PAHs to the atmosphere from the activities related to the burning of agricultural residues is appreciated.

Discrepancies between modelled and observed B(a)P air concentrations for Catalonia

Though the application of scenarios with the decreased input from agricultural sources allowed to improve in general the level of agreement between modelling results and measurements, there are two groups of monitoring sites in Catalonia, for which modelled values still differ significantly from the observed concentrations. In particular, ***the model tends to overestimate B(a)P air concentrations observed in Barcelona and underestimate concentrations measured in mountainous and rural areas of North-eastern part of Catalonia*** as shown in Fig. 4.10.

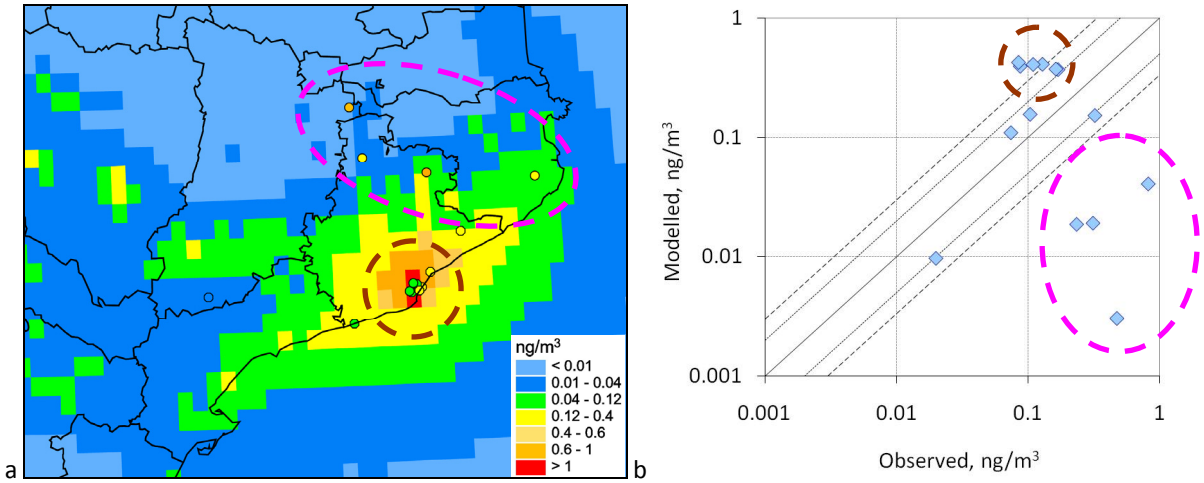


Fig. 4.10*. Modelled B(a)P air concentrations over Catalonia for 2014, obtained with application of emissions according to the Scenario 2 (a) and their comparison with measured air concentrations (b), ng/m^3. Brown circle indicates monitoring sites in the area of Barcelona, while magenta circle indicates monitoring sites in the North-eastern part of Catalonia*

Possible reasons of disagreement between the modelled and observed concentrations in these two cases can be spatial allocation of emissions and deficiencies of modelling approach related to insufficient spatial resolution and complex terrain conditions.

The spatial allocation of emissions from one of the most important sectors, namely, Residential combustion, in the area of Catalonia is shown in Fig. 4.11a. The spatial distribution of emissions from this sector contradicts with the distribution of observed concentrations, negatively correlating with measurements, which can be explained by the allocation of higher emission values in the area of Barcelona and lower emissions in rural areas.

According to the national studies the city of Barcelona is characterised by relatively low concentrations of B(a)P. Domestic and residential heating activities in the city, as well as public transport, are not contributing substantially due to the use of natural gas [*Querol et al.,* 2016; *Viana et al.*, 2016]. Thus higher emissions from the residential combustion can be expected in the rural areas rather than in the area of Barcelona. The studies of air quality in Barcelona city indicated considerable influence of air pollution transport from rural areas [*van Drooge et al.,* 2014, 2015].

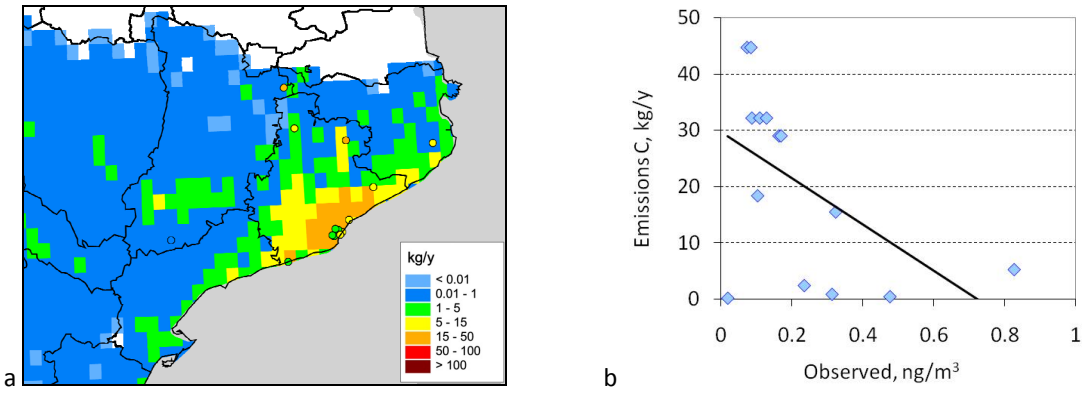


***Fig. 4.11**. Spatial distribution of B(a)P emissions from Residential combustion sector (a), kg/y, and scatter plot with values of B(a)P emissions versus measured B(a)P air concentrations (b) at monitoring sites in Catalonia in 2014*

To evaluate the sensitivity of modelling results to spatial distribution of emissions, data on releases from the sector Residential combustion were modified. Re-allocation of emissions from this sector was made in the assumption that emission sources in the rural areas can be more significant due to more extensive use of biomass or wood burning comparing to urban areas of Barcelona, where the natural gas is applied. Spatial distributions of original and modified emission fluxes are shown in Fig. 4.12a and 4.12b. Experimental model run with re-allocated emissions have shown better agreement between modelled and observed values for Barcelona city (Fig. 4.13), though the model still tends to overpredict measured concentrations. Therefore model assessment of PAH pollution of this area requires more detailed analysis of spatial distribution of emissions and co-operation with national experts.

The level of disagreement between the modelled concentrations and measurements of sites located in mountainous and rural areas, namely, Manlleu (ES1397A), Bellver de Cerdanya (ES1348A), la Bisbal (ES1559A), and Berga (ES1851A), was not changed significantly in the experimental model simulations. High levels of B(a)P concentrations may be related to the influence of local meteorological conditions (e.g. frequent inversions) [*Viana et al*., 2016], which were not rather well captured by the model and the effect of local emission sources possibly missing in the emission inventory. Thus, further analysis of the differences and improvement of model performance for these sites would require application of finer spatial resolution for modelling and more detailed emission data for this area.

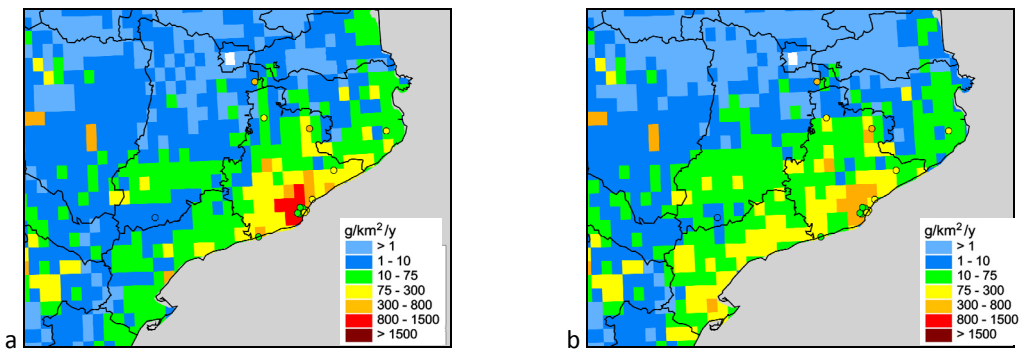


***Fig. 4.12**. Spatial distributions of original and modified allocation of B(a)P emission fluxes, g/km^2/y over Catalonia for 2014*

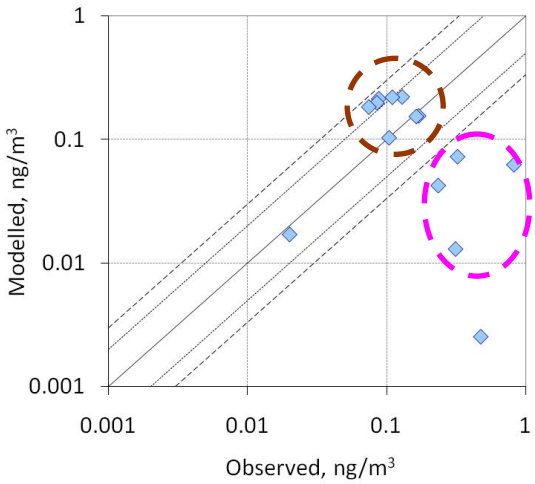


***Fig. 4.13**. Modelled B(a)P air concentrations over Catalonia for 2014, obtained with application of modified allocation of B(a)P emission fluxes, in comparison with measured B(a)P air concentrations, ng/m^3*

4.4. Concluding remarks and further activities

Preliminary analysis of PAH pollution levels in Spain has been performed on the basis of available information on emissions, observed PAH concentrations, and modelling results. According to available measurements, spatial distribution of B(a)P pollution levels in Spain is characterized by relatively higher levels of B(a)P pollution in its northern areas and lower in other areas of the country. Comparison of model predictions with measurements indicates overestimation of observed B(a)P air concentrations for central and western parts of the country, which can be attributed to possible uncertainties in the reported national PAH emissions as well as uncertainties in modelling approach and measurements.

In order to explore the reasons of discrepancies, data on PAH emissions and measurements were analyzed. National PAH emission inventory of Spain is characterized by noticeably high contribution of emissions from field burning of agricultural residues (about 70%), while contribution of residential combustion is less important. This pattern of emissions distinctly differs from the inventories of other EMEP countries, where residential combustion is dominating source category (except for Portugal and Cyprus). Besides, spatial variations of emissions are not consistent with variations of observed concentrations.

Experimental model simulations with several emission scenarios and analysis of observed levels of PAH pollution have indicated possible uncertainties in estimates of PAH emissions. In particular, model simulations with experimental scenarios of emissions have indicated that decreasing of the input of agricultural sources have led to improved bias and spatial correlation between observed and modelled B(a)P air concentrations for most part of monitoring sites in Spain. Thus, further refinement of emissions related to the burning of agricultural residues and analysis of their uncertainties is appreciated.

Further activities with respect to evaluation of PAH pollution levels in Spain, using model simulations and results of national studies, in co-operation with national experts from Spain will include:

- Analysis of factors affecting levels of B(a)P air concentrations (meteorological conditions, levels of concentrations of atmospheric reactants, parameters of model parameterizations of processes governing B(a)P transport and removal from the atmosphere).
- Evaluation of PAH pollution levels in Spain using the EMEP GLEMOS and CHIMERE models as well as inter-comparison of modelling results and comparison with national measurements.
- Sector specific model simulations to evaluate contribution of different source categories in different regions of Spain.
- Source-receptor modelling to evaluate the transport of B(a)P between the administrative sub-regions of Spain as well as contributions of surrounding countries.

It is planned to prepare joint report of MSC-E and national experts in modelling, monitoring, and developing of emission inventories, to summarize the outcome of model evaluation and analysis of B(a)P pollution levels in Spain.

5. ASSESSMENT OF PCDD/Fs, PCBs, AND HCB POLLUTION IN THE EMEP REGION

Model assessment of environmental pollution by PCDD/Fs, PCBs, and HCB has been carried out using combined global and regional-scale model simulations. The outcome of the assessment includes estimates of spatial distribution and temporal changes of PCDD/Fs, PCBs, and HCB air concentrations and deposition fluxes. Transboundary transport of pollution 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 modelling results. Results of model simulations have been compared with measurements of the EMEP monitoring sites. Brief overview of progress and outcome of this study is presented below. Detailed information on modelling results and their analysis are given in the internet on the MSC-E web site (www.msceast.org).

5.1. Emission data for model assessment

Assessment of PCDD/F and HCB pollution in the EMEP domain has been carried out on the basis of gridded emission data provided by CEIP. Detailed description of reported PCDD/F and HCB emissions of the EMEP countries, as well as gap-filling methods used for the elaboration of GNFR emission inventory for 2015, can be found in the Technical report of CEIP [*Tista et al.,* 2017].

Spatial distributions of PCDD/F and HCB emissions from anthropogenic sources in the EMEP domain, used in model simulations for 2015, are presented in Figs 5.1a and 5.1b, respectively.

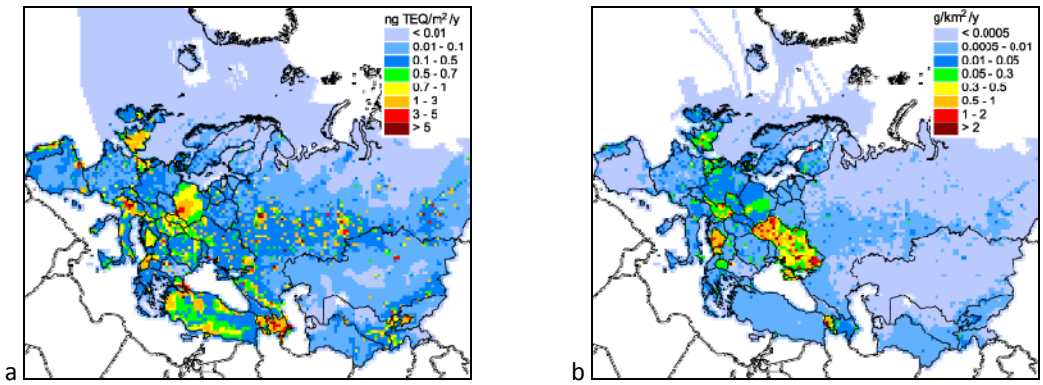


Fig. 5.1. *Spatial distribution of PCDD/F (a) and HCB (b) emissions in the EMEP countries in 2015 with resolution 50x50 km²*

According to national inventories and expert estimates of CEIP total annual anthropogenic emissions in the EMEP region for 2015 account for about 5.9 kg TEQ for PCDD/Fs and 848 kg for HCB. ***PCDD/F and HCB emissions for 2015 are almost 40% and 90% higher than the corresponding emissions for 2014, provided in the previous submission of emission data*** [*Tista et al*., 2016].

Changes of national total PCDD/F and HCB emissions in the EMEP countries from 2014 to 2015 in absolute values (E_{2015}-E_{2014}) are illustrated in Fig 5.2. Positive values in the diagram indicate increase of emissions from 2014 to 2015, while negative values denote a decrease of emissions.

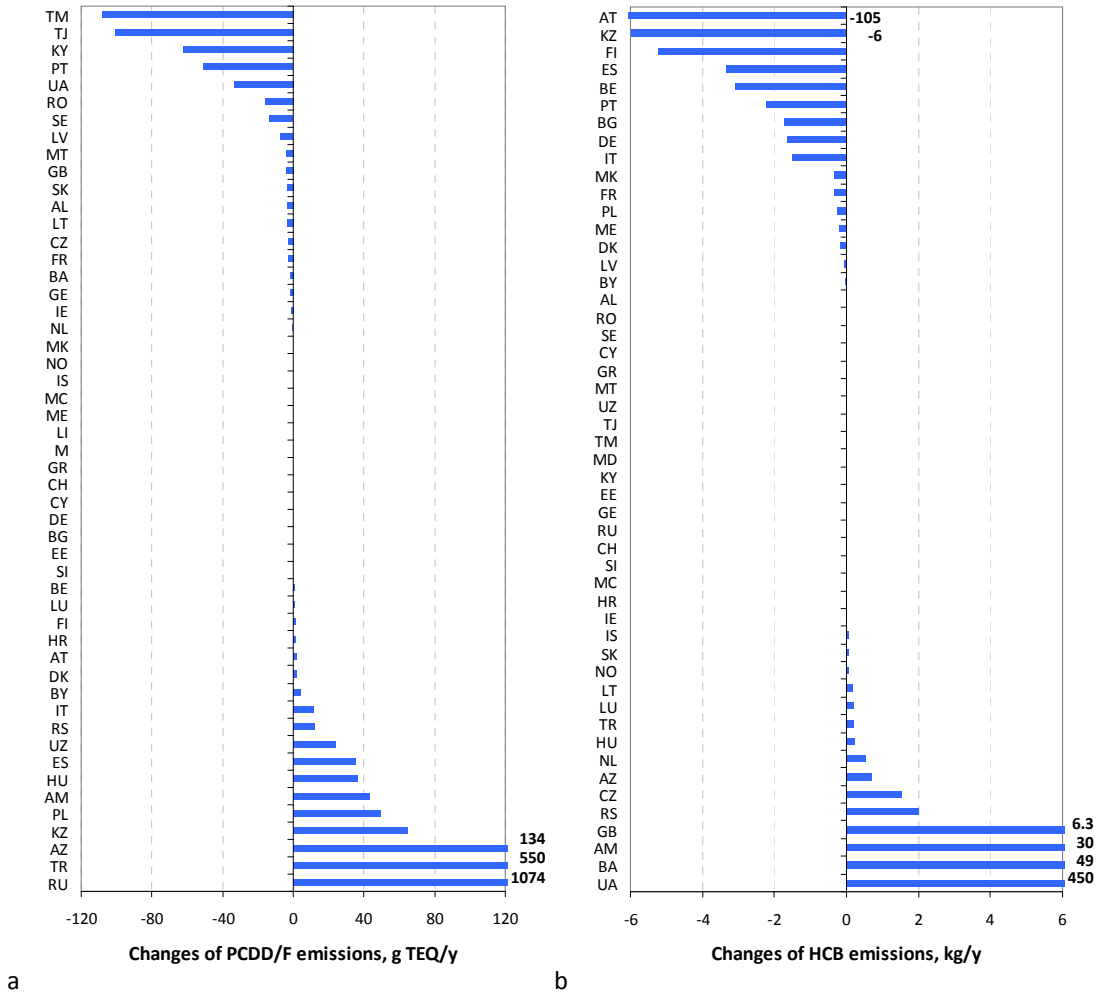


Fig. 5.2. *Changes of annual total emissions of PCDD/Fs, g TEQ/y (a) and HCB, kg/y (b) from 2014 to 2015 (E_{2015}-E_{2014}). Negative values denote decrease of emissions, and positive ones denote increase of emissions*

The most significant increase of PCDD/F emissions in 2015 can be indicated for the Russian Federation, Turkey, Azerbaijan, and Kazakhstan, amounted to a factor of 16, 6.4, 2.5, and 1.6, respectively. The largest decline of emissions is noted for Kyrgyzstan, Turkmenistan, Tajikistan, and Portugal (by a factor of 5.3, 3.8, 3.5, and 1.7, respectively). Changes of emissions for the Russian Federation and Turkey are caused by the application of expert estimates, made by CEIP on the basis of the information from literature [*Treger*, 2011; *Wang et al.*, 2016]. For Azerbaijan, recalculations of emissions have been made in all sectors using updated emission factors of the EMEP/EEA Emission Inventory Guidebook 2016 instead of factors applied before [*IIR of Azerbaijan*, 2017].

Considering relative changes of PCDD/F emissions from 2014 to 2015, the largest increase is noted for Armenia, Turkey, and the Russian Federation (by a factor of 16, 6, and 2.5). Besides, significantly higher emissions in 2015 (by more than 50%) are noted for Hungary, Azerbaijan, and Luxembourg. The most significant decline can be mentioned for Malta, Kyrgyzstan, Turkmenistan, and Tajikistan (by a factor of 175, 5.3, 3.8, and 3.5). The other EMEP countries are characterized by less significant changes.

For HCB the most noticeable increase of emission in 2015 can be indicated for Ukraine, Bosnia and Herzegovina, and Armenia (Fig. 5.2b). National total emissions of these countries were changed from

104 kg to 553 kg, from 0.001 kg to 49 kg, and from 0.005 kg to 30 kg, respectively. Significantly lower estimate of HCB emissions for 2015, comparing to 2014, is noted for Austria (36 and 141 kg respectively).

The highest relative increase of HCB emissions was estimated for Serbia, Ukraine, Iceland, Lithuania, and Azerbaijan (by a factor of 9.7, 5.3, 2.5, 1.8, and 1.6, respectively). The largest relative decline of emissions is noted for Kazakhstan, Spain, Austria, and Portugal (by a factor of 7.6, 7.1, 3.9, and 2.9, respectively). Significant change of emission for Ukraine is caused by the use of national emission of this country, reported to the Stockholm Convention on POPs. Annual HCB emissions of Bosnia and Herzegovina, and Armenia for 2015 were extrapolated from the estimates of emissions for 1990 and 1995 made by *Pacyna et al.* [2003] [*Tista et al.*, 2017]. The sharp decrease of HCB emissions in Austria by 74% between 2014 and 2015 can be explained by unintentional release of HCB in 2014 during incineration of contaminated material at low temperature [*IIR of Austria*, 2017].

For the evaluation of global-scale transport and fate experimental emission scenario for PCDD/Fs, based on the UNEP SC inventory of dioxins and furans emissions, was used in model simulations [*Gusev et al.*, 2014; *Shatalov et al.*, 2014]. Global gridded emissions of PCDD/Fs to the atmosphere and soil were prepared using the national emission inventories reported by countries to the UNEP SC. Model simulations of HCB global-scale transport were carried out on the basis of experimental emission scenario of historical HCB releases during the period covering several recent decades [*Shatalov et al.,* 2010]. Spatial distributions of PCDD/F and HCB emissions, applied in global-scale model simulations for 2015, are shown in Fig.5.3.

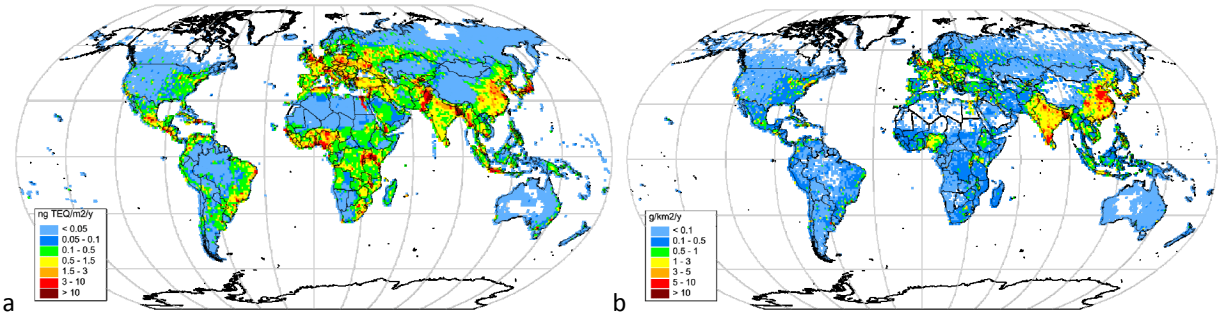


Fig. 5.3. *Spatial distribution of global annual emissions of PCDD/Fs, ng TEQ/m^2/y (a) and HCB, g/km^2/y (b) with spatial resolution 1°x1°, applied in the model simulations for 2015*

Emission data for the model simulations of PCB transport and fate were based on the officially submitted data by the EMEP countries, and available expert estimates. As it was mentioned in previous status report [*Gusev et al*., 2016], national inventories of PCB emissions are provided for the total releases of PCBs without splitting into particular congeners. For modelling purposes speciated PCB emission is required to evaluate transport and fate of individual indicator PCB congeners. Taking this into account, expert estimates of global PCB congener specific emissions of *Breivik et al.* [2007] were used in the modelling for 2015. The indicator congener PCB-153 has been selected to characterize transboundary transport and pollution levels of PCBs.

Gridded data on PCB-153 emissions for the EMEP domain were prepared using officially submitted spatial distribution of PCB emissions of 20 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 and on global scale, applied in the model simulation for 2015, is presented in Fig. 5.4.

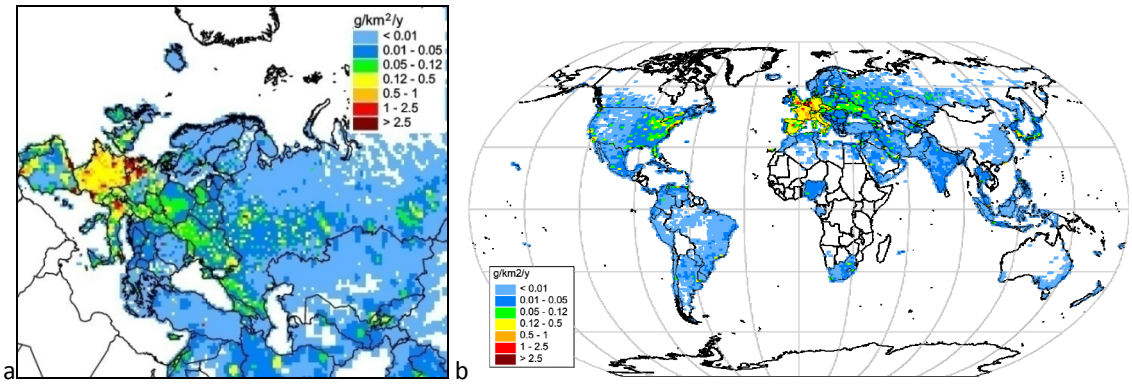


Fig. 5.4. *Spatial distribution of PCB-153 emissions in the EMEP domain with resolution 50x50 km^2 (a) and over the global domain (b) with resolution 1^{o}x1^o used in the model simulations for 2015*

5.2. Pollution levels in the EMEP region and on global scale in 2015

Model assessment of PCDD/F, PCB-153, and HCB pollution levels in the EMEP domain was carried out using global-scale modelling and nested regional-scale modelling. Model simulations of pollution on global scale for 2015 were used to set up boundary conditions for regional model simulations. Initial conditions for the evaluation of pollution levels in 2015 have been obtained using long-term spin-up model runs.

Global-scale levels of POP pollution

Spatial distributions of global-scale annual mean PCDD/F, PCB-153, and HCB air concentrations generated for 2015 are presented in Fig. 5.5. Modelling results show relatively high PCDD/F air concentrations in Africa and South Asia, while levels of pollution in Europe, North and South America, and Australia are lower. For PCB-153 elevated air concentrations are noted for the European region. For other regions less significant air concentrations are estimated. Model predictions for HCB indicate relatively high annual mean air concentrations in Eastern and Southern Asia, and low concentrations in the European countries. These patterns of air concentrations reflect in general the spatial distribution of anthropogenic emissions.

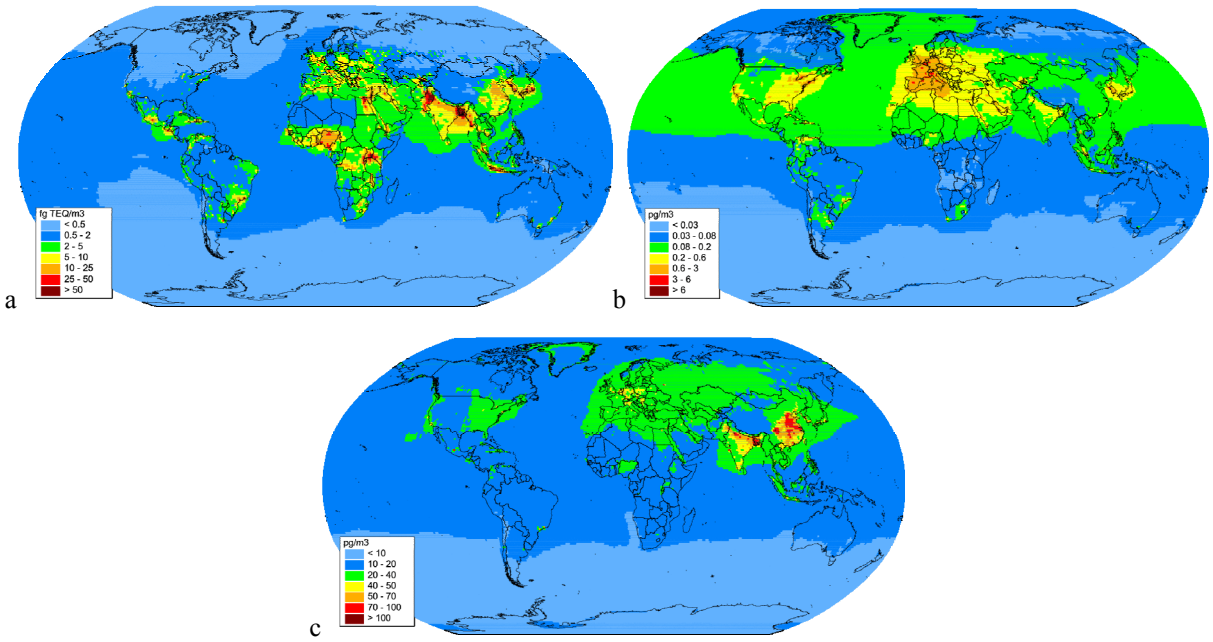


Fig. 5.5. *Spatial distribution of modelled annual mean PCDD/F, fg TEQ/m^3 (a), PCB-153, pg/m^3 (b), and HCB, pg/m^3 (c) air concentrations for 2015*

Comparison of global-scale model predictions with available measurements of PCDD/Fs, PCB-153, and HCB air concentrations was carried out at previous stage of work [*Shatalov et al.*, 2015] and is not presented here. It was shown that the model tended to slightly underestimate PCDD/F concentrations measured at rural and background sites in Europe, South America, and East Asia. More significant underprediction of observed concentrations was obtained for monitoring sites in North America. Reasonable (within a factor of 2-3) agreement of modelling results with observed air concentrations was obtained for PCB-153. Results of model simulations for HCB were close to measured HCB concentrations for most of the regions. However, the model tended to underpredict observed air concentrations at some of the monitoring sites in North America, Europe and in the Arctic.

Pollution levels in the EMEP region

Model estimates of annual mean air concentrations and deposition fluxes of PCDD/Fs, PCB-153, and HCB in the EMEP region for 2015 are shown in Fig. 5.6, 5.7, and 5.8, respectively. Elevated levels of PCDD/F annual mean air concentrations (12-40 fg TEQ/m^3 and higher) are estimated for Azerbaijan, Poland, Slovakia, Hungary, Romania, and Albania. Areas of relatively high air concentrations can also be noted for northern Italy, Serbia, Ukraine, the Russian Federation, and Kazakhstan. For PCB-153 elevated annual mean air concentrations (1-8 pg/m^3 and higher) are indicated for countries in Western and Central Europe, whereas for Northern Europe and the EECCA countries lower levels of concentrations are obtained. Model predictions of HCB annual mean air concentrations are generally higher for countries of Central and Eastern Europe (about 20 pg/m^3) with largest levels of air concentrations in Ukraine (20-40 pg/m^3). Other areas are characterized by low levels of air concentrations. Spatial distribution of HCB air concentrations is more homogeneous comparing to PCDD/Fs and PCB-153 due to its higher stability in the atmosphere.

Spatial distribution of annual deposition fluxes of PCDD/Fs and PCB-153 is in general similar to that of air concentrations. For HCB relatively high levels of deposition fluxes are estimated for the countries of Northern Europe as well as for the northern and eastern parts of the Russian Federation.

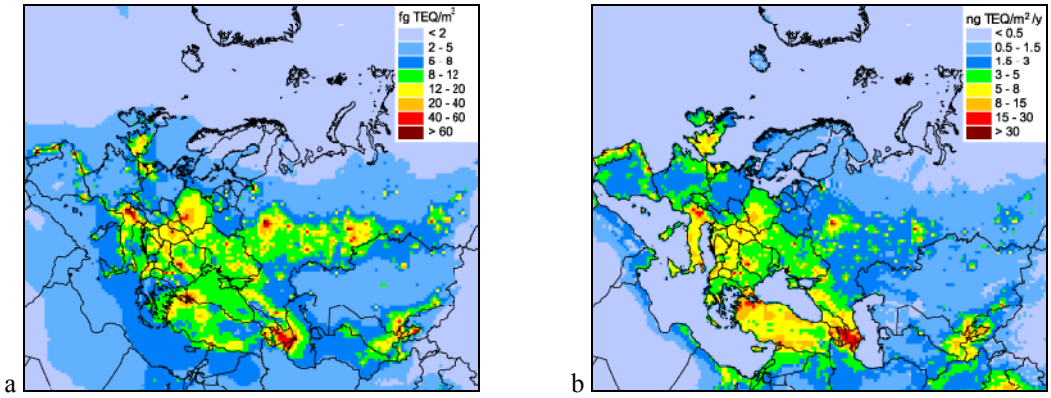


Fig. 5.6. *Spatial distributions of PCDD/F air concentrations (a) and deposition fluxes (b) in the EMEP region in 2015*

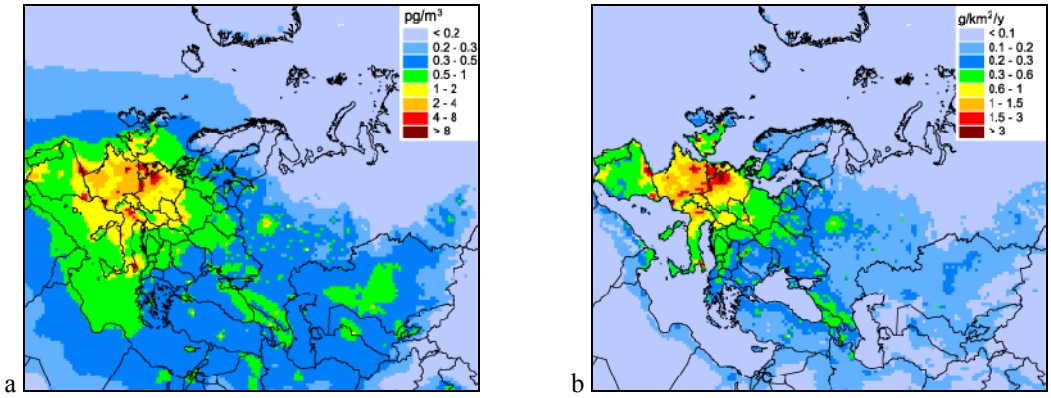


Fig. 5.7. *Spatial distributions of PCB-153 air concentrations (a) and deposition fluxes (b) in the EMEP region in 2015*

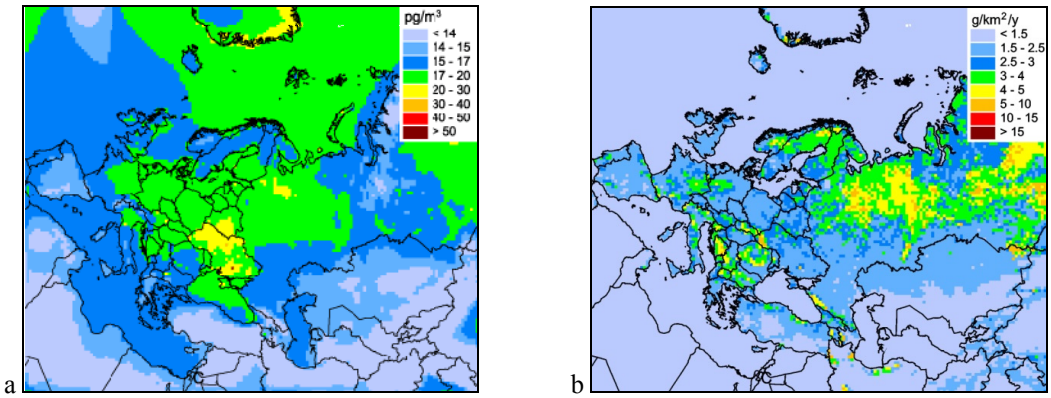


Fig. 5.8. *Spatial distributions of HCB air concentrations (a) and deposition fluxes (b) in the EMEP region in 2015*

Changes of pollution levels between 2014 and 2015

To evaluate inter-annual variability of POP pollution levels in the EMEP region, results of the model simulations for 2015 were compared with the results for 2014. Variations of model estimates of pollution levels for individual EMEP countries are caused by changes of emission data and meteorological conditions, applied in model simulations for 2015 and 2014. Relative differences in pollution levels between these two years are shown in Fig. 5.9 on the example of PCDD/Fs annual mean deposition fluxes, calculated for each EMEP country.

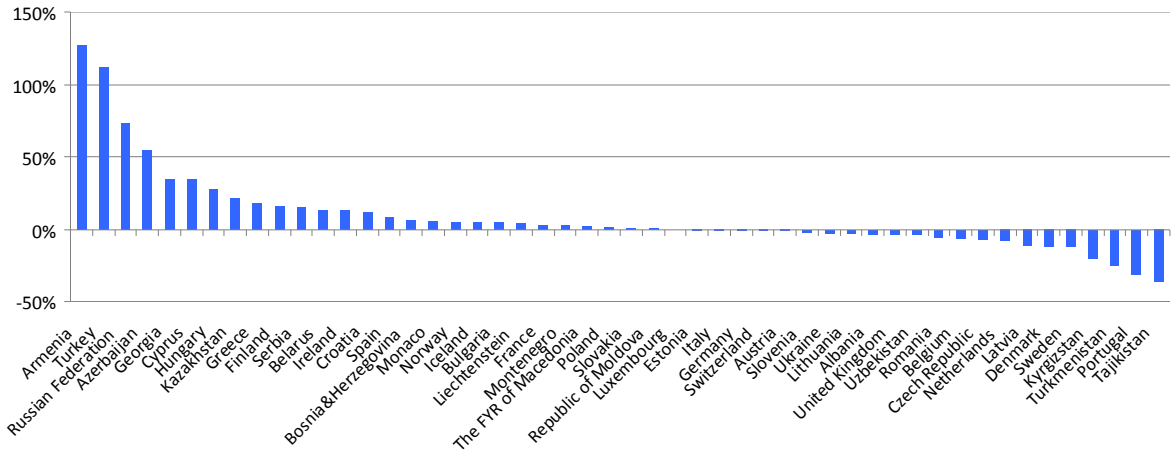


Fig. 5.9. *Changes (in %) of annual mean PCDD/F deposition fluxes in the EMEP countries between 2015 and 2014, $(D_{2015}-D_{2014})/D_{2014}$. Negative values denote decrease, and positive ones denote increase*

Annual mean deposition fluxes of PCDD/F in the EMEP countries varied from 2014 to 2015 from about 40% decrease to about 130% increase. The most significant decline of deposition is estimated for Tajikistan followed by Portugal, Turkmenistan, and Kyrgyzstan, which can be attributed to decreasing national emissions of these countries. Increase of deposition in 2015 (more than 50%) is obtained for Armenia, Turkey, the Russian Federation, and Azerbaijan. The other countries are characterized by lower changes of annual mean PCDD/F deposition fluxes.

5.3. Transboundary transport of pollution in 2015

Transboundary transport of PCDD/Fs, PCB-153, and HCB within the EMEP region was evaluated for 2015 taking into account contributions of anthropogenic and secondary emission sources. Contributions of non-EMEP emission sources were estimated using global-scale model simulations. According to modelling results, relative contributions of the EMEP anthropogenic emission sources to deposition of PCDD/Fs in the EMEP countries varied from 16% to 62%, for secondary emission sources from 30% to 80%, and for non-EMEP emission sources from 1% to 50% (Fig. 5.10). For PCB-153 relative contributions of the EMEP anthropogenic emission sources were in the range 5-45%, secondary emissions about 45-90%, and non-EMEP emission sources about 2-45% depending on a country. In case of HCB, the share of the EMEP anthropogenic emissions in the deposition of HCB to the EMEP countries

was not significant, varying from less than one percent to 12%, while secondary emissions and non-EMEP emission sources contributed about 30-70%.

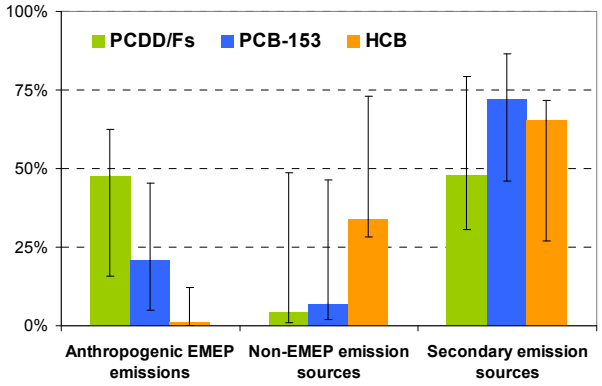


Fig. 5.10. *Contributions of three source groups (anthropogenic EMEP, non-EMEP and secondary emission sources) to deposition over the EMEP countries in 2015 for PCDD/Fs (a), PCB-153 (b) and HCB (c). Whiskers denote the range of contributions across the EMEP countries*

Model estimates of transboundary transport and contributions of different types of emission sources are illustrated by modelling results for PCDD/Fs. Annual PCDD/F deposition fluxes to the EMEP countries are shown in Fig. 5.11. Elevated levels of PCDD/F deposition fluxes in the EMEP countries (exceeding 10 ng TEQ/m^2/y) are obtained for Azerbaijan and Armenia. Significant deposition fluxes (about 5 ng TEQ/m^2/y) are also noted for Albania, Turkey, Georgia, Hungary, Slovakia, Italy and some other countries. Relatively low levels of deposition are characteristic of the Scandinavian countries as well as the Russian Federation, Kazakhstan, and Malta.

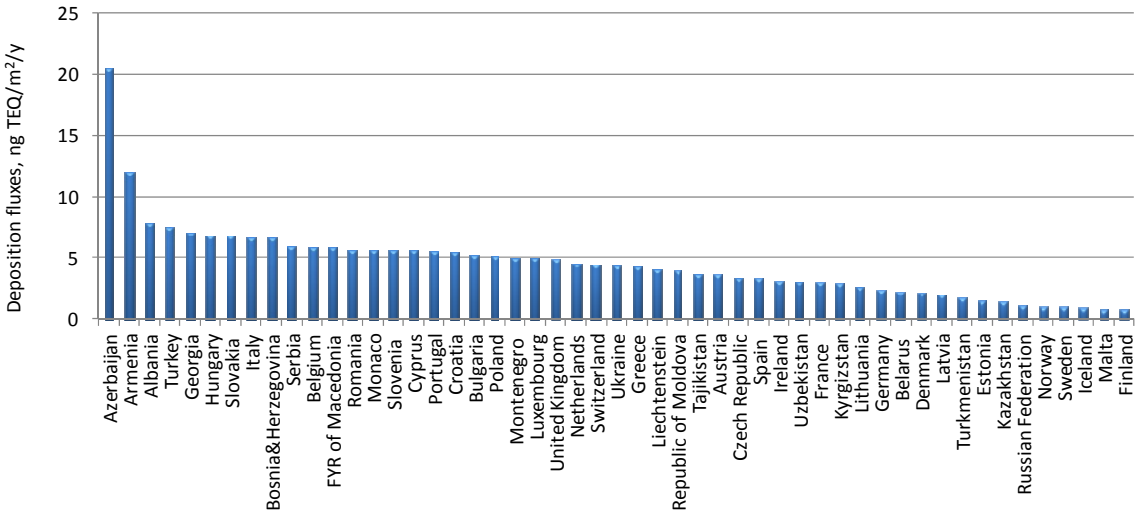


Fig. 5.11. *Annual deposition fluxes of PCDD/Fs in the EMEP countries calculated for 2015, ng TEQ/m^2/y*

Transboundary transport significantly contributes to the pollution of the EMEP countries by dioxins and furans. Model estimates of contributions of national emission sources and transboundary transport to

anthropogenic deposition of PCDD/Fs in the EMEP countries for 2015 are shown in the Fig. 5.12. ***For 45% of the EMEP countries the fraction of PCDD/F deposition to their territories, caused by the transport from external emission sources, exceeds the fraction of deposition from their national emissions.*** This can be noted for countries with relatively small territory or relatively low national emissions (e.g. Lichtenstein, Iceland, Luxembourg, and Cyprus). At the same time, for countries with relatively high PCDD/F emission or located far from major emission sources (e.g. Azerbaijan, Portugal, Turkey, Spain) contribution of transboundary transport is low.

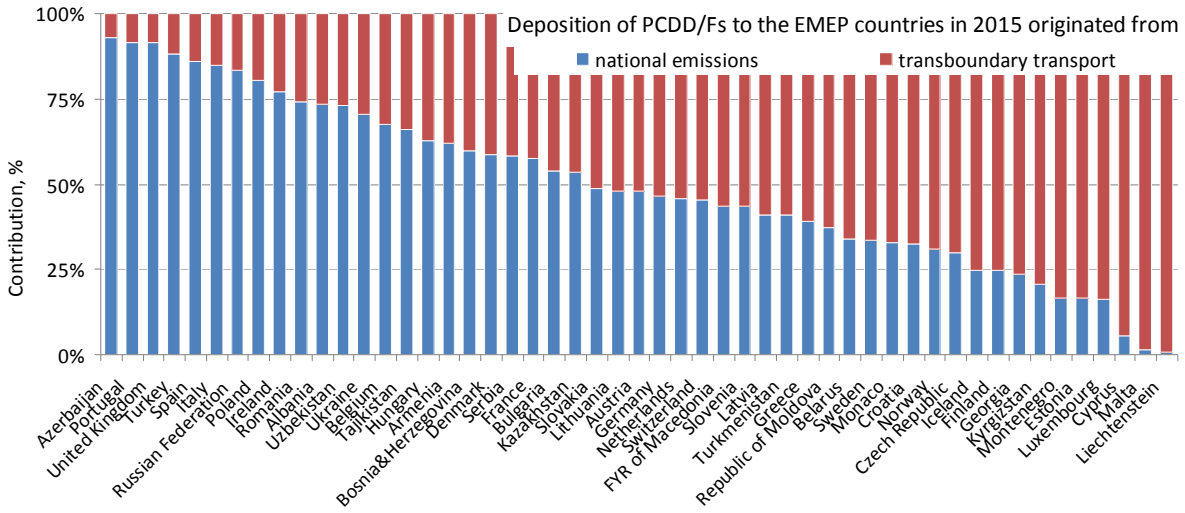


Fig. 5.12. *Relative contributions of national emission sources and transboundary transport to anthropogenic deposition of PCDD/Fs over the EMEP countries in 2015*

Transboundary transport of pollution can also be characterised by the ratio between the fractions of total anthropogenic deposition, originated from national emission sources of a country, and fallen out within and outside its boundaries (Fig. 5.13). ***For 18 countries the fraction of PCDD/Fs deposited outside their territories in 2015 is higher than the fraction of PCDD/Fs deposited to the countries.***

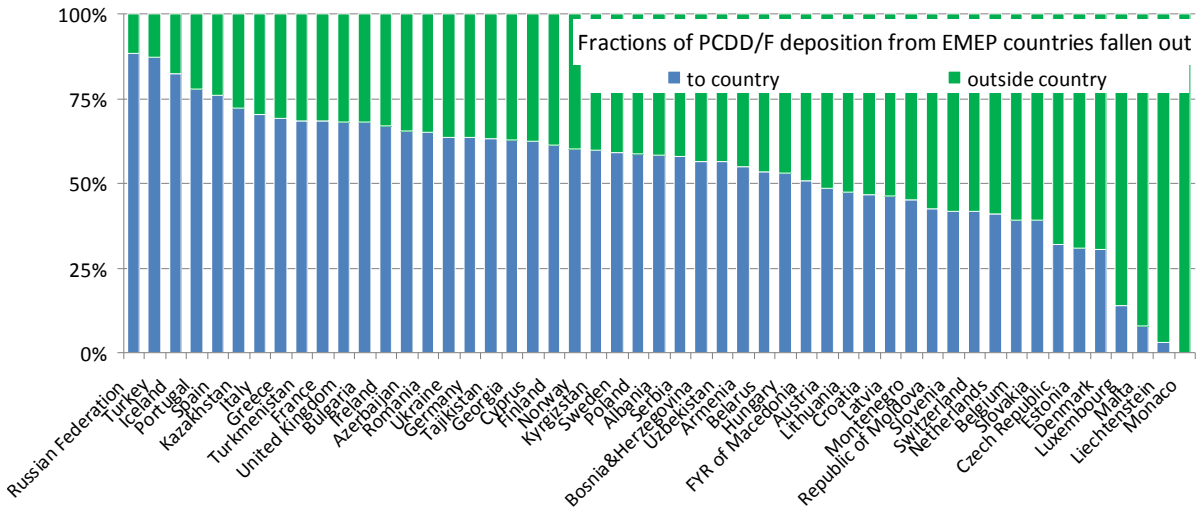


Fig. 5.13. *Fractions of PCDD/F deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in 2015*

Model estimates for 2015 also indicate that about 50% of annual PCDD/F deposition from anthropogenic emission sources within the EMEP region is provided by the emissions of 4 countries, namely, the Russian Federation, Turkey, Kazakhstan, and Ukraine.

5.4. POP pollution of the Arctic

Assessment of POP pollution levels in the EMEP region, carried out by MSC-E in framework of its operational activity, includes also information on the Arctic pollution by POPs. In accordance with the agreement between CLRTAP and AMAP, reached at the joint meeting held in Potsdam (Germany) in 2016, MSC-E prepared information on POP pollution levels in the area of the Arctic, covered by the EMEP region.

Spatial distributions of PCB-153 and PCDD/F annual mean air concentrations in the Arctic region, evaluated for 2015, are demonstrated in Fig. 5.14.

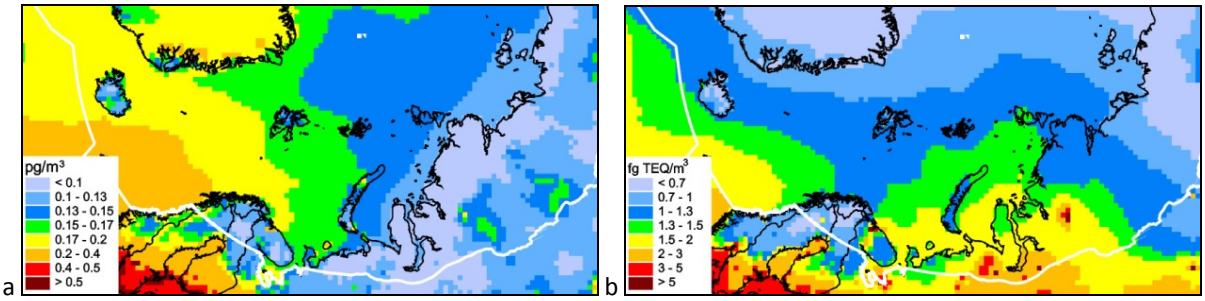


Fig. 5.14*. Spatial distribution of modelled annual mean air concentrations of PCB-153 (a), and PCDD/Fs (b) over the Arctic region covered by the EMEP domain calculated for 2015. White line denotes the boundary of the AMAP domain*

Model predictions indicate declining levels of pollution in northward direction. Elevated levels of PCB-153 air concentrations are noted for Greenland and Northern Atlantic. For PCDD/Fs relatively high concentrations can be seen western Siberia. Lower levels of PCB-153 and PCDD/F concentrations were estimated for the areas of the high Arctic.

Evaluation of pollution of the Arctic takes into account contributions of EMEP anthropogenic, EMEP secondary and non-EMEP sources. According to model estimates substantial contributions to PCDD/F and PCB-153 to deposition in the Arctic is made by non-EMEP emission sources (about 30%). Contribution of secondary sources of PCDD/Fs makes up 36%, and of PCB-153 – 62%. EMEP anthropogenic sources of PCDD/Fs and PCB-153 contributed to deposition levels in the Arctic 34% and 5%, respectively. Estimates of relative contributions to deposition over the Arctic region in 2015 from the anthropogenic emission sources of the EMEP countries are illustrated in Fig. 5.15.

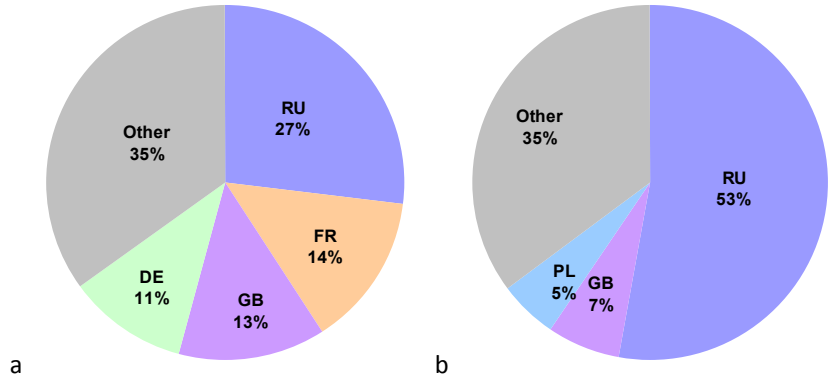


Fig. 5.15. *Relative contributions to annual deposition of PCB-153 (a) and PCDD/Fs (b) in 2015 over the part of the Arctic region covered by the EMEP domain, originated from anthropogenic emissions sources of the EMEP countries*

The largest contribution to PCB-153 and PCDD/F deposition is made by the emission sources of the Russian Federation (27% and 53%, respectively). Relatively high contributions to PCB-153 deposition are also made by emissions of France (14%), the United Kingdom (13%), and Germany (11%), and for PCDD/Fs by the emission sources of the United Kingdom (7%) and Poland (5%).

5.5. Comparison of modelling results with measurements

To verify modelling results on PCB-153 and HCB, comparison of modelled air concentrations with measurements of the EMEP monitoring network for 2015 was carried out. Air concentrations of PCDD/Fs are not regularly measured by the EMEP network. Model predictions of PCDD/F pollution levels on global and regional scales were evaluated against measurements, collected from various sources, at previous stages of work and can be found in the reports of the Centre [*Shatalov et al.*, 2015].

Monitoring of PCB-153 and HCB in 2015 was carried out at 7 EMEP monitoring sites located in Czech Republic, Finland, Norway, and Sweden (Chapter 1). Annual mean modelled and measured PCB-153 air concentrations are presented in Fig. 5.16a. ***The model reasonably well described observed spatial distribution of PCB-153. For most of the monitoring sites the difference between measured and modelled concentrations does not exceed a factor of 2. At the same time for monitoring site SE0014R model prediction underestimates annual mean observed concentration by more than a factor of 2.*** Similar underestimation was seen in the comparison of modelled and measured concentrations for 2014 [*Gusev et al.*, 2016]. The model showed lower air concentrations comparing to measurements for warm months, which might be explained by the underestimation of volatilization of PCB-153 for the location of the monitoring site. Further analysis of reasons of this disagreement is required in co-operation with national experts.

Modelled air concentrations of HCB for 2015 and available measurements of EMEP monitoring sites are shown in Fig 5.16b. It can be noted that amount of sites, provided measurements of HCB in air for 2015 and 2014, decreased from 12 in 2014 down to 6 in 2015. Comparison of modelled and measured air

concentrations of HCB for 2014 indicated that for more than a half of monitoring sites agreement between modelling results and measurements was within a factor of 2 [*Gusev et al.*, 2016].

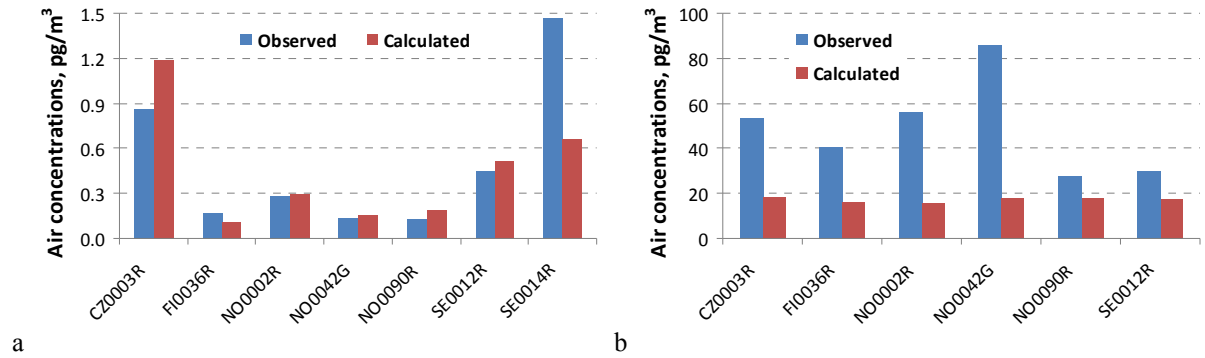


Fig. 5.16. *Comparison of annual mean modelled PCB-153 (a) and HCB (b) air concentrations with measurements of the EMEP monitoring sites for 2015, pg/m^3*

The model tended to underestimate observed HCB concentrations with more significant deviations (exceeding a factor of 2) for the sites CZ0003R, NO0002R, and NO0042R. Similar differences between the modelled and measured concentrations are obtained for 2015. Model predictions are lower than observed HCB air concentrations by 40% for SE0012R and by a factor of 4.7 for NO0042R.

The underestimation of HCB measurements by the model can be explained by the incompleteness of HCB emissions data reported by the EMEP countries. In particular, reported national inventory of HCB emissions in Germany indicated that some of the emission source categories might have releases of HCB (e.g. chemical industry, metal industry, and cement production). However, they are not currently covered in the inventory due to missing measurements leading to underestimation of total amount of HCB emitted to the atmosphere [*Strogies et al.*, 2017]. Along with emissions from industrial sources and waste incineration, residential wood combustion can be important source of HCB emissions. Experimental study of POP emissions from small scale combustion in Estonia indicated significant uncertainties in the available emission factors of HCB and the need of their further refinement to take into account particular conditions in the countries [*Maasikmets et al.*, 2016].

Additional reasons of the disagreement between modelled and observed HCB air concentrations can be attributed to the uncertainties of measurements of HCB air concentrations and deficiencies of modelling approach. As it was mentioned in Chapter 1, HCB is characterized by relatively high volatility that is known to cause the effect of breakthrough in course of measurements. Besides, relatively high levels of HCB air concentrations, observed at Scandinavian monitoring sites and in the Arctic, may indicate the influence of intercontinental transport and ongoing emissions in areas outside the EMEP region. Therefore updating of information on global HCB emissions and their temporal changes is needed. Finally, ***to improve assessment of HCB pollution in the EMEP countries, further analysis of available measurements and model predictions is required in co-operation with experts in monitoring and evaluation of HCB emissions.***

6. CO-OPERATION AND DISSEMINATION OF INFORMATION

Co-operation with subsidiary bodies, Parties to the Convention as well as various national and international organizations and programmes is essential for the assessment of POP pollution of the EMEP region.

6.1. Task Force on Measurements and Modelling (TFMM)

In the framework of cooperation with TFMM, MSC-E participated in 18th meeting of the Task Force held in May 2016 in Prague, the Czech Republic. The outcome of recent research activities and model developments, performed by the Centre, was presented and discussed. In particular, the progress in the transition of POP modelling to the new EMEP grid and necessary adaptation of GLEMOS modelling system was outlined. Pilot simulations of B(a)P pollution on the new EMEP grid showed in general better agreement with measurements comparing to the results for older EMEP grid. At the same time, for some of the stations model predictions noticeably differed from the observed pollution levels. Thus, the need of further refinement of GLEMOS model parameterization of gas-particle partitioning, degradation, and deposition processes was emphasized.

Preliminary results of country-specific case study of B(a)P pollution in Spain were demonstrated during the meeting. To analyze spatial variations of B(a)P concentrations over the country, fine resolution modelling using detailed emissions data and measurements of B(a)P at national monitoring network was carried out. Preliminary results of experimental model simulations with several emission scenarios and analysis of observed levels of PAH pollution indicated possible uncertainties in estimates of PAH emissions in Spain. Besides, the representative of Spain informed participants about ongoing refinement of national inventory of PAH emissions. Continuation of this activity is planned to include updating of the EMEP modelling results for Spain as well as results of national modelling and their inter-comparison. In addition, examination of uncertainties of national PAH emissions is appreciated.

As an extension of activities in the framework of country-scale pollution studies, it was suggested to perform analysis of B(a)P pollution levels in France and Poland. Important topic, relevant to further improvement of pollution assessment, can be the application of modelling and monitoring data fusion approaches in course of future meetings of the Task Force.

6.2. UNEP Stockholm Convention

Co-operation and exchange of information on POP pollution with UNEP Stockholm Convention has been continued. Activities related to the refinement of national POP emission inventories under the Stockholm Convention are of importance for the evaluation of emissions of the EMEP countries. In particular, national inventories are being revised due to further development of the UNEP Standardized Toolkit and recalculations of national emission data. Provided data comprise emissions of PCDD/Fs and in some cases of other POPs to the atmosphere and other compartments and this is additional source of information on POP emissions in the EMEP region. Besides, it is of importance for updating of the

scenario of global PCDD/F emission and for the assessment of environmental pollution on the global scale.

Collaboration with experts involved in the UNEP Stockholm Convention Global Monitoring Plan (GMP) activities on the collection and analysis of global-scale monitoring data on POP concentrations in air and other compartments is important for the evaluation of POP intercontinental transport and long-term changes of pollution. The GMP is established as a global framework for collection and analysis of information on POP content in the environmental media [*Mogulova and Priceputu*, 2016]. Ongoing work on GMP implementation in the countries on the basis of regional projects is planned to further improve capacities for sampling of POPs in air and surface water and produce new monitoring data.

6.3. Helsinki Commission

Long-range atmospheric transport is an important pathway of POPs to the Baltic Sea and other regional seas in the EMEP region. In cooperation with other EMEP Centres, MSC-E regularly performs model assessments of atmospheric load of various pollutants, including POPs, to the Baltic Sea. 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 2016 assessment of POP pollution load to the Baltic Sea included evaluation of long-term changes and source apportionment of B(a)P deposition fluxes, and verification of modelling results against measurements. Outcome of this study 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 [*Bartnicki et al.*, 2016].

Following various measures, performed in the EMEP countries in early 1990-s in order to reduce emissions from industrial sources, atmospheric deposition of B(a)P to the Baltic Sea decreased substantially (about 40%) in the period 1990-2000 (Fig. 6.1a). However the subsequent period, namely, 2001-2014, is characterized by stabilization of emissions and levels of deposition to the sea. According to the model estimates the most significant changes of deposition (> 50%) took place in the Western Baltic and the Bothnian Bay sub-basins. Other sub-basins of the Baltic Sea were characterized by somewhat lower decrease of deposition (about 30-40%).

Spatial distribution of annual B(a)P deposition fluxes to the Baltic Sea in 2014 is presented in Fig.6.1b. Relatively higher levels of atmospheric load were estimated for the western sub-basins (the Sound and the Western Baltic). Source apportionment of B(a)P deposition indicates that HELCOM countries contributed about 60% to the pollution of the Baltic Sea. Among these countries the largest contribution to total deposition was made by Poland (24%) and Germany (10%). Significant input of B(a)P was also originated from other EMEP countries (about 40%).

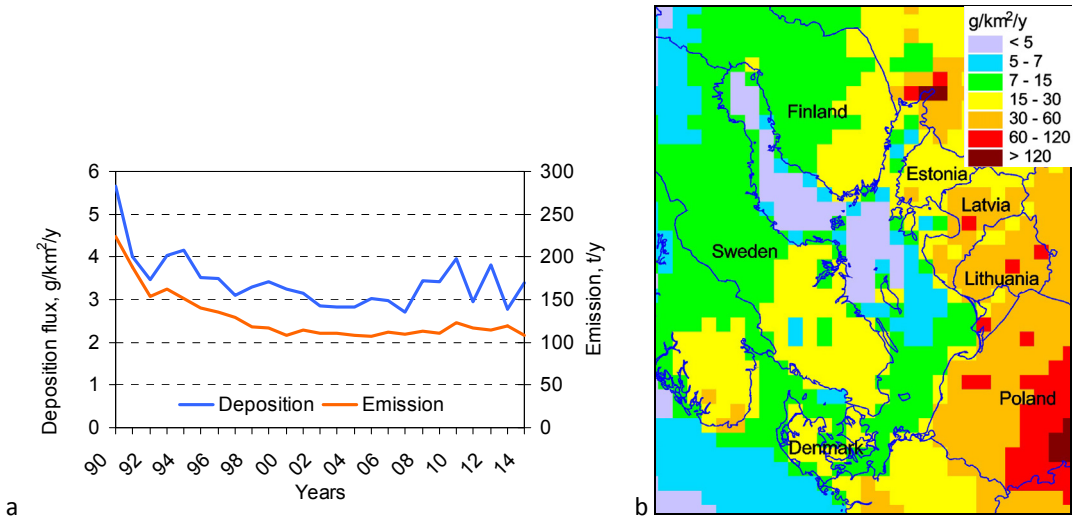


Fig. 6.1. *Changes of annual B(a)P emissions of HELCOM countries and annual deposition to the Baltic Sea in the period 1990-2014 (a) and spatial distribution of B(a)P deposition fluxes in 2014, g/km²/y (b)*

Seasonal variations of B(a)P pollution levels are characterized by significant difference between the cold and warm periods of the year. In particular, according to measurements and model estimates this difference can be up to an order of magnitude and higher (Fig. 6.2 a,b). Large seasonal variability of B(a)P pollution is caused by several factors including intra-annual changes of emissions, especially from the residential combustion sector, and effect of more intensive degradation during the warmer period. Modelled and observed levels of concentrations and deposition fluxes closely describe seasonal changes of B(a)P pollution for most part of the year 2014. At the same time, model predictions underestimate observed pollution levels for January, which indicates the need of further refinement of seasonal changes of B(a)P emissions.

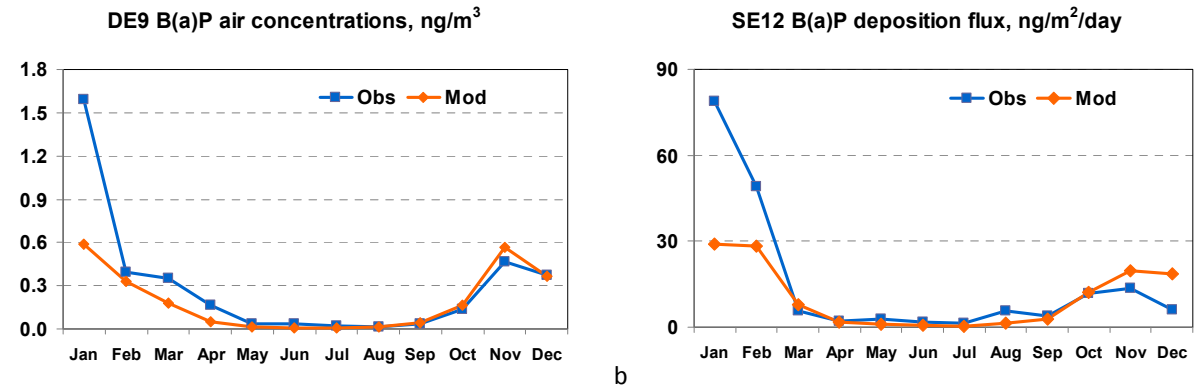


Fig. 6.2. *Comparison of monthly mean modelled and observed B(a)P air concentrations (ng/m³) for the EMEP site DE9 (a) and monthly mean deposition fluxes (ng/m²/day) for the EMEP site SE12 (b)*

Results of the model evaluation of B(a)P atmospheric input to the Baltic Sea were discussed at the meeting of the HELCOM PRESSURE group held in Warsaw in 2016. Particular attention was paid to the stabilization of long-term changes of B(a)P air concentrations and deposition fluxes during the recent decade as well as to the contribution of residential combustion sources to the pollution levels in the Baltic Sea region.

6.4. Parties to the Convention

MSC-E continued collaboration with Parties to the Convention in the field of POP pollution assessment. Model assessment of PAH pollution with fine spatial resolution in the framework of country-specific case studies is started for Spain in accordance to the recommendation of the second joint session of the Working Group on Effects and the Steering Body to EMEP. Objective of this study is to perform analysis of B(a)P pollution levels in Spain using national emission data with fine resolution, measurements of monitoring network of Spain and results of EMEP GLEMOS model and CHIMERE model. Preliminary outcome of the study is presented in this report and directions of further work are outlined. Results of this activity are also important for the ongoing transition of POP modelling to the new EMEP grid with finer spatial resolution.

To support national study of POP pollution in Italy, regional-scale modelling for the evaluation of POP air concentrations in the Mediterranean region was carried out. Results of the EMEP model simulations will be used as information on boundary concentrations in national-scale modelling, performed by National Agency for New Technologies, Energy and the Environment of Italy (ENEA). The outcome of the activities of ENEA is used by national authorities of Italy to analyze and improve national environmental and human health protection policy.

In accordance with the agreement between ENEA and MSC-E, the Centre carried out model assessment of pollution levels of selected POPs (PAHs, PCDD/Fs, PCB-153, and HCB) in the EMEP domain for 2015 and generated gridded data on air concentrations with high temporal resolution for the agreed area surrounding Italy. Modelling results were verified by the comparison of modelled air concentrations with observed air concentrations for 2015, derived from EMEP/CCC database (EBAS). The work was carried out under financial support of ENEA.

Along with this, the Centre collaborated with national experts from Karlsruhe Institute of Technology (KIT) in Germany developing a model system MoRE (Modelling of Regionalized Emission) for the evaluation of emissions of different pollutants, including heavy metals, nutrients, PAHs, to surface water bodies. MoRE is an instrument for modelling emissions into surfaces water bodies and for identifying relevant emission pathways. MoRE is based on the model concept MONERIS [*Behrendt et al.*, 2000] and has been adapted for the modelling of pollutant emissions [*Fuchs et al.,* 2012]. Following the request of national experts, MSC-E provided gridded data on atmospheric deposition fluxes of B(a)P to inland waters of the country for 2011-2014.

MAIN CHALLENGES AND DIRECTIONS OF FUTURE RESEARCH

This Status Report presents a summary of current activities carried out by EMEP Centres, MSC-E and CCC, in the field of POP pollution assessment. Main emphasis in the report is given to the evaluation of PAH pollution levels at regional and country scales. Besides, the progress in the transition of operational POP modelling to the new EMEP, evaluation of POP pollution levels, and co-operation with EMEP countries and international organizations is described. Directions of future work and main challenges, need to be addressed in accordance with the recommendations of the Ad hoc Policy Review Group [ECE/EB.AIR/WG.5/2017/3], are outlined below in this section.

- Adaptation of the GLEMOS model to the new EMEP grid and pilot modelling with finer spatial resolution, carried out on the example of PAHs (B(a)P), has shown generally better agreement between the modelled and observed pollution levels comparing to the results for the old EMEP grid. At the same time, for better performance of the GLEMOS model, further refinement of model parameterizations of gas-particle partitioning, degradation, and deposition processes, taking into account interaction of PAHs with aerosols and atmospheric reactants, is needed.

- High levels of B(a)P air concentrations and exceedances of EU target value, especially in urban areas of the EMEP countries, has been indicated as an important air pollution issue during the recent Steering Body meeting. Country-specific case studies on the basis of detailed emission and monitoring data can be used for the analysis of PAH pollution and population exposure. Besides, these studies can indicate potential uncertainties in emission inventories as well as in modelling approach. Detailed assessment of PAH pollution is planned to be continued for several countries, namely, Spain, France, and Poland, on the basis of fine resolution modelling, monitoring and modelling data fusion approaches, and analysis of pollution levels in urban areas in co-operation with national experts and TFMM.

- In order to improve model estimates of POP cycling in the environment and influence of secondary emission sources on the pollution levels in the EMEP region, further refinement of GLEMOS model parameterizations of POP air-surface exchange is needed. This work is planned to include evaluation of PCDD/F and PCB intercontinental transport and fate, applying multi-compartment modelling, as well as analysis of factors affecting POP fate and distribution in the terrestrial and aquatic environmental compartments.

- Quality of national inventories of POP emissions, reported by EMEP countries, is one of the most important factors affecting model assessment of transboundary transport of pollution. Though the completeness and consistency of officially submitted data on POP emissions is gradually improving, there are still significant uncertainties in estimates of POP releases from specific source categories (e.g. residential combustion, biomass burning), speciation of POP emissions (e.g. of PAHs, PCBs, PCDD/Fs), as well as their spatial distribution and temporal variations. Besides, refinement of information on intra-annual variability and vertical distribution of POP emissions for individual source categories is required in co-operation with TFEIP, CEIP, and national experts.

- Comparability and consistency of EMEP monitoring data on POPs is an important issue for the analysis of spatio-temporal variability of POP pollution levels. Existing differences in sampling and analytical methodologies can be a complicating factor for the comparison of data from different sampling sites. For instance, a special challenge is monitoring of HCB in air, which has been shown to undergo breakthrough in high volume air samples. This effect may lead to underestimation of its concentrations and to limited comparability across the sites within the EMEP network. Thus, there is a need for continued focus on harmonization of sampling strategies to enhance the spatial comparability of measurements of this pollutant as well as other measured POPs.

- Collaboration and exchange with information on POPs between EMEP and international organization (e.g. UNEP Stockholm Convention, AMAP, HELCOM, and OSPAR) is essential for further progress in the assessment of POP pollution in the EMEP region. Complementary use of POP monitoring data for the atmosphere obtained by active and passive sampling, and measurements of POPs in surface media (e.g. soil and water compartments) is of importance for the analysis of POP spatial and temporal trends. Thus, strengthening cooperation with relevant activities of international organizations to collect global-scale measurements of POP pollution levels is highly appreciated.

REFERENCES

Aas W., and Bohlin-Nizzetto P. [2017] Heavy metals and POP measurements 2015, Norwegian Institute for Air Research. EMEP/CCC-report x/2017.

Barber J.L., Sweetman A.J., van Wijk D., Jones K.C. [2005] Hexachlorobenzene in the global environment: emissions, levels, distribution, trends and processes. *Sci. Tot. Environ*., 349, 1-44.

Bartnicki J., Gusev A., Aas W., Benedictow A. [2016] Atmospheric Supply of Nitrogen, Cadmium, Mercury, Benzo(a)pyrene and PBDEs to the Baltic Sea in 2014. Summary Report for HELCOM. MSC-W Technical Report 1/2016.

Behrendt H., Huber P., Kornmilch M., Opitz D., Schmoll O., Scholz G., Uebe R. [2000] Nutrient Emissions into River Basins. UBA-Texte, 23/00, 1st ed; Federal Environment Agency (Umweltbundesamt): Berlin.

Bi et al. [2005] Size distribution of n-alkanes and polycyclic aromatic hydrocarbons (PAHs) in urban and rural atmospheres of Guangzhou, China. *Atmospheric Environment* 39 (2005) 477–487.

Breivik K., Sweetman A., Pacyna J.M., Jones K.C. [2007] Towards a global historical emission inventory for selected PCB congeners - A mass balance approach-3. An update. Science of the Total Environment, vol. 377, pp. 296-307.

Denier van der Gon H.A.C., van het Bolscher M., Visschedijk A.J.H. and P.Y.J.Zandveld [2005] Study to the effectiveness of the UNECE Persistent Organic Pollutants Protocol and costs of possible additional measures Phase I: Estimation of emission reduction resulting from the implementation of the POP Protocol, TNO report B&O-A R 2005/194.

EMEP/CCC [2014] Manual for sampling and chemical analysis. Kjeller, Norwegian Institute for Air Research (EMEP/CCC-Report 1/2014) URL: http://www.nilu.no/projects/ccc/manual/index.html [Accessed June 2017].

EMEP/EEA Guidebook [2013] EMEP/EEA air pollutant emission inventory guidebook. http://www.eea.europa.eu//publications/emep-eea-guidebook-2013.

EU [2004] Directive 2004/107/EC of the European Parliament and of the Council of 15 Dec. 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air. Off. J. Eur. Union., L 23, 3-16.

Fuchs S., Wander R., Rogozina T., Hilgert S., Scherer U. [2012] Methodical Optimization of Model Approaches on Pollutant Balancing in River Basin Districts to Promote the Implementation Strategy for the Water Framework Directive. Final report on the Project FZK: 370 822 202/01 (unpublished).

Gusev A., V.Shatalov, and O.Rozovskaya [2014] PilotModelling of PCDD/F Transport and Fate on Global Scale and within the European Region. *Organohalogen Compounds*. Vol. 76, 958-961.

Gusev A., O.Rozovskaya, V.Shatalov, W.Aas, P. Nizzetto, M. Modlík, P. Machálek, R. Srnenský [2016] Persistent Organic Pollutants: assessment of transboundary pollution on regional and global scales. EMEP Status Report 3/2016.

Halse A.K., Schlabach M., Eckhardt S., Sweetman A.J., Jones K.C., Breivik K. [2011] Spatial variability of POPs in European background air. *Atmos. Chem. Phys*., 11: 1549-1564.

Hays M.D., N. D. Smith, J. Kinsey, Y. Dong, and P. Kariher [2003] Polycyclic aromatic hydrocarbon size distributions in aerosols from appliances of residential wood combustion as determined by direct thermal desorption—GC/MS. *Aerosol Science* 34 (2003) 1061–1084.

Hung H., Kallenborn R., Breivik K., Su Y., Brorstrøm-Lunden E., Olafsdottir K., Thorlacius J.M., Leppanen S., Bossi R., Skov H., Manø S., Stern G., Sverko E., Fellin P. [2010] Atmospheric monitoring of organic pollutants in the arctic under the arctic monitoring and sassessment programme (AMAP). *Sci. Tot. Environ*., 408, 2854-2873.

IIR of Austria [2017] Umweltbundesamt. Environment agency Austria. Austria's Informative inventory report (IIR) 2017. Submission under the UNECE CLRTAP. Report Rep-0609, Vienna 2017.

IIR of Azerbaijan [2017] The Ministry of Ecology and Natural Resources. Informative inventory report of Azerbaijan 2017. Submission under the UNECE CLRTAP.

Ilyin I., O.Rozovskaya, O.Travnikov. W. Aas [2017] Assessment of heavy metal transboundary pollution on regional and national scales, transition to the new EMEP grid. EMEP Status Report 2/2017.

Ilyin I., Rozovskaya O., Travnikov O., Varygina M., Aas W. and Pfaffhuber K.A. [2016] Assessment of heavy metal transboundary pollution, progress in model development and mercury research. EMEP Status Report 2/2016, 75p.

Ilyin I., O.Rozovskaya, O.Travnikov, M.Varygina, W.Aas [2014] Heavy Metals: Transboundary Pollution of the Environment. EMEP Status Report 2/2014.

Allen J. O., N. M. Dookeran, K. A. Smith, A. F. Sarofim, K. Taghizadeh, and A. L. Lafleur [1996] Measurement of Polycyclic Aromatic Hydrocarbons Associated with Size-Segregated Atmospheric Aerosols in Massachusetts, *Environ. Sci. Technol*. 1996, 30, 1023-1031.

Keyte I.J., Harrison R.M., Lammel G. [2013] Chemical reactivity and long-range transport potential of polycyclic aromatic hydroarbons – a review, *Chem. Soc. Rev*.,42 (24), 9333 – 9391.

Lobscheid A.B., McKone T.E., Vallero D.A. [2007] Exploring relationships between outdoor air particulate–associated polycyclic aromatic hydrocarbon and PM2.5: a case study of benzo(a)pyrene in California metropolitan regions, Atmospheric Environment, 41, pp. 5659-5672.

Ma J., Hung H., Tian C., Kallenborn R. [2011] Revolatilization of persistent organic pollutansts in the Arctic induced by climate change. *Nat. Clim. Change*, 1, 255-260.

Maasikmets M., H. Kupri, E. Teinemaa, K. Vainumae, T. Arumae, O. Roots, V. Kimmel [2016] Emissions from burning municipal solid waste and wood in domestic heaters. *Atmospheric Pollution Research*, 7, 438-446.

Magulova K. and A. Priceputu [2016] Global monitoring plan for persistent organic pollutants (POPs) under the Stockholm Convention: Triggering, streamlining and catalyzing global POPs monitoring. *Environmental Pollution,* 217, (2016), Pages 82-84.

Melymuk L., Bohlin P., Sáňka O., Pozo K., Klánová J. [2014] Current challenges in air sampling of semivolatile organic contaminants: sampling artifacts and their influence on data comparability. *Environ. Sci. Technol*., 48, 14077-14091.

Melymuk L., Bohlin-Nizzetto P., Prokes R., Kukucka P., Klánová J. [2016] Sampling artifacts in active air sampling of semivolatile organic contaminants: comparing theoretical and measured artifacts and evaluating implications for monitoring networks. *Environ. Poll*., 217, 97-106.

Pacyna J.M., K. Breivik J. Münch J. Fudala [2003] European atmospheric emissions of selected persistent organic pollutants, 1970-1995. *Atmos. Environ*., vol. 37, pp.S119-S131.

Perea J.L. [2016] Informative inventory report of Spain. 1990-2014. Submission to the Secretariat of the Geneva Convention and EMEP Programme. Spanish Ministry of Agriculture, Food and Environment. March, 2016.

Phoothiwut S. and S. Junyapoon [2013] Size distribution of atmospheric particulates and particulate-bound polycyclic aromatic hydrocarbons and characteristics of PAHs during haze period in Lampang Province, Northern Thailand. Air Qual Atmos Health (2013) 6:397–405.

Querol et al. [2016] PM speciation and source apportionment. Report 4, 12/2016. AIRUSE LIFE 11 ENV/ES/584.

Schlabach M., Aas W., Hung H. M., Rania F., and Su Y. [2011] NCP, AMAP and EMEP POP laboratory comparison 2010. Norwegian Institute for Air Research. EMEP/CCCReport 7/2011.

Shatalov V., Ilyin I., Gusev A., Rozovskaya O., Travnikov O. [2015] Heavy Metals and Persistent Organic Pollutants: development of multi-scale modelling and trend analysis methodology. EMEP/MSC-E Technical report 1/2015.

Shatalov V., Ilyin I., Gusev A., Rozovskaya O., Travnikov O. [2014] Heavy Metals and Persistent Organic Pollutants: Model Assessment of Pollution and Research Activities. EMEP/MSC-E Technical report 4/2014.

Shatalov V., Ilyin I., Gusev A., Rozovskaya O., Sokovykh V. and Travnikov O. [2013] Heavy Metals and Persistent Organic Pollutants: Model Assessment of Pollution and Research Activities. MSC-E Technical Report 1/2013. 68 p.

Shatalov V., A. Gusev, S. Dutchak, O. Rozovskaya, V. Sokovykh, N. Vulykh, W. Aas, K. Breivik [2010] Persistent Organic Pollutants in the Environment. EMEP Status Report 3/2010.

Shen H., Y. Huang, R. Wang, D. Zhu, W. Li, G. Shen, B. Wang, Y. Zhang, Y. Chen, Y. Lu, H. Chen, T. Li, K. Sun, B. Li, W. Liu, J. Liu, and S. Tao [2013] Global Atmospheric Emissions of Polycyclic Aromatic Hydrocarbons from 1960 to 2008 and Future Predictions. *Environ. Sci. Technol*., 47 (12), 6415–6424.

Strogies M., P. Gniffke, K. Hausmann, K. Juhrich [2017] German Informative Inventory Report 2017 (IIR 2017), Umwelt Bundesamt, (http://iir-de.wikidot.com/welcome:welcome).

Su Y. and Hung H. [2010] Inter-laboratory comparison study on measuring semi-volatile organic chemicals in standards and air samples. *Environ. Poll*., 158, 3365-3371.

Tang X. L., X. H. Bi, G. Y. Sheng, J. H. Tan and J. M. Fu [2006] Seasonal Variation of the Particle Size Distribution of n-alkanes and Polycyclic Aromatic Hydrocarbons (PAHs) in Urban Arosol of Gangzhou, China, Environmental Monitoring and Assessment 117: 193–213.

Theakston F. [2000] Air quality guidelines for Europe. Second edition. WHO Regional Publications, European Series, No. 91, ISBN 92 890 1358 3.

Tista M., Mareckova K. and R.Wankmueller [2016] Methodologies applied to the CEIP GNFR gap-filling 2016. Part II: Persistent organic pollutants (Benzo(a)pyrene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Indeno(1,2,3-cd)pyrene, Dioxin and Furan, Hexachlorobenzene). Technical report CEIP 02/2016.

Tista M., Wankmueller R. and K.Mareckova [2017] Methodologies applied to the CEIP GNFR gap-filling 2017. Part II: Persistent organic pollutants (Benzo(a)pyrene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Indeno(1,2,3-cd)pyrene, Dioxin and Furan, Hexachlorobenzene). Technical report CEIP 02/2017.

Travnikov O. and Ilyin I. [2005] Regional Model MSCE-HM of Heavy Metal Transboundary Air Pollution in Europe. EMEP/MSC-E Technical Report 6/2005. (http://www.msceast.org/reports/6_2005.pdf).

Treger Y.A. [2011] Persistent organic pollutants. Problems and ways of their decisions (in russian). Вестник МИТХТ 6/5, pp.87-97.

Van Drooge, B., Fontal, M., Bravo, N., Fernández, P., Fernández, M., Muñoz-Arnanz, J., Jiménez, B., Grimalt, J., [2014] Seasonal and spatial variation of organic tracers for biomass burning in PM1 aerosols from highly insolated urban areas. *Environ. Sci. Pollut. Res. Int.* 21, 11661–70.

Van Drooge B.L., Grimalt J.O. [2015] Particle size-resolved source apportionment of primary and secondary organic tracer compounds at urban and rural locations in Spain. *Atmos. Chem. Phys*. 15, 7735–7752.

Venier M., H. Hung, W. Tych, and R.A. Hites [2012] Temporal Trends of Persistent Organic Pollutants: A Comparison of Different Time Series Models, *Environ. Sci. Technol*., 46, 3928−3934.

Venkataraman C., S. Thomas and P. Kulkarni [1999] Size Distributions of Polycyclic Aromatic Hydrocarbons – gas/particle partitioning to urban aerosols, *J. Aerosol Sci. Vol*. 30, No. 6, pp. 759Ð770.

Venkataramant C. and S. K. Friedlander [1994] Size Distributions of Polycyclic Aromatic Hydrocarbons and Elemental Carbon. 2. Ambient Measurements and Effects of Atmospheric Processes, *Environ. Sci. Technol.* 1994, 28, 563-572.

Viana et al. [2016] Contribution of residential combustion to ambient air pollution and greenhouse gas emissions. ETC/ACM Technical Paper 2015/1. February 2016

Wang B., Fiedler H., Huang J., Deng S., Wang Y. and G.Yu [2016] A Primary Estimate of Global PCDD/F Release Based on the Quantity and Quality of National Economic and Social Activities (Supporting Information). *Chemosphere* 151:303-9. doi: 10.1016/j.chemosphere.2016.02.085.

Zhang Y. and S. Tao [2009] Global atmospheric emission inventory of polycyclic aromatic hydrocarbons (PAHs) for 2004. *Atmos Environ*, 43 (2009) pp 812–819.

ANNEX A

PROPOSALS FOR THE WORKPLAN FOR 2018-2019 AND UPDATED MANDATE OF MSC-E

This annex presents proposals of MSC-E for the future mandatory work and research activity on assessment of heavy metal and POP pollution level assessment. There are some items e.g., country-scale assessment of heavy metals and POPs in Russia, which inclusion in the proposal is under discussion.

Proposal of MSC-E Mandate

(a) Prepare data on anthropogenic emissions of heavy metals and POPs on regional (EMEP domain) and global scales including auxiliary parameters (e.g. emission height, temporal variation, chemical composition etc.) as input for operational modelling based on gridded emission dataset provided by Centre for Emission Inventories and Projections (CEIP) and expert estimates;

(b) Generate meteorological data for operational and research modelling on different scales (global, regional) based on datasets of the European Centre for Medium-Range Weather Forecasts (ECMWF);

(c) Prepare input data required for modelling of heavy metals and POPs on regional and global scales, including wind suspension of mineral dust as well as atmospheric concentrations of chemical reactants and particulate matter;

(d) Collect and process measurement data for evaluation of model performance from various monitoring networks and databases (e.g. EBAS, AirBase, GMOS, UNEP SC GMP Data Warehouse, etc.);

(e) Update the modelling tools with new findings and improved parameterizations developed by the Centre in its research activities in accordance with the bi-annual work-plan and cooperation with scientific community;

(f) Perform simulations of heavy metals and POPs dispersion on a global scale for evaluation of intercontinental transport and initial and boundary conditions for regional-scale assessment of pollution levels;

(g) Perform further testing and evaluation of model performance in simulations of air concentration and deposition levels as well as source-receptor relationships of heavy metals and POPs on the new EMEP grid;

(h) Perform operational model assessment of heavy metal (Pb, Cd, and Hg) and POP (PAHs, PCBs, PCDD/Fs, and HCB) pollution levels over the EMEP domain;

(i) Perform quality assurance and quality control of modelling results through evaluation against measurements from the EMEP and other monitoring networks;

(j) Provide support of Parties to the Convention with use of the model assessment results and access to the modelling tools.

(k) Contribute to the work of the subsidiary bodies and task forces:

- Task Force on Measurements and Modelling (TFMM): Continue cooperation on the evaluation of model performance and improvement of modelling approaches in the field of heavy metal and POP pollution assessment; present and discuss results of the national scale case studies and other research activities on heavy metal and POP pollution with fine resolution;
- Task Force on Hemispheric Transport of Air Pollution (TFHTAP): Cooperate on assessment of intercontinental transport of Hg and POPs and its impact on pollution levels in the EMEP countries;
- Working Group on Effects: Continue collaboration with ICP-Vegetation on evaluation of heavy metal pollution levels in Europe using modelling results and measurements in mosses and develop cooperation with other International Cooperative Programmes; provide support of the Coordination Centre for Effects (CCE) with information on ecosystem-specific deposition heavy metals and POPs for assessment of critical load exceedances; contribute to the Task Force on Health with information on toxic substances (PAHs, PCDD/Fs and others);

(l) Cooperate on dissemination of information and data exchange with international bodies including UNEP, AMAP, Stockholm Convention, Minamata Convention, HELCOM, etc.;

(m) Prepare annual Status Reports and individual country reports for the EMEP countries and make results of model calculations available online at the MSC-E website; develop and maintain a website in Russian to facilitate access to information by countries in Eastern Europe, the Caucasus and Central Asia;

(n) Report on its activities and deliverables to the Steering Body to EMEP and Working Group on Effects and participate in annual meetings of the relevant Task Forces (TFMM, TFHTAP).

Proposal for the workplan of MSC-E future research activities for 2018-2019

Title	Description/Objectives	Deliverables	Collaboration
Country-scale assessment of HM and POP pollution (Case Studies)	1) Assessment of country-specific HM and POP pollution using detailed national emission and monitoring data for Poland (Cd, BaP), Spain (BaP), France (BaP), UK (Pb, Cd - ?), Russia (Pb, Cd - ?). 2) Evaluation of pollution levels in high-emission and high-impact (e.g. urban) areas using data fusion approaches. 3) Analysis of factors affecting quality of HM and POP pollution modelling with fine spatial resolution.	1) Model estimates of pollution levels with high spatial resolution; 2) Analysis of discrepancies between modelled and observed pollution levels; 3) Estimates of contributions of LPS, national, regional, and non-EMEP emissions as well as key source categories to pollution; 4) Estimates of air pollution levels in urban areas based on combined use of modelling results and measurements 5) Recommendations for improvement of model assessment quality in the EMEP region. 6) Overview of main results of Case Studies (since 2011)	TFMM, Poland (IEP-NRI), Czech Republic (CHMI), Spain (CIEMAT), France (INERIS), UK (CEH), Russia (IGCE)
Evaluation of multi-compartment intercontinental transport of Hg and POPs	1) Assessment of Hg, PCDD/Fs and PCB pollution of the EMEP countries and other regions (e.g. the Arctic) from regional and global sources 2) Analysis of the key factors affecting POP and Hg accumulation in and exchange between the environmental media 3) Evaluation of secondary emissions of selected POPs (PCDD/Fs, PCBs) and Hg and their contribution to pollution of the EMEP countries.	1) Source apportionment of Hg, PCDD/Fs, and PCB pollution levels in the EMEP countries including regional and global sources 2) Evaluation of Hg deposition to sensitive ecosystems (e.g. wetlands, in-land waters and catchment areas, marginal seas, the Arctic) 3) Contribution to EMEP Global Mercury Assessment 2018 4) Development of model parameterizations for Hg aquatic chemistry and air-surface exchange processes 5) Evaluation and refinement of model parameterizations of PCDD/F and PCB processes in terrestrial and aquatic compartments	TFHTAP, TFMM, WGE/ICP-Waters, UNEP, AMAP, Stockholm Convention, Minamata Convention
Analysis of effectiveness of implementation of Protocol on POPs	Contribution to evaluation of stricter measures for mitigation of B(a)P pollution levels	1) Analysis of long-term trends of B(a)P pollution levels in the EMEP countries 2) Assessment of B(a)P pollution levels with focus on densely populated areas and comparison with air quality guidelines (health effects) 3) Evaluation of the key source categories contribution to B(a)P pollution levels 4) Projections of future B(a)P pollution levels on the basis of emission scenarios (if available)	TFTEI