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Persistent Organic Pollutants in the Environment

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EXECUTIVE SUMMARY

In accordance with the EMEP Work-plan for 2010, Meteorological Synthesizing Centre East (MSC-E) and Chemical Coordinating Centre (CCC) continued the investigations of pollution of the EMEP region by persistent organic pollutants (POPs). The outcomes of these studies are summarized in this Status Report.

Investigation of hemispheric/global transport of POPs is an important field of activity aimed at evaluation of the contribution of global emission sources to the pollution levels with the EMEP domain. In the framework of this activity MSC-E has continued to study POP intercontinental transport and to develop the Global EMEP Multi-media Modelling System (GLEMOS) for POPs. Pilot global-scale model simulations demonstrated the operability of this system and indicated the areas of further improvement. Elaboration of emission scenarios for hemispheric HCB modelling permitted to improve the agreement of model estimates with available measurements of HCB air concentrations. Modelling experiments indicated essential role of historic emissions for the evaluation of current HCB pollution levels.

The work under the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) to assess intercontinental transport of pollutants is of particular importance for the elaboration of the EMEP global modelling approach. Among the priorities of the Task Force activity in the recent years were the multi-model experiments to quantify global scale source-receptor relationships and the compilation of the HTAP 2010 Assessment Report. Major activities of the MSC-E in this respect were connected with the preparation of contributions to the HTAP Assessment 2010, coordination of the TF HTAP multi-model experiments for POPs, and participation in the discussions of the Assessment Report content on POPs.

Evaluation of PAH, PCDD/F, PCB and lindane pollution levels and transboundary transport within the EMEP domain for 2008 was carried out on the basis of measurements, emission data, and modelling.

In 2008 there were twenty EMEP monitoring sites measuring POPs, which were three more comparing to the previous year 2007, and twelve of them conducted measurements of POP concentrations in both compartments (air and precipitation). In addition to the EMEP passive air sampling campaign, the Global Atmospheric Passive Sampling (GAPS) network is a key global program producing comparable global-scale data for POPs. Besides, EMEP has started a new laboratory intercomparison together with the Northern Contaminants Program (NCP) in Canada coordinated by the Laboratory Services Branch (LaSB) of the Ontario Ministry of the Environment (MOE).

Emission data for model assessment of POP long-range transport within the EMEP region were compiled on the basis of official information submitted by the EMEP Centre on Emission Inventories and Projections (CEIP) and unofficial expert estimates. Gridded emission datasets for modelling purposes were prepared by MSC-E. For the evaluation of intercontinental transport emissions of PCDD/Fs, PCB-153 and γ-HCH within the Northern Hemisphere were compiled.

Evaluation of environmental pollution in the EMEP region was carried out on the basis of regional/hemispheric models.

Modelling and measurement information showed that the highest levels of B[a]P concentrations were characteristic of the central, eastern, and southern parts of Europe. The most significant concentrations were obtained for the eastern part of the Ukraine. Lower concentrations were seen in the Northern and Western Europe. Air concentrations of B[a]P are subject to strong seasonal variability. The information on seasonal variability of emissions from the EMEP countries is hence highly desirable.

Model simulations of PCDD/Fs were performed for the mixture of 17 toxic congeners based on the physical-chemical properties of PCDD/F indicator congener 2,3,4,7,8-PeCDF. The most significant concentrations were found in some regions of the Southern and Eastern Europe. Similar to B[a]P, seasonal variations of emissions and variability of air concentrations inside the grid cells can essentially affect the results.

Modelling of PCB-153 was made on the basis of the global emission inventory prepared by *K. Breivik et al.* [2007]. Modelling results indicated elevated levels of PCB-153 concentrations in several regions of the Western and Southern Europe (western Germany, the Netherlands, northern France and northern Italy).

The environmental levels of lindane (γ-HCH) have been significantly decreased within the European region during two recent decades due to the reduction of its application. In particular, non-zero lindane emissions for at least one year in the period 2000 – 2008 were reported only by five European countries: the United Kingdom, Spain, Belgium, Croatia, and Romania. Elevated annual mean air concentrations of γ-HCH (50 pg/m^3 and above) in 2008 were predicted by the model for the United Kingdom and Spain. Reduced influence of primary γ-HCH emission sources increase the importance of the contributions of secondary emissions and intercontinental transport to the contamination of the EMEP region.

This year the country-to-country matrices for 2008 were calculated for PAHs, PCDD/Fs, and, for the first time, for PCBs. The contribution of transboundary transport in the EMEP countries was typically 20 – 70% of deposition originated from primary anthropogenic emission sources, depending on country size, geographical location and domestic emissions. For some countries the contribution of non-EMEP sources exceeded 20%. Model simulations showed significant contribution of secondary emission sources (re-emission) to the contamination of the EMEP countries.

Reasonable level of agreement was obtained between the modelling results on B[a]P, PCB-153 and γ-HCH and regular measurements of the EMEP monitoring network . Particularly, the deviations between the modelled and measured annual mean air concentrations were about a factor of two-three. Correlation coefficients between the monthly mean computed and observed air concentrations were in the range from 0.6 to 0.8. The agreement of modelling results with measured annual means of γ-HCH air concentrations was basically within a factor of three. The measurements of PCDD/F concentrations are stipulated by EMEP on a voluntary basis at supersites only. However, the work on the comparison of calculated PCDD/F concentrations with measurements obtained on national basis is now ongoing.

To improve the assessment of the EMEP domain pollution by POPs and to reduce its uncertainties the elaboration of an approach to integrate monitoring, emission inventories, and modelling has been started. This integrated approach allows performing complex analysis of the information on the pollution provided by monitoring and modelling activities and to indicate the areas of their further improvement, in particular, necessity to improve emission data, refine modelling approaches, apply fine resolution modelling, and conduct specific monitoring campaigns. In the framework of this work a special modelling tool to analyze the differences between the model estimates and measurements was developed. This tool represents a simplified version of the adjoint model based on the backward trajectory approach. The tool was tested at the comparison of modelled and measured B[a]P concentrations for 2008.

The changes in meteorological and environmental parameters due to future climate change can essentially affect the fate of POPs in the environment. Investigation of the influence of meteorological and environmental parameters on the atmospheric transport of POPs has been initiated this year. Preliminary

results showed that the most valuable factors determining long-range transport of POPs were temperature, vegetation cover, and precipitation intensity.

Development of size-segregated approach to describe the deposition of particle-bound POPs and evaluation of the sensitivity of model estimates to the accounting for particle size distribution has been continued. It was found that the use of particle size-segregated deposition scheme led to better spatial correlation of annual mean modelled and measured B[a]P concentrations in air. Further work in order to evaluate and improve the size-segregated description of POP transport and removal is required and planned to be performed in the future.

MSC-E has continued investigation of behavior and transport of new POPs in the environmental compartments. The evaluation of long-range transport of perfluorooctane sulphonate anion (PFOS) and related substances has been performed this year. Perfluorooctane sulphonate anion is a representative of a large class of chemicals including different organic compounds. Two different groups of chemicals can be distinguished from this class − PFOS in the ionic form and the so-called PFOS-related substances. It was found that the main pathway of dispersion of the chemicals of the first group was the transport with sea currents. For modelling of environmental transport of these substances the model modification is required since classical models based on the partitioning in the system air/octanol/water are inapplicable for this purpose. On the opposite, modelling long-range transport for the substances of the second group with the help of existing models is possible. However, for correct description of PFOS levels in the environment both their direct transport in the marine environment and atmospheric transport of their precursors should be investigated.

MSC-E and CCC closely cooperated with TFMM, TFHTAP, international organizations and programmes (AMAP, EU, HELCOM, OSPAR, UNEP, WMO, etc.), and national experts. Special attention was paid to strengthening cooperation with EECCA countries.

Next year EMEP Centres will continue improving the evaluation of pollution levels and transboundary transport of POPs in the EMEP domain and at the hemispheric/global scale. In particular, attention will be paid to the elaboration of EMEP global multi-media modelling system, further development of integrated measurement/modelling/emission approach, investigation of uncertainties of POP measurements using results from the EMEP and AMAP laboratory intercomparison together with field intercomparison between passive and active sampling.

CONTENTS

INTRODUCTION

This Status Report describes the progress in activities of the EMEP Centres − Meteorological Synthesizing Centre-East (MSC-E) and Chemical Coordinating Centre (CCC) − in the evaluation of contamination of the EMEP region by persistent organic pollutants (POPs). During 2010 investigations of POP long-range atmospheric transport and transboundary fluxes on the basis of measurements, emission data and modelling were continued. These activities were performed in accordance with the EMEP Work-plan for 2010 [*ECE/EB.AIR/2009/7*].

The work on the investigation of POP long-range transport was carried out in two directions. The first was the evaluation of pollution levels and deposition fluxes in the EMEP region for 2008. The second direction was the investigation of POP transport and environmental fate on the hemispheric/global scales.

In order to improve the understanding of POP long-range transport at hemispheric/global scales, MSC-E participated in the work of the EMEP Task Force on Hemispheric Transport of Air Pollutants (TF HTAP). Particularly, MSC-E took part in preparation of the Part C on POPs within the HTAP 2010 Assessment Report and coordinated the intercomparison of POP models within the TF HTAP multi-model intercomparison study. Besides, MSC-E continued the development of the Global EMEP Multi-media Modelling System (GLEMOS) working out the modules describing POP fate in the environment including the processes in the main environmental compartments (soil, seawater) and exchange between these compartments and the atmosphere.

The evaluation of the pollution within the EMEP domain for 2008 was performed for the following POPs: polycyclic aromatic hydrocarbons (B[a]P), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCB-153), lindane (γ-HCH). Input information on emissions for model assessment was based on the most recent officially submitted emission totals and information on spatial distribution of emissions along with available unofficial emission estimates. For the evaluation of contributions of non-EMEP sources to the pollution of the EMEP domain and of re-emissions due to historical accumulation, available emission data for PCDD/Fs, PCB-153 and γ-HCH within the Northern Hemisphere were compiled. Evaluation of transboundary fluxes in the EMEP region (country-to-country matrices) was carried out for B[a]P, PCDD/Fs and PCB-153.

The work on elaboration of integrated measurement/modelling/emission approach to the evaluation of POP contamination levels was initiated. For this work the above-described model estimates of POP pollution together with the corresponding emission data and measurements obtained at the EMEP monitoring network were used. The information on the observed levels of POPs in air and in precipitation for 2008 was available from twenty EMEP stations located in the northern, western, and central parts of Europe, which were three more than in 2007. To link the monitoring information with emission data with the help of the comparison between model predictions and measurement results, a special modelling tool based on the usage of backward trajectory approach was developed. It was tested on the analysis of the agreement between measurements and calculations for B[a]P.

Further development of MSCE-POP model was continued in the following directions. The work on the methodology of ranking meteorological and environmental parameters with respect to their influence on POP fate in the environment was performed. The methodology was tested at two substances: B[a]P and β-endosulfan. The results obtained can be useful in the investigation of climate change scenarios from the viewpoint of their influence to POP transport and accumulation in the environment. Evaluation of the sensitivity of POP model to the application of size-segregated parameterization of deposition was done. Due to the necessity of further refinement of emission data for HCB, calculations of the transport and

accumulation of this pollutant in the environment were performed on the basis of scenarios of historic emissions.

In the field of evaluation of POP pollution levels within the EMEP region, the EMEP Centres closely co-operated with the subsidiary bodies to the Convention, international organizations and programmes as well as with national experts. Special attention was paid to the collaboration with EECCA countries.

Detailed information on the work fulfilled during this year are presented in the Technical Reports of the EMEP Centres [*Ilyin et al.*, 2010; *Jonson and Travnikov*, 2010; *Aas and Breivik*, 2010] as well as on the Internet www.msceast.org and www.emep.int.

Below the content of the report is briefly outlined.

Chapter 1 is devoted to the description of MSC-E activities related to the evaluation of the global transport of POPs and its contribution to the pollution of the EMEP domain. The progress in further development of the global multimedia approach in the framework of the GLEMOS is outlined along with pilot model simulations for PCB-153. Modelling of HCB long-range transport and fate in the period of 1945-2008 based on the scenarios of historic emissions is presented. The contribution of MSC-E to the activity of TF HTAP directed to the assessment of POP intercontinental transport is described.

Chapter 2 presents the results of evaluating pollution levels and transboundary transport of selected POPs in the EMEP domain. Here the emission data used for modelling, available monitoring information and obtained modelling results are discussed. In conclusion, the analysis of agreement between measurement data and calculation results with the help of the backward trajectory approach is given.

Chapter 3 describes the progress in the development of the EMEP multicompartment POP transport model (MSCE-POP) achieved in 2010. The investigation of meteorological and geophysical factors affecting the fate of POPs in the environment in connection to possible climate change is presented. The sensitivity of modelling results to the application of size-segregated approach to describe the deposition of particle-bound POPs is examined. Continuation of the work on POP-like substances is described.

Chapter 4 highlights the co-operation of MSC-E and CCC with CLRTAP subsidiary bodies, international organizations and programmes, and national experts.

Chapter 5 contains a brief description of future activities related to POPs proposed by CCC and MSC-E for 2011.

The main results of the EMEP Centres work in the field of the evaluation of pollution levels and transboundary transport of POPs are summarized in Conclusions.

The report is complemented by an Annex containing detailed matrices of transboundary fluxes for 2008 calculated using MSCE-POP model for PCDD/Fs, B[a]P and PCB-153.

Acknowledgements

The authors of the report are grateful to Tom Harner, Matthew MacLeod, Anne Hollander, and other contributors for co-operation in preparation of POP part of the HTAP 2010 Assessment Report.. Substantial contribution to the technical preparation of the report was made by Irina Strizhkina (MSC-E).

1. ASSESSMENT OF POP INTERCONTINENTAL TRANSPORT

Evaluation of the intercontinental transport constitutes an significant task in the assessment of POP pollution of EMEP region. For some POPs their global transport considerably contributes to the pollution levels and comprises essential exposure pathway. To investigate the role and extent of the influence of global sources the EMEP has initiated the development of relevant modelling approach starting from hemispheric scale and moving recently to the global scale modelling. The status of this activity is described briefly in this report.

Further investigation of hemispheric transport of air pollution is an important field of work aimed at evaluation of the intercontinental transport of global pollutants, such as mercury and POPs, and providing the EMEP countries with the information on its role in regional and local scale pollution. In framework of this activity MSC-E continued to develop the Global EMEP Multi-media Modelling System (GLEMOS) for POPs and performed pilot global-scale model simulations in order to test its operability and to indicate potential problems. Further studies of global transport and fate for POPs with the essential LRTP and persistence were carried out for HCB elaborating scenarios of its historic emissions and investigating its environmental fate and the role of primary and secondary emission sources.

The work under the TF HTAP to assess intercontinental transport of pollutants is of particular importance for the elaboration and further improvement of the global modelling approach being developed within EMEP. Among the major priorities of the Task Force in recent years were the multi-model experiments to quantify global scale source-receptor relationships and the preparation of the HTAP 2010 Assessment Report. Starting from the beginning of this work, MSC-E participated in these activity collaborating with POP experts from different countries and international organizations. Among the main Centre`s tasks this year were the preparation of contributions to the Assessment Report, coordination of the TF HTAP multi-model experiments for POPs, and participation in the discussions on the content of the Assessment Report.

1.1. Development of POP modules for GLEMOS

The GLEMOS is being developed at the MSC-E to study global dispersion of pollution and contribution of intercontinental transport to the concentrations and deposition in various regions. This activity was started several years ago and is carried out in cooperation with MSC-West. The GLEMOS is an Eulerian type chemistry transport model that meets the following main requirements [*Tarrasón and Gusev*, 2008]:

- multi-pollutant;
- multi-media;
- flexible model domain;
- variable spatial resolution;
- modular structure;
- computationally efficient.

This year the major efforts in the developing and adopting of the GLEMOS to study POP long-range transport were directed to the elaboration of the modules required for the implementation of the multi-media approach within this system. The state of the GLEMOS development and preliminary modelling results are presented in this section. More detailed information in this respect can be found in [*Jonson and Travnikov*, 2010].

Several modules representing the atmosphere, soil and ocean (seawater) along with the description of the processes occurred with POPs in these media were elaborated. In addition, a module of inter-media POP exchange was developed. For operability testing of implemented POP modules a series of global-scale one year model simulations with spatial resolution $1^0 \times 1^0$ and $5^0 \times 5^0$ was performed for PCB-153. This PCB congener has been selected since its air concentrations in gaseous and aerosol phases are comparable in magnitude that facilitates the analysis of the quality of atmospheric processes modelling. Besides, the data on PCB-153 emissions and measurements were available. The following input information was used in test model runs: atmospheric emissions distribution compiled on the basis of the global emission inventory of PCBs [*Breivik et al.,* 2007] and meteorological data calculated by Global Environmental Multiscale Model (GEM) [*Côté et. al.,* 1998a*, Côté et. al.,* 1998b].

Multimedia approach in GLEMOS: current state of development

The structure and processes implemented in current GLEMOS version for global-scale POP modelling are illustrated in Fig. 1.1. Several phases of POPs (shown as rectangles) are considered in each medium. These phases are involved in different physical-chemical processes (highlighted blue), namely, transport, degradation, phase partitioning, and inter-media exchange (arrows).

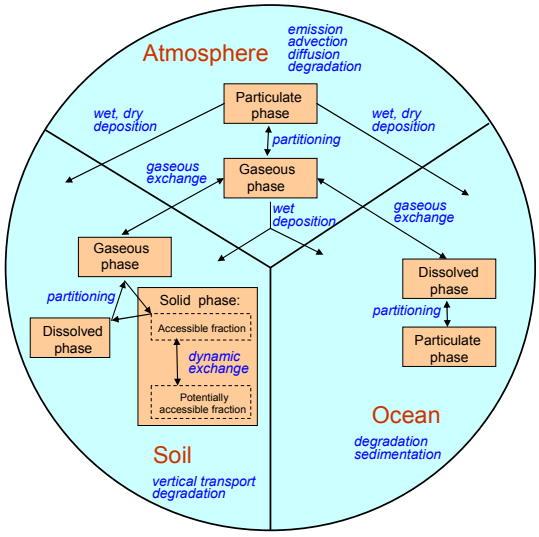

Fig. 1.1. *The scheme of the main processes related to POPs in the current version of the GLEMOS model (the dissolved phase includes also POPs sorbed on the dissolved organic matter)*

The modules of environmental media (atmosphere, soil, and ocean) were constructed on the basis of corresponding modules of the regional MSCE-POP model. The same parameterizations of physical-chemical processes have been used. The detailed description of these parameterizations can be found in [*Gusev et al.,* 2005]. It should be noted that seawater module for the GLEMOS modelling system was implemented in a simplified way neglecting the advection and diffusion processes in the marine environment at current stage of the development. Additional simplification is related to the influence of vegetation, which is currently not included. However, it is planned to develop more complex version of the multimedia approach within the GLEMOS system at further stages of the work.

The module of inter-media exchange developed for the GLEMOS model differs from those of the regional and hemispheric MSCE-POP models. The description of this component of the GLEMOS is presented below in more detail.

Traditionally the method of splitting into physical processes is used in air quality modelling. This method consists of the transformation of the initial evolutionary problem with complex operators to a sequence of tasks with the operators of a simpler structure. This approach simplifies the scheme of modelling but

can reduce the accuracy of calculations. At present splitting into physical processes is employed to describe mass exchange between media in the hemispheric version of MSCE-POP model too. But the new exchange module developed for global-scale modelling is based on simultaneous consideration of all the processes of intermedia interactions in each vertical model column. The system of differential equations describing exchange processes is formulated as follows:

$$\frac{\partial C_i}{\partial t} = \sum_j \alpha_{ij} C_j \qquad (1.1)$$

where *Ci* are the concentrations of pollutant in model layers involved in intermedia exchange; it can be one of atmospheric layers or upper soil/ocean layer (Fig. 1.2);

α_{ij} are the functions of media parameters characterizing exchange processes between i-th and j-th layers.

The processes considered in the current version of the exchange module are the following:

- wet deposition of aerosol and gaseous phases from air to soil and ocean;
- dry deposition of particulate phase from air to soil and ocean;
- gaseous exchange between air and soil/ocean.

The system (1.1) is solved by the Euler method:

$$C_i(t+\Delta t) = C_i(t) + \Delta t \sum_j \alpha_{ij}(t) C_j(t),$$

where Δt is the integration time step. It is chosen one and the same for all exchange processes that is the main difference of this approach from one realized in the regional MSCE-POP model version. Maximum allowed time step is chosen from the condition that each of the concentrations does not fall more than twice for the period Δt.

The new exchange module was tested separately and as the part of the GLEMOS model and showed reasonable results. All the test calculations described below were performed using the new exchange module.

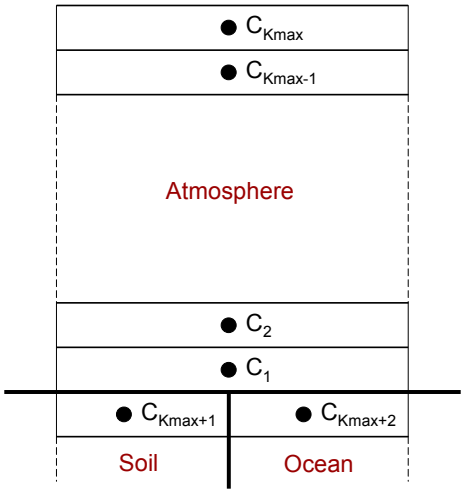

Fig. 1.2. *Vertical column of model cells involved in cross-media exchange (Kmax is the number of model layers in atmosphere)*

Pilot results of PCB-153 global-scale modelling using GLEMOS

Long-term modelling of PCB-153 transport and fate with 5°×5° spatial resolution

Model simulations for the period of time from 1930 to 2005 on $5^0 \times 5^0$ global grid were performed. Along with the model testing the purpose of these computations was preliminary investigation of the processes of PCB-153 accumulation in media and the processes of intermedia exchange.

For the period considered two scenarios of anthropogenic atmospheric emissions were used:

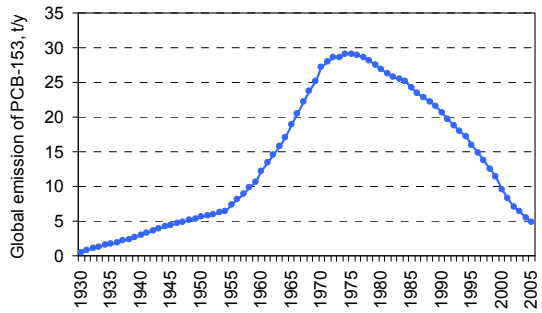

Fig. 1.3. *Time trend of PCB-153 global atmospheric emissions, t/y*

1. Historical emissions compiled on the basis of "maximum" and "default" scenarios of the global inventory [*Breivik et al.,* 2007] (Fig. 1.3).

2. One and the same emissions for each year (equal to that of 2005 in the first scenario).

The latter scenario was constructed to study the conditions of the quasi-steady state distribution of pollution within the three-media system being currently realized in the GLEMOS model.

The analysis of modelling results was mainly focused on the integral characteristics of environmental contamination, namely, total annual mass content in media and total intermedia annual mass fluxes. This approach allows the identification of the most common patterns of PCB-153 behavior in the environment.

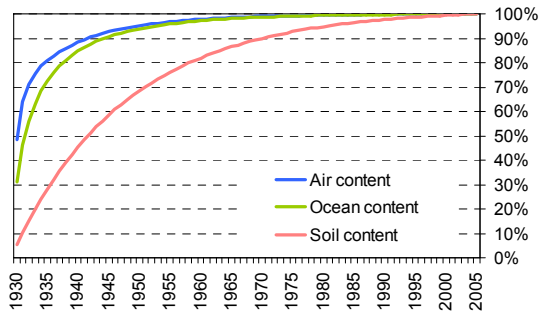

Fig. 1.4. *Time trends of PCB-153 content in environmental media (constant emission scenario), %*

The results of **model simulations with constant emissions** indicated the soil as the most inertial medium with respect to dynamics of PCB-153 content (Fig. 1.4). The accumulation of PCB-153 in soil was the slowest one comparing with that in other media. Assuming that total PCB-153 content in soil in 2005 according to the considered scenario is 100%, we can conclude that 90% level was reached in 1971. The corresponding 90% level of content in air was achieved in 1942, and for ocean – in 1945.

The dynamics of PCB-153 accumulation in the environmental media can be described as a superposition of two exponential-type processes: fast (characteristic of the initial stage of accumulation) and slow (being important at final stage). Reasonable approximation of accumulation process is:

$$M = M_{\infty}\left[1-(\alpha_{fast}\exp(-\frac{t}{T_{fast}})+\alpha_{slow}\exp(-\frac{t}{T_{slow}}))\right], \qquad (1.2)$$

where M is pollutant mass content in a media, M_{∞} is equilibrium mass content, α_{fast}, α_{slow} are the shares of each process in total mass, T_{fast}, T_{slow} are accumulation time scales.

The values of α and T for each of three media calculated using the least squares method on the basis of long-term PCB-153 modelling results (Fig. 1.4) are placed in Table 1.1.

Table 1.1. *Description of accumulation dynamics of PCB-153 in the environmental media*

Medium	T_{fast}, years	α_{fast}	T_{slow}, years	α_{slow}	R2 - coefficient of multiple determination of approximation
Air	1.0	0.68	11.0	0.32	0.999
Ocean	1.6	0.52	9.7	0.48	0.999
Soil	10.6	0.09	20.0	0.91	1.000

It is seen that fast process is essential for air (68% of equilibrium mass). Characteristic times are substantially different (1 and 11 years). For seawater characteristic times are more close to each other (1.6 and 9.7 years), the shares of the processes are comparable. Finally, fast process in soil is almost negligible (9% of equilibrium mass), so only one time scale is characteristic of this media (20 years).

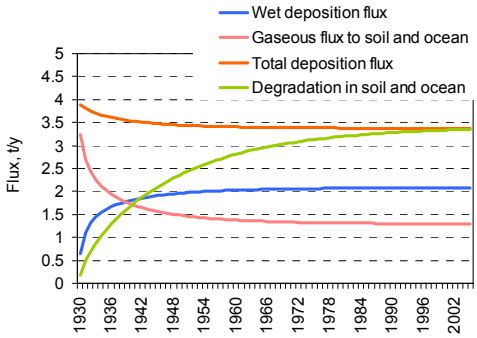

Fig. 1.5. *Input-output mass balance of PCB-153 in soil for time period from 1930 to 2005 (scenario of constant emission), t/y*

High soil capacity for accumulation can be explained as follows. The direction and intensity of air-soil gaseous exchange are determined by the difference of volume concentrations of gaseous phases in these media. At the same time the proportion of gaseous component of PCB-153 in soil is much less then the shares of dissolved and solid phases, and the balance between these phases is assumed to be established instantly due to the process of partitioning. So, getting into soil, the most part of gaseous substance is immediately sorbed on solid and dissolved organics. The condition of soil saturation is the equality of total deposition flux from air (it decreases in time – red curve in Fig. 1.5) and the velocity of total soil degradation (increases in time – green curve). The ratio between wet deposition and gaseous fluxes from air is of interest. First years gaseous flux prevails because of low soil concentrations of PCB-153 (Fig. 1.5). As soil is saturated in time, gaseous flux weakens, wet deposition becomes dominant.

The results **of modelling with historical emission** confirm the conclusion about high inertia of soil. Maximum total content of PCB-153 in this medium takes place in 1989, i.e. 14 years after emission maximum (Fig. 1.6). In 2005 the mass of PCB-153 in soil amounts to 74% of maximum level whereas air emission drops down to 16% this time (Table 1.2). Air and ocean react to emission change much faster (Fig. 1.6 and Table 1.2).

Table 1.2. *The characteristics of time trends of PCB-153 emissions and media content*

Medium / emission	Characteristics of time trend	
	Year of maximum	Ratio of 2005 to maximum
Air	1978	31%
Soil	1989	74%
Ocean	1979	36%
Emissions	1975	16%

Soil contains more than 90% of total PCB-153 mass in the environment since 1942 (Fig. 1.7). The share of seawater in PCB-153 mass balance is several percent. The share of air is the least and amounts to 0.4% in 2005. This does not lead to the conclusion that the atmosphere is an inessential medium since all intermedia exchange is realized through air.

The most part of PCB-153 mass is removed from the environment due to soil degradation (Fig. 1.8). This process is responsible for ~80% of total removal in 1970 and ~90% in 2005. The contributions of each ocean and air degradation amount to ~5% in 2005. The relative importance of the removal from air decreases from year to year.

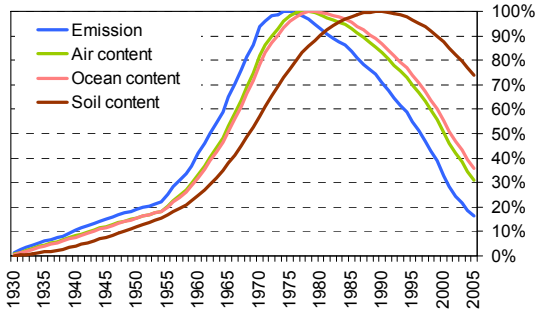

Fig. 1.6. *Time trends of PCB-153 content in environmental media, %*

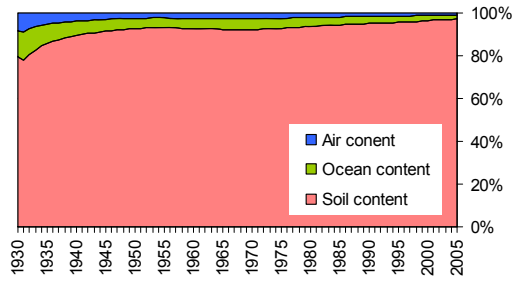

Fig. 1.7. *The percentage ratio of PCB-153 content in environmental media for time period from 1930 to 2005, %*

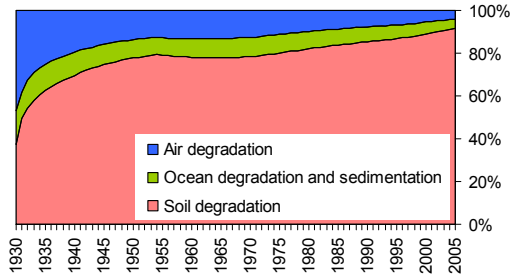

Fig. 1.8. *The percentage ratio of different ways of PCB-153 removal from environmental media for time period from 1930 to 2005, %*

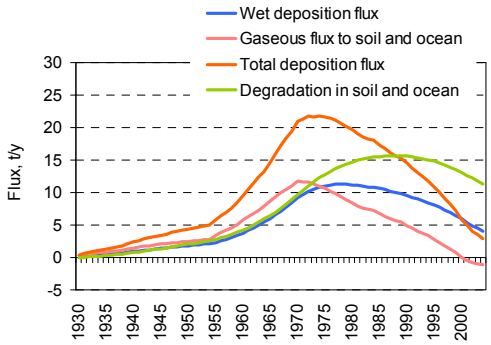

Fig. 1.9. *Input-output mass balance of PCB-153 in soil for time period from 1930 to 2005, t/y*

The balance of PCB-153 mass input-output for soil is presented in Fig. 1.9. The curves of total flux from air and soil degradation intersect in 1989 which is the year of soil content maximum. Since 1989 degradation prevails resulting in soil concentration reduction. In 2000 air-soil gaseous flux changes its direction. The reason of this is slow reduction of PCB-153 soil content in comparison with air content.

It should be mentioned that the ratio of input-output fluxes of PCB-153 in soil changes with high intensity during the last 10 years of calculation period. This means that the system is unstable, quasi-steady state seems to be unreachable in the nearest future.

Main results of long-term modelling of PCB-153 global contamination on $5^0 \times 5^0$ grid are as follows:

1. The accumulation in soil is the most essential. It contains more than 90% of total PCB-153 mass in the environment. Soil degradation is responsible for ~ 90% of total removal of PCB-153. Air and ocean compartments in sum account for less than 10% of pollutant mass and removal flux.

2. Soil is the most inertial medium. It follows the changes of anthropogenic emissions with significant delay. The accumulation time scale for soil (T_{soil} = 20 years) exceeds time scales for other media.

3. The three-media system under consideration had not reached quasi-steady state by 2005.

Modelling of PCB-153 transport with 1°×1° spatial resolution

The main purpose of $1^0 \times 1^0$ modelling of PCB-153 atmospheric transport was the evaluation of the capability of GLEMOS to reproduce POPs environmental contamination levels at the current stage of model development.

Three annual model simulations with historical emissions [*Breivik et al.,* 2007] were carried out: for 2001, 2005 and 2008. Spin-up from 1930 on $5^0 \times 5^0$ global grid was employed. Obtained results were compared against observations and the similar results of MSCE-POP regional model. Some of the results are presented below to illustrate the actual state of the art.

The atmosphere. The spatial distribution of annual mean air concentration of PCB-153 in the surface air layer is presented in Fig. 1.10. Elevated levels of PCB-153 in air occurred in the regions with maximum anthropogenic emissions: they are Europe and Northern America. Maximum concentrations (over 5 pg/m^3) take place in the Central Europe. Over the most part of the Southern Hemisphere PCB-153 air concentrations are relatively low (0.2-0.8 pg/m^3).

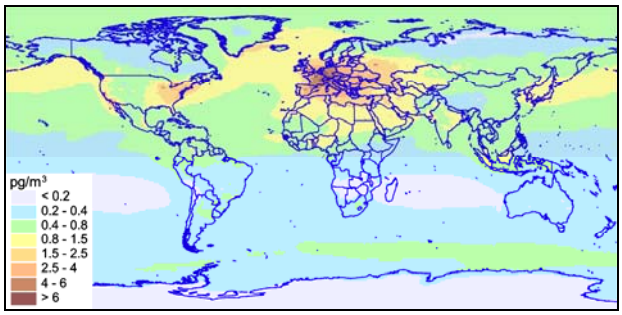

Fig. 1.10. *Spatial distributions of PCB-153 annual mean concentration in surface air for 2008, pg/m^3*

The results of modelling for different years were compared to measurement data on PCB-153 concentrations in air and precipitation obtained at the EMEP monitoring network. Annual and monthly mean concentrations were considered. Here some results for air concentrations in 2008 are discussed.

Seven monitoring sites measured concentrations of PCB-153 in air in 2008. The scatter plot of annual mean concentrations is shown in Fig. 1.11. Most of the points (pairs of measured/calculated values) on the scatter plot are very close to the regression straight line. That is the evidence of significant spatial correlation between the model calculations and measurements. The linear correlation coefficient amounts to 0.89. Calculated air concentrations of PCB-153 exceed measured ones about 35% on the average.

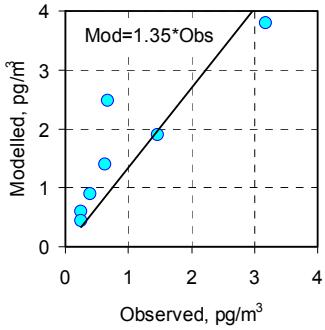

Fig. 1.11. *Scatter plot of annual mean air concentrations of PCB-153, calculated by the GLEMOS model and measured at the EMEP monitoring sites*

The temporal changes of monthly mean air concentrations of PCB-153 are in accordance with measurements for most of the EMEP sites. Fig. 1.12 presents examples for two stations with relatively high level of PCB-153 air concentrations: they are the Czech site CZ03 and the Swedish site SE14. In both cases the model reproduced summer elevation of pollutant air concentrations. The coefficients of linear correlation of measured and modelled monthly mean values amount to 0.85 for CZ03 and 0.91 for SE14.

Calculated PCB-153 air concentrations for 2001 and 2005 as well as concentrations in precipitation for all the years considered are in reasonable agreement with the EMEP measurements too.

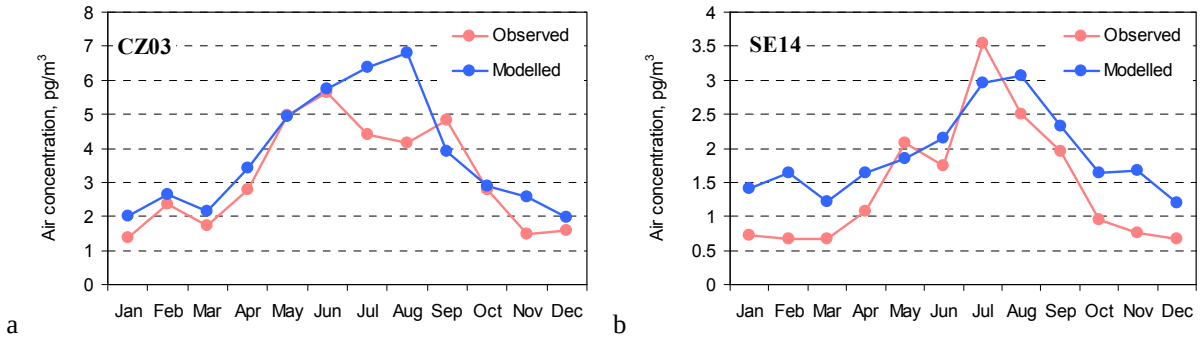

Fig. 1.12. *Monthly mean PCB-153 concentrations in air in 2008 calculated by the GLEMOS model and measured at two EMEP monitoring sites: CZ03 (a) and SE14 (b)*

Ocean. Spatial distribution of PCB-153 concentrations in surface layer of seawater is given in Fig. 1.13. There is evident latitudinal dependence of contamination level. The highest values of PCB-153 content in seawater (more then 1 pg/L) were obtained for high-latitude regions. The reason for this is strong temperature dependence of the gaseous flux from air to water: lower air temperatures correspond to higher air-water flux. It is remarkable that seawater PCB-153 concentrations in the Antarctic region are several times higher than those in equatorial regions.

Measurement data on PCB-153 concentrations in seawater of different campaigns were employed for the validation of POPs modules of GLEMOS (Table 1.3). It should be noted that this comparison is of preliminary character since currently the simplified version of the seawater module is implemented in the GLEMOS model. For the comparison with measurements related to different years calculation results for 2001 were scaled in accordance with the temporal trend of PCB-153 total ocean mass content (Fig. 1.4). As it is seen from the Table 1.3, the model reproduced measured levels of PCB-153 content in seawater reasonably well with the exception of few cases (Bering and Chuckchi Seas). The agreement between calculated and measured concentrations is mostly within a factor of five and for 7 of 10 regions within a factor of three.

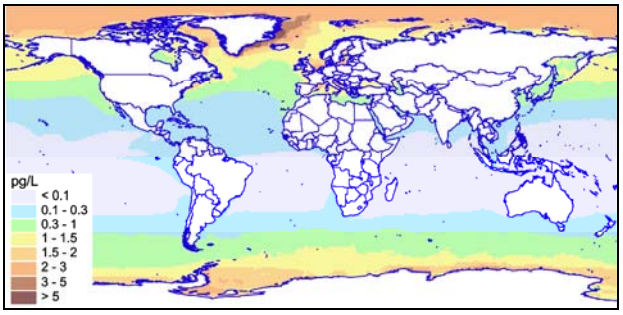

Fig. 1.13. *Spatial distributions of PCB-153 annual mean concentration in seawater for 2008, pg/L*

Table 1.3*. Comparison of measured and modelled concentrations of PCB-153 in seawater*

Location	Year	Conc. in seawater, pg/L (averaged over all samples within a geographical region)		Reference
		Measured	Modelled	
Norwegian Sea	1985	15	4.62	[*AMAP*, 1998]
Barents Sea	1992	1.14	3.69	[*AMAP*, 1998]
Baffin Bay	1998	5.8	1.35	[*AMAP*, 2004]
Beaufort Sea	1999	3.9	2.83	[*AMAP*, 2004]
North Sea	1988	4.77	4.49	[*Schulz-Bull et al.*, 1991]
Bering Sea	1990	0.36	7.35	[*Iwata et al.*, 1993]
Chukchi Sea	1990	0.25	4.62	[*Iwata et al.*, 1993]
Japan Sea	1995	0.33	1.52	[*Kannan et al.*, 1998]
Barents Sea	1996	6.7	3.91	[*PWGSC*, 2003]
Beaufort Sea	1999	5.8	3.35	[*PWGSC*, 2003]
Mean		4.40	3.69	-

Soil. Figure 1.14 shows global-scale spatial distribution of calculated annual mean PCB-153 concentrations in the top 5 cm of soil. Elevated level of pollution is a characteristic of the Europe and North America. In the most polluted central part of Europe soil concentrations exceed 100 pg/g. The range 20-70 pg/g can be assumed as background for Europe, 3-20 pg/g – for North America.

The distributions of annual mean PCB-153 concentrations in soil for the EMEP region calculated by the GLEMOS model (a) and the MSCE-POP model (b) are presented in Fig. 1.15. The both maps look similar as a whole, but there are some substantial differences in several regions (e.g. Greenland). The global model gives higher level of PCB-153 soil concentrations in the most polluted regions of Europe than the regional one.

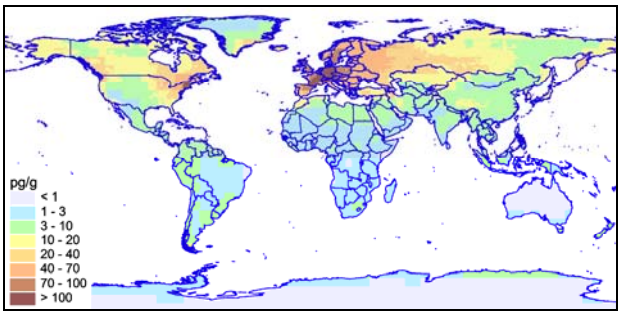

Fig. 1.14. *Spatial distributions of PCB-153 annual mean concentration in soil for 2008, pg/g*

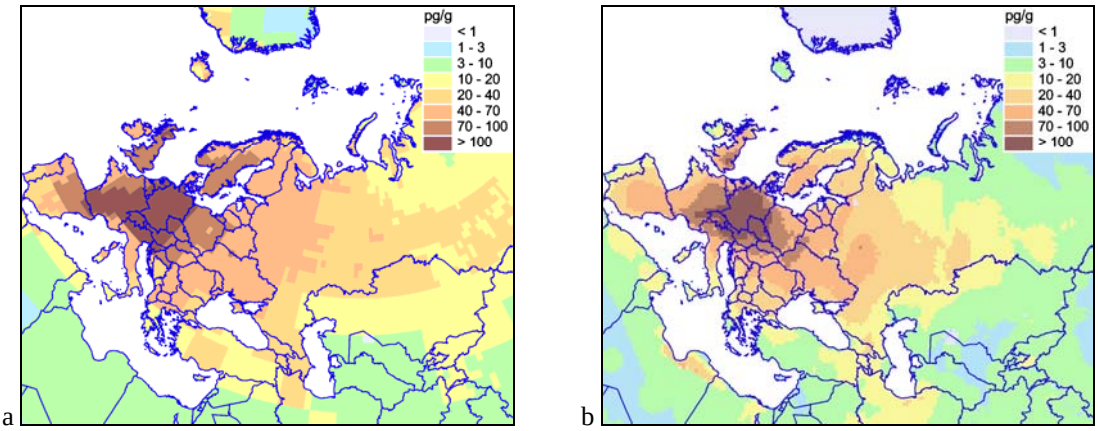

Fig. 1.15. *Spatial distributions of PCB-153 annual mean concentration in upper soil layer for 2008 calculated by GLEMOS model (a) and MSCE-POP model (b), pg/g*

Modelled concentrations of PCB-153 in upper soil layer for 2001 were compared with surface soil measurements of PCB-153/132 performed at 191 background sites worldwide in 1998 [*Meijer et al.,* 2003]. To reduce significant dispersion of observational data (spread in measured values exceeds one order of magnitude in some $1^0 \times 1^0$ model grid cells), they were grouped the following way: the series of measurements related to one and the same cell was replaced by the arithmetic mean of these measurements. The resulting scatter plot of observed and calculated soil concentrations is given in Fig. 1.16. In general, measured values exceed calculated ones substantially. High vertical gradient of PCB-153 soil concentration near ground surface seems the main reason for this: maximum levels of concentration can not be reproduced by the model due to limited vertical spatial

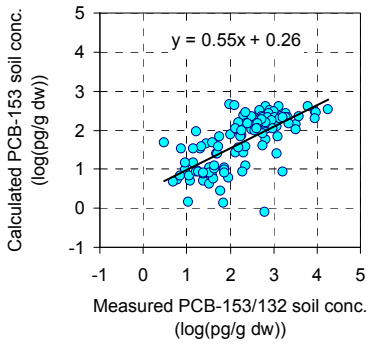

Fig. 1.16. *Scatter plot of calculated soil concentrations of PCB-153 in the upper soil layer and measured soil concentrations of PCB-153/132 [Meijer et al., 2003] (dw – dry weight of soil)*

resolution of the model grid. At the same time, there is good correlation between measured and modelled values in logarithmic scale (Fig. 1.16). Taking into account significant scatter of the observed soil concentrations, the results of the comparison can be considered as reasonable.

Thus, pilot version of the GLEMOS modelling system for POPs with multimedia approach was elaborated. The system was supplied with three modules representing the atmosphere, soil, and seawater compartments and the description of relevant processes governing POP fate. The simplified description of POP fate within the seawater in the model requires implementation of POP transport with sea currents and diffusion processes. Further improvement of the multimedia approach within the GLEMOS modelling system is required. Particularly, the description of POP fate in vegetation and the interaction of this compartment with the atmosphere and soil is needed.

Preliminary model simulations of PCB-153 global transport and fate performed by the developed model revealed the consistency of model estimates of PCB-153 air concentrations with measurements of the EMEP monitoring network. In general, the computed concentrations in upper soil and seawater surface layer were in a reasonable agreement with available measurements. At the same time the differences obtained demonstrated the need of further work on the refinement of processes descriptions implemented in the model.

1.2. Evaluation of HCB long-range transport

Comparing to other POPs considered in this assessment the evaluation of HCB pollution levels exhibits more complications due to the limited information on the sources and levels of its current emissions and also the role of secondary emission sources. In previous assessments [*Gusev et al.,* 2008; 2009] the studies of HCB pollution were mainly performed using the construction of emission scenarios and subsequent model simulations. Based on the analysis of modelled and observed concentrations it was indicated that the levels of HCB emission in Europe were likely more significant than that officially reported by the European countries which led to the essential underestimation of measured concentrations by the model. This year these studies were continued with model simulations of HCB fate in the period 1945-2008 and the evaluation of the role of primary and secondary HCB emission sources.

To study the HCB fate in the environment simple scenarios of its historical emission were constructed. Following available information the application of HCB in various activities was started from 1945 and reached its maximum in 1980-s. The major source of its release into the environment in that period was the application in the agriculture as a fungicide. Additional sources contributing to the emissions of HCB were the production of chlorinated solvents and pesticides, wastes and sewage sludge incineration, metals smelting, sintering process, steel manufacturing, production of magnesium and cement as well as combustion of fossil fuel. Following the information on agricultural use of HCB in *Food and Agriculture Production Yearbook* [1989] *N. Vulykh et al.* [2001] estimated that the global HCB emissions in 1980-s could ranged approximately from 7 kt to 18 kt per year with average annual emission about 12 kt. It should be noted that these estimates are subject of essential uncertainties as they do not cover all the countries where the HCB was applied. Starting from 1980-s the agricultural use of HCB was banned in many countries world-wide resulting in considerable decrease of HCB use and emissions. According to the estimates of *R.E.Bailey et al.* [2001] annual global emissions of HCB in the middle of 1990-s were about 23 t per year varying in the range from 12 t to 92 t. Using these estimates three simple scenarios (maximum, average, and minimum) of HCB emission temporal development in period 1945-2008 were elaborated (Fig. 1.17).

For the period 1995-2008 it was assumed that global emissions were gradually decreasing by about 10% a year on average. The spatial distribution of global emission was prepared with the spatial resolution 1×1 degree using the distribution of cropland area [*Li*, 1999] for the emissions from agricultural use and population density [*Li*, 1996] for other sources of HCB emissions.

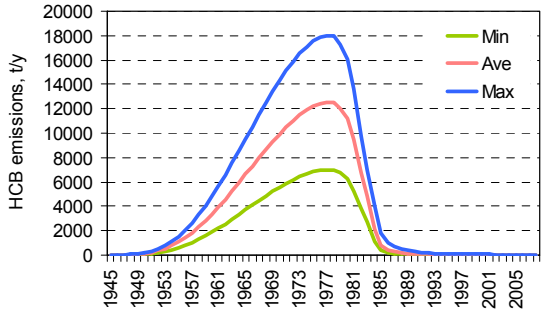

Fig. 1.17. *Estimates of historic and current HCB emissions under the three emission scenarios (minimum, average, and maximum) for the period 1945-2008, t/y*

Model simulations based on the constructed emission scenarios allowed to predict the fate of HCB in various environmental compartments and to compare the dynamics of its content with the observed levels of concentrations in media and estimates of other models. In this study initial attempt to analyze the environmental distribution of HCB is presented which will be further evaluated in subsequent investigations. At current stage the hemispheric MSCE-POP model was applied to simulate the fate of HCB within the Northern Hemisphere. Predictions of burdens of HCB in the seawater and soil are shown in Fig. 1.18.

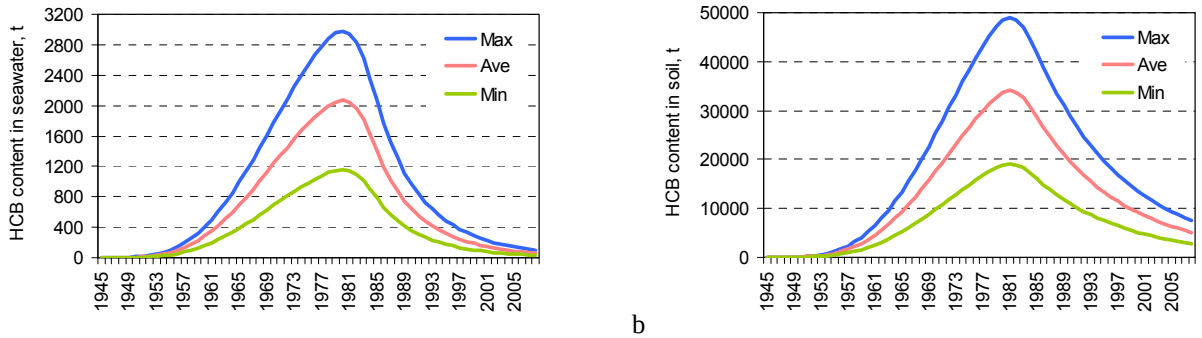

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Fig. 1.18. *Dynamics of burdens of HCB in the seawater (a) and soil (b) under the three emission scenarios (maximum, average, minimum), t*

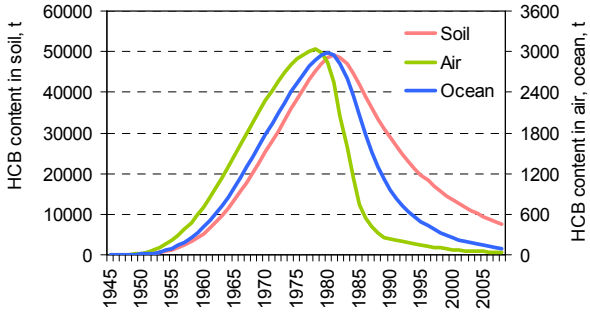

Fig. 1.19. *Comparison of HCB content dynamics in the atmosphere, soil, and ocean waters obtained under the maximum HCB emission scenario, t*

Following the increasing global application of HCB in period 1945-1980 its content in the atmosphere and soil increases with similar rates. At the same time, the rates of HCB content decrease in air and soil, after the banning of HCB use in agriculture starting from 1980-s, are substantially different. Difference in the decrease rates of HCB in the atmosphere, soil, and ocean water is illustrated in Fig. 1.19. Changes of HCB air content almost follow the emission decrease, while the decrease of HCB in soil is substantially slower. Particularly, up to 1990 the mass of HCB in air decreased by more than 90%, whereas in soil it decreased by only about 40% and in 2000 by about 70% of the maximum content in 1980-s.

Model evaluation of HCB fate indicated that the major part of HCB mass in the 2008 is stored in soil (more than 90%). Other media contain only small share of the total environmental burden. These estimates of HCB distribution in the environment are close to other modelling studies. In particular, the

fate of HCB within the Great Lakes region was estimated using the CHEMGL model [*Zhang et al.,* 2003]. Their prediction of mass distribution indicated that soil contained almost 80% of total HCB mass in the environment compartments of the studied region. Similar estimates were obtained by *M.MacLeod and D. Mackay* [1999] using equilibrium criterion model. Their results indicated that when HCB was emitted to soil only (representing agricultural use) more than 90% of the total mass was distributed in soil. Similarly, *J.L.Barber et al.* [2005] in their global mass balance calculations of HCB fate found that major part of HCB in the environment was likely accumulated in soil. Calculated total amount of HCB in the environment presented in [*Barber et al.,* 2005] and estimates obtained in this study are comparable differing by about a factor of two. At the same time, essential differences in the amount of HCB in the particular compartments can be noted. Particularly, larger amount of HCB in the seawater, vegetation, and sediments compartments was obtained by *J.L.Barber et al.* [2005], which can be explained by different modelling approaches and physical-chemical properties of HCB used.

The spatial distribution of annual mean HCB concentrations in surface air and upper soil layer are presented in Fig. 1.20. The accumulation of HCB in soil compartment and slow rate of decrease can play an essential role in forming the presently observed levels of HCB in the atmosphere. It can be seen that significant levels of air concentrations were predicted for the European part of Russia where elevated levels of HCB in soil can be seen.

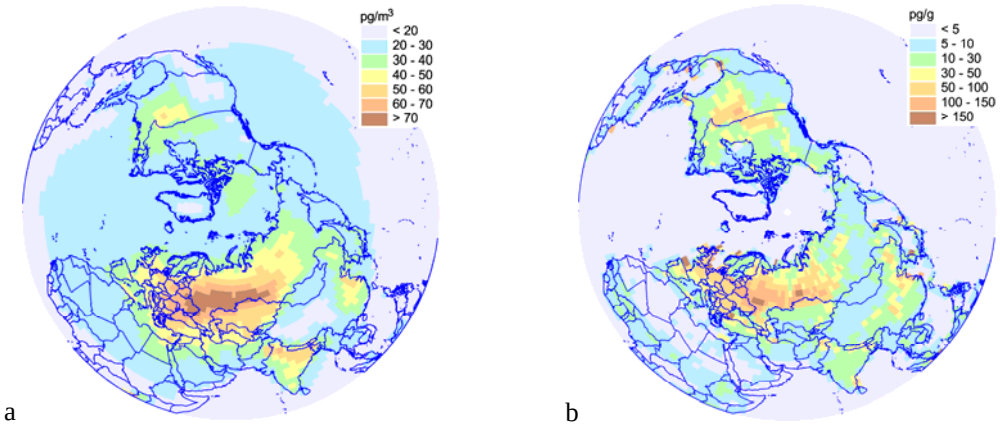

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Fig. 1.20. *Spatial distribution of annual mean HCB concentrations in surface air (a), pg/m^3 and upper soil layer (b), pg/g*

Comparison of model simulation results with observed trends of HCB air concentrations at 9 monitoring stations of EMEP network (CZ3, DE1, DE3, DE8, DE9, FI96, NO1, NO42, and NO99) showed generally reasonable agreement between the modelled and measured concentrations for major part of the period 1991-2008 (Fig. 1.21). It can be seen that modelling results fit measurements of 8 monitoring sites in 1991-2005. For the end of the period, 2005-2008, measurements of NO42 and NO1 indicated slow increase of HCB air concentrations which contradicted with obtained modelling results and can be connected with the deficiencies of constructed emission scenario for this period. Thus, more detailed inventory of HCB emissions and its temporal behavior is required to refine these model estimates and to improve the agreement between the measured and calculated levels of pollution.

Essential difference is found between the model predictions and observed concentrations at the CZ3 site. Measurements of this site demonstrate essential interannual variations which were not predicted by the model. Some of the observed concentrations (for 1997, 1999-2001, 2006, 2007) are in line with model results while for other years observed concentrations are essentially higher. This site is located in the Central Europe with dense population distribution and may be influenced by nearby sources of HCB emissions not well captured by the model and emission inventory used.

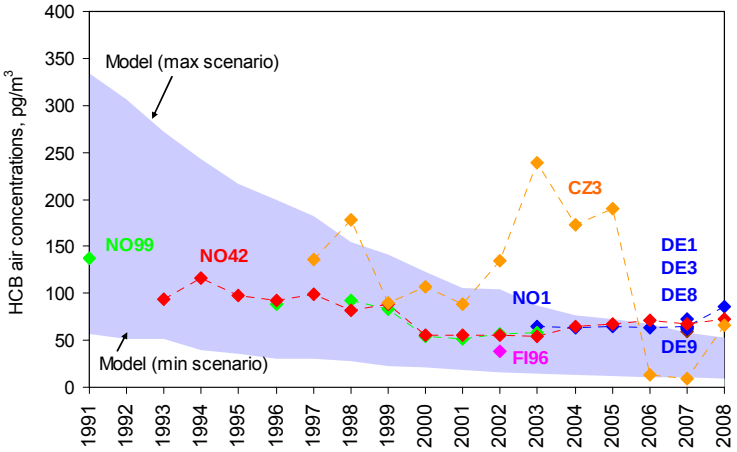

Fig. 1.21. *Modelled HCB annual mean concentrations in surface air (averaged over the stations used for the comparison) obtained under maximum and minimum emission scenarios versus the air concentrations observed at the EMEP monitoring stations in period 1991-2008, pg/m^3. Model predictions are shown as the grey band over the diagram, measurements of monitoring sites are illustrated as colored solid rhombs connected with dotted lines*

On the whole, model simulations with the constructed scenarios of HCB emission permitted to evaluate HCB fate in main environmental compartments in line with other modelling studies. Elaboration of scenarios of historic and current HCB emissions resulted in better agreement of modelling results with available measurements and demonstrated essential role of the historic emissions for the evaluation of HCB pollution levels. Further investigations of HCB long-range transport and fate on global scale taking into account regional inventories of HCB emissions and the use of more detailed spatial resolution for particular regions are required.

1.3. TF HTAP multi-model experiments for POPs

The TF HTAP multi-model intercomparison study was organised to support the developing of better understanding of air pollution intercontinental transport. It includes several modelling groups for different pollutants which perform model simulations on the basis of common methodology. In order to contribute to this activity and to support the development of the EMEP global modelling approach the MSC-E initiated the intercomparison of global/hemispheric POP models following the methodology used within TF HTAP. This year the Centre continued coordination of this work and led the analysis of multi-model experiments aimed at the evaluation of source-receptor relationships for selected POPs.

The study is organised to estimate the effect of emission changes in the selected four HTAP source regions, namely, Europe (EU), North America (NA), East Asia (EA), and South Asia (SA), on the pollution levels in the receptors. The same four regions and the Arctic were considered as the receptors in the analysis of modelling results. Three POP models participated in this intercomparison representing multimedia box models (SimpleBox [*Brandes et al.*, 1996; *Den Hollander et al.*, 2004]), spatially resolved multimedia box models (BETR-Global [*MacLeod et al.*, 2005]), and multimedia chemistry transport model (MSCE-POP [*Gusev et al.*, 2005]). Following the common methodology applied within the TF HTAP intercomparison study the sensitivity of pollution levels to the 20% emission reduction in four HTAP source regions was evaluated. The source-receptor relationships were estimated for three PCB congeners (PCB-28, PCB-153, and PCB-180), and α-HCH, characterized by significant long-range transport potentials and residence times in the atmosphere and other environmental compartments.

Estimates of source-receptor relationships

The effect of regional 20% emission reductions on the annual mean POP concentrations (average of the three models) and the range of variability between the models results are shown in Fig. 1.22. In most cases the largest effect on the pollution levels resulted from local emissions change (6-16 %). The response to the emission perturbation varied across the receptor regions as well as the selected POPs. The differences were mainly caused by the uneven spatial distribution of emissions (different relative contributions of the source regions to the total emission) and the effect of prevailing transport pathways. Particularly, for α-HCH the most marked effect on concentrations in the receptor region (decrease from 2% to 13%) resulted from reduction of SA emissions, which had the largest contribution to the total emission. Similar influence was obtained by the models on PCB pollution levels due to the changes of emissions in the EU region. The lowest effect resulted from the reduction of α-HCH in the EA and NA, and for PCBs in the EA due to their relatively low contributions to the total emissions. It should be noted that emission perturbations were applied only to the anthropogenic component of the emission.

The models were quite consistent in simulating the change in concentrations in air in response to regional emission changes. In most of the cases the differences between the mean response and the model estimates were within a factor of 2-3. In some cases however, especially when the effect of emission changes was low, these differences were higher. The BETR-Global provided generally larger decreases of concentrations in response to emission reduction, which indicated more pronounced atmospheric transport of pollution. Among the reasons for this might be the lower spatial resolution of the BETR-Global in comparison to the MSCE-POP. The smallest effect of emission changes in most of cases was predicted by the SimpleBox.

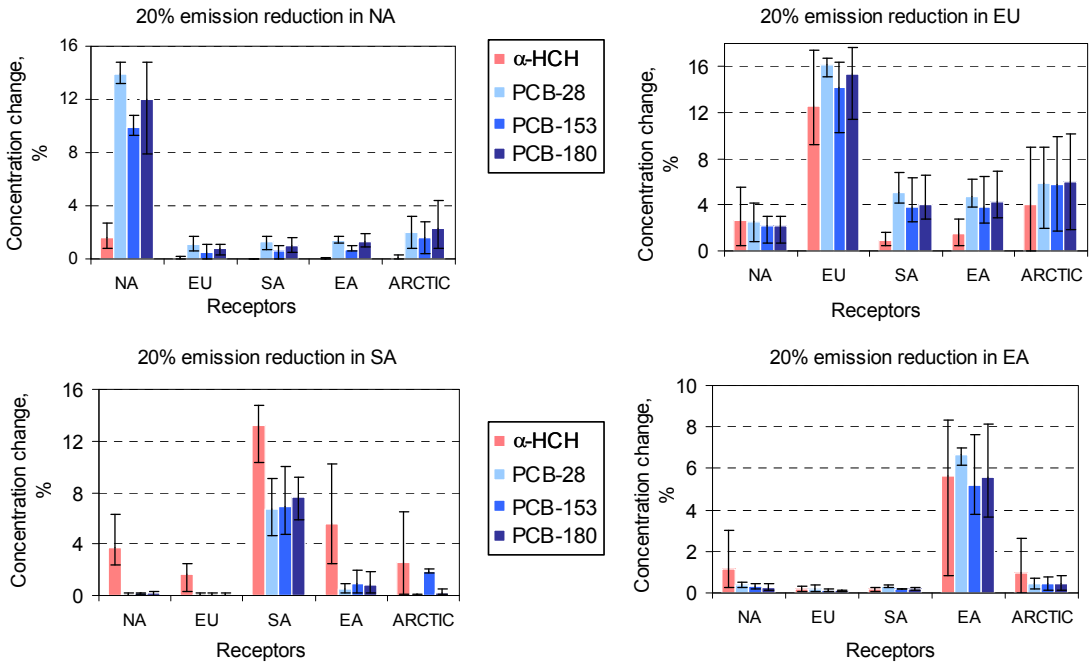

Fig. 1.22. *Relative decreases (in %) of annual mean surface air concentrations in the receptor regions due to 20% emission reductions in four HTAP regions. The bars represent the average change over the models participated. The whiskers show the maximum and minimum estimates of response to the emission reduction obtained by the models*

The influence of emission reduction for the selected source-receptor pairs, representing the zonal transport of pollution in eastward direction and meridional transport to the Arctic, is shown in Fig. 1.23. The strongest SR relationships for PCBs can be seen for pairs EU->SA and EU->EA (4-5% decrease of concentrations). Other SR relationships (NA->EU, EA->NA, and SA->EA) were comparatively weaker (about 1% and lower). For α-HCH the strongest SR relationship was obtained for the pair SA->EA (about 6%). Arctic pollution was mostly sensitive to the changes of emissions in the EU region (for all selected POPs) followed by the NA region (for PCBs) and the SA region (for α-HCH). Comparing the results for PCB congeners, generally higher response to the emission changes can be noted for PCB-28 (especially for MSCE-POP and SimpleBox) than for the heavier congeners (PCB-153, PCB-180) explained by its higher volatility. This can be seen in cases of NA->EU, EA->NA, EU->SA, and EU->EA pairs of sources and receptors.

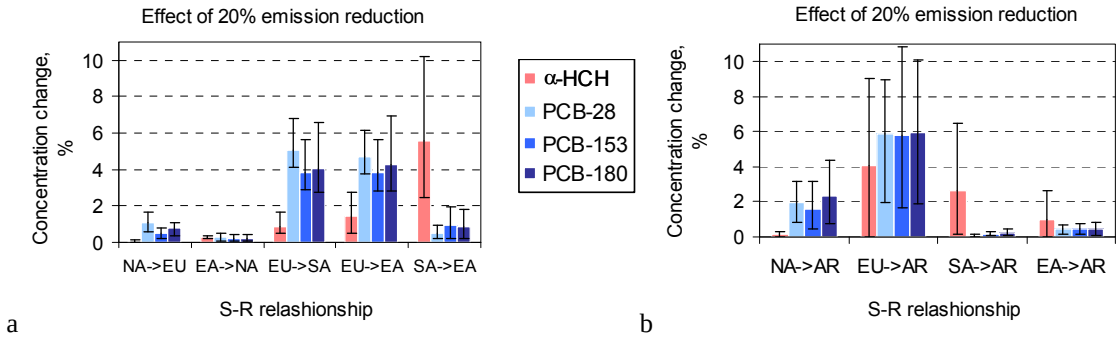

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Fig. 1.23. *Relative decreases (in %) of annual mean surface air concentrations due to 20% decrease of emission for the selected source-receptor pairs. The bars represent the average change over the models participated. The whiskers show the maximum and minimum estimates obtained by the models. The changes are shown for the source-receptor relationships between selected four HTAP regions (a) and between these regions and the Arctic (b)*

Relative importance of emission changes in the HTAP source regions for the selected receptors was additionally studied using experiments with the evenly distributed emissions of arbitrary amount. Similar model simulations were performed with 20% emission reduction in the HTAP source regions for 2001. In this experiment one-year model simulations showed mostly the influence of anthropogenic emission sources of the current year and contribution of secondary sources was minimized.

Estimates of source attribution were obtained extrapolating the effect of 20% emission changes in the HTAP source regions up to the 100%. Contributions of distant and local emission sources (averaged for considered POPs) to the pollution estimated by the BETR-Global and the MSCE-POP models are shown in Fig. 1.24. The highest contribution of foreign sources was obtained for the EA region (about 25%) followed by the NA and the EU regions (about 20%), while the lowest contribution was for the SA region (about 10%). Differences between the estimates of contributions provided by the models were not large, however, the contribution of domestic sources were typically larger in the MSCE-POP model results.

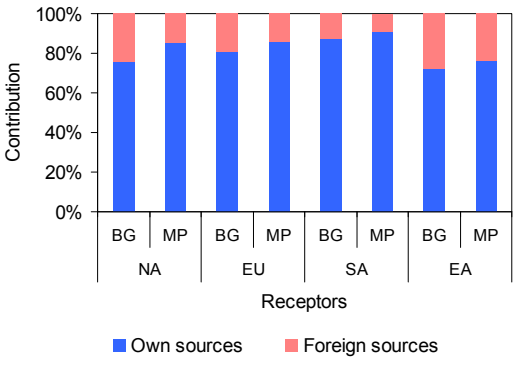

Fig. 1.24. *Estimates of contributions of foreign and own emissions to air concentrations over receptor regions averaged for the selected POPs obtained by the two models BETR-Global (BG) and MSCE-POP (MP)*

Intercontinental transport of POPs

Participated models provided consistent estimates of air concentration distribution on annual level and major transport pathways of selected POPs (Fig. 1.25). The spatial distribution pattern is shown on the example of the MSCE-POP and the BETR-Global models. The areas of elevated concentrations reflect the major source regions and transport pathways. The modelled air concentrations showed the prevalence of α-HCH transport from the EU and the SA regions which affected downwind regions. For PCB-28 marked evidence of the transport from the EU and the NA regions to the Arctic and to the EA region over Eurasia can be seen. Heavier PCB congeners were characterised by weaker transport to the Arctic and the EU influence on other regions. Differences in illustrated air concentration patterns can be originated from slightly different spatial patterns of emissions, models resolution, and modelling approaches used.

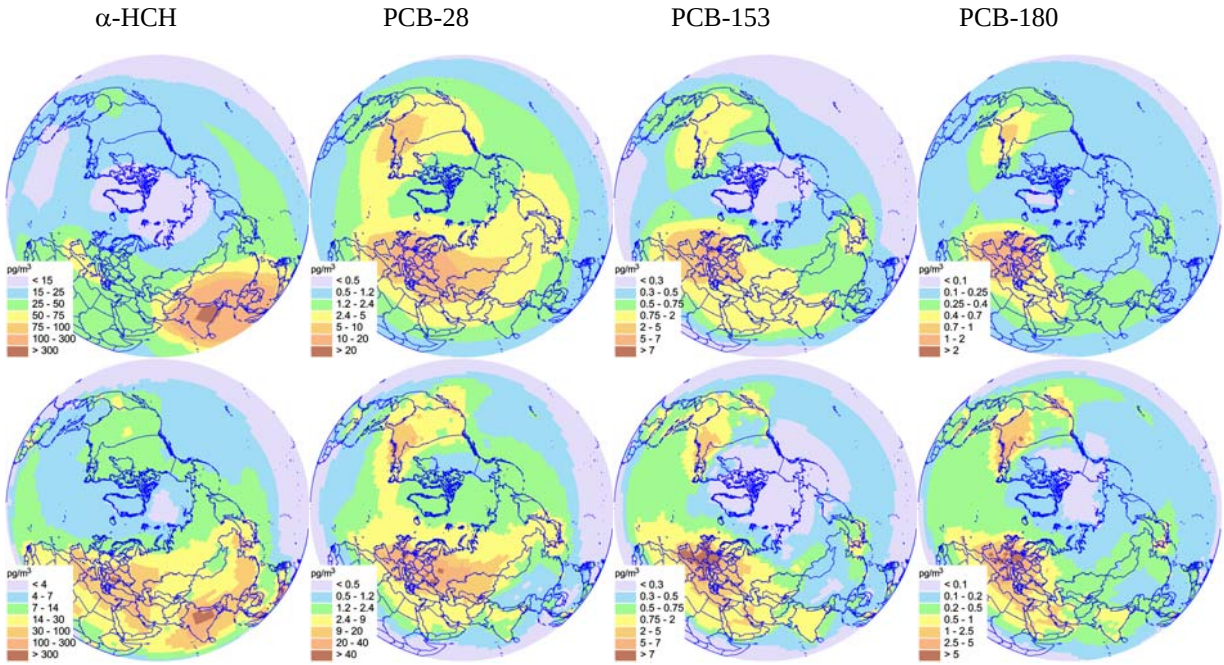

Fig. 1.25. *Spatial distribution of modelled annual mean air concentrations of α-HCH, PCB- 28, 153, 180 obtained by BETR-Global (upper row) and MSCE-POP (lower row) models, pg/m^3*

Comparison of modelled and measured concentrations

Modelling results of MSCE-POP and BETR-Global models were compared with available measurements of the EMEP and AMAP monitoring networks. The scatter plots in Fig. 1.26 illustrate the level of agreement between the modelled and measured annual mean air concentrations of selected POPs. On the whole, better agreement for both models was found for the PCB-153, PCB-180, and α-HCH. For these chemicals the most of predicted air concentrations were within a factor of 3 of the observed levels. Larger discrepancies (a factor of 4 and higher) were obtained for PCB-28, especially for the monitoring sites in Kosetice (the Czech Republic), Rorvik and Aspvreten (Sweden). The overestimation of the air concentrations can be explained by the uncertainties of PCB-28 emission estimates or larger significance of secondary sources less well represented in the model. At the same time both models provided reasonable agreement with measurements of remote sites in the Arctic. Significant underestimation of observed PCB-153 and PCB-180 in air by the BETR-Global can be noted

for the Kosetice site. This might be the effect of low spatial resolution used in the model and the necessity to use finer resolution to better resolve measured levels of pollution in the Central Europe, where ongoing sources are indicated by the emission inventory.

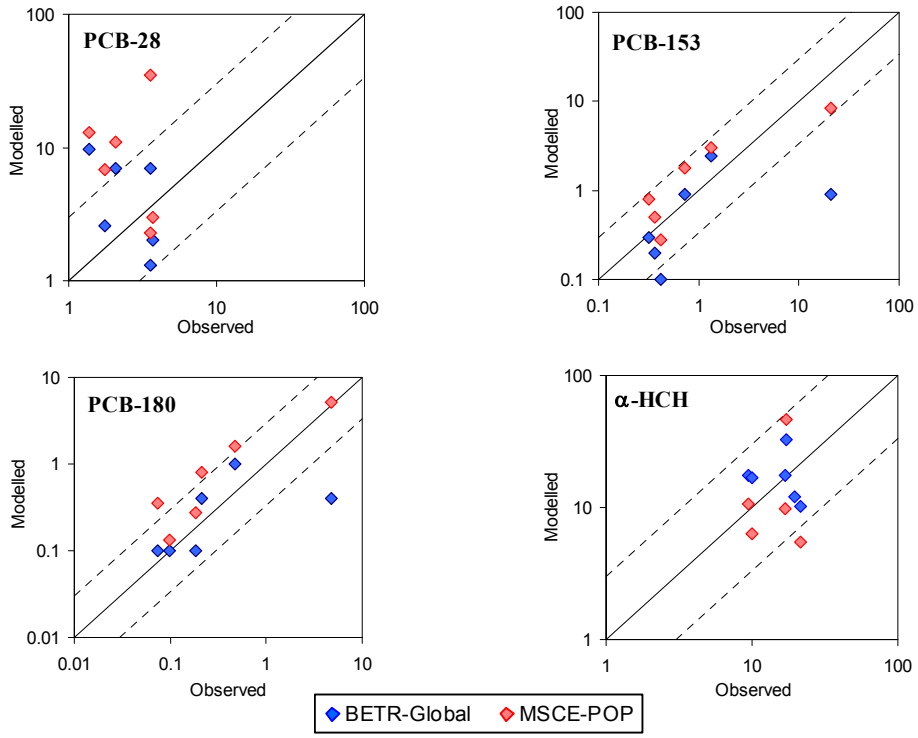

Fig. 1.26. *Comparison of BETR-Global and MSCE-POP model predictions with annual mean air concentrations of PCB-28, PCB-153, PCB-180 and α-HCH in 2001 observed at monitoring sites of EMEP and AMAP networks. Dashed lines show the area of agreement within a factor of 3, pg/m^3*

Main modelling results, obtained in framework of this study, along with the analysis and conclusions are included in the HTAP 2010 Assessment Report, being prepared currently. More detailed description of this study can also be found in [*Jonson and Travnikov*, 2010].

1.4. Contribution to HTAP 2010 Assessment Report

The TF HTAP Assessment 2010 is aimed to summarize the information on hemispheric/global transport of air pollution and to present the current state of understanding of its mechanisms and governing processes. The information on POPs in the Assessment covers recent advances in monitoring of POP levels and trends, availability of emission inventories and modelling tools aimed at evaluation of POP long-range transport, as well as the results of modelling studies illustrating POP long-range transport and source-receptor relationships.

The MSC-E actively participated in the preparation of the Part C of the Assessment devoted to POPs leading compilation of the chapter on modelling of POP fate and source-receptor relationships on global scale, and contributing to the Conceptual Overview of POP intercontinental transport.

Modelling of POP intercontinental transport

Providing the information on the status of the development of POP models it is shown that there is essential progress in elaboration of the modelling approaches describing POP fate in the environment with widely varying levels of detail. Available set of POP models include multimedia box models, trajectory models, and spatially resolved multicompartment chemistry transport models designed for different applications − from screening of a large number of substances with respect to their long-range transport potential and environmental persistence to the detailed evaluation of pollution levels and trends and source-receptor relationships.

Developed POP models have been evaluated against available measurements of non-polar POPs concentrations in the environment. Evaluation studies reveal reasonable agreement between observed and predicted POP concentrations in the atmosphere. Deviations between the modelled and observed concentrations for most of the studied POPs are typically within a factor of three to four or better, however, in some cases the differences can be substantial indicating essential uncertainties both in emission inventories and in modelling approaches.

Modelling studies of POP long-range transport at global (intercontinental transport) and regional (transboundary transport) scales have been performed for a subset of legacy POPs (PAHs, PCBs, PCDD/Fs, HCHs, DDT) and new POPs (brominated flame retardants and fluorinated acids). For most of these POPs a significant role of intercontinental transport was indicated. These studies show the prevalence of POP intercontinental transport with westerly winds within the Northern Hemisphere indicating also migration of POPs to the Arctic region. For example, modelling indicated that PCBs in air in the Arctic originated mostly in Europe and North America. A significant contribution from Asian sources of HCHs to the pollution of other parts of the world was shown. Regional studies of POP long-range transport showed important contributions of distant sources (through the intercontinental transport). Particularly, more than 50% of the PCBs deposited to the Great Lakes were attributable to distant emission sources. The North American sources can have noticeable contribution to the pollution of the EMEP region by PCDD/Fs and B[a]P.

The major processes governing intercontinental transport of POPs at relatively short time scales (monthly or annual basis) are atmospheric transport, gas-particle partitioning, degradation in the atmosphere and deposition. Phase partitioning in the atmosphere, temperature dependence of POP physical-chemical properties and processes are essentially important for the description of intercontinental transport of POPs. Besides the information on the dynamics of aerosols, as POP carriers, and variations of reactive atmospheric species are of importance for the description of POP atmospheric transport.

It should be noted that the evaluation of the source-receptor relationships for POPs can be more complicated compared to the traditional pollutants, because of the significance of the secondary emission sources (re-emission fluxes). The exchange of POPs between the atmosphere and underlying surface, leading to the accumulation in environmental media and subsequent re-emission, and transport with ocean currents play an important role in the determining of pollution levels in locations remote from sources, especially for those chemicals that are cycling in the environment for decades. The role of secondary emission sources can increase while primary emissions are decreasing or ceasing due to emission control measures. Evaluation of the re-emission is subject of essential uncertainties as it depends on the historical emissions and requires simulations of long periods of time. Further work on the development and improvement of model parameterization of gaseous exchange between the atmosphere and underlying surface and improving information about degradation in all the environmental media is needed. Particularly, for correct simulation of POP environmental levels in soil and seawater, further refinement of POP phase partitioning in these media is important. Additionally, the

POP fate in soil, seawater, and vegetation is not sufficiently understood. Thus the extension of available monitoring information (long-term trends, global scale monitoring campaigns, etc.) is required for the improvement of modelling approaches and refinement of model estimates of intercontinental transport. In this respect the sharing of obtained measurements and providing the access to the data of monitoring networks is of importance.

With respect to the evaluation of POP candidate substances it is stressed that both mass balance models and multicompartment chemistry transport models can be used to support the evaluation of environmental hazard associated with new substances. These models evaluate LRTP and environmental persistence of chemicals taking into account main environmental processes such as media exchange and degradation (multimedia approach). The results of model evaluation of new substances are used in the negotiation process as supporting information for revision of the POP Protocol under CLRTAP. In particular, metrics of LRTP and environmental persistence derived from multimedia models are recognized to be relevant for the evaluation of new substances by the Task Force on POPs. At the same time wider use of approaches for the evaluation of new substances based on the application of multimedia POP models of different complexity along with half-life criteria is recommended.

Integrated approach to assessment of POP pollution transport

The Assessment Report presents an important issue for further development of the approach to the assessment of pollution by POPs. Particularly, separate consideration of emission data and the results of evaluation of POP pollution levels by monitoring and modelling cannot provide sufficient information on POP contamination and may lead to considerable uncertainties. There is essential potential for complementing and integrating of the monitoring activities, development of emission inventories, and modelling of POP long-range transport and fate within the assessment and to reduce the uncertainties.

Assessment of environmental contamination by POPs is complicated by the peculiarities of their physico-chemical properties and fate in the environment. Particularly, there is wide range of physical-chemical properties of POPs, and they have potential to intermedia exchange and accumulation in environment. Thus for some POPs contamination levels can be determined not only by current but also by historical emissions. Generally, the assessment of environmental pollution by POPs should provide the following information:

- primary and secondary emissions to air and other media and their trends;
- estimates of spatial and temporal contamination trends based on monitoring and modelling;
- attribution of sources of pollution at global and regional scales;
- predictions of future pollution levels based on future emission scenarios;
- estimates of the effect of future climate changes on the pollution levels.

This information can be provided in framework of further improvement of monitoring, emission inventories, and modelling, and integrating their results. Monitoring of POPs provides measured levels of pollution and their trends. However, spatial and temporal coverage of currently available measurements of POPs limits the analysis of the pollution levels and trends in various environmental compartments at regional (e.g., Europe) and global scales. In this respect essential role in the POP pollution assessment belongs to the modelling of POP long-range transport and fate. Model predictions based on the developed emission inventories provide consistent information on the pollution variations

in space and time, and contributions of particular sources and source categories to the pollution levels. Nesting approaches developed in a number of contemporary global models permit to link the assessment of pollution on global, regional, and local scales. Models are indispensable for the analysis of future contamination based on the scenarios of future emissions and climate change, as well as for the development and improvement of emission inventories and the planning and organization of monitoring networks and field measurement campaigns. Monitoring is important for further development of models especially in areas where certain gaps in knowledge exist. In particular, specially designed field studies and monitoring experiments at the super-sites can provide valuable information for further development of modelling approaches and thus for the improvement of the understanding of POP fate. Taking this in to account integrated approach is an important direction to further improve the assessment of environmental contamination by POPs and to reduce its uncertainties.

Findings and recommendations

The HTAP 2010 Assessment Report provides key findings regarding the significance of global POP transport and recommendations for future work in this area which is particularly important for further activity of the EMEP directed to the evaluation of POP pollution and transboundary transport between the EMEP countries.

Long-range transport as an exposure pathway

- Taking into account the environmental persistence of POPs, long-range transport constitutes an important pathway for human exposure and ecosystem impacts. For many POPs the main route of human exposure is through food.

- The main processes governing intercontinental transport of POPs at relatively short time scales are atmospheric transport, gas-particle partitioning, degradation and deposition. For POPs which have been cycling in the environment since decades the exchange between the atmosphere and underlying surface and transport with ocean currents play an important role in determining pollution levels in remote areas.

- Research by the appropriate national and international bodies with respect the health effects of legacy and emerging chemicals for which long-range transport results in significant exposure is needed. It is a priority to continue to improve our understanding of the fate and transport of POPs through continued efforts in monitoring and processes research, modelling, and inventory of emission sources on global scale.

Monitoring

- Monitoring have provided important information on the behaviour of POPs in the environment and their transport around the globe. Both intensive field studies and sustained monitoring have helped evaluate the transport and fate of POPs. The spatial coverage and resolution of air monitoring have increased considerably due to the adoption of passive air sampling methods.

- At the same time monitoring of POPs in other media, e.g. ocean, snow, soil, and vegetation, are still limited and studies of deposition and degradation processes are required. Sustained monitoring of POPs in environmental compartments and human media is needed for understanding and evaluation of POPs impact on ecosystems and human health.

- Passive air sampling should be sustained to ensure the ability to develop spatial and temporal trends and can be expanded to cover regions where measurements of POPs are currently not available.

- Integrated monitoring of various media is essential for studies of partitioning and intermedia exchange processes of POPs and are important for validation of modelling approaches.

Modelling and process understanding

- Available emission inventories and modelling approaches for POPs provide consistent information on the pollution levels, their spatial and temporal variations, and contributions of particular sources and source categories. Nesting approaches developed in a number of contemporary models permit to link the assessment of pollution on global, regional, and local scales. Besides, models are useful for the analysis of future levels of pollution and their changes due to regulatory activities or projected climate changes.

- Evaluation of models reveals reasonable agreement between available measurements and model predictions of POP concentrations in the atmosphere. Deviations between the modelled and observed concentrations for most POPs are typically within a factor of three to four.

- At the same time modelling of POP distribution in other compartments, namely, soil, vegetation, seawater, snow and ice, is still uncertain and further studies of the uncertainties in physical-chemical properties of some POPs are required.

- There is a need to further study the soil-air partitioning and the role of organic matter content, to apply micrometeorologically based techniques for studying gaseous exchange fluxes, to investigate the role of surface microlayer in the exchange and accumulation of POPs in media, and to improve measurements of physical-chemical properties for model parameterization.

Primary and secondary emission sources

- To quantify and predict source-receptor relationships and transboundary transport of POPs the information on emissions and their temporal development is required. At present there are global and regional emission inventories for some POPs (PCBs, HCHs, HCB, PAHs, PCDD/Fs) elaborated. At the same time, the development of emission inventories is a challenging work and there are still essential gaps in information on emissions sources.

- Most of available inventories of POP emissions quantify primary emissions only, while secondary emissions, formed as a result of accumulation in the environmental media, are also important emission source and may supersede primary emissions in future. However, evaluation of re-emission can be subject of essential uncertainties and requires information on historical emissions.

- Although global emission inventories for some POPs are available at present, further work on elaboration of emission inventories for other POPs and reducing uncertainties of already developed inventories is highly appreciated.

- Dynamic models, which include description of mode of release and relevant air-surface exchange processes reflecting the life-time of POPs in the environment, need to be applied to estimate secondary emissions contribution.

- Development of emission inventories for POPs requires strengthening of co-operation between different scientific communities and international organizations.

Emerging substances

- Environmental observations, process-oriented field and laboratory studies, and modelling can play important role in the screening process of new substances. Both mass balance models and multicompartment chemistry transport models can be used to support the evaluation of environmental risk associated with new substances.

- There is a need to conduct process-oriented research and to adapt monitoring and analytical techniques to target new POPs. Transport models need to be parameterized for these chemicals and new emission inventories be developed.

Effects of climate change

- Projected climate changes have potential to affect mostly all POP pathways, in particular, in the atmosphere, hydrosphere, cryosphere, biosphere, and soil. There is evidence that climate change phenomena, i.e. elevated temperatures, sea-ice reduction, and extreme climate events, can remobilize previously deposited and accumulated POPs.

- For investigation of the effect of climate change on POP fate in the environment scenarios of future climate change including meteorological data for modelling and of future emissions of POPs are required.

For the prediction of POP long-range transport the coordinated use of multimedia modelling, monitoring of POP content and trends in the environment, refinement of the understanding of both primary and secondary emissions, as well as degradation processes of POPs, are essential.

Taking into account the complexity of POP pollution evaluation it is of importance to strengthen co-operation and information exchange between various international programs and bodies.

2. EVALUATION OF POP TRANSPORT AND POLLUTION LEVELS IN EMEP DOMAIN

2.1. Emission data for model assessment

POP emissions were compiled on the basis of official data submitted by the EMEP Centre on Emission Inventories and Projections (CEIP) and unofficial expert estimates for the following pollutants: PAHs (indicator compound B[a]P), PCDD/Fs (the mixture of 17 toxic congeners), PCBs (indicator congener PCB-153) and lindane (γ-HCH). Gridded information for these POPs was prepared by MSC-E for the EMEP domain with spatial resolution 50×50 km^2. For the evaluation of intercontinental transport the spatial distribution of PCDD/Fs, PCB-153 and γ-HCH emissions within the Northern Hemisphere with resolution $2.5^0 \times 2.5^0$ were constructed.

This year pilot calculations of HCB transport were made on the basis of emission scenarios. Detailed information on HCB emission scenarios is presented in Section 1.2 of the report.

Information on POP emission changes in the period of 1990-2008 for each EMEP country and spatial distribution of annual emissions for 2008 can be found in the Internet on the MSC-E web site [*http://www.msceast.org/*]. Officially submitted data on POP emissions are available at the web site of CEIP [*http://www.emep-emissions.at/ceip/*].

POLYCYCLIC AROMATIC HYDROCARBONS (PAHs)

Gridded emissions for B[a]P within the EMEP domain for 2008 were prepared using officially submitted PAH emissions. For the European countries, which did not report their emissions, unofficial data of TNO emission inventory [*Denier van der Gon et al.*, 2005; *MEPA*, 2007] were used. The B[a]P emissions in the Asian part of Russia were estimated using official emission data for the European part of the country and the ratio between the population of the European and the Asian parts of the country. The B[a]P emission values for Tajikistan, Turkmenistan and Uzbekistan were taken from the global atmospheric emission inventory of PAHs prepared by *Y.Zhang and S.Tao* [2009]. For Kazakhstan and Kyrgyzstan unofficial emission data from TNO emission inventory [*Denier van der Gon et al.*, 2005] were applied. The spatial distribution of B[a]P emissions in the Central Asian countries and the Asian part of Russia was determined on the basis of data on population density [*Li*, 1996] obtained from the web site of Canadian Global Emissions Interpretation Centre [*http://www.ortech.ca/cgeic*].

The spatial distribution of B[a]P emissions for 2008 is illustrated in Fig. 2.1. Elevated levels of B[a]P emissions (50 – 300 g/km^2/y) can be noted for the central, southern and eastern parts of Europe. Countries of the Northern and Western Europe, Russia, and the Central Asian countries are characterised by relatively low emission fluxes (5 - 50 g/km^2/y).

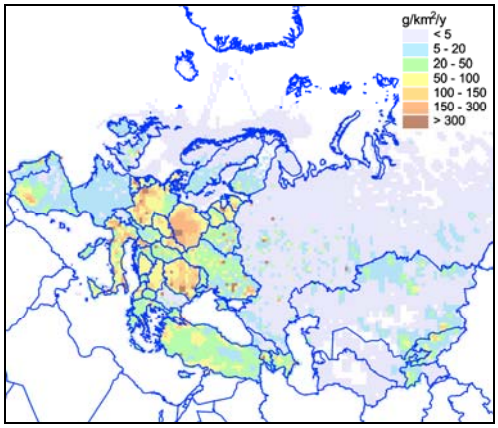

Fig. 2.1. *Spatial distribution of B[a]P emissions in 2008 over the EMEP domain with resolution 50×50 km², g/km²/y*

Total B[a]P emission within the EMEP grid in 2008 used for model calculation is estimated as 507 tonnes. Maximum contribution to the total B[a]P emission within the EMEP domain in 2008 was made by the Ukraine (20%) followed by Poland (9%), Italy (9%), and Romania (9%).

Officially reported information on uncertainties of PAH emissions for 2008 is available for Denmark, Finland, France, Sweden and the United Kingdom. According to the data submitted by countries, the uncertainty of the Danish B[a]P emissions is 939% [*Nielsen et al.*, 2010], whereas for the UK uncertainty of B[a]P emissions is in the range of -60% to 200% [*Murrells et al.*, 2010]. The uncertainty of French and Swedish PAH-4 emissions is 75% [*CITEPA*, 2010] and 583% [*SEPA*, 2010], respectively. For the Finnish PAH-4 emissions the uncertainty is in the range from -78% to 187% [*SYKE*, 2010].

POLYCHLORINATED DIBENZO(P)DIOXINS AND DIBENZOFURANS (PCDD/Fs)

This year preparation of annual PCDD/F emission values for 2008 used for modelling was carried out on the basis of official information submitted by CEIP. In absence of officially reported information unofficial data of emission inventories [*Denier van der Gon et al.*, 2005; *Pulles et al.,* 2006] were applied. The information about spatial distribution of dioxin emissions at least for one year of the period 1990-2008 was provided by 26 countries (Austria, Belarus, Belgium, Bulgaria, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Iceland, Ireland, Italy, Latvia, Lithuania, Netherlands, Norway, Poland, Slovakia, Spain, Sweden, Switzerland and the United Kingdom). The gridded 2008 emissions were prepared by CEIP and MSC-E. For Austria, Belarus, Croatia, Estonia, Ireland, Italy, Latvia, Lithuania and Spain the gridded data provided by CEIP were used. For the rest 41 countries the gridded data were prepared by MSC-E.

The PCDD/F emission for the Asian part of Russia was estimated using officially reported rates for the European part of the country and data on population density similar to PAHs. The PCDD/F emissions of Tajikistan, Turkmenistan and Uzbekistan for 2008 were taken from the unofficial inventory of PCDD/F emissions in the Central Asian countries made in the framework of the global International POPs Elimination Project (IPEP) [*Hodjamberdiev,* 2006]. The latest available information on PCDD/F emission in the USA was taken from the dioxin and furan inventories prepared by [*UNEP*, 1999] for 1995. The spatial distribution of PCDD/F emissions in the Central Asian countries and the Asian part of Russia was constructed on the basis of data on population density [*Li*, 1996].

The spatial distribution of PCDD/F emissions in the EMEP domain for 2008 is shown in Fig. 2.2. Significant levels of PCDD/F emissions (0.5 - 5 ng TEQ/m^2/y) can be seen in countries of the Central, Southern, and Eastern Europe. Other parts of Europe, in particular, the Northern and Western Europe, are characterised by lower emission fluxes varying from 0.001 - 0.5 ng TEQ/m^2/y.

The total emissions of PCDD/Fs in 2008 used for model calculations within the Northern Hemisphere were estimated to 9.6 kg TEQ, including 6.8 kg TEQ/y within the EMEP domain and 2.8 kg TEQ/y in

North America. The most significant contributions to the total annual emission within the EMEP region were made by the Russian Federation (11%), Turkey (11%), the Ukraine (11%), and Poland (4%).

Officially reported information on uncertainties of PCDD/F emissions for 2008 is available for Denmark, Finland, France, Sweden and the United Kingdom. The uncertainty of Danish, French and Swedish dioxin emissions is 627% [*Nielsen et al*., 2010], 60% [*CITEPA*, 2010] and 116% [*SEPA*, 2010], respectively. For the Finnish dioxin emissions the uncertainty is in the range from -40% to 56% [*SYKE*, 2010], whereas the uncertainty of the UK dioxin emissions is in the range from -50% to 200% [*Murrells et al*., 2010].

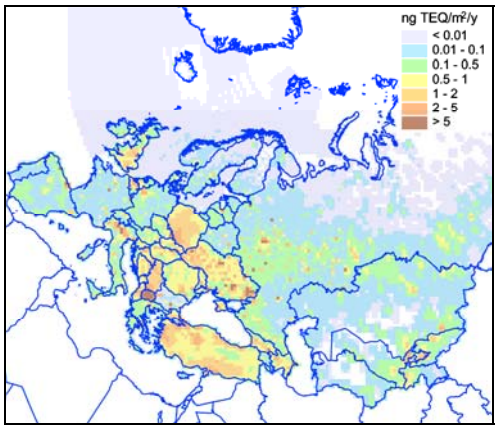

Fig. 2.2. *Spatial distribution of PCDD/F emissions in 2008 over the EMEP domain with resolution 50×50 km^2, ng TEQ/m^2/y*

POLYCHLORINATED BIPHENYLS (PCBs)

PCB modelling requires the information on congener composition of PCB mixture. Since official data do not include such information, emission data for modelling of PCB long-range transport and deposition were prepared on the basis of unofficial global inventory of PCB usage and emissions [*Breivik et al*., 2007]. However, spatial distributions of PCB emissions delivered by countries were taken into account.

The emission inventory provides consistent set of historical and future emissions of 22 individual PCB congeners from 1930 up to 2100. The spatial distribution of annual PCB emissions on global scale with resolution 1°×1° was prepared. Three scenarios of global PCB emissions, namely, minimum scenario, default scenario, and maximum scenario, describe the range of emission variations estimated as an order of magnitude [*Breivik et al*., 2002 a,b]. Computations with the hemispheric MSCE-POP model using the three emission scenarios allowed evaluating what emission data could be used as input information for modelling to get reasonable description of pollution levels within the EMEP region. Using the results of these computations, the emission values between the maximum and default emission scenario were selected for the investigation of PCB long-range transport and deposition.

The indicator congener PCB-153 was selected for the evaluation of pollution levels for 2008 on regional and hemispheric scales. The spatial distribution of PCB-153 emissions within the EMEP region was prepared using the data of emission inventory of *K.Breivik et al*. [2007] and spatial distribution of PCB emissions officially submitted by EMEP countries (Fig. 2.3). For the evaluation of PCB-153 long-range transport within the Northern Hemisphere the gridded emissions with resolution 2.5°×2.5° were arranged (Fig. 2.4).

It can be seen that the most significant levels of PCB-153 emission fluxes are characteristic of the EMEP region (0.01 – 2 g/km^2/y). Other regions are characterised by comparatively lower annual emissions (less than 0.005 g/km^2/y).

Officially reported information on uncertainties of PCB emissions for 2008 is available for Finland, France and the United Kingdom. According to the submitted information, the uncertainty of French PCB emissions is 31% [*CITEPA*, 2010]. Uncertainty of Finnish PCB emissions is in the range from - 64% to 115% [*SYKE*, 2010], and the uncertainty of the UK PCB emissions is in the range from - 40% to 90% [*Murrells et al*., 2010].

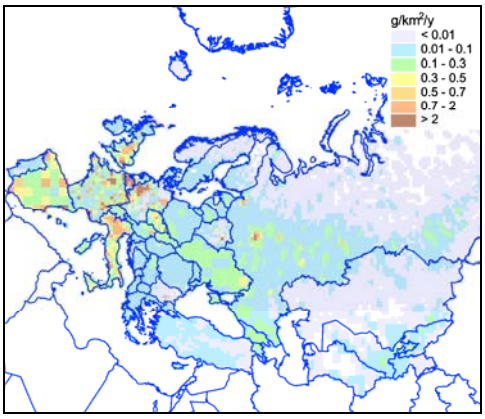

***Fig. 2.3**. Spatial distribution of PCB-153 emissions over the EMEP domain in 2008 with resolution 50×50 km², g/km²/y*

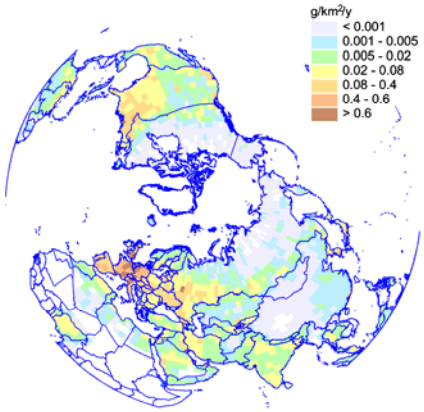

***Fig. 2.4**. Spatial distribution of PCB-153 emissions within the Northern Hemisphere in 2008 with resolution 2.5⁰×2.5⁰, g/km²/y*

LINDANE (γ-HCH)

The compilation of emission data for lindane within the EMEP region was based on the official emission data complemented by unofficial expert estimates. Taking into account that the usage of lindane was severely restricted in Europe and that 27 countries reported no application of lindane, no emission of γ-HCH in 2008 was assumed for these countries in the model simulations. Data on emissions or usage of γ-HCH outside the EMEP domain for the period 1990-2008 were compiled from several sources and covered only part of the Northern Hemisphere being thus subject of significant uncertainties. Three groups of emission sources were considered, namely, European, North American, and Chinese emission sources. For the EMEP region official information on emissions was complemented by unofficial estimates on γ-HCH usage in the period 1990-1996 [*Pacyna et al.*, 1999] and expert estimates of γ-HCH emission for 2000 [*Denier van der Gon et al.*, 2005]. The γ-HCH emission values in North America and China were estimated on the basis of information from [*Li et al.*, 1996; *Macdonald et al.*, 2000; *Li et al.*, 2001; and *Li,* 2004].

The spatial distribution of γ-HCH emissions within the EMEP domain was obtained from the officially submitted data (Belgium, Germany, and Spain) and from unofficial expert estimates [*Pacyna et al.*, 1999; *Denier van der Gon et al.*, 2005]. The gridded γ-HCH emissions for North America were prepared by *Y.F.Li* [2004]. Spatial distribution of γ-HCH emissions in China was produced using the cropland area data available in the Canadian Global Emissions Interpretation Centre [*http://www.ortech.ca/cgeic*].

The uncertainty of the United Kingdom lindane emissions, officially reported for 2008, is in the range from -100% to 400% [*Murrells et al.*, 2010].

2.2. Assessment of POP pollution levels

This section presents a brief information on modelled pollution levels within the EMEP domain for selected POPs, namely, PAHs (B[a]P), PCDD/Fs, PCBs (PCB-153), and γ-HCH. Detailed description of pollution levels and the comparison of modelling results with measurements can be found in the MSC-E Technical report [*Ilyin et al.*, 2010] and country-specific reports are available at MSC-E web site.

BENZO[A]PYRENE (B[a]P)

The simulation of the environmental levels for B[a]P was performed by means of the regional version of MSCE-POP model for 2008. Taking into account the fact that this pollutant exists in the atmosphere mainly in the particulate phase, it can be concluded that the influence of non-EMEP sources and secondary sources (re-emissions) will not be essential for the considered pollutant.

The spatial distribution of modelled B[a]P annual mean air concentrations for 2008 is shown in Fig. 2.5. The model predicted low B[a]P concentrations for remote areas (about 0.001 – 0.02 ng/m^3), in particular, in the northern part of the Scandinavian Peninsula, northern areas in the UK, Iceland and Spitzbergen. Moderate values of concentrations (0.02 – 0.08 ng/m^3) were obtained for the United Kingdom, Sweden, and wide areas in Finland, France, Spain, and the Russian Federation. Elevated levels of B[a]P air concentrations were obtained in the central, eastern, and southern parts of Europe (about 0.08 – 1 ng/m^3). The most significant concentrations (more than 1 ng/m^3) can be seen in the eastern part of the Ukraine.

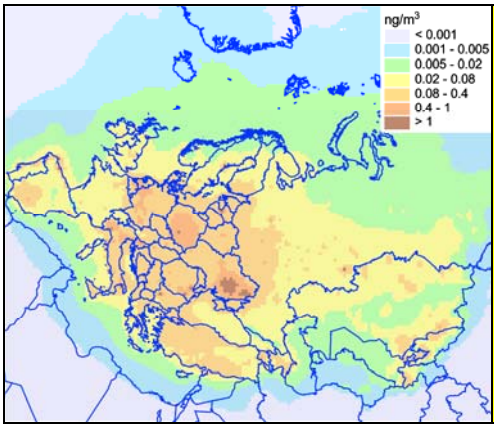

Fig. 2.5. *Spatial distribution of B[a]P concentrations in air in 2008, annual means, ng/m^3*

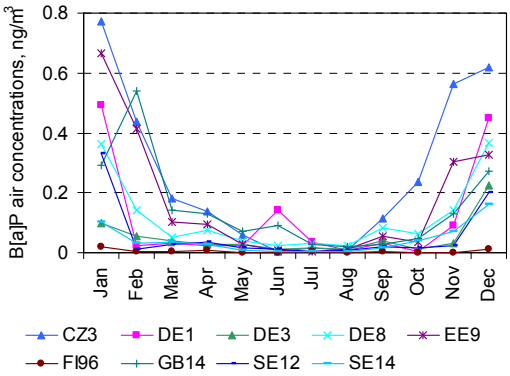

Fig. 2.6. *Seasonal variations of B[a]P concentrations at a number of EMEP sites, ng/m^3*

It should be stressed that Fig. 2.5 presents annual means of B[a]P concentrations in Europe. At the same time air concentrations for this pollutant are subject to high seasonal variations. This can be illustrated by measured B[a]P concentrations at a number of EMEP measurement sites in different months of 2008 (Fig. 2.6). The difference between B[a]P concentration levels in winter and summer months can be as much as an order of magnitude. Besides, the model with 50×50 km^2 resolution calculates concentrations averaged over grid cells. Due to variability of concentrations inside grid cells, concentrations at particular locations could exceed calculated concentrations.

While interpreting the obtained results it also should be taken into account that the contamination levels can be essentially higher due to uncertainties in emission data. From this viewpoint reporting emission uncertainties by countries is highly appreciated. With this information, calculations using maximum and minimum emission scenarios can be made to characterize possible range of predicted contamination levels.

So, it can be concluded that concentration levels in the central, eastern and southern Europe can exceed the level of 1 ng/m^3 during some time periods, which is MPL (maximum permissible level) for the considered pollutant accepted in a number of countries. For refinement of evaluation of air concentrations at particular locations simulations with improved seasonal variations and finer spatial resolution are required. Additional information on emission uncertainties can help in the evaluation of ranges of predicted contamination levels.

The comparison of the calculations made for 2008 with that performed for 2007 in the previous year show that, in spite of the fact that average concentrations in the EMEP domain was enlarged by about 12%, changes of the contamination levels within particular countries are more essential and can reach 10 – 20% and higher, being either positive or negative. Changes in air concentrations for a number of EMEP countries in comparison to the changes in emission totals of these countries are shown in Table 2.1.

Table 2.1. *Changes in B[a]P air concentrations for some EMEP countries in comparison to the changes in emission totals*

Country	Emission totals, tonnes/year			Air concentrations, ng/m^3		
	2007	2008	Difference, %	2007	2008	Difference, %
France	5.97	5.01	-16.2%	0.06	0.05	-22.2%
Luxembourg	0.38	0.37	-4.8%	0.24	0.20	-14.6%
Portugal	2.97	3.01	1.4%	0.08	0.07	-8.0%
Spain	19.31	18.16	-5.9%	0.09	0.08	-12.0%
Belgium	4.60	4.18	-9.1%	0.34	0.29	-15.6%
…	…	…	…	…	…	…
Armenia	2.35	2.40	2.2%	0.16	0.17	10.3%
the Ukraine	99.88	100.68	0.8%	0.58	0.67	15.3%

Average air concentrations in some countries were decreased up to about 20% compared to that calculated for 2007. However, the concentrations in the other countries were increased up to about 20% and higher. The reason of these changes is both the changes in emissions and in meteorological conditions.

The concentration reduction in countries with comparably large emissions is conditioned in a great extent by the reduction of domestic emissions. Thus, emission reduction in France (16.2%) approximately corresponds to the reduction of mean annual concentrations in this country (22.2%). However, even for such countries concentration reduction does not always correlate with emission reduction. For example, emission total in Portugal *increased* by 1.4% whereas air concentrations *decreased* by 8.8%. This can be explained by the influence of the change in meteorological condition. Below the primary analysis of the influence of meteorological variability to the concentration levels is shown.

For countries with small territory and comparably low emissions the influence of changes in meteorology is prevailing. Thus, emission reduction in Luxembourg (from 385 to 366 kg/y) amounts to about 5% whereas the reduction of mean annual concentrations reaches 14.6%. To reveal the reasons of such a difference, back trajectory approach was applied. Fig. 2.7 shows the spatial distribution of probabilities for the trajectories arriving to Luxembourg during a year to pass through EMEP grid cells.

It is evident, that the atmospheric transport in 2007 comes from contaminated regions of Eastern Europe (e. g. from Poland) in higher extent than in 2008. This tendency is even more pronounced for particular months (see Fig. 2.8).

Thus, meteorological variability can lead to the changes of air concentrations in particular countries by ±10 – 20%. It should be taken into account that country averaged air concentrations were used in the above analysis. The changes of air concentrations inside the country due to the meteorological variability can be even more significant.

The application of back trajectory technique to the investigation of the influence of meteorological variability to pollution levels in the EMEP countries is planned to be continued in future.

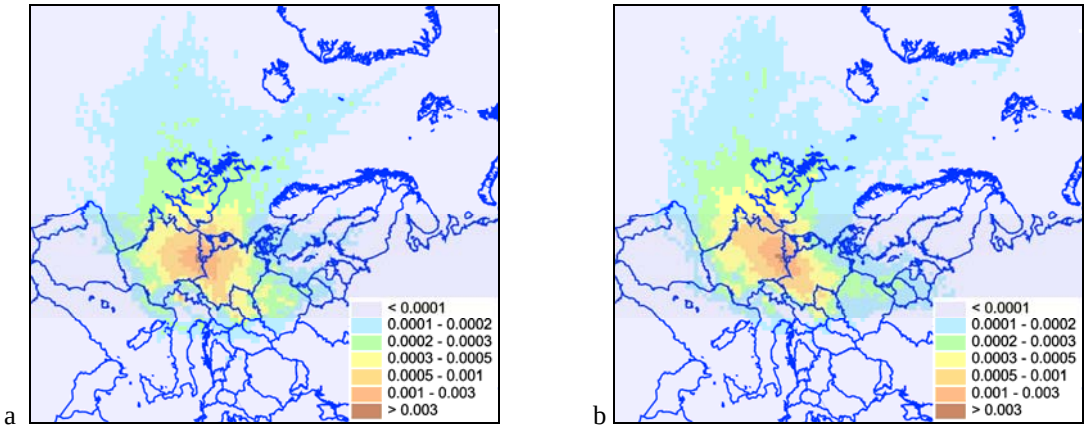

Fig. 2.7. *Spatial distribution of probabilities for the trajectories arriving to Luxembourg (a) in 2007, (b) in 2008 to pass through EMEP grid cells*

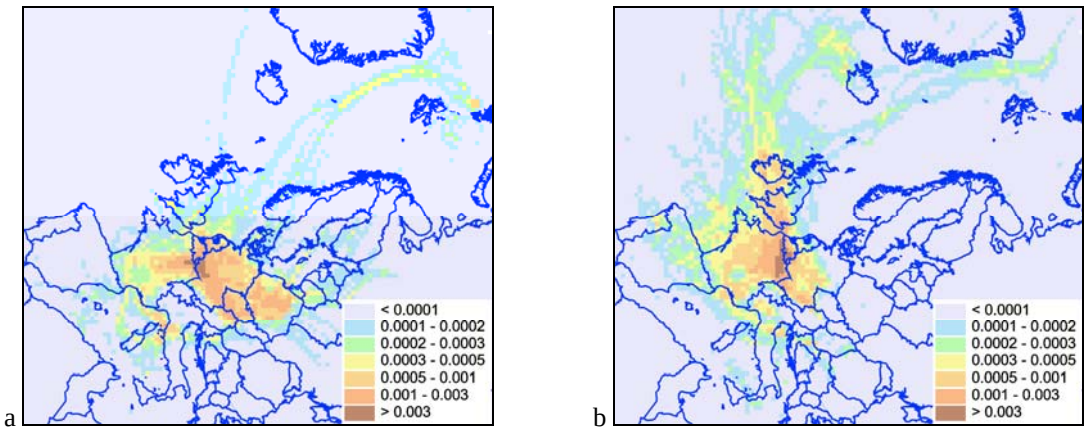

Fig. 2.8. *Spatial distribution of probabilities for the trajectories arriving to Luxembourg: (a) in October 2007, (b) in October 2008 to pass through EMEP grid cells*

POLYCHLORINATED DIBENZO(p)DIOXINS AND DIBENZOFURANS (PCDD/Fs)

Evaluation of transboundary transport of PCDD/Fs for 2008 was carried out for the mixture of 17 toxic PCDD/F congeners. Model simulations were based on the physical-chemical properties of PCDD/F indicator congener 2,3,4,7,8-PeCDF. Differences between the modelling results based on the physical-chemical properties of indicator congener and the results of simulations using the properties of all 17 toxic congeners did not exceed 20% for air contamination levels and were within a 5% range for country-to-country deposition matrix [*Dutchak et al.,* 2004]. Taking into account the ability of PCDD/Fs to be accumulated in various environmental media (soil, seawater, vegetation) and be transported at hemispheric level, initial and boundary conditions for these calculations were

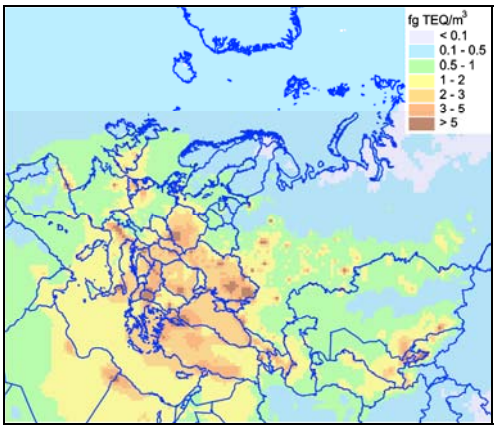

Fig. 2.9. *Spatial distribution of modelled PCDD/Fs concentrations in air in 2008, annual means, fg TEQ/m^3*

obtained by application of the hemispheric version of MSCE-POP model for the period from 1970 to 2008. On hemispheric scale the influence of PCDD/F emission sources of the USA and Canada was taken into account. For other regions outside the EMEP domain emission data for PCDD/Fs are currently not available. Therefore the contribution of non-EMEP emission sources of PCDD/Fs in this model simulation can be underestimated.

The spatial distribution of modelled PCDD/F annual mean air concentrations for 2008 is shown in Fig. 2.9. The model predicted low PCDD/F concentrations for remote areas (less than 0.5 fg TEQ/m^3), in particular, in the most part of the Scandinavian Peninsula, northern areas in the UK, Iceland and Spitzbergen. Moderate values of concentrations (0.5 – 2 fg TEQ/m^3) were obtained for the United Kingdom, France, Spain, and the Russian Federation. Elevated levels of PCDD/F air concentrations were obtained in the central, eastern, and southern parts of Europe (about 2 – 5 fg TEQ/m^3). The most significant concentrations (more than 5 fg TEQ/m^3) can be seen in eastern part of Ukraine and some regions of the Southern and Eastern Europe.

It should be stressed that strong seasonal variations of air concentrations are characteristic of PCDD/Fs since main emission sectors for these pollutants are combustion processes. The submission of the information on PCDD/F emission seasonal variations by countries is therefore strongly desirable. Further, all measurement data on PCDD/F contamination levels are reported to the EMEP on voluntary basis which makes it hard to evaluate possible uncertainties in modelling results. At present the comparison of calculated contamination levels of PCDD/Fs with available measurements at three Swedish and Finnish measurement sites in the framework of Swedish EPA project is ongoing. This work is aimed at the refinement of model assessment of the contamination of the EMEP region by PCDD/Fs.

The comparison of calculations for 2008 with the calculations made in the previous year for 2007 shows that the average of air concentrations over the EMEP domain had been decreased by about 8%. However, similar to the case of B[a]P, the changes of air concentrations in particular countries were found to be more essential (- 30% - 20%). For example, air concentrations in France, Portugal and Spain decreased by about 20%, whereas air concentrations in Kyrgyzstan, the Republic of Moldova and Latvia increased by 10% - 20% in comparison with calculations for 2007. The difference again can be explained by changes in meteorology and in spatial distribution of emissions.

POLYCHLORINATED BIPHENYLS (PCBs)

Investigation of PCB intercontinental transport and pollution levels for 2008 was carried out for the EMEP region with resolution 50×50 km^2. Modelling was made for one indicator congener PCB-153 on the basis of the global emission inventory of PCBs prepared by *K. Breivik et al.* [2007]. Due to essential persistence of PCBs in the environment and their accumulation in seawater and soil compartments, the contributions of non-EMEP sources and secondary emission sources to the contamination of the EMEP region occur to be essential. To take this contributions into account, hemispheric simulations were performed for a sufficiently long period preceding the year 2008. During the hemispheric model run initial and boundary conditions for regional modelling of PCB-153 for 2008 were prepared.

Annual mean air concentrations of PCB-153 for 2008 are shown in Fig. 2.10. The calculated contamination levels in the remote regions (Scandinavian Peninsula, northern parts of the UK and the Russian Federation) are found to be from 0.1 to 1 pg/m^3. Moderate levels of contamination (1 – 3 pg/m^3) were calculated for the Eastern Europe, wide areas in Spain and France. Elevated levels of PCB-153 concentrations (3 – 5 pg/m^3 and more) were obtained for some regions of the Western and Southern Europe (western Germany, the Netherlands, northern France and northern Italy).

The comparison of calculations for 2008 with those made for 2007 in the previous year shows slight increase of air concentrations averaged over the entire EMEP domain (about 2.5%). However, due to changes in meteorology and spatial distribution of emissions, more essential changes of air concentrations in particular EMEP countries take place. Thus, essential decrease of air concentrations (10% - 20%) was obtained for Finland, Estonia, Hungary, Belarus, Slovenia, the Czech Republic, Croatia, Austria, France, Spain and Portugal. The increase of air concentrations in comparison with 2007 (10% - 20%) was found for Cyprus, Kazakhstan, the Russian Federation, the Ukraine, Ireland, the Netherlands, Uzbekistan, Tajikistan and Kyrgyzstan.

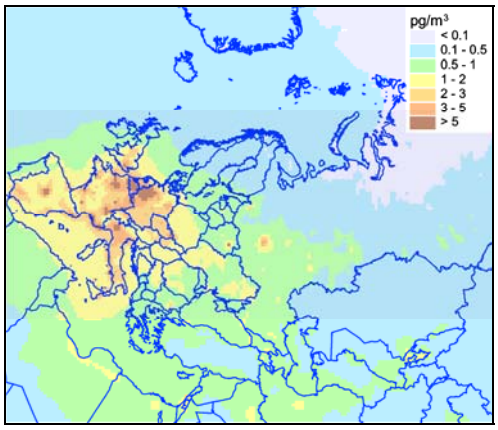

Fig. 2.10. *Spatial distribution of PCB-153 concentrations in air in 2008, pg/m^3*

LINDANE (γ-HCH)

The environmental levels of lindane (γ-HCH) have been significantly decreased within the EMEP region during two recent decades due to the reduction of its application. In particular, non-zero lindane emissions for at least one year in the period 2000 – 2008 were reported only by five European countries: the United Kingdom, Spain, Belgium, Croatia, and Romania. In other European countries, according to the officially reported information, there was no application of lindane or its emission was insignificant.

For the interpretation of the results of model calculations it should be taken into account that the data on emissions or usage of γ-HCH outside the EMEP region for the period 1990-2008 were compiled from several sources and covered only part of the Northern Hemisphere being thus subject of significant uncertainties. Only two groups of non-EMEP emission sources were considered in the calculations: North American, and Chinese ones.

Model simulation of γ-HCH long-range transport and accumulation in the environmental compartments within the Northern Hemisphere was performed with the hemispheric MSCE-POP model for the period of 1985-2008. Modelling of γ-HCH distribution in the EMEP region for 2008 was carried out by regional MSCE-POP model with allowance to initial and boundary conditions obtained by hemispheric model.

Spatial distribution of annual mean γ-HCH air concentrations within the EMEP region is presented in Fig. 2.11. Elevated γ-HCH air concentrations (50 pg/m^3 and above) in 2008 are characteristic of the United Kingdom and Spain. Moderate levels of air concentrations were obtained for the countries of the Western and Central Europe (5 – 20 pg/m^3). The countries of the Eastern Europe and Central Asia are characterized by particularly low γ-HCH air concentrations (about 5 pg/m^3), which is mainly connected with the absence of information on the emissions for these regions.

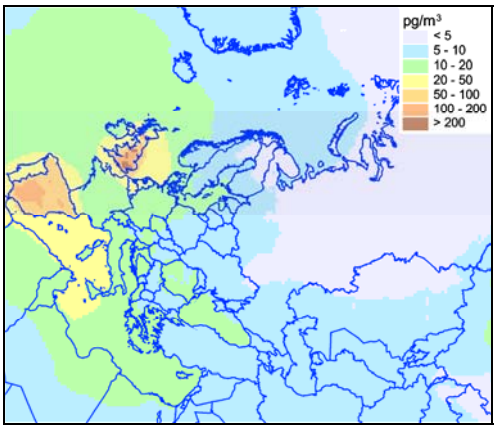

Fig. 2.11. *Spatial distribution of modelled γ-HCH concentrations in air in 2008, pg/m^3*

Similar to the above considered pollutants, in spite of little change of air concentrations averaged over the entire EMEP domain (about 4%), changes of air concentrations in particular EMEP countries are more essential (from -15% to about 20%).

2.3. POP transboundary pollution

This year the evaluation of transboundary transport of PAHs (B[a]P) and PCDD/Fs (the mixture of 17 toxic congeners) within the EMEP region was continued. The country-to-country matrix for 2008 was calculated with the use of the regional MSCE-POP model at the EMEP domain with resolution 50×50 km^2. Emission data used for the modelling purposes are described in Section 2.1. In addition, first attempt to evaluate the transboundary transport for PCB-153 was performed. These estimates for PCB-153 should be considered as preliminary since they were performed using expert estimates of emissions complemented for several countries by the officially reported information on spatial distribution of emissions. It should be noted that official emissions of PCBs do not contain the congener composition which is important for modelling of PCB pollution levels. Due to high long-range transport potential and persistence in the environment, contributions of non-EMEP emission sources and secondary sources (re-emissions) are expected to be essential for this pollutant. These peculiarities of PCB-153 can leave an imprint at the evaluation of source-receptor relationships for this pollutant.

Estimates of deposition due to transboundary transport of pollution include contributions of emission sources of European countries, distant sources outside the EMEP grid, and re-emission. However, since B[a]P exists in the atmosphere mainly in the particulate phase, the contributions of remote emission sources for this pollutant seem to be comparatively small.

A present the following modelling approach is implemented in the MSCE-POP model for the evaluation of transboundary transport of POPs. Only the anthropogenic emissions of the current year, transported through the atmosphere, are considered as a contribution of a given country to the transboundary deposition flux over another country. The pollutant that was accumulated by the media and then re-volatilised to the atmosphere is considered as the contribution of re-emission and the origin of this part of deposition is not tracked. In principle, it is possible to track the contribution of emissions of each country during multiple cycles of accumulation and re-emissions. However, the contribution of emissions of the current year in re-emission flux is negligible in comparison with the contributions of emissions of a long period proceeding the considered year, so tracking of contributions of various countries including accumulation and re-emission processes can be performed in the course of long-term calculations. Such calculations require reliable data on historical emissions. From the other hand, the current approach calculates the part of transboundary transport which is subject to regulations on the political level.

Detailed description of the approach can be found in the Technical report [*Gusev et al.*, 2009]. The information on the calculated source-receptor relationships for 2008 is presented in the Annex A.

BENZO[A]PYRENE (B[a]P)

Anthropogenic emission of B[a]P within the EMEP domain for 2008 was accounted for about 507 tonnes. Total deposition of B[a]P to the European and the Central Asian countries in 2008 from anthropogenic emission sources amounted to 197 tonnes. This figure is in between the result obtained for 2006 (231 tonnes, [*Gusev et al.*, 2008]) and the result obtained for 2007 (175 tonnes, [*Gusev et al.*, 2009]). These changes can be attributed to the interannual variability of meteorological parameters and

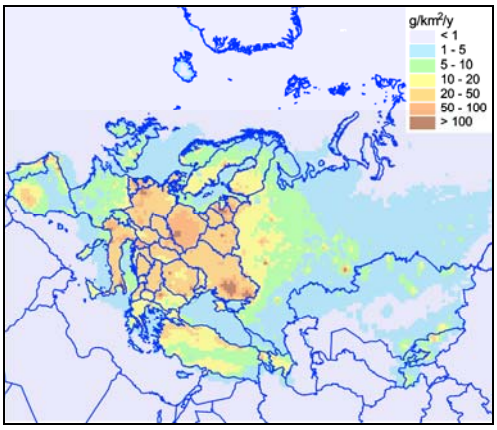

Fig. 2.12. *Spatial distribution of annual deposition flux of B[a]P calculated for 2008, g/km²/y*

increase of B[a]P emissions. Total deposition to the EMEP domain from all sources (anthropogenic and re-emission) made up 223 tonnes. The contribution of re-emission sources in 2008 is accounted for about 20%.

Spatial distribution of calculated annual B[a]P deposition fluxes within the EMEP domain in 2008 is given in Fig. 2.12. It is seen that B[a]P deposition fluxes over the European countries varied from 1 g/km^2/y to over 100 g/km^2/y. Essential values of deposition fluxes (20 – 100 g/km^2/y and higher) were characteristic of the Central, Southern, and Eastern Europe. In other parts of Europe (Scandinavian Peninsula, France, Spain, Portugal and the United Kingdom) deposition fluxes typically did not exceed 10 g/km^2/y. Deposition fluxes of B[a]P calculated for the Central Asian countries were comparatively lower (about 1 – 10 g/km^2/y and lower).

Modelling of long-range transport and deposition of B[a]P within the EMEP domain allows comparing the contributions of national emission sources, transboundary transport, and re-emission to the total deposition. Estimates of total annual deposition of B[a]P over the European and the Central Asian countries in 2008 are presented in Fig. 2.13 along with contributions of different emission sources (national sources, transboundary transport and re-emissions). It can be seen that transboundary transport of B[a]P is a significant source of pollution for the European countries. Its contribution can vary from almost 100% for Monaco, Iceland and Switzerland to about 6% for Spain (see Fig. 2.14). For 7 countries the contribution of B[a]P transboundary transport exceeds 75%, and for 25 countries – 50%. Transboundary transport and its contribution to total deposition depends on a number of factors like the size of a country territory, peculiarities of meteorological conditions, and magnitude of domestic emission of a given country. In particular, for countries with significant national emission, in comparison with the emissions of surrounding countries, the contribution of transboundary transport is typically low, like for instance, for the Ukraine (13%), Germany (16%) and Poland (19%). At the same time for countries with relatively small territory or low emission the contribution of transboundary transport can be essential (Switzerland – 94%, Malta – 81%).

The contribution of a particular country to the transboundary transport of pollution can be characterised by the fractions of total deposition originated from its national emission sources which is deposited inside and outside its territory. This information is shown for each EMEP country as B[a]P deposition in tonnes per year in Fig. 2.15. The fraction (%) of total deposition from national emission sources which is deposited outside the country (export) is shown in Fig. 2.16. It is seen that essential part of pollutant emitted from the sources of the European and the Central Asian countries is transported outside their boundaries. In particular, this fraction varies from almost 100% for Monaco to about 11% for Turkey.

For 18 countries the fraction of deposition occurred outside their boundaries exceeds 50% of total deposition from anthropogenic sources. The most essential contribution to the transboundary transport of B[a]P in 2008 was made by emission sources of the Ukraine (about 14 tonnes) followed by Poland (10 tonnes) and Romania (6 tonnes).

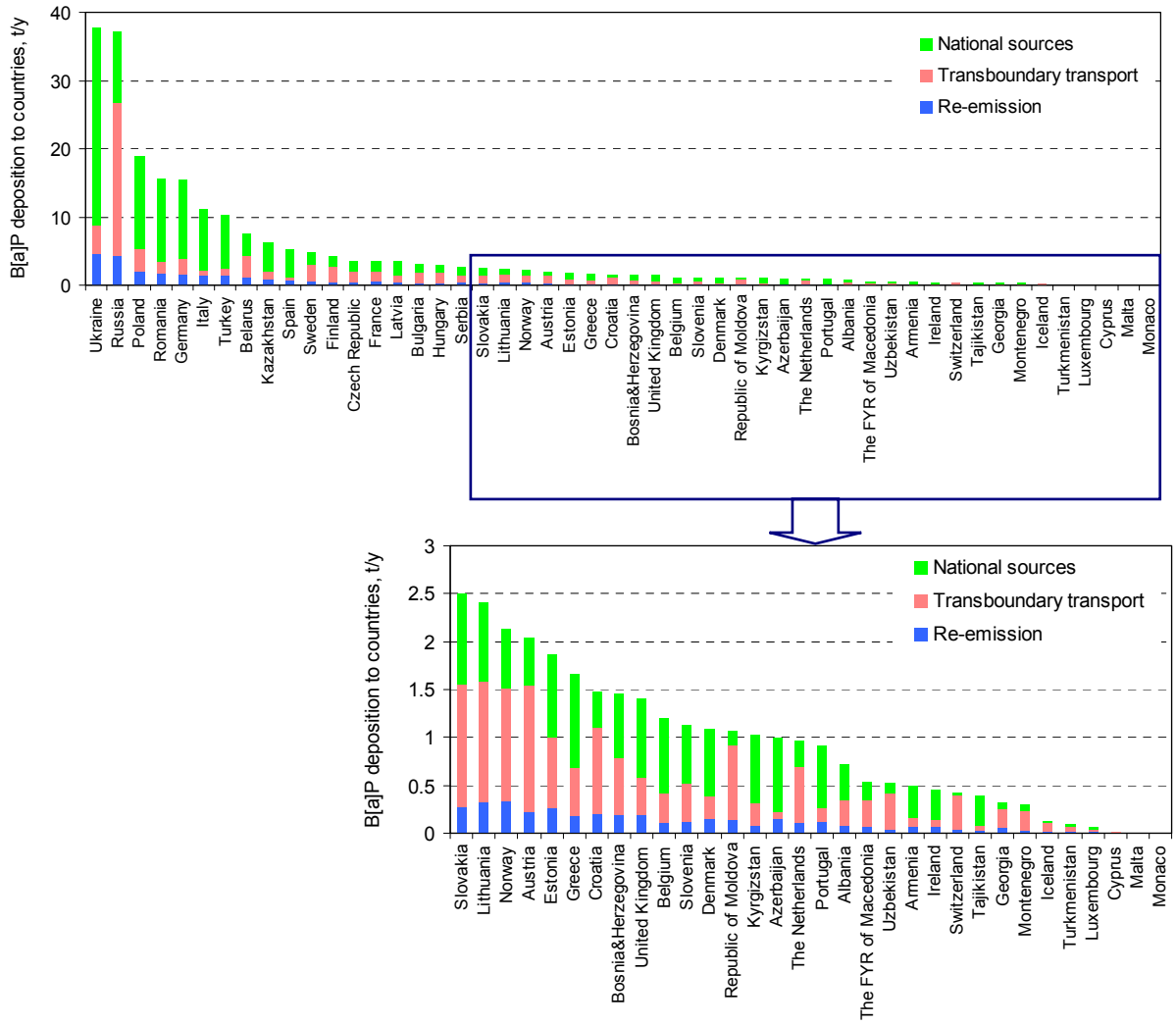

Fig. 2.13. *Total annual deposition of B[a]P over the European and the Central Asian countries in 2008 and contributions of national emission sources, transboundary transport within the EMEP region, and re-emission, t/y*

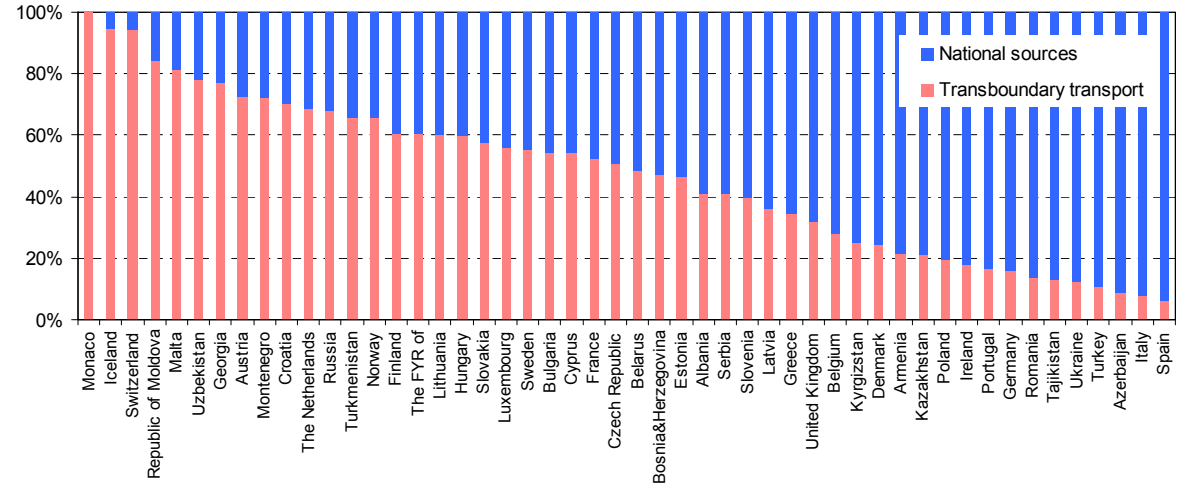

Fig. 2.14. *Relative contribution of transboundary transport and national emission sources to total deposition of B[a]P in the European and the Central Asian countries for 2008 (excluding contribution of re-emission), %*

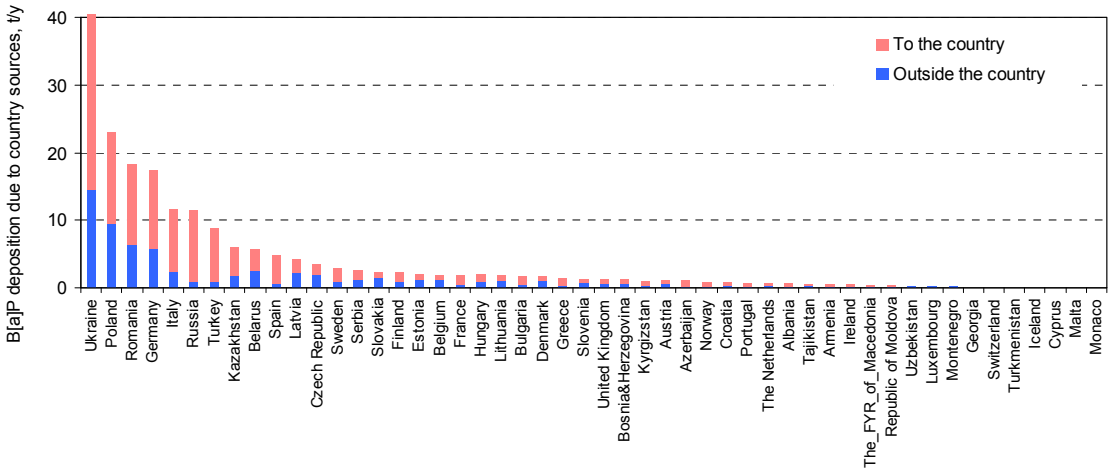

Fig. 2.15. *Fractions of B[a]P deposition originated from countries emission sources which are deposited to and outside their territories in 2008, t/y*

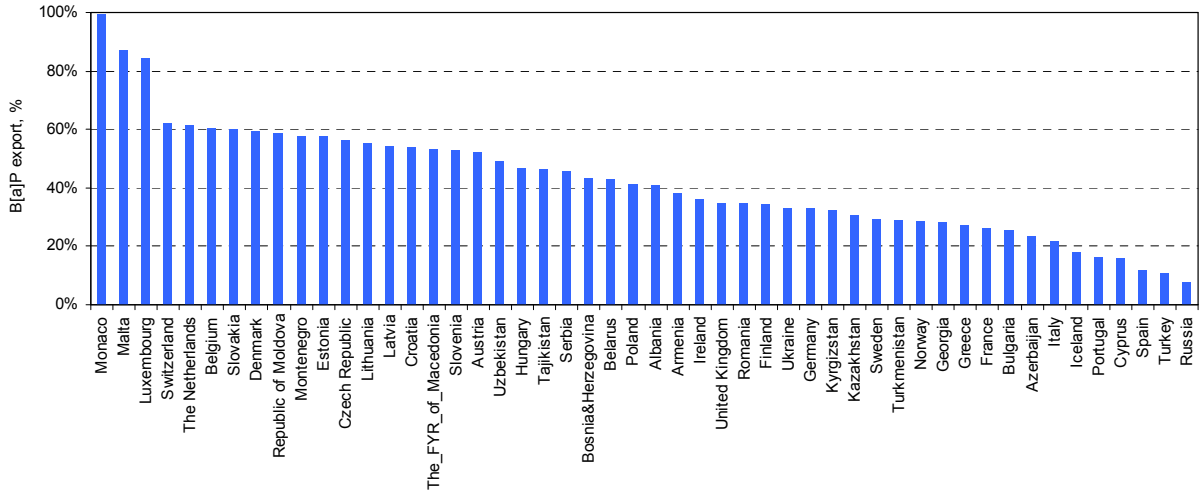

Fig. 2.16. *Fractions of B[a]P deposition originated from countries emission sources and occurred outside their territories in 2008 (export), %*

On the basis of country-to-country calculations more detailed information on import and export was prepared for each EMEP country. The information on import consists of fractions of deposition to the country originated by emission sources of all other European countries (import charts). Two examples of import charts (for Germany and Finland) are presented in Fig. 2.17.

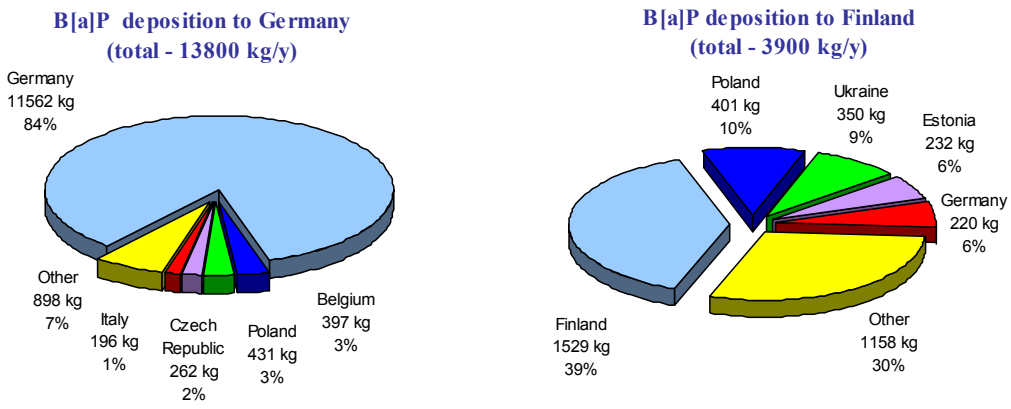

Fig. 2.17. *Import of B[a]P deposition for Germany and Finland for 2008, kg*

It is seen that the most part of B[a]P deposition to Germany (84%) is determined by its domestic sources followed by Poland and Belgium (3% each), the Czech Republic (2%) and Italy (1%). All other EMEP countries determine 7% of deposition to Germany. Another situation takes place in Finland where only about 40% of depositions are due to the domestic sources. Other contributors to the contamination of Finland are Poland (10%), the Ukraine (9%), Estonia and Germany (6% each) and other EMEP countries (about 30% altogether).

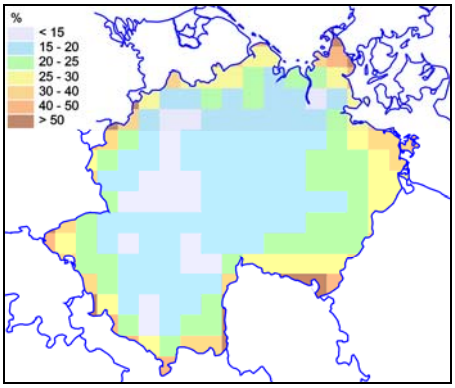

Fig. 2.18. *Contributions of transboundary transport of B[a]P in Germany, %*

However, even if the total deposition over a country as a whole is determined mostly by its own sources, transboundary transport can play essential role in some parts of its territory. For example, spatial distribution of the contributions of external sources in Germany is presented in Fig. 2.18. It is seen that the contribution of transboundary transport nearby the country boundaries can reach 40% and more.

Similar, the information on export contains the fractions of deposition, originated by emission sources of considered country, which take place over the territory of the country itself and over the territories of other EMEP countries. Export charts for Germany and Finland are shown in Fig. 2.19.

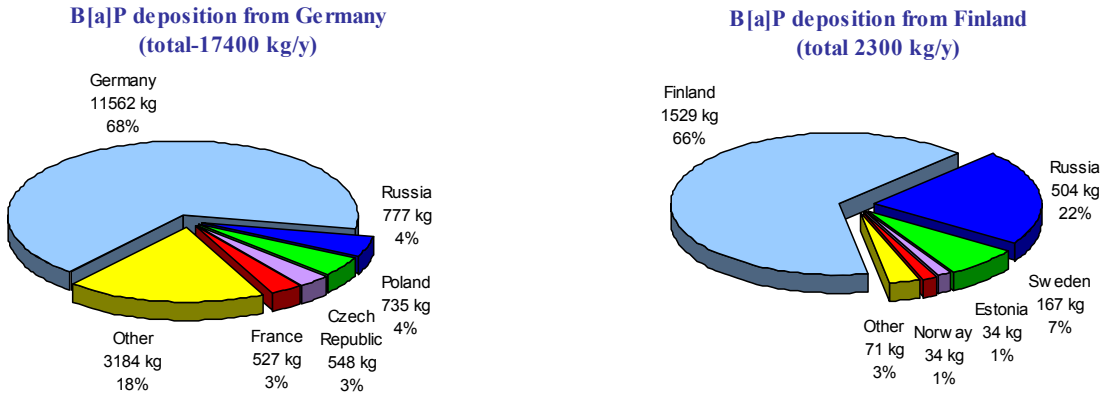

Fig. 2.19. *Export of B[a]P deposition for Germany and Finland for 2008, kg*

In particular, model calculations show that both for Germany and Finland 60 – 70% of deposition originated from national emission sources are occurred over the countries themselves. The rest 30 – 40% of deposition due to national sources are distributed between the territories of the EMEP countries. For Germany main receptors are the Russian Federation, Poland, the Czech Republic and France, and for Finland – the Russian Federation, Sweden, Estonia and Norway.

Export and import charts for all the European and the Central Asian countries can be found at the MSC-E web site www.msceast.org.

POLYCHLORINATED DIBENZO(p)DIOXINS AND DIBENZOFURANS (PCDD/Fs)

Model simulations of transboundary transport and deposition of PCDD/Fs within the EMEP region were carried out taking into account the contributions of national emission sources of the European and the Central Asian countries, non-EMEP emission sources (USA and Canada), and re-emission. Figure 2.20 presents the total annual deposition of PCDD/Fs over the EMEP domain for 2008 split into the contributions of different groups of emission sources (national sources, transboundary transport inside EMEP domain, non-EMEP sources, and re-emission). Transboundary transport and its contribution to total deposition depends again on a number of factors like the size of a country territory, peculiarities of meteorological conditions, and magnitude of domestic emission of a given country.

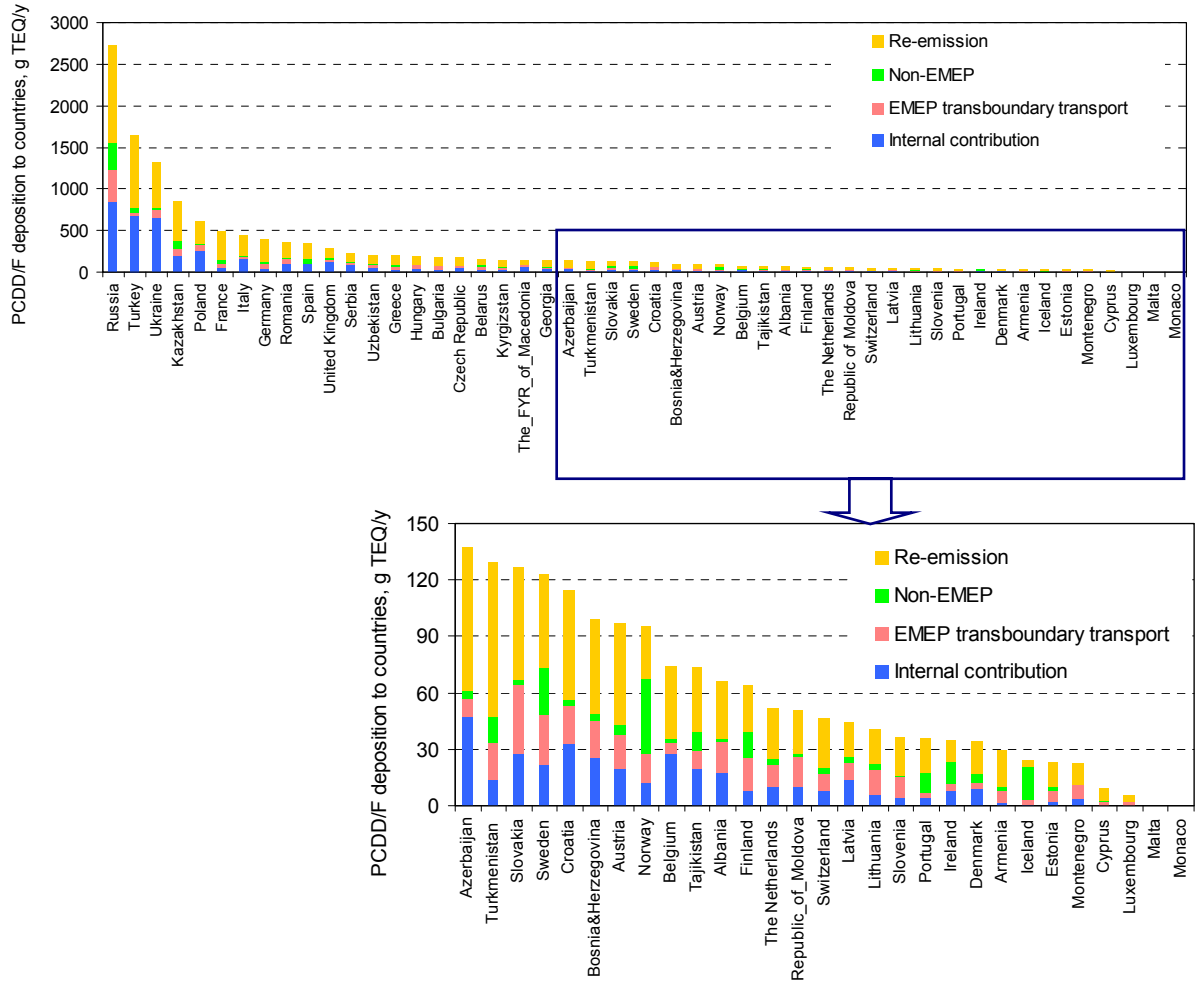

Fig. 2.20. *Total annual deposition of PCDD/Fs over the European and the Central Asian countries in 2008 and contributions of national emission sources, transboundary transport within the EMEP region, transport from non-EMEP sources and re-emission, g TEQ/y*

Relative importance of PCDD/F transboundary transport from anthropogenic emission sources of the European and the Central Asian countries comparing to the contribution of national sources is shown in Fig. 2.21 (here re-emission contribution is excluded). Contribution of transboundary transport to the deposition varies from almost 100% for Cyprus to about 7% for the United Kingdom. For 20 countries

the contribution of PCDD/F transboundary transport exceeds 50%. The contribution of transboundary transport for the countries with significant national emissions of PCDD/Fs (Turkey, the Ukraine) and countries situated upwind to the major emission sources (the United Kingdom, Spain) is comparatively low being about 7%-12%.

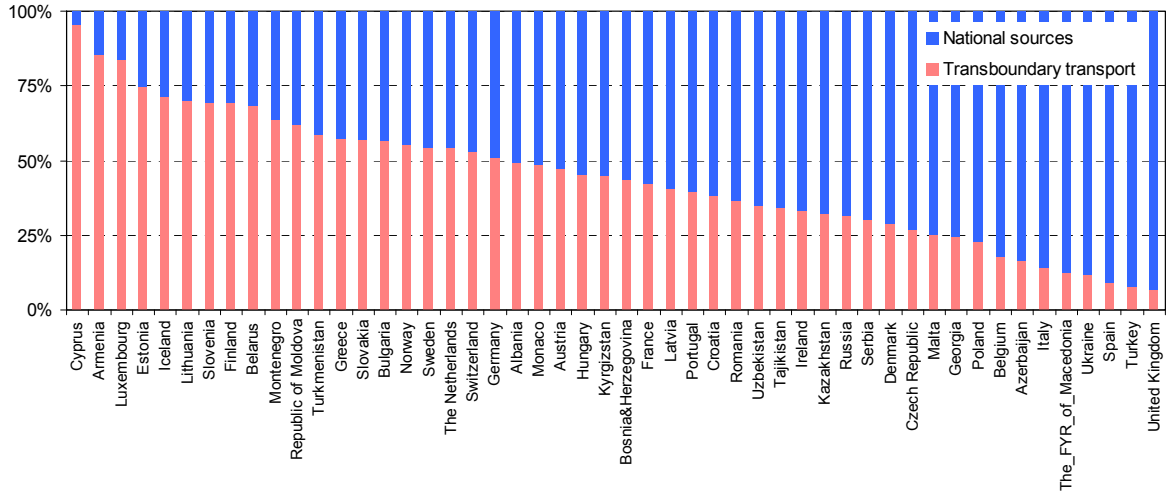

Fig. 2.21. *Relative contribution of transboundary transport and national emission sources to total deposition of PCDD/Fs in the European and the Central Asian countries for 2008 (excluding contribution of re-emission)*

Except for the transboundary transport from EMEP countries considerable contributions for some countries are made by non-EMEP sources. In particular, for 15 countries the contribution of non-EMEP sources exceeds 20%. So, for PCDD/Fs global transport can play essential role for the evaluation of contamination levels in the EMEP countries.

The fractions of total deposition originated from national emission sources which is deposited inside and outside country's territory are shown for each EMEP country in g TEQ/y in Fig. 2.22. The fraction (%) of total deposition from national emission sources which is deposited outside the country (export) is shown in Fig. 2.23. Essential part of the emitted PCDD/Fs from the sources of the European and Central Asian countries is transported outside their boundaries. The highest contribution to the transboundary transport of PCDD/Fs in 2008 is made by emission sources of the Ukraine (about 270 g TEQ) followed by the Russian Federation (127 g TEQ) and Poland (110 g TEQ). The fraction of total deposition occurred outside a country territory varies from almost 100% for Monaco and Malta to about 13% for Turkey. For 12 countries the fraction of total deposition from national emission sources deposited outside their territory exceeds 50% (see Fig. 2.23).

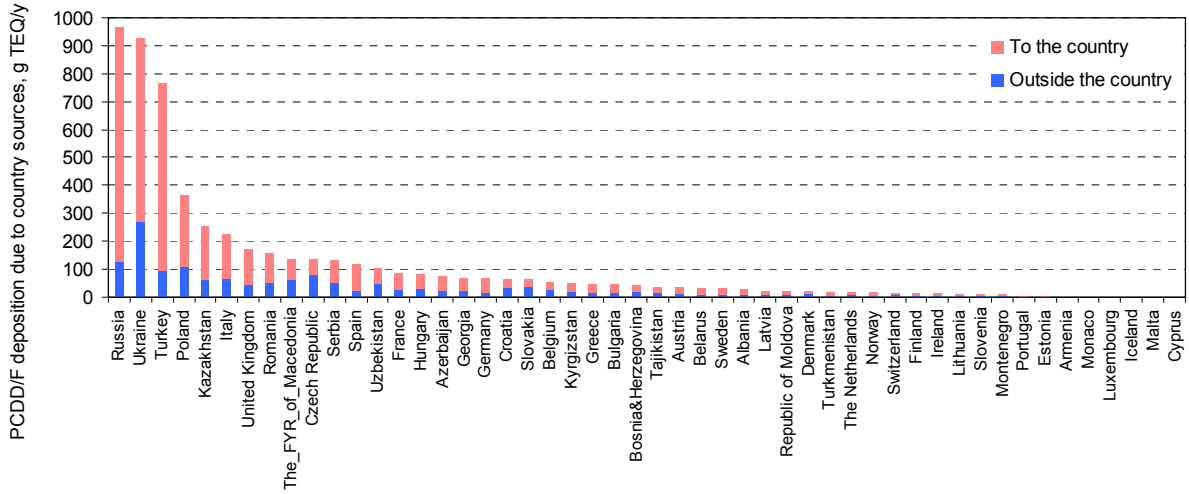

Fig. 2.22. *Fractions of PCDD/F deposition originated from countries emission sources which are deposited to and outside their territories in 2008, g TEQ/y*

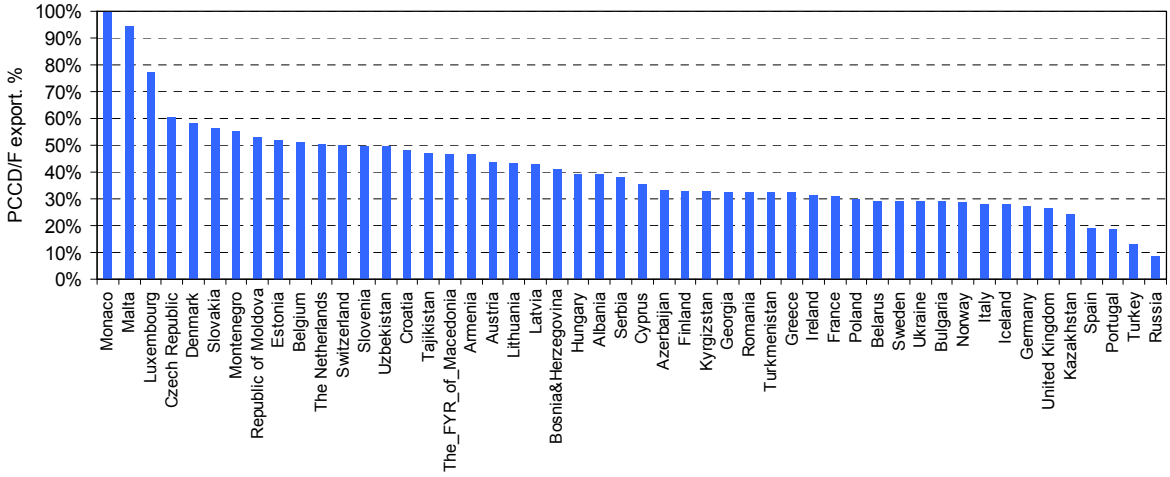

Fig. 2.23. *Fractions of PCDD/F deposition originated from countries emission sources and occurred outside their territories in 2008 (export), %*

Similar to the case of B[a]P, import and export charts are generated for all the European and the Central Asian countries. This information can be found at the MSC-E web site www.msceast.org.

POLYCHLORINATED BIPHENYLS (PCBs)

This year pilot evaluation of transboundary transport of PCB-153 within the EMEP region is done for 2008. Calculations are carried out taking into account the contributions of national emission sources of the European and the Central Asian countries, non-EMEP emission sources and re-emission.

To begin with, the contributions of anthropogenic emission sources to depositions of PCB-153 to the European countries are considered (with contribution of re-emissions excluded). Figure 2.24 presents the total annual deposition of PCB-153 over the EMEP region (with re-emissions excluded) for 2008 split into the contributions of different groups of emission sources.

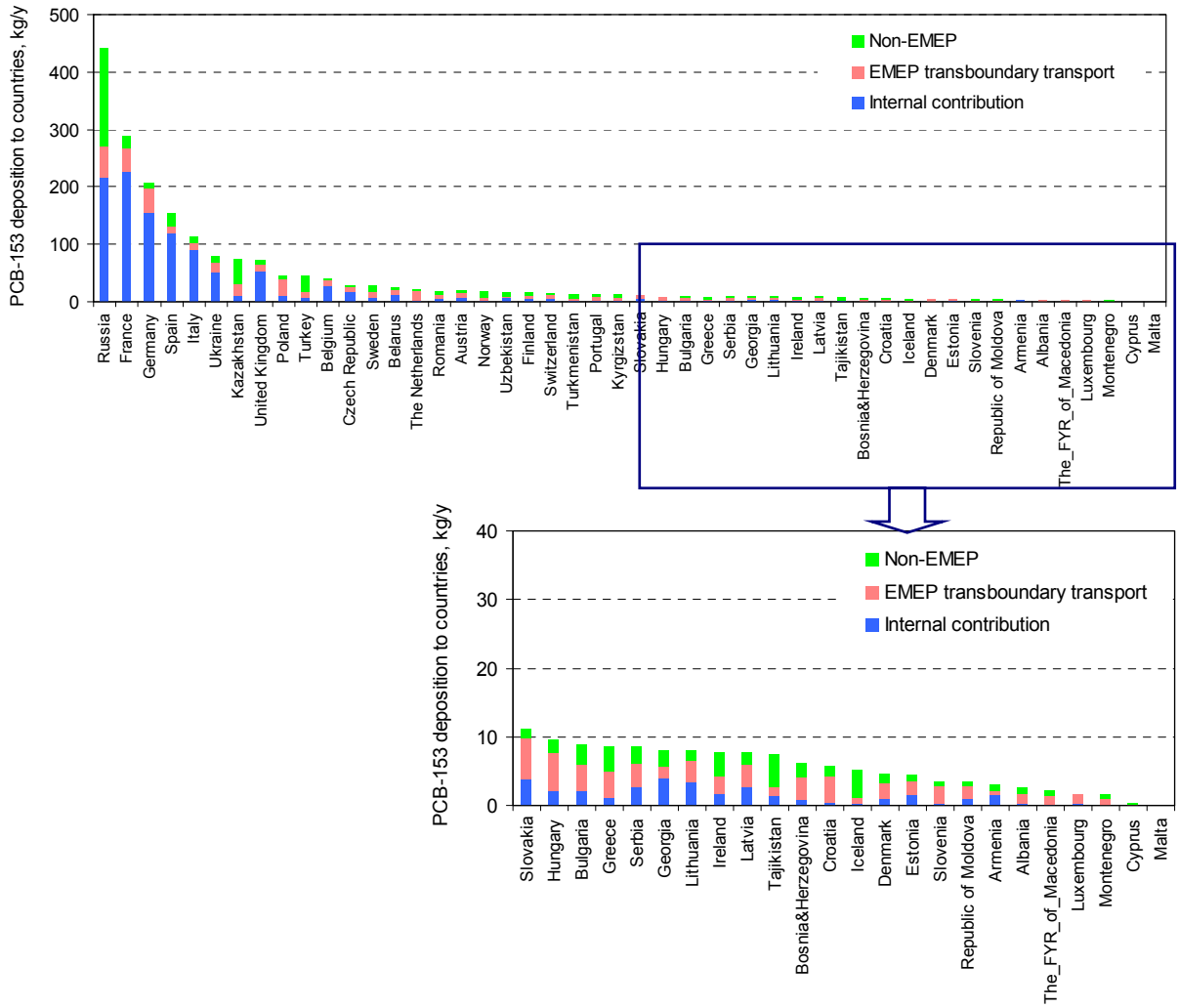

Fig. 2.24. *Total annual deposition of PCB-153 (with re-emissions excluded) over the European and the Central Asian countries in 2008 and contributions of national emission sources, transboundary transport within the EMEP region and transport from non-EMEP sources, kg/y*

For PCB-153 calculations show high contributions (mainly from 20% to 50%) of non-EMEP sources to the contamination of the European countries (Fig. 2.25).

These contributions vary from 60% – 80% for Iceland, Turkmenistan, Tajikistan, Azerbaijan, Norway, Turkey, Kazakhstan and Kyrgyzstan to 3% – 6% for the Netherlands, Germany and Belgium. It occurs that high contribution of non-EMEP sources is characteristic of countries with low emissions located near the boundary of the EMEP domain.

Relative importance of PCB-153 transboundary transport from anthropogenic emission sources of the European and the Central Asian countries comparing to the contribution of national sources is shown in Fig. 2.26. Contribution of transboundary transport to the deposition varies from almost 95% for Montenegro to about 8% for Spain. For 34 countries the contribution of PCB-153 transboundary transport exceeds 50%, and for 12 countries – 75%.

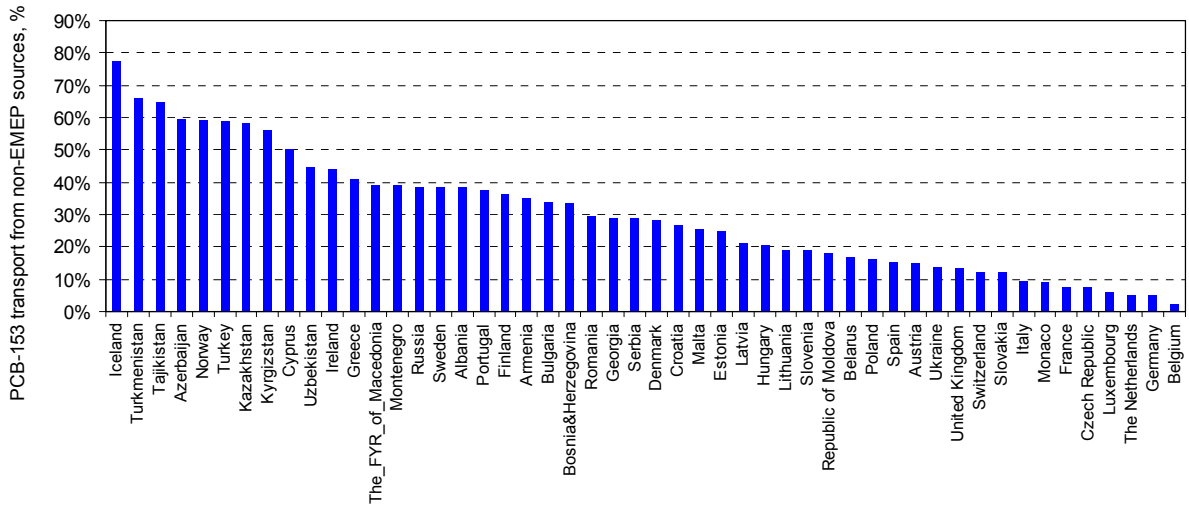

Fig. 2.25. *Relative contribution of non-EMEP sources to the contamination of European countries by anthropogenic sources, %*

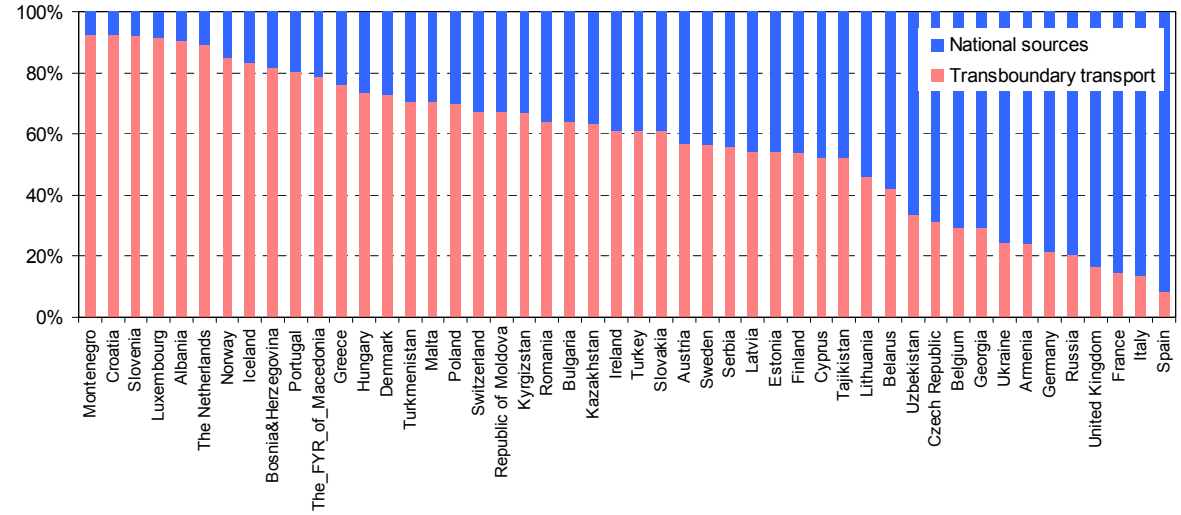

Fig. 2.26. *Relative contribution of transboundary transport and national emission sources to total deposition of PCB-153 in the European and the Central Asian countries for 2008 (re-emissions excluded), %*

The fractions of total deposition originated from national emission sources which is deposited inside and outside country's territory for each EMEP country are shown in Fig. 2.27. The fraction (%) of total deposition from national emission sources which is deposited outside the country (export) is shown in Fig. 2.28. Essential part of the emitted PCB-153 from the sources of the European and the Central Asian countries is transported outside their boundaries. The most essential contribution to the transboundary transport of PCB-153 in 2008 is made by emission sources of France (about 76 kg) followed by Germany (64 kg) and Italy (35 kg). The fraction of total deposition occurred outside a country territory vary from 97% for Malta to about 19% for Turkey. For 18 countries the fraction of total deposition from national emission sources deposited outside their territory exceeds 50%.

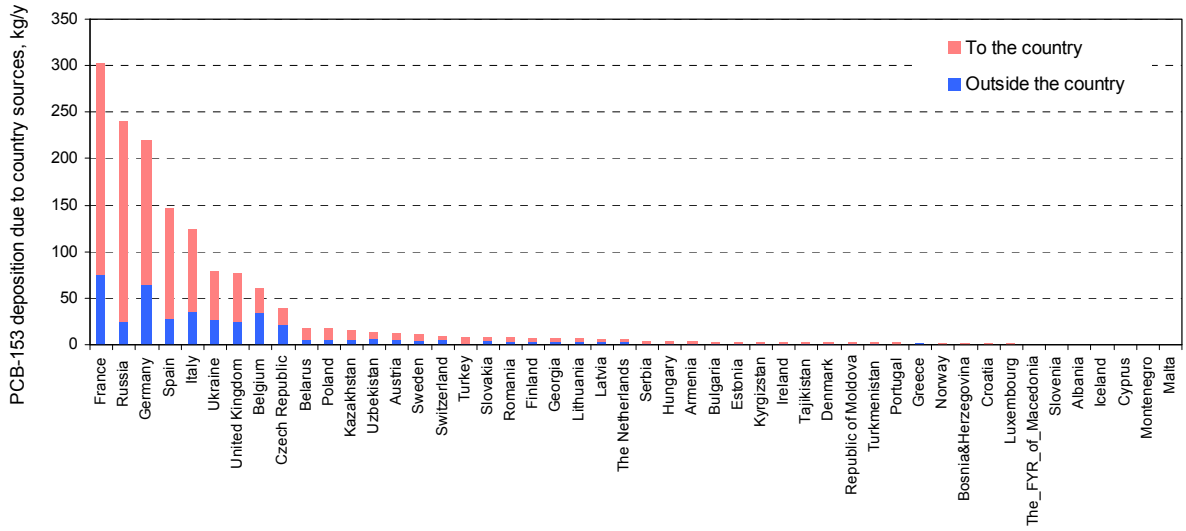

Fig. 2.27. *Fractions of PCB-153 deposition originated from countries emission sources which are deposited to and outside their territories in 2008, kg/y*

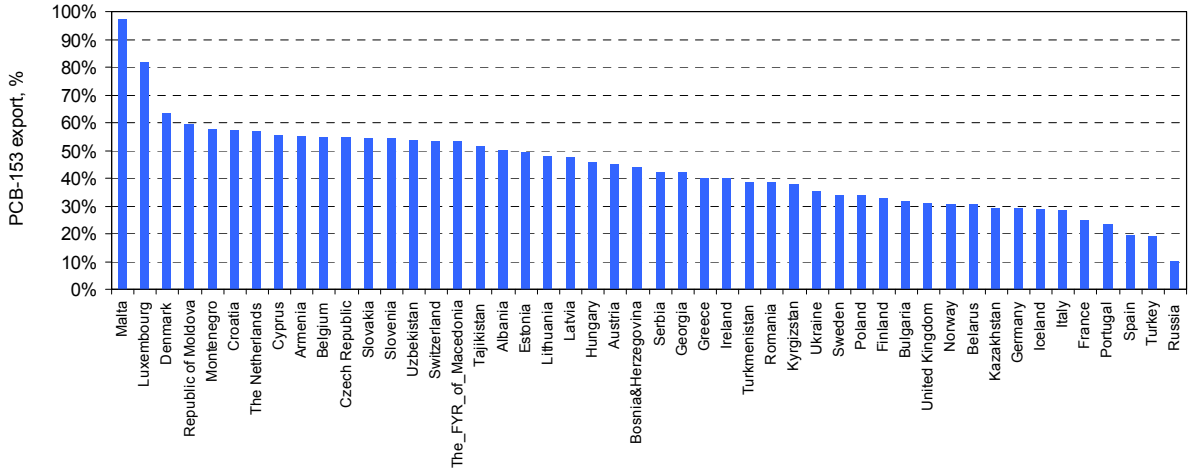

Fig. 2.28. *Fractions of PCB-153 deposition originated from countries emission sources and occurred outside their territories in 2008 (export), %*

Model calculations show significant contributions of re-emissions to the contamination of the European countries (from 40% to 80% for different countries). This can be explained by the rapid decrease of PCB-153 emissions in Europe since the last decades (PCB-153 emissions in Europe have decreased about three times since 1970). This fact together with high persistence of PCB-153 in soil (with half-life about 6 years according to model parameterization) leads to the increased contribution of secondary emission sources (re-emissions) to the contamination of the European countries. These results are preliminary and further work on the evaluation of PCB-153 accumulation in media is required. Some literature sources report lower influence of re-emissions on PCB contamination levels.

More detailed information on source-receptor relationships for PCB-153 can be found at MSC-E web site www.msceast.org.

2.4. Measurement of POPs in EMEP

EMEP measurements of POPs

POPs were included in the EMEP's monitoring program in 1999. However, earlier data has been available and collected, and the EMEP database thus also includes older measurements [*http://ebas.nilu.no*]. A number of countries have been reporting POPs within the EMEP area in connection with different national and international programmes such as HELCOM, AMAP and OSPAR. Data from the open scientific literature are also used for model validation and complements the EMEP data. Detailed information about the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [*Aas and Breivik*, 2010].

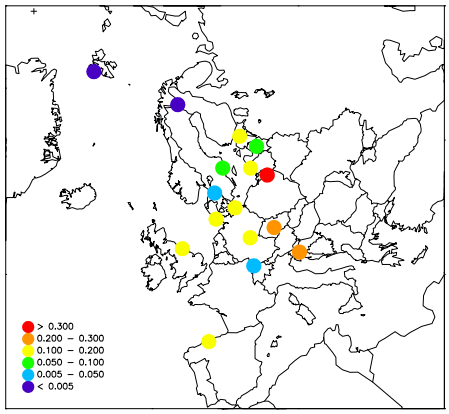

Fig. 2.29. *Spatial distribution of the annual average concentrations of benzo-a-pyrene (B[a]P) in 2008, ng/m^3*

In 2008 there were twelve sites measuring POPs in both compartments, and altogether there were twenty measurement sites, which are three more than in 2007 (Table 2.2). Furthermore there are five sites in Spain delivering campaign data of PAHs. Most of the additional measurements are PAH and more specifically benzo-a-pyrene (B[a]P) which is required to monitor in accordance to the EUs air quality directive [*EU*, 2004]. B[a]P which is a by-product of incomplete combustion processes is the most frequently measured POP component in EMEP. These results are therefore highlighted herein, whereas additional results for other compounds can be found in the data report [*Aas and Breivik*, 2010]. The spatial pattern of the average annual concentration level of B[a]P is shown in Fig. 2.29, and the concentrations seem to decrease when moving towards more remote areas in Europe. Notable differences in air concentrations can be seen between some adjacent sites suggesting that some EMEP sites may be influenced by local emissions of PAHs. In general, elevated concentrations of POPs are often seen in central parts of Europe [*Aas and Breivik*, 2010] reflecting proximity to major sources areas in Europe [*Breivik et al.*, 1999; *Breivik et al.,* 2007].

Table 2.2. *Measurement sites and program in 2008*

Country	Code	Name	POPs in air and aerosol	POPs in precipitation
Belgium	BE0014R	Koksijde		Pecticides, HCHs
Czech Republic	CZ0003R	Kosetice	PAHs, PCBs, pesticides, HCHs	PAHs, PCBs, pesticides, HCHs
Germany	DE0001R	Westerland	PAHs	PAHs, PCBs, pesticides, HCB
	DE0003R	Schauinsland	PAHs	PAHs, PCBs, pesticides, HCB
	DE0008R	Schmücke	PAHs	PAHs, PCBs, pesticides, HCB
	DE0009R	Zingst	PAHs	PAHs, PCBs, pesticides, HCB
Estonia	EE0009R	Lahemaa	PAH (Benzo[a]pyrene)	
Spain	ES0008	Niembro	PAHs	
Finland	FI0096R	Pallas	PAHs, PCBs, pesticides, HCHs	PAHs, PCBs, HCHs
Great Britain	GB0014	High Muffles	PAHs, PCBs	
Iceland	IS0091R	Storhofdi	PCBs, pesticides, HCB, HCHs	PCBs, pesticides, HCB, HCHs
Latvia	LV0010R	Rucava	PAH (Benzo[a]pyrene)	
	LV0016R	Zoseni	PAH (Benzo[a]pyrene)	
Netherlands	NL0091R	De Zilk		γ-HCH
Norway	NO0042G	Spitsbergen	PAHs, PCBs, pesticides, HCHs, HCB	
	NO0001R	Birkenes	PCBs, HCB, HCHs	PCBs, HCB, HCHs
Poland	PL0005R	Diabla Gora	PAHs	PAHs
Sweden	SE0012R	Aspvreten	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs
	SE0014R	Råö	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs
Slovenia	SI0008R	Iskbra	PAHs	PAHs

Even though the spatial coverage has improved, there is still a need for more monitoring sites, especially in south – southeast of Europe to fulfill the goal of the EMEP monitoring strategy [*UN-ECE*, 2009]. There are, however, some positive developments in this region. At the EMEP sites in Moldova and Kazakhstan there will be one year of air and aerosol measurements of key POPs (PAHs, PCBs, pesticides, HCB, HCHs) from June 2009, a campaign financed by the Norwegian Ministry of Foreign Affairs. The first results are expected to be published in the next year status report.

Passive air sampling

To gain new insight into the spatial patterns of POPs in European background air a passive air sampling (PAS) campaign using polyurethane foam disks (PUF) was carried out in various European countries during late summer 2006 as presented in last year status report [*Gusev et al.,* 2009] and are being published by *A.K.Halse et al.* [2010]. The data from this campaign are openly available at [*http://ebas.nilu.no*]. PAS have become increasingly popular and have shown to be complementary to active air sampling, especially for semi-volatile organic compounds [e.g. *Shoeib and Harner*, 2002, *Jaward et al*., 2003; *Harner et al*., 2006, *Pozo et al.*, 2009]. In addition to the EMEP initiative, the Global Atmospheric Passive Sampling (GAPS) network is a key global program for producing comparable global-scale data for POPs. This program started in December 2004 and has three month sampling periods at more than fifty sites on all the continents [*Pozo et al*., 2006 and 2009; *Lee et al.,* 2007] Furthermore, a two year POP passive measurements campaign was initiated in April 2009 by RECETOX (MONET-EUROPE) which aims to collect monthly samples at a number of EMEP sites to support relevant efforts under the Stockholm Convention on POPs, both also in support of EMEP.

Uncertainty in POP measurements

It is difficult to quantify the uncertainty in the POP measurements since it depends on several factors (methodology, sample handling and preparation etc) and which components in question. *S.J.Hayward et al.* [2010] compared high and low volume active samplers with two different types of passive samplers and they concluded that the annually averaged air concentrations determined by the different systems are within a factor of 2.5 for all pesticides except two. This is similar to what was observed for the sites with active sampling in parallel to the EMEP passive campaign [*Halse et al*., 2010; *Gusev et al.,* 2009]. The comparability between two identical samplers is usually better, but just using different laboratory may decrease the comparability significantly. In the POP laboratory intercomparison performed in 2000-2002. [*Manø and Schaug*, 2003] large uncertainties was seen for some labs –a bias of factor 2 or 3 for some components for some laboratories, though several laboratories were also within 20% of the expected value for most of the species that was included in the intercomparison (PAHs and organochlorine compounds). Due to these large uncertainties in the analytical performance across the EMEP network, it has therefore been a wish to follow up on this intercomparison to check whether there have been any improvements in the comparability between the European laboratories. Since POPs are global pollutants, in addition to the fact that laboratory intercomparison is a costly and difficult exercise, EMEP has joined forces with the Northern Contaminants Program (NCP) in Canada, which is coordinated by the Laboratory Services Branch (LaSB) of the Ontario Ministry of the Environment (MOE), to perform a new laboratory intercomparison. It will build on experience from the QA/QC program developed in the project Intercontinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA) lead by Environment Canada. In addition to EMEPs own QA/QC objectives, this coordination will also allow for the assessment of data comparability between air monitoring programs, especially important also for future effectiveness evaluation of the Stockholm Convention on POP. All the laboratories participating in EMEP, HELCOM, OSPAR or AMAP have been invited to participate. This interlaboratory study aims to assess variability in analyzing standards/sample

for four classes of trace organic chemicals, namely, polycyclic aromatic hydrocarbons (PAHs); polybrominated diphenyl ethers (PBDEs); polychlorinated biphenyls (PCBs); and organochlorine pesticides (OCPs) Three samples were distributes in June 2010, two injection-ready analytical standards and one real air sample extract, and the first results are expected during autumn 2010.

2.5. Analysis of agreement between measurements and calculations and development of integrated monitoring/modelling/emission approach

Traditionally approaches applied to the evaluation of environmental pollution by POPs are based on monitoring of concentration levels and/or application of models to evaluate chemical dispersion based on available emission inventories. However, the information obtained by each of these three types of activity (monitoring, modelling and emission inventories) is subject to its own uncertainties.

As shown in Section 2.1 the range of emission data uncertainties can reach several times in some cases. The estimates of the intrinsic uncertainties of modelling were evaluated in the course of the EMEP model review (see [*Gusev et al.*, 2005; *Shatalov et al.*, 2005]). Particularly, the uncertainties due to variability of meteorological parameters were estimated as about 40%, and the uncertainties due to pollutant-relative parameters – as about 20%. As indicated in Section 2.4 measurement data are also subject to uncertainties depending on methodology both in sampling and laboratory analysis.

To reduce these uncertainties, there might be a need to integrate these activities more closely under the monitoring/modelling/emission approach (integrated approach). The general idea of such an approach is that, at the first stage, adjustment of all three components of the assessment process (monitoring, modelling and emission inventories) is performed on the basis of the analysis of agreement between measurement data and modelling results. At the second stage, in order to perform refined assessment, adjusted models and emissions (as well as scenarios of emissions) and monitoring data are used to evaluate contamination levels, source-receptor relationships, etc. The work on elaborating the integrated approach under EMEP on the basis of back trajectory analysis is now ongoing.

The key point in such analysis is the interpretation of the comparison of modelling results with measurements from the point of view of the uncertainties in emissions, monitoring, and modelling. The tool for such interpretation based on back trajectory approach is elaborated under EMEP. In essence, this tool is a simplification of the use of adjoint models for the analysis of the agreement between measurements and calculation results. In comparison with the usage of direct adjoint approach, this tool requires much less computational resources and is suitable for the operational modelling.

The constructed tool can be used to accomplish the following tasks:

- Evaluation of representativeness of monitoring sites and QA/QC.
- Locating emission sources having maximal influence on the agreement between model calculations and measurements at the given site or at a number of sites.
- Constructing emission scenarios lying in the range of emission uncertainties and leading to the improvement of agreement between measurements and modelling results.
- Evaluating the obtained emission scenarios.
- Improvement of model parameterizations.

Usage of emission scenarios can select the “hot spots” in emission inventories for further investigation by the experts on emissions.

This section presents just a brief description of the integrated approach. More detailed information can be found in Technical Report [*Ilyin et al.*, 2010].

Basic tool for emission/measurements/modelling approach

The central point of emissions/monitoring/modelling approach is the analysis of the comparison of the agreement between calculations and available measurements. For this analysis the so-called *influence function* $\{\gamma_{ij}\}$, say, for monthly averages of air concentrations[1] at the locations of measurement sites taking part in the comparison, can be applied. By the influence function we mean a set of numbers γ_{ij} determined in each grid cell (i, j) such that for any emission distribution E_{ij} the following relation takes place:

$$\overline{c} = \sum_{i,j} \gamma_{ij} E_{ij} \ , \qquad (2.1)$$

where $\overline{c}$ is air concentration at the considered location averaged over the given period (month). The numbers γ_{ij} should depend on the chosen location and on period of averaging but not on the emission distribution itself. The numbers γ_{ij} can be obtained by adjoint models. Here we use the approximation of the influence function on the basis of back trajectory calculations.

This approximation is based on a simplified model of POP transport taking into account the processes of advection and removal from the atmosphere. However, the results of evaluation of monthly means of air concentrations at the EMEP sites obtained by this function are sufficiently close to the predictions made by the MSCE-POP model. For the illustration, the comparison of monthly means of B[a]P air concentrations in 2008 at CZ3 and GB14 calculated with the help of the influence function with those calculated by the full MSCE-POP model are given.

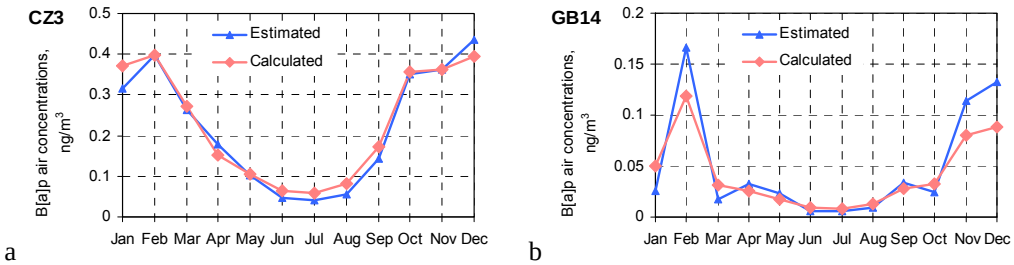

Fig. 2.30. *Comparison of monthly means of B[a]P air concentrations at CZ3(a) and GB14(b) calculated by the influence function and by the MSCE-POP model, ng/m^3*

The introduced notion of influence function can be applied to the analysis of agreement between measurements and modelling at one or several monitoring sites. The examples of its application are given in the next subsection.

The method of approximation is described in the Technical Report [*Ilyin et al.*, 2010].

[1] Monthly averages are chosen just to be definite; the approach described below can be applied to various periods of averaging. Similar, after slight modifications, deposition fluxes can be investigated instead of air concentrations.

Analysis of the agreement between measurements and calculations

This subsection contains the analysis of the agreement between measurements and model calculations for three pollutants: B[a]P, PCB-153 and γ-HCH. The presentation of the analysis for all these pollutants goes as follows. First, calculated and measured annual averages of air concentrations, concentrations in precipitation and deposition fluxes are considered. Then the analysis of seasonal variations of these parameters is performed.

Benzo[a]pyrene (B[a]P). For B[a]P measured concentrations in air are available for 17 EMEP measurement sites. Among them 6 sites perform in parallel the measurements of B[a]P concentrations in precipitation and 3 sites– of deposition fluxes. The comparison of calculated and measured annual means of air concentrations, concentrations in precipitation and deposition fluxes is given in Fig.2.31a,b and c, respectively.

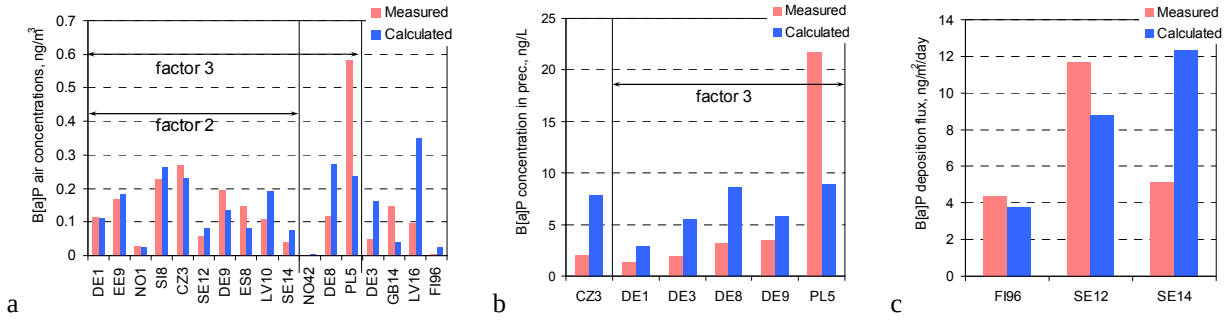

Fig. 2.31. *Comparison of calculation results with measurements for B[a]P: (a) – air concentrations, ng/m^3; (b) – concentrations in precipitation, ng/L; (c) – deposition flux (annual averages), ng/m^2/day*

The calculated air concentrations agree with measurements within a factor of three at 13 sites (about 75% of the total number of sites). The maximum ratio between calculated and measured air concentrations is 4.7. This value is obtained at site FI96, where measured annual mean of B[a]P concentration is as low as 0.005 ng/m^3. On the average, the calculated values of air concentrations exceed measurements 1.7 times.

The agreement between calculated and measured B[a]P concentrations in precipitation is mostly within a factor of three. The only exception is the site CZ3, where calculated-to-measured ratio equals 4. Modelled concentrations in precipitation exceed measurements 3.2 times on the average.

Finally, measured and calculated values of deposition fluxes differ not more than 2.4 times. At that, calculation results are lower than measurements at two sites (FI96 and SE12) and higher – at SE14.

The discrepancies in average calculated-to-measured ratio for air (1.7), precipitation (3.2) and deposition flux (2.4) can be conditioned by uncertainties in model description of deposition process and by usage meteorological parameters (precipitation intensity and temperature) averaged over 50×50 km^2 EMEP model grid cell instead of their values directly at site locations. For the investigation of deposition process more simultaneous measurements of air concentrations, deposition fluxes and concentrations in precipitation complemented by meteorological parameters are required.

The correlations between measured and modelled monthly means of air concentration are summarized in Table 2.3.

Table 2.3. *Correlation coefficients between monthly averages of calculated and measured air concentrations at 17 EMEP sites*

CZ3	0.88	**ES8**	-0.44	**NO42**	0.76
DE1	0.56	**FI96**	0.82	**PL5**	0.86
DE3	0.68	**GB14**	0.85	**SE12**	0.62
DE8	0.65	**LV10**	0.73	**SE14**	0.92
DE9	0.77	**LV16**	0.77	**SI8**	0.86
EE9	0.77	**NO1**	0.95		

Correlation coefficients between calculated and measured monthly means of air concentrations are 0.6 – 0.8 and higher. No correlation is obtained at the site ES8. To explain this fact, we remark that beginning from August 13, 21 values of air concentrations measured at this site were below the detection limit. The plot of raw measurement data in comparison with model calculations is presented in Fig. 2.32.

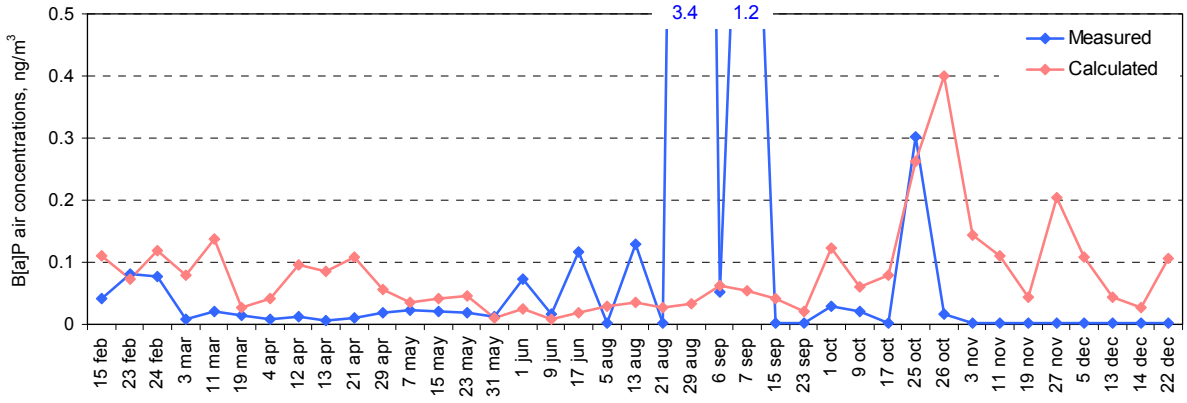

Fig. 2.32. *Comparison of raw measurements with calculated B[a]P air concentrations at ES8, ng/m^3*

If outstanding values of air concentrations (3.4 ng/m^3 at August 29 and 1.2 ng/m^3 at September 7) and concentrations below the detection limit are ignored, the agreement of measured and calculated data becomes much better at least in the order of magnitude. In this case correlation coefficient between measurements and model predictions becomes 0.3 and the averages agree within a factor of two.

However, in spite of high values of correlations between measured and modelled air concentrations, it occurs that seasonal variations of this parameter are underestimated by model calculations. This can be illustrated by the comparison of calculations and measurements at sites CZ3 and EE9 (Fig. 2.33).

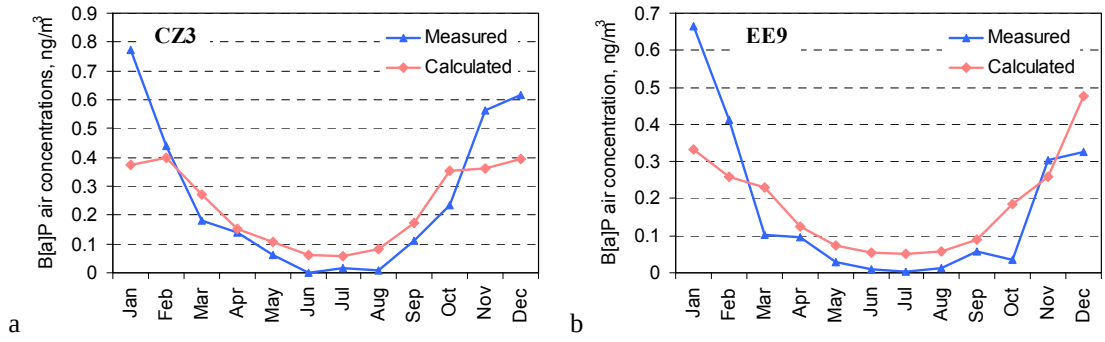

Fig. 2.33. *Comparison of monthly means of calculated and measured air B[a]P concentrations at EMEP measurement sites CZ3 (a) and EE9(b), ng/m^3*

Two calculation experiments showing how the change in emission data (including their seasonal variability) can refine the agreement between model results and monitoring data are performed.

1. Emissions are enlarged in each grid cell where this enlargement leads to the refinement of the agreement between measurements and modelling. Standard seasonal variations of emissions are taken (as assumed in MSCE-POP model).

2. Emissions are modified as for the first scenario but more pronounced seasonal variations are assumed (see Fig. 2.34).

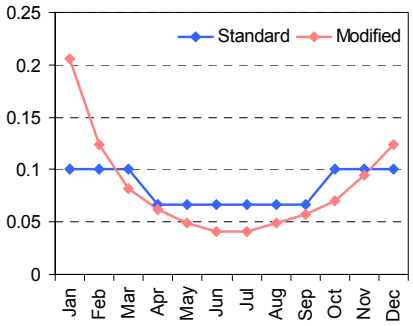

Fig. 2.34. *Fractions of annual B[a]P emission totals emitted in each month of 2008 according to standard and modified emission scenario*

The choice of the cells for which the emissions should be enlarged is done on the basis of the methodology described above with the help of the influence function (see formula (2.1) above). The comparison of air concentrations estimated with the help of the influence function for these two scenarios with standard calculations and measurements for CZ3 is shown by plots in Fig. 2.35.

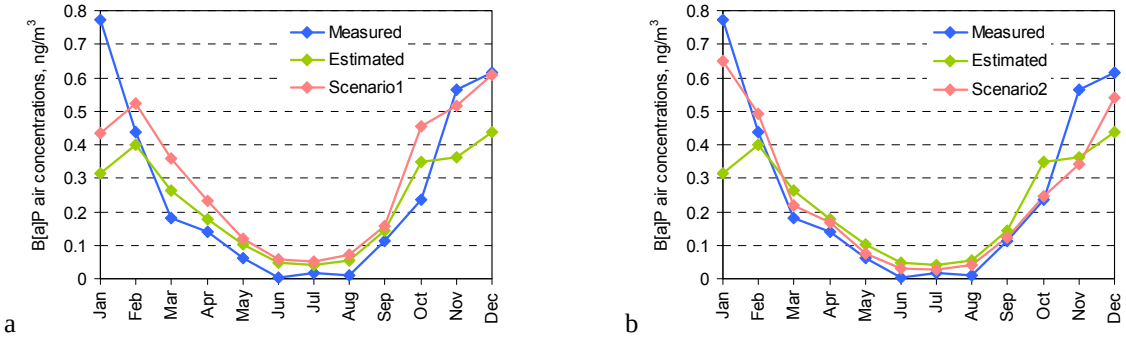

Fig. 2.35. *Comparison of two emission scenarios at CZ3 with measurements and standard calculations (a) – scenario 1, (b) – scenario 2, ng/m^3*

Figure 2.36 shows the results of similar calculation experiment carried out at the site EE9.

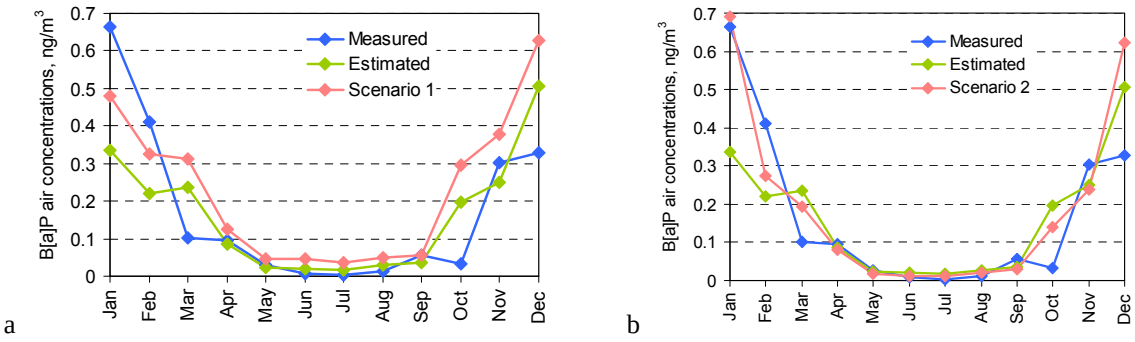

Fig. 2.36. *Comparison of two emission scenarios at EE9 with measurements and standard calculations (a) – scenario 1, (b) – scenario 2, ng/m^3*

For both considered sites the application of Scenario 1 leads to the refinement of the agreement between measurement and modelling results, at least in winter months. Correlation coefficient between measurements and calculation results enlarges from 0.84 to 0.86. At the same time, the refinement of seasonal variations is not sufficient. On the opposite, the application of Scenario 2 (which includes the modification of emission seasonal variations) refines the agreement between calculation results and measurement data essentially (correlation coefficient becomes 0.96). This exemplifies the application of the integrated approach showing possible directions of the improvement of emission inventory for B[a]P.

Polychlorinated biphenyls (PCB-153). The comparison of measured and calculated annual means of PCB-153 air concentrations, concentrations in precipitation and deposition fluxes is demonstrated in Fig. 2.37.

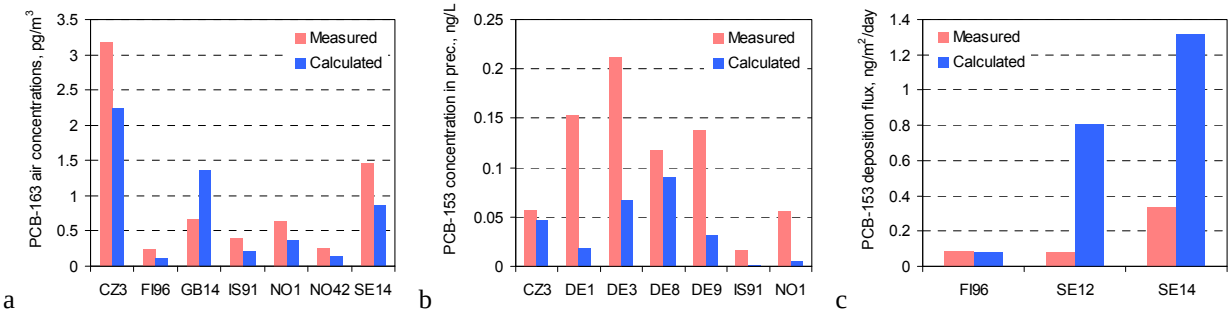

Fig. 2.37. *Comparison of calculation results with measurements for PCB-153: (a) – air concentrations, pg/m^3; (b) – concentrations in precipitation, ng/L; (c) – deposition fluxes (annual averages), ng/m^2/day*

Modelled annual means of air concentrations for PCB-153 agree with measured ones within a factor of two. The correlation coefficient between calculations and measurements is 0.89. The value of calculation-to-measurement ratio is 0.79 on the average, indicating slight underestimation of air concentration by the model. Calculation-to-measurement ratio for the PCB-153 concentrations in precipitation is 0.35 with correlation coefficient 0.53.

Model estimates of deposition fluxes agree well with measurements at the site FI96 and overestimate measured ones at SE12 and SE14. The difference can be connected with the methodology of measurements of dry and wet deposition fluxes or with uncertainties of model parameterization of dry and wet deposition.

The comparison of seasonal variations of calculated and measured PCB-153 air concentrations show generally high correlation between monthly averages of calculations and measurements. Correlation coefficients range from 0.64 to 0.9 with exception for the sites NO1 (K_{corr} = 0.21) and NO42 (K_{corr} = 0.19). To explain these two cases, we consider the plots of raw measurement data in comparison with calculations for these two sites (Figs. 2.38 and 2.39).

The discrepancies between model calculations and measurements at NO1 are found for two periods: from January 23 to March 12 and from April 2 to April 23. For the rest time the model reasonably reproduces PCB-153 air concentrations. For NO42, there exist two episodes where calculations strongly differ from measurement data and from other measurements at this site: June 15 and August 17 and 24. The discrepancies between calculations and measurements at these two sites may be explained by the influence of local emission sources. For more detailed analysis of the situation the trajectory approach is planned to be applied in future.

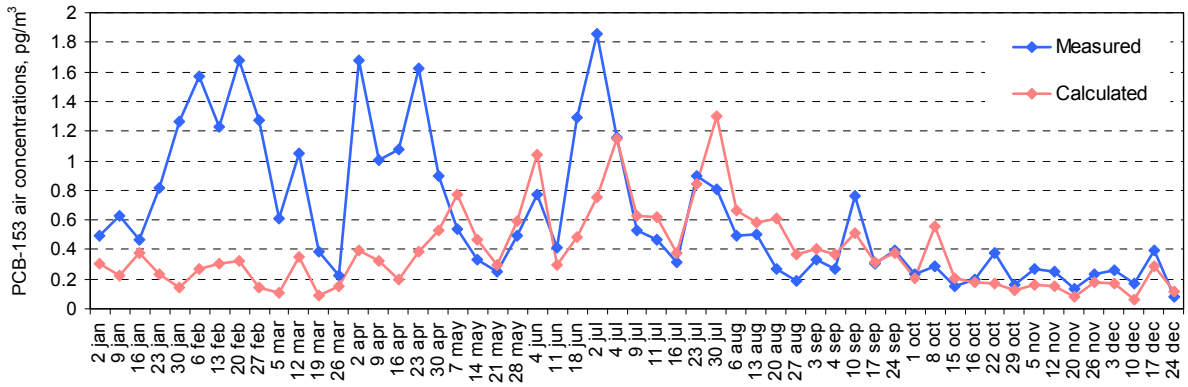

Fig. 2.38. *Comparison of raw measurements with calculated PCB-153 air concentrations at NO1, pg/m^3*

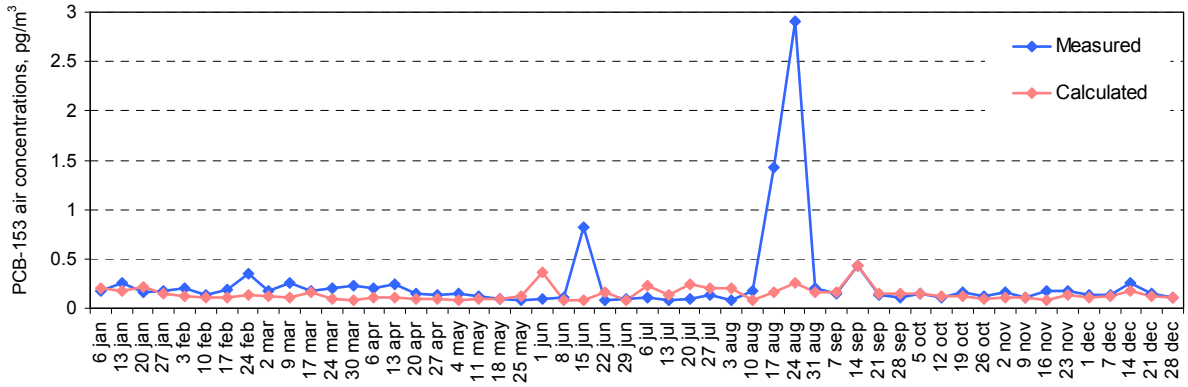

Fig. 2.39. *Comparison of raw measurements with calculated PCB-153 air concentrations at NO42, pg/m^3*

The discrepancies between calculated and measured PCB-153 deposition fluxes at sites SE12 and SE14 are shown in Fig. 2.40.

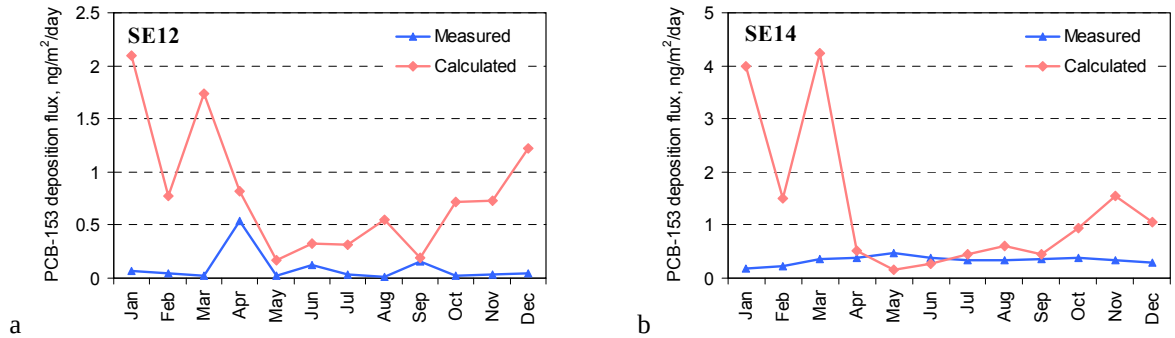

Fig. 2.40. *Comparison of measured and calculated PCB-153 deposition fluxes at SE12 and SE14, ng/m^2/day.*

It is seen that overestimation of PCB-153 deposition flux takes place mainly in winter months. It can be conditioned by the uncertainties in model description of scavenging with snow. So, further refinement of model parameterization of deposition process is required.

Lindane (γ-HCH). The comparison of measured and calculated annual means of PCB-153 air concentrations, concentrations in precipitation and deposition fluxes is presented in Fig. 2.41.

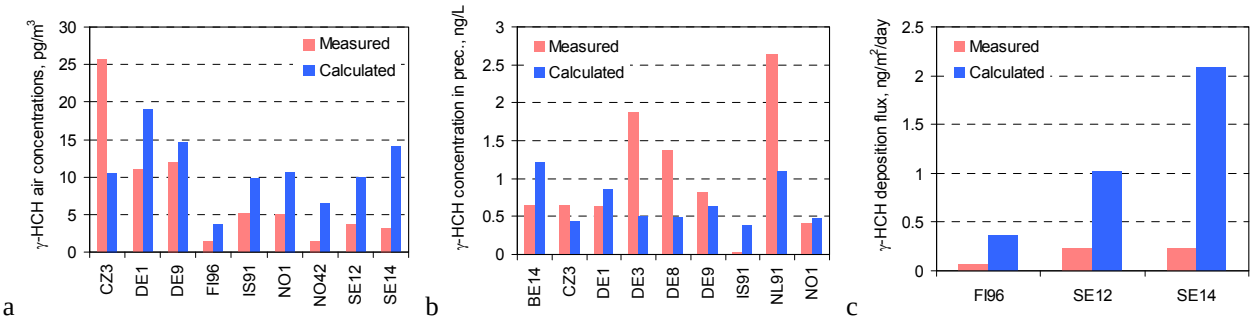

Fig. 2.41. *Comparison of calculation results with measurements for γ-HCH: (a) – air concentrations, pg/m^3; (b) – concentrations in precipitation, ng/L; (c) – deposition fluxes (annual averages), ng/m^2/day*

The agreement between calculated and measured annual means of γ-HCH air concentrations is within a factor of three at all the sites except for NO42 and SE14 where calculation-to-measurement ratio slightly exceeds four. The overestimation of air concentrations by the model can be conditioned by the uncertainties of emission data (emission totals, spatial distribution of emissions and their seasonal variations). According to the official data, emissions of γ-HCH are very low. Hence, contamination by γ-HCH is supported by the pollutant mass previously accumulated in the environment (particularly, in soil). So, for the refinement of model parameterization of γ-HCH accumulation in soil more measurements in this medium are required. Calculated values of γ-HCH concentrations in precipitation occur to be lower than measured ones at almost all monitoring sites (Fig. 2.41b). For most of the monitoring sites calculated and measured γ-HCH concentrations in precipitation agree with measured ones within a factor of three. The comparison of measured and calculated deposition fluxes at the EMEP sites FI96, SE12 and SE14 shows that the model strongly overestimates observed deposition fluxes. This overestimation is even stronger than the overestimation of air concentrations at the corresponding sites.

The above comparison shows that the improvement of model description of dry and wet deposition is desirable for further refinement of the description of the contamination of the EMEP region by γ-HCH.

The consideration of monthly means of γ-HCH air concentrations shows that about 65% of calculated and measured monthly averages of air concentrations agree within a factor of three. Correlation coefficients of calculated monthly averages with measured ones are in the range from 0.4 to 0.95. The differences between measured and calculated values of air concentrations are conditioned by the fact that the model overestimates their seasonal variations at most of the sites (see, for example, the comparison of calculated and measured air concentrations at DE1 and SE12 presented in Fig. 2.42). This overestimation may be due to the uncertainties in model description of temperature dependence of γ-HCH physical-chemical parameters (vapor pressure and the Henry coefficient) or due to the uncertainties in the description of γ-HCH accumulation in soil.

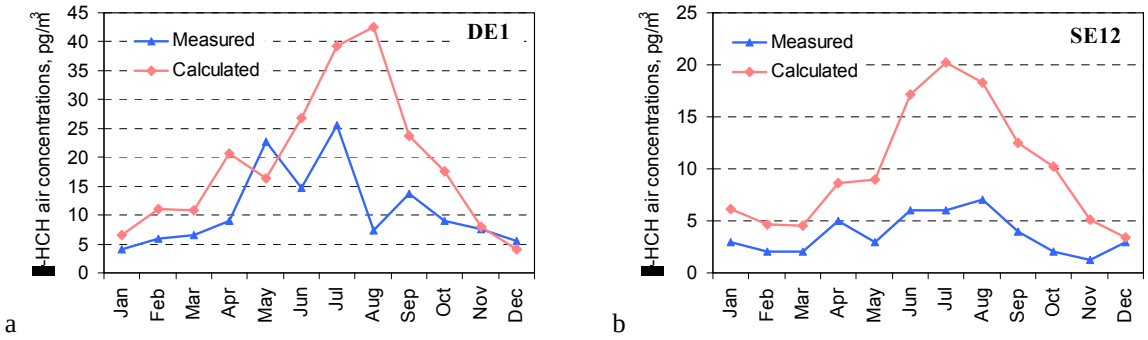

Fig. 2.42. *Comparison of monthly means of calculated and measured air γ-HCH concentrations at EMEP measurement sites DE1 (a) and SE12(b), pg/m^3*

It can be concluded that calculated air concentrations of B[a]P, PCB-153 and γ-HCH agree with measurements mostly within a factor of three.

For the improvement of the agreement between model calculations and measurements for concentrations in precipitation and deposition fluxes further refinement of model description of deposition processes is needed. For the investigation of these processes more simultaneous measurements of air concentrations, deposition fluxes and concentrations in precipitation complemented by meteorological parameters are required.

For more realistic description of seasonal variations of pollution the data on emission seasonal variations are required.

The elaboration of the integrated monitoring/modelling/emission approach based on the application of back trajectory technique has been started. The application of this approach to the analysis of the agreement between measurements and calculation results allows determining hot spots in emission inventories and measurement data and main directions of further model modification. Further development of the approach is required.

3. POP MODEL DEVELOPMENT

In 2010 the work on further development of the multicompartment POP transport model (MSCE-POP) was continued. This year main attention was paid to the evaluation of the influence of meteorological parameters on POP fate in the environment, evaluation of model sensitivity to the application of size-segregated parameterization of POP deposition in particulate phase, and investigation of POP candidate substances.

3.1. Evaluation of influence of meteorological parameters on POP long-range transport

The changes in meteorological and environmental parameters due to future climate change can essentially affect the fate of various pollutants (POP and heavy metals) in the environment. So, for the evaluation of the influence of climate change on transport and accumulation of substances, it is important to investigate the influence of meteorological parameters (temperature, precipitation amount, etc.) and environmental parameters (particularly land cover) on the atmospheric transport of pollutants. For the investigation of this influence it is necessary to select a target parameter distributing the pollution by the substance under consideration and a set of factors which are able a priori affect the target parameter. The method of such an investigation was described earlier in Technical Report [*Gusev et al.,* 2009] in the context of investigation of POP-like candidate substances. Here the work on evaluation of the dependence of POP transport distance on the meteorological parameters is continued. The investigation of another target parameter (total deposition flux to selected EMEP countries) is performed in the framework of the evaluation of HM pollution [*Ilyin et al.*, 2010a]. Two POPs are chosen for the analysis: benzo[a]pyrene (well-known POP already included into the consideration under CLRTAP Protocol on POPs) and β-endosulfan (POP candidate being at present under consideration by the Task Force on POPs). These two pollutants are different from the point of view of their physical-chemical properties. For example, B[a]P is mainly present in the atmosphere in particulate phase whereas for β-endosulfan the gaseous phase is more essential (up to 60% in summer months).

A target parameter for the investigation is transport distance (*TD*) of the considered pollutant. This parameter is one of the key parameters for the description of the pollutant's fate in the environment. It characterizes the ability of a POP to be transported to sufficiently long distance from the source where it is released into the environment. The value of *TD* is defined as an average distance at which the pollutant concentration drops 1000 times in comparison to that near the source [*Vulykh et al.*, 2006]. Below the dependence of TD on selected meteorological and environmental parameters (factors) is examined.

The following factors were included into the investigation:

- temperature *T*,
- precipitation amount *P,*
- fraction of water bodies *W* in the area over which the transport of the pollutant occurs,
- fraction of forest *F* in this area,
- fraction of grass *G* in this area.

For the analysis, calculations of pollutant transport from conventional point sources located in Finland (30.1ºE, 62.7ºN), Italy (10.5ºE, 45.3ºN) and the UK (1.2ºW, 53.7ºN) are performed. These three locations represent different climatic zones in Europe. Namely, the source in the UK is located in the seashore area, the Finnish source is situated in the continental part of northern Europe, and the Italian source is located in southern Europe. For the comparability of the results the intensity of each point source is taken one and the same – 1 t/y. For the comparison of the results between various years model simulations are done for several years (1996, 2001, 2003, and 2004) with different meteorological conditions.

For each source and each month of the considered years the target parameter *TD* and monthly values of the above factors are calculated in the following way. Using the model meteorology the area covered by trajectories originated from the considered source is calculated. Then the value of each factor is determined as an average over this area with weights equal to the time which trajectories of wind speed spend in the considered grid cell within the considered month. So, the values of the factors *W*, *F* and *G* depend not only on the land cover information but also on the absolute values and directions of the wind speed during the considered month. Thus, the input data for the analysis is the set {(TD_i, T_i, P_i, W_i, F_i and G_i), i = 1, ..., 48}, where i is the month number.

We note that for the investigation both the entire set of input data described above and the subset of this set can be chosen. In this study we shall use four data sets corresponding to each particular year. Under such an approach the influence of the considered factors to the target parameter within one and the same year is investigated. To investigate the stability of the obtained results the investigation of the data set including all considered years simultaneously is additionally performed for each pollutant and each conventional source.

Below we present a short description of the results of the analysis for B[a]P and β-endosulfan. This description consists of general description of the input data followed by the investigation of factors influencing the behavior of these pollutants in the environment. The detailed presentation can be found in the Technical Report [*Ilyin et al.*, 2010].

BENZO[A]PYRENE (B[A]P)

General description of the input data. The comparison of seasonal variations of *TD* for B[a]P, predicted by the model for the considered years, is presented in Fig. 3.1.

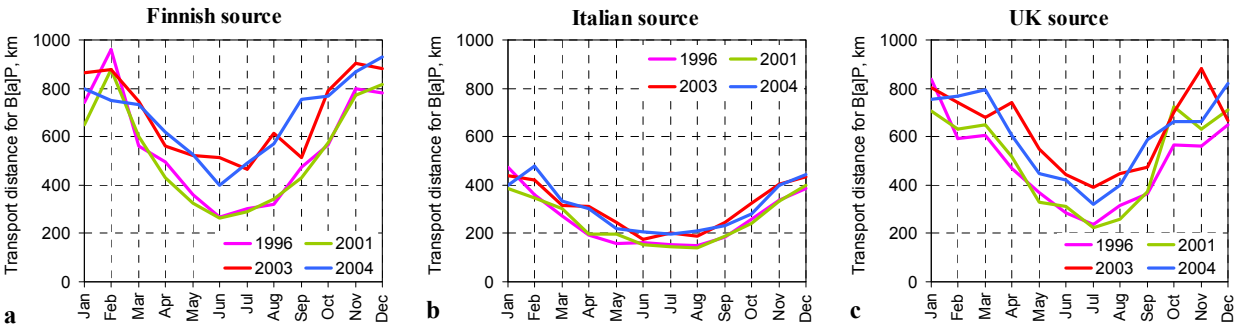

Fig. 3.1. *Monthly based values of the TD for B[a]P calculated on the basis of simulations of B[a]P transport from Finnish (a), Italian (b) and UK (c) sources, km*

The following peculiarities of *TD* seasonal variations can be marked:

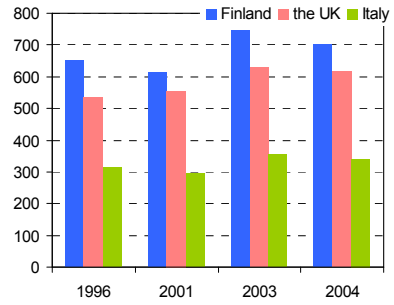

Fig. 3.2. *Annual based values of the TD for B[a]P for different sources and years*

1. Absolute values of the *TD* differ between chosen sources. Namely, the highest ability to long-range transport is calculated for Finland, and the lowest – for Italy (see Fig. 3.2).

2. There is a difference between values of the *TD* calculated for one and the same source in different years (Fig. 3.2).

3. Seasonal variations of B[a]P transport distance for all sources and years have one and the same shape with higher values in cold months and lower values in warm months. However, seasonal variations for each particular pollutant and source locations have their own peculiarities.

Results of the analysis. To illustrate the method applied for the investigation, it is exemplified below by the analysis of calculations made with Finnish source for 1996. Seasonal variations for this year are represented in Fig. 3.1a.

The method is based on successive regression analysis of the input data. It is realized as step-by-step procedure. At each step next (in importance) factor is selected. Table 3.1 summarizes the results obtained at all steps of the analysis. Each of the four subsequent rows of the table corresponds to one of the steps of the analysis and consists of:

- The list of factors selected at the given step in the order of their importance.
- Regression coefficients α_T, α_P, α_W, α_F, α_G.
- The intercept S of the regression relation.
- Coefficient of multiple determination for the constructed regression relation.

Table 3.1. *The results of regression analysis of B[a]P transport distance calculated on the basis of the transport from the Finnish source in 1996*

Factors involved	Regression coefficients						Coefficient of multiple determination R^2
	Temp.	Precip.	Water	Forest	Grass	Intercept	
	αT	αP	αW	αF	αG	S	
T	-17.9					600	0.65
T, F	-19.9			-3780		1890	0.78
T, F, G	-21.7			-4190	2293	1160	0.81
T, F, G, W	-21.9		1800	-2420	5140	-910	0.82

At the first step regression relations between the target parameter *TD* and each of the factors *T*, *P*, *W*, *F* and *G* separately are calculated. Comparing the coefficients of multiple determination obtained by these calculations the factor of main priority is chosen as the factor for which the multiple determination coefficient is maximal. For Finnish source in 1996 the temperature *T* occurs to be the most priority factor for explaining the variability of the transport distance (multiple determination factor equals 0.65). The first row of the Table 1 corresponds to this step. The corresponding regression relation is:

$$TD = \alpha_T \cdot T + S = -17.9 \cdot T + 600 \qquad (3.1)$$

approximating *TD* with coefficient of multiple determination 0.65.

At the second step regression relations between *TD* and pairs of factors (*T*, *P*), (*T*, *W*), (*T*, *F*) and (*T*, *G*) are calculated. The factor of the second priority is selected as the second element of the pair for which the multiple determination coefficient is maximal. In the considered case forest fraction *F* is a factor of second priority. The results of this step are summarized in the second row of Table 3.1. The corresponding regression relation is:

$$TD = \alpha_T \cdot T + \alpha_F \cdot F + S = -19.9 \cdot T - 3780 \cdot F + 1890 \qquad (3.2)$$

Application of this relation allows enlarging the multiple regression coefficient up to 0.78.

So, if the contamination is transported along the trajectories coming through forest areas, the transport distance is lower than in the case when no forest appears along the transport trajectories. Due to prevailing of vegetation-covered areas in Finland, this factor is one of the factors of main priority.

Similar procedure is applied to select the factors of next priorities. The following two rows describe the two next factors (grass and water fractions *G* and *W*, respectively) for Finland in 1996. The coefficient of multiple determination for the final regression relation is 0.82.

The comparison of the values of *TD* for B[a]P calculated by the model for 1996 on the basis of the Finnish source with that calculated by the regression relations constructed at steps 1 – 4 is displayed in Fig. 3.3.

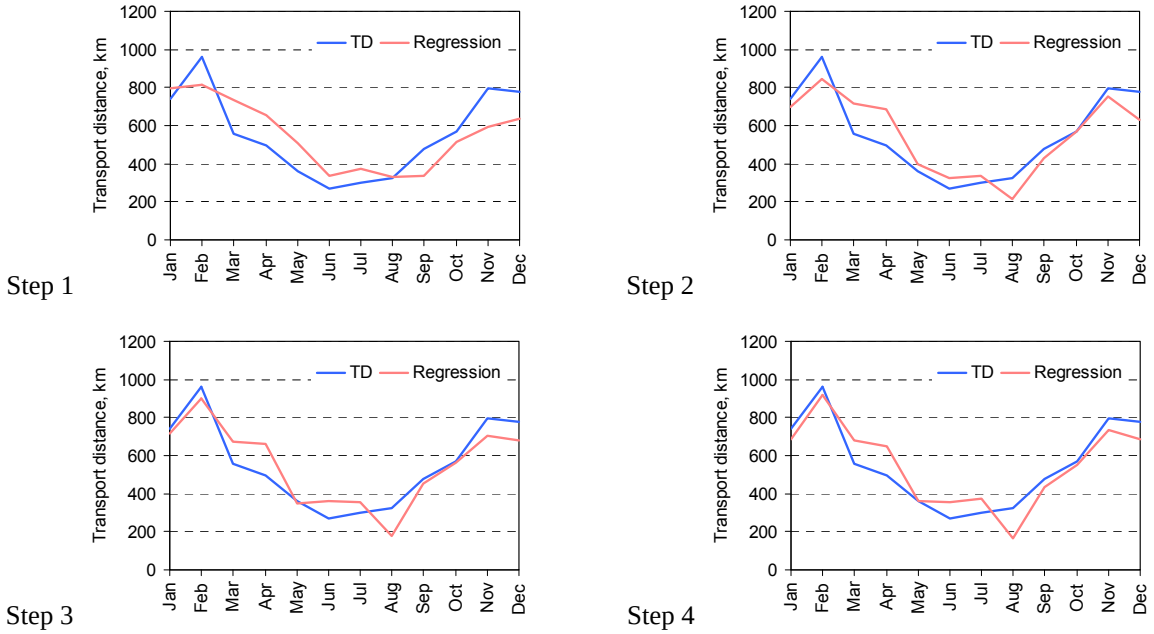

Fig. 3.3. *The comparison of the initial TD for 1996 with that calculated by the regression relations (Finnish source), km*

The comparison of the plots corresponding to different steps of approximation shows that at the first stage (consideration of the main priority factor only) the approximation takes into account just a general tendency of the *TD* variations. Inclusion of the factors of second and third priority subsequently improves the approximation. The inclusion of the fourth factor leads to almost negligible improvement. This can be seen also from the consideration of coefficients of multiple determination R^2 obtained at each approximation step (last column in Table 3.1). The inclusion of the second factor changes this coefficient from 0.646 to 0.778 whereas the inclusion of the fourth factor leads to the minimum change of R^2 from 0.807 to 0.816. The same is true for all other years also.

Further, the analysis shows that the factor of the first priority is temperature followed by forest fraction for all considered years. The regression coefficients for these factors are negative, which indicates that the ability of B[a]P to long-range transport decreases with the increase of the temperature and fraction of the area covered by forests.

To examine the stability of ranking of factors between different years the analysis was applied to all years together. The results are summarized in Table 3.2.

Table 3.2. *The results of regression analysis of B[a]P transport distance calculated on the basis of the transport from the Finnish source for all years together*

Factors involved	Regression coefficients						Coefficient of multiple determination R^2
	Temp.	Precip.	Water	Forest	Grass	Intercept	
	αT	αP	αW	αF	αG	S	
T	-13.4					638	0.47
T, F	-13.4			-2290		1386	0.53
T, F, P	-13.0	-23.6		-2388		1457	0.54
T, F, P, G	-13.0	-23.4		-2385	-91	1490	0.54

The comparison of the results obtained for particular years with the result using all set of data shows that the two main factors are the same for all considered sets of data: the temperature and the forest fraction of the trajectory-covered area. The contribution of changes in precipitation amounts occurring within the selected period is minimal.

The plot comparing the variations of the TD with the results of calculations by regression relation, taking into account four priority factors within four considered years, is presented in Fig. 3.4.

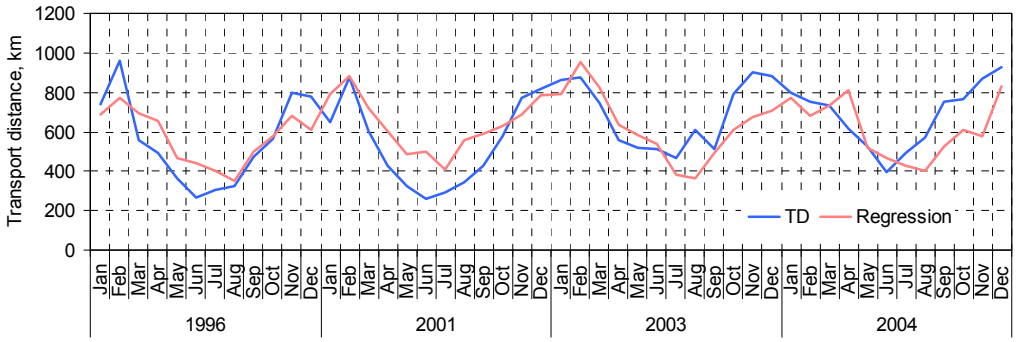

Fig. 3.4. *The comparison of the initial TD for years 1996, 2001, 2003 and 2004 with that calculated by the regression relations (Finnish source), km*

The differences between the values of *TD* calculated by the model and those obtained by the regression relation can be explained by several reasons. First, the set of factors chosen for the analysis may be incomplete. Second, regression analysis does not take into account the spatial variations of the factors within the area covered by the trajectories originated from the source. However, regression relations explain about 55% of total variations of the *TD*, which is reasonable for such kind of the analysis.

Now the description of the results of the analysis obtained on the basis of simulations of B[a]P transport from the Italian source is presented. Factor ranking together with coefficients of multiple determination are shown in Table 3.3.

Table 3.3. *The results of regression analysis of B[a]P transport distance calculated on the basis of the transport from the Italian source for particular years*

1996		2001		2003		2004	
Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2
G	0.64	G	0.76	G	0.88	G	0.46
G, T	0.76	G, T	0.86	G, W	0.89	G, T	0.58
G, T, W	0.82	G, T, W	0.90	G, W, T	0.90	G, T, F	0.69
G, T, W, P	0.83	G, T, W, P	0.93	G, W, T, P	0.90	G, T, F, W	0.74

The results obtained for Italian source show higher multiple determination coefficient than that obtained for Finnish source. Coefficients of multiple determination for regression relations using four factors range from 0.74 to 0.93 for the considered four years. The main priority factor is found to be grass fraction. Temperature is the factor of second priority for three from four considered years. The factor of the third priority is mostly water fraction. Precipitation and forest fraction are less important.

Thus, similar to the case of Finnish source, for Italy the two main factors are vegetation cover and temperature. Due to the peculiarities in geographic location of the source, the type of vegetation mostly influencing long-range transport is grass.

The results of the analysis obtained on the basis of simulations of B[a]P transport from the UK source are summarized in Table 3.4.

Table 3.4. *The results of regression analysis of B[a]P transport distance calculated on the basis of the transport from the UK source for particular years*

1996		2001		2003		2004	
Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2
F	0.50	T	0.57	T	0.57	T	0.70
F, P	0.68	T, F	0.70	T, F	0.63	T, P	0.75
F, P, T	0.78	T, F, P	0.77	T, F, W	0.66	T, P, F	0.76
F, P, T, W	0.81	T, F, P, W	0.83	T, F, W, G	0.68	T, P, F, W	0.82

Similar to the two above considered cases, temperature and vegetation cover are the two factors of highest priority for almost all years considered. However, in case of the UK, precipitation amount can be also a priority factor for the description of the variability of *TD*. For two years (1996 and 2004) it is a factor of the second priority. The values of multiple regression coefficient for relations using four factors ranges from 0.68 to 0.83 between the considered years.

β-ENDOSULFAN

General description of the input data. Seasonal variations of the *TD,* calculated on the monthly basis for β-endosulfan, are presented in Fig. 3.5.

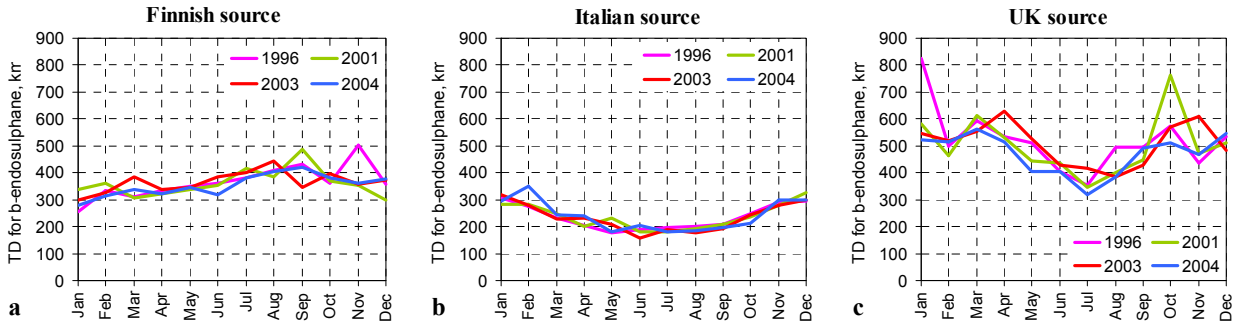

Fig. 3.5. *Monthly based values of the TD for β-endosulfan calculated on the basis of simulations of transport from Finnish (a), Italian (b) and UK (c) sources*

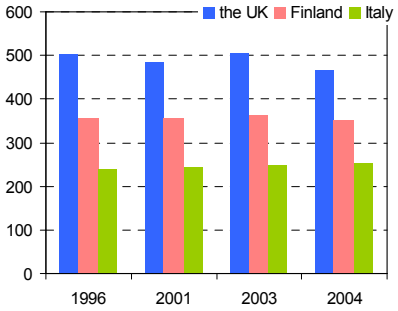

Fig. 3.6. *Annual based values of the TD for β-endosulfan for different sources and years*

Similar to the case of B[a]P, absolute values of the *TD,* calculated for different sources, are different (though the difference is less than for B[a]P). However, in contrast to the B[a]P, here maximum *TD* value has been obtained for the UK (see Fig. 3.6). Another essential difference between these two pollutants is that the shape of *TD* seasonal variations is different for different source locations. Namely, for Italy and the UK the shape of the *TD* seasonal variations is similar to that obtained for B[a]P (higher values in cold months and lower – in warm ones). But for Finland the situation is the opposite: higher values of the *TD* are characteristic for warm months.

Results of the analysis. Here the analysis of the influence of the above factors to the transport distance is applied to explain the differences in its values. The results obtained on the basis of calculations made with Finnish source for all the considered years are summarized in Table 3.5.

Table 3.5. *The results of regression analysis of β-endosulfan transport distance calculated on the basis of the transport from the Finnish source for particular years.*

1996		2001		2003		2004	
Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2
F	0.52	W	0.58	T	0.65	T	0.46
F, T	0.66	W, T	0.87	T, F	0.76	T, F	0.63
F, T, W	0.80	W, T, P	0.88	T, F, W	0.84	T, F, W	0.73
F, T, W, G	0.84	W, T, P, F	0.88	T, F, W, P	0.87	T, F, W, P	0.75

For three of the four considered years main factors affecting the *TD* are temperature and forest fraction. For one of the years the water fraction is considered to be a factor of main priority.

However, there is essential difference between the results obtained for B[a]P and β-endosulfan. For the latter pollutant regression coefficients for temperature for Finnish source are found to be positive. This means that the transport distance enlarges with the increase of temperature. The explanation of this fact may be as follows.

The removal rate of a substance from the atmosphere is a crucial parameter determining the ability of this substance to be transported over long distances. This parameter is determined mainly by degradation and deposition to the underlying surface. Both these processes are temperature-dependent. When the temperature is growing, the fraction of the gaseous phase of the pollutant increases, and degradation becomes faster. However, the rates of wet and dry deposition of gaseous phase decrease with temperature. So, the removal from the atmosphere is determined by two concurrent processes, and the overall removal rate depends on the relation between the rates of these processes. From the obtained results it follows that in the range of temperatures characteristic for Finland (typically from − 15 to +15 ºC) main removal processes are wet and dry deposition of gaseous phase which leads to the inverse character of temperature dependence of the *TD* (higher values of the *TD* in warm months, see Fig. 3.5.a). We remark that for Italy and the UK, where positive temperatures prevail, regression coefficients for the temperature are found to be negative similar to the case of B[a]P. That is why the character of seasonal variations of the *TD* for β-endosulfan is similar to that obtained for B[a]P (higher *TD* values in cold months).

The results of the analysis obtained on the basis of simulations of β-endosulfan transport from the Italian source are given in Table 3.6. The Table shows the factor ranking together with coefficients of multiple determination.

Table 3.6. *The results of regression analysis of β-endosulfan transport distance calculated on the basis of the transport from the Italian source for particular years.*

1996		2001		2003		2004	
Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2
G	0.81	G	0.79	W	0.86	G	0.41
G, T	0.83	G, P	0.87	W, P	0.89	G, F	0.64
G, T, F	0.83	G, P, W	0.90	W, P, T	0.90	G, F, P	0.71
G, T, F, W	0.83	G, P, W, T	0.91	W, P, T, F	0.91	G, F, P, W	0.76

The main priority factor is found to be grass fraction for most of the years similar to the result obtained during the investigation of B[a]P. The factor of second priority is temperature (for one of four considered years), precipitation amount (for two of considered years) and forest fraction (for one of the considered years). The factor of the third priority can be water fraction, temperature or precipitation.

Thus, similar to the case of Finnish source, for Italy the two main factors are vegetation cover and temperature. In some meteorological situation water fraction can strongly affect the values of the TD.

The results of the analysis obtained on the basis of simulations of β-endosulfan transport from the UK source are summarized in Table 3.7.

Similar to the two above considered cases, temperature and vegetation cover are the two factors of highest priority for almost all years considered. However, in the case of the UK, precipitation amount can be also a priority factor for the description of the variability of *TD*. For two years (1996 and 2004) it is a factor of the second priority. The consideration of multiple determination coefficients shows that in this

case the inclusion of all four selected factors leads to the improvement of the agreement between the *TD* values calculated on the basis of regression relation and the values calculated bi the model. The values of multiple regression coefficient for relations using four factors range from 0.68 to 0.83 between the considered years.

Table 3.7. *The results of regression analysis of β-endosulfan transport distance calculated on the basis of the transport from the UK source for particular years*

1996		2001		2003		2004	
Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2	Factors involved	Coefficient of multiple determination R^2
F	0.50	T	0.57	T	0.57	T	0.70
F, P	0.67	T, F	0.70	T, F	0.66	T, P	0.75
F, P, T	0.79	T, F, P	0.77	T, F, W	0.72	T, P, G	0.75
F, P, T, W	0.81	T, F, P, W	0.83	T, F, W, G	0.78	T, P, G, F	0.76

Concluding, it can be mentioned that for B[a]P, the most valuable factors determining its long-range transport are temperature and vegetation cover. The type of vegetation cover can be different depending on the geographical location of the source considered. For particular locations, precipitation intensity may essentially influence the ability to the long-range transport.

For β-endosulfan, the most valuable factors determining its long-range transport are again temperature and vegetation cover. However, in contrast to the B[a]P case, the dependence of the transport distance *TD* for β-endosulfan on temperature is different for different temperature ranges. Namely, for lower temperature range (taking place in Finland) transport distance increases with the increase of temperature. This leads to the inverse seasonal variations of the *TD* (higher values of the *TD* in warmer months). For particular locations, water fraction may essentially influence the ability to the long-range transport.

Projected climate changes can essentially affect the POP transport. To reveal the influence of climate change on the POP fate in the environment, the projections of changes both of meteorological parameters (temperature, precipitation) and geophysical parameters (particularly vegetation cover) are strongly desirable.

3.2. Sensitivity of POP model to application of particle size-segregated deposition parameterization

The current version of the MSCE-POP regional model treats particulate phase of POPs as mono-disperse fraction with fixed diameter [*Gusev et al.,* 2005]. The main purposes of the research described in this section was to evaluate the feasibility of operational modelling of POP atmospheric transport taking into account particle size distribution and to estimate the sensitivity of modelling results to the diameter of carrier particles. This research is a continuation of the work on the size-segregated approach for modelling of POPs dispersion and removal presented in previous Status Report [*Gusev et al.,* 2009].

To examine the influence of particle size distribution on deposition processes experimental calculations of the atmospheric transport of benzo[a]pyrene within the EMEP domain were carried out for 2008. B[a]P was chosen for modelling experiments because of its considerable particle-bound fraction.

For the sensitivity evaluation the wet and dry deposition schemes of the MSCE-POP model were changed to the size-segregated ones. In case of dry deposition the experience gained in the development of the parameterization of aerosol dry deposition in the MSCE-HM regional model [*Travnikov and Ilyin,* 2005] was used. For wet deposition of B[a]P a simple expression of the dependence between the washout ratio in the lowest model layer and aerosol particle diameter was employed. It is a quasi-linear function in double logarithmic scale (Fig. 3.7). This expression is based on literature data on aerosol wet deposition parameterizations [*Tsyro and Erdman,* 2000; *Sportisse*, 2007, etc.]. For particle diameter d_p = 0.84 μm adopted in the current monodisperse version of the MSCE-POP regional model for B[a]P the new parameterization gives the same value of washout ratio (W= 5×10^4) as the old one. The minimum at d_p = 0.2 μm (see Fig. 3.7) imitates the so-called "Greenfield gap" – a well known feature of below-cloud washout consisting in weaker scavenging of the particles with intermediate diameters (d_p ~ 0.05 - 1 μm).

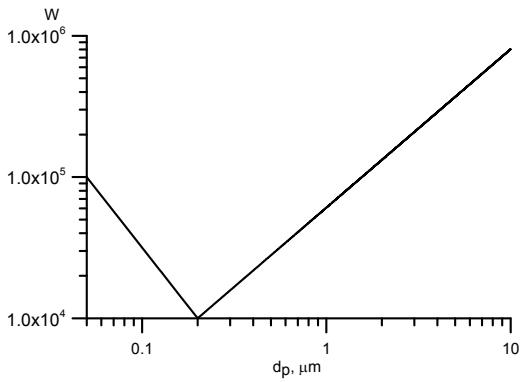

Fig. 3.7. *Washout ratio (W) as a function of aerosol particle diameter (d_p)*

Several runs of the MSCE-POP model with different parameterizations of deposition (old and new) and different B[a]P particle phase size distributions (mono-disperse and size-segregated) were done (Table 3.8). The variants with fixed particles diameter d_p = 0.84 μm (runs 1,2) were considered to assess the effect of deposition parameterization change just as it was. For the "size-segregated" run 8 it was supposed that at the moment of emission the number concentration of carrier particles had lognormal distribution with mass median diameter MMD = 0.84 μm and standard deviation σ = 2. The continuous lognormal function was approximated by a quasi-constant 5-bin distribution as it shown in Fig. 3.8. To investigate the sensitivity of calculation results to the diameter of carrier particles five scenarios with mono-disperse aerosol distribution were examined (runs 3-7). For these runs particles diameters were chosen equal to MMDs of five bins mentioned above.

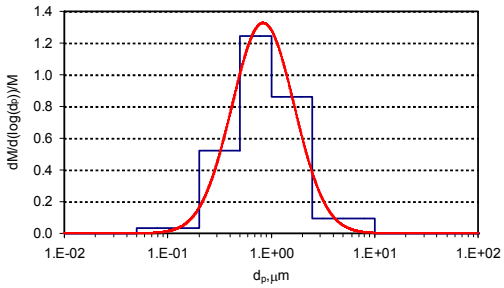

Fig. 3.8. *Initial B[a]P mass distribution on particle sizes(d_p) adopted in this work*

Table 3.8. *Characteristics of model runs*

N	Name of run	Deposition parameterization	Particle size distribution
1	Base	old	Mono-disperse with dp = 0.84 μm
2	New removal schemes	new	Mono-disperse with dp = 0.84 μm
3-7	New removal schemes	new	Mono-disperse with dp = 0.10 μm; 0.32 μm; 0.71 μm; 1.58 μm; 5.00 μm
8	Size-segregated	new	Quasi-constant 5-bin (Fig. 2) with MMD = 0.84 μm

The change of parameterization of deposition (run 2) resulted in the increase of total deposition flux of B[a]P over the major part of the EMEP domain (Fig. 3.9a) as a consequence of the growth of dry deposition velocities to the most types of land-cover (wet deposition velocities were equal to each other in the runs 1, 2 because the washout ratio had one and the same value for d_p = 0.84 μm – see the description of new wet deposition parameterization presented above). However, dry deposition velocity over the urban areas decreased when new parameterization was used. Therefore, in regions with high

fraction of urban area (e.g., the United Kingdom, east of Ukraine, Belgium) the total deposition flux decreased. The inclusion of particle size distribution (run 8) led to the slight increase of total deposition over the most part of the model domain (compare Fig. 3.9ab). Air concentrations calculated in the experimental model runs 2, 8 decreased in comparison to the results of the base run over the most part of the model domain (Fig. 3.10). As a whole, the spatial distributions of air concentrations and deposition fluxes of B[a]P for the size segregated run 8 did not differ much from those for mono-disperse run with d_p = 0.84 μm (run 2). Generally, the differences between the calculated values of air concentrations and deposition obtained in the base run and analogous results of size-segregated run did not exceed 20% for the most polluted regions of the Europe.

The comparison of annually averaged modelling results against the EMEP measurements of B[a]P concentrations in air and precipitation did not show essential differences associated with the introduction of the size-segregated deposition schemes comparing to the currently used ones (Table 3.9). At the same time, the effect of parameterization change can be more essential considering finer temporal and spatial resolution. It can be mentioned also that the size-segregated parameterization of dry deposition provided better spatial correlation of annual mean modelled and measured B[a]P concentrations in air than the old one (R_{corr} = 0.52 vs. 0.48 for the base case).

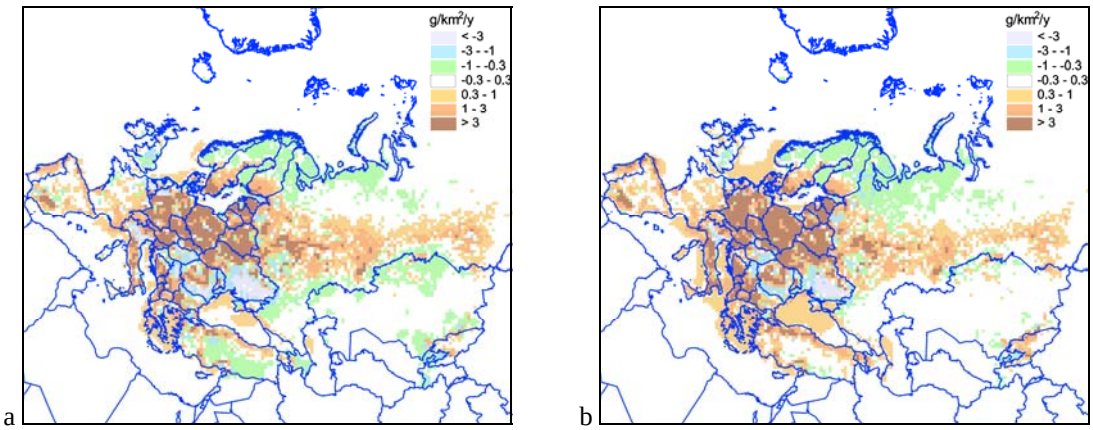

Fig. 3.9. *Differences in spatial distributions of annual total deposition flux of particle B[a]P over the EMEP domain in 2008: a – difference between the run with new parameterization of deposition (run 2) and the base run 1; b – difference between the size segregated run 3 and the base run 1*

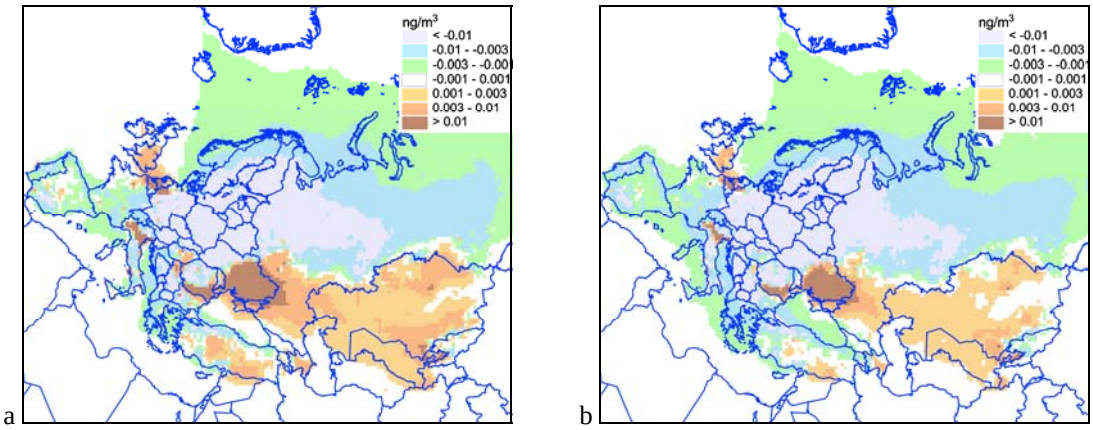

Fig. 3.10. *Differences in spatial distribution of annual air concentration of B[a]P over the EMEP domain in 2008: a – difference between the run with new parameterization of deposition and the base run; b – difference between the size segregated run and the base run*

Table 3.9. *Statistics for different model runs based on yearly mean values*

Name of run	Concentration in air (17 sites)		Concentration in precipitation (6 sites)	
	<BIAS>, ng/m^3	Rcorr(spatial)	<BIAS>, ng/L	Rcorr(spatial)
Base	0.007	0.48	1.05	0.54
New removal schemes	-0.011	0.52	0.22	0.54
Size-segregated	-0.014	0.51	0.67	0.54

Using performed model simulations the dependence of modelling results on the carrier particle diameter (d_p) was investigated (model runs 3-7 with mono-disperse particle size distribution). The increase of aerosol diameter resulted in the considerable growth of dry and wet deposition of particle-bound B[a]P. Total dry deposition increased nearly five times with changing d_p from 0.1μm to 5μm (Fig. 3.11), wet deposition – nearly 3 times. The minimum of wet deposition at d_p= 0.32 can be explained by the presence of minimum in washout ratio curve (Fig. 3.7). At the same time, B[a]P content in air decreased as shown in Fig. 3.12) and total mass of particle phase of B[a]P in air fell almost five times.

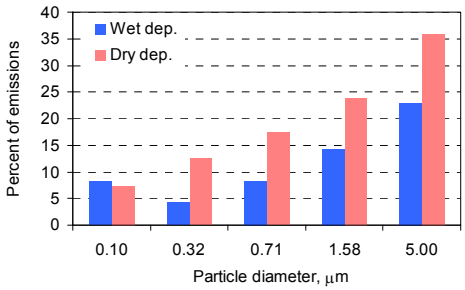

Fig. 3.11. *Annual wet and dry deposition of aerosol phase of B[a]P to EMEP domain in percent of emitted mass*

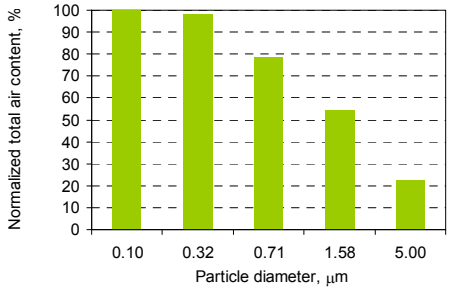

Fig. 3.12. *Annual mean total aerosol phase mass of B[a]P in the whole EMEP domain normalized to the results of the model run with d_p =0.01μm*

Thus, the size-segregated parameterizations of wet and dry deposition of POPs in particulate phase were developed and tested in framework of several modelling experiments. The introduction of the particle size-segregated approach is important development of the MSCE-POP model as aerosol particles play important role in the long-range transport and removal of POPs from the atmosphere. At the same time, further work to evaluate and improve the description of size-resolved POP dispersion and removal is required.

3.3. Evaluation of POP candidate substances

The investigation of long-range transport of perfluorooctane sulphonate anion (PFOS) and related substances has been begun this year.

During last several years dangerous properties of these chemicals for human health and the environment were under consideration in OECD, UNEP (the Stockholm Convention) and CLRTAP. As a result, perfluorooctane sulfonic acid, its salts and perfluorooctane sulphonil fluoride were included to the Stockholm Convention. The inclusion of PFOS to the CLRTAP Protocol on POPs was considered and approved by the Executive Body for the Convention [ECE/EB.AIR91].

Perfluorooctane sulphonate anion with chemical formula $C_8F_{17}SO_2$ is a representative of a large class of chemicals including different organic compounds − salts, acids, ethers, etc. At least two different groups

of chemicals can be distinguished from this class − PFOS in the ionic form and the so-called PFOS-related substances.

First group contains perfluorooctane sulphonate anion, its salts and other derivatives which exist almost by 100% in the ionic form under the conditions of natural waters (neutral pH). The most important property for this group is high enough solubility. For example, the solubility of potassium salt of PFOS in pure water is about 600 mg/L, and about 12 mg/L in seawater [*Environmental and health assessment.*., 2003].

Low values of vapor pressure lead to low volatility of these compounds. Due to high solubility, they are characterized by slow exchange at air/water interface. These properties allow concluding that the long-range atmospheric transport is not characteristic for substances from this group. At the same time these substances are characterized by high persistence in seawater (half-life of potassium salt of PFOS is reported to be more than 40 years [*Brooke et al*., 2004]).

Manufacturing and consumption of these substances determine the main root of their emissions − mostly to surface waters due to discharge of sewage. The direct emission to the atmosphere is negligible. So, the main pathway of long-range transition of these chemicals is the transport with sea currents.

It should be also taken into account that the behavior of the substances from this group in the environment strongly differs from the behavior of “traditional” POPs. Besides, it is difficult (or impossible) to measure some physical-chemical properties of these substances (e.g., octanol/water partitioning coefficient cannot be measured due to surface-active properties). For modelling of environmental transport of the substances of this group the model modification is required since classical models based on the partitioning in the system air/octanol/water are inapplicable for this purpose [*Mackay*, 2001].

The second group (PFOS-related substances) consists of such compounds as perfluorosulphoamides and their derivatives, acrylates polymers, etc. containing PFOS as a structural element. The characteristic feature of these chemicals is the ability to generate PFOS during their degradation; they are named PFOS precursors. The number of such substances is evaluated by various researchers as 100 − 200. In [*Brooke et al*., 2004] 96 substances are listed as compounds potentially degrading to PFOS. At the same time the products with higher molecular mass (polymers) are stable and do not produce PFOS [*Environmental and health assessment.*., 2003].

The substances from this group are as a rule neutral and are more volatile than the ionic forms. For the comparison, vapor pressure of potassium salt of PFOS (ionic substance) is $3 \cdot 10^{-4}$ Pa whereas for N-ethyl perfluorooctane sulphonamido ethanol (N-EtFOSE − PFOS-related substance) this parameter equals 0.35 Pa [*Environmental and health assessment.*., 2003; *Lei et al.*, 2004]. The half-life of N-EtFOSE in the atmosphere can reach 50 days [*Lei et al.*, 2004].

The analysis of measurement results for PFOS-related substances shows the presence of such substances as N-methyl perfluoroocnane sulphonamido ethanol (N-MeFOSE), N-ethyl perfluorooctane sulphonamido ethanol (N-EtFOSE) in the atmosphere of remote regions [*Dreyer et al*., 2009; *Shoeib et al.*, 2006; *Piekarz et al.*, 2007].

Thus, modelling long-range transport with the help of existing models is possible for the substances of this group.

However, for correct description of PFOS levels in the environment both their direct transport in the marine environment and atmospheric transport of their precursors should be further investigated.

4. COOPERATION

The work on evaluation of POP contamination in the EMEP region is performed in close collaboration with subsidiary bodies to the Convention, various international organizations and national experts. Main directions of these activities are presented in this section. In addition, co-operation with the Task Force on HTAP concerning participation in the HTAP 2010 Assessment Report and multi-model HTAP intercomparison are described in detail in Sections 1.3 and 1.4 of the present report.

4.1. HELCOM

This year MSC-E along with CCC and MSC-W continued the co-operation with the Baltic Marine Environment Protection Commission (HELCOM). In accordance with the Memorandum of Understanding between the HELCOM and the United Nations Economic Commission for Europe on cooperation in the field of monitoring of air pollutants EMEP Centres prepared an annual joint report on the evaluation of airborne pollution load to the Baltic Sea [*Bartnicki et al*., 2009]. MSC-E contributed to the report with the evaluation of atmospheric input of PCDD/Fs to the Baltic Sea. Officially reported emission data on PCDD/Fs to EMEP for 1990-2007 were used in simulations for the Helsinki Commission. Modelling results on the pollution of the Baltic Sea by PCDD/Fs along with the contributions of surrounding countries were presented. The report was welcomed and endorsed by the Contracting Parties at the HELCOM MONAS 12 meeting in October 2009 and was recommended to be published on the EMEP and HELCOM websites.

Along with the joint report, the indicator fact sheets with updated information on the temporal variations of PCDD/F emissions to air and their deposition over the Baltic Sea in the period from 1990 to 2007 were prepared. These indicator fact sheets are available in the Internet at the HELCOM web site [*www.helcom.fi*].

Annual emissions of dioxins and furans have decreased in HELCOM countries during the period from 1990 to 2007 by 21%. The most significant drop of PCDD/F emissions can be seen in Finland (67%) and Denmark (58%). Some decrease of emission can also be noted for Lithuania (46%), Sweden (39%), Germany (27%), Russia (18%), Poland (17%), and Estonia (15%). For some of the HELCOM countries the level of PCDD/F emissions in 2007 is higher than emission of 1990, in particular, in Latvia which emission for 2007 is higher than that for 1990.

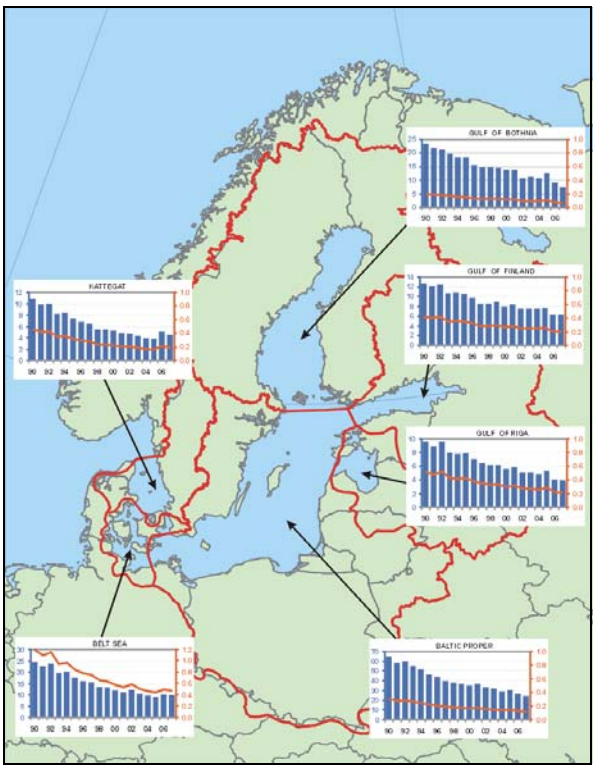

Fig. 4.1. *Computed net annual deposition of PCDD/Fs to the six sub-basins of the Baltic Sea for the period 1990-2007 in t/y as bars (left axis) and deposition fluxes in ng TEQ/m^2/y as lines (right axis)*

The level of annual atmospheric deposition of PCDD/Fs to the Baltic Sea has decreased in the period 1990-2007 by 62%. The most significant drop in PCDD/F deposition over individual sub-basins took place in the Gulf of Bothnia (68%) and the Baltic Proper (63%) (Fig. 4.1). For other sub-basins the decrease of deposition varies from 51% to 61%. The highest level of PCDD/F atmospheric deposition fluxes (0.47 ng TEQ/m^2/y) over the Baltic Sea in 2007 can be seen in its southern-western part (the Belt Sea) while the lowest one (0.06 ng TEQ/m^2/y) over the Gulf of Bothnia. In other sub-basins the level of deposition fluxes varies from about 0.11 to 0.21 ng TEQ/m^2/y. Among the HELCOM countries the most essential contributions to deposition over the Baltic Sea belong to Poland, Russia, and Denmark.

4.2. Co-operation with national experts

This year essential attention is paid to the examination of the environmental fate and transport of polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs). One of the basic difficulties in the investigation of this group of substances is the lack of measurement data needed to the interpretation of modelling results and validation of model assumptions and parameterization. To overcome partly this difficulty MSC-E together with the UMEÅ University, the Stockholm University and Institute for Applied Environmental Research (Sweden) participates in the Swedish EPA research program “BalticPOPs”. In the framework of this project it is planned to evaluate the contamination of the Baltic Sea and its subregions with the help of model evaluation and available measurements at three Scandinavian sites (Aspvreten, Pallas and Vindeln). On the basis of this research it is supposed to further develop the integrated approach taking into account emissions, modelling and measurements for PCDD/Fs. The results of the project will be published next year.

MSC-E continues its participation in the EU HEIMTSA project launched in 2007 under the 6th Framework Programme of the European Commission (EC) in collaboration with Universität Stuttgart (Germany). The project aims to support the Environment and Health Action Plan (EHAP) by extending health impact assessment (HIA) and cost benefit analysis (CBA) methods and tools so that environment and health impacts of policy scenarios in key sectors can be evaluated reliably at the European level. The project includes the investigation of PCDD/F, PCBs, B[a]P and trifluralin. This research, carried out in the frame of the INTARESE/HEIMTSA mega case study, is based on linking of two multimedia models: MSCE-POP and PANGEA.

Special attention this year was paid to strengthening cooperation with EECCA countries. In particular, Memorandum of Understanding between MSC-E and Institute for National Resources Use and Ecology of the National Academy of Science of Belarus was signed. The main aim of the Memorandum is a complex of joint works within scientific research activities in the framework of transboundary air pollution by POPs, creation of scientific basis of the compliance with international agreements concerning the protection of atmospheric air and strengthening of scientific potential of Parties.

5. FUTURE ACTIVITIES

Annual activities

Preparation of meteorological data for operational modelling based on the European Centre for Medium-Range Weather Forecasts (ECMWF) analysis and continue work on update/development of the meteorological drivers.

Preparation of anthropogenic POP emissions data as input for operational modelling on the basis of gridded emission dataset provided by CEIP.

Calculations of PAH, PCBs, PCDD/Fs and γ-HCH air concentrations and ecosystem-dependent depositions over the EMEP domain in 2009 with resolution 50×50 km^2.

Computation of country-to-country deposition matrix for PAHs, PCDD/Fs and PCBs for 2009.

Estimation of PAHs, PCBs, PCDD/Fs, and γ-HCH deposition on the regional seas.

Calculation PCBs, PCDD/Fs and γ-HCH dispersion on a hemispheric/global scale for evaluation of contributions of intercontinental transport to regional pollution levels.

Evaluation new measurement data of POPs from the EECCA region to assess the relative importance of the different pollutants and the main source regions.

Analysis of the agreement between measured and modelled pollution levels with the help of back trajectory approach together with national experts on emission inventories and monitoring.

Preparation of individual country status reports in English and Russian for the EECCA countries.

Contribution to the work of the subsidiary bodies to the Convention and EMEP Task Forces:

- WGSR – support the work of TF on POPs in the evaluation of new POP candidates;
- TFMM, TFHTAP: Continuation of the co-operation in the field of POP modelling on both regional and global scales.

Cooperation with international bodies: AMAP, UNEP, the Stockholm Convention on POPs, EU (in particular as regards the EU regulation on registration, evaluation, authorization and restriction of chemical substances (REACH)), WMO, EEA, HELCOM, the OSPAR Commission and national experts.

Dissemination of results (e.g. via status reports, technical notes, the website, publication in peer-reviewed journals).

Research and development activities

Continuation of the analysis of the effect of altering meteorological parameters associated with the climate change on POP pollution levels in Europe.

Usage the results from the EMEP and AMAP laboratory intercomparison of POPs in 2010 together with field intercomparison between passive and active sampling to assess the uncertainties in the POP measurements.

Exploration of the use of passive POP measurements to validate the EMEP model and other transport models to evaluate source contributions.

Continuation of the elaboration of the integrated monitoring/modelling/emission approach on the basis of back trajectory analysis and simplified adjoint models.

Investigation of seasonal variations of emissions for B[a]P and PCDD/Fs on the basis of calculations with different emission scenarios.

Continuation of the investigation of HCB contamination within the EMEP domain on the basis of experimental calculations with emission scenarios worked out with the help of available data.

Further development and testing the global modelling system GLEMOS for POPs including improvement of the modular architecture, elaboration of the multi-media approach and refinement of the pollutant specific processes.

Participating in the TFHTAP model intercomparison studies.

Participating in elaboration of the criteria and metrics for evaluation of modelling results in co-operation with TFMM.

CONCLUSIONS

This Status Report presents the progress in the evaluation of the pollution levels and transboundary transport of POPs within the EMEP region and at the hemispheric/global scales achieved in 2010. The main conclusions of the work carried out by CCC and MSC-E are summarized below.

POP intercontinental transport

The pilot version of the GLEMOS for POPs was elaborated. Particularly, the modules representing the atmosphere, soil, and seawater compartments were included as well as the description of intermedia exchange processes. Further improvement of the multimedia approach within the GLEMOS is connected with the inclusion of vegetation compartment and implementation of POP transport with sea currents.

Preliminary model simulations of PCB-153 global transport and fate performed by the multimedia version of the GLEMOS revealed the consistency of model estimates of PCB-153 air concentrations with measurements of the EMEP monitoring network. Besides, the computed concentrations in upper soil and seawater were in a reasonable agreement with available measurements. At the same time, the differences obtained demonstrated the need of further work on the refinement of process parameterizations implemented in the model.

Elaboration of emission scenarios and HCB modelling experiments permitted to improve the agreement of model estimates with available measurements. Model simulations demonstrated essential role of the historic emissions for the evaluation of current HCB pollution levels. Further investigations of HCB long-range transport and fate on global scale, taking into account available regional inventories of HCB emissions and the use of more detailed spatial resolution for particular regions, are needed to refine the description of current and future levels of pollution.

Intercomparison of POP models, designed on the basis of the TF HTAP methodology and aimed at evaluation and quantifying POP intercontinental transport, showed concentrations and deposition in receptor regions to be sensitive to the changes of POP emissions on the global scale. Model simulations, performed for the selected POPs, have demonstrated that the contribution of intercontinental transport to the pollution levels in the HTAP receptor regions can reach almost 30%.

Contamination levels in the receptors were most sensitive to the emission reductions in regions with high emission rates. Particularly, 20% reduction of PCB emissions in Europe and North America and similar decrease of α-HCH in the South Asia had the greatest influence on receptor regions, including the Arctic.

The HTAP 2010 Assessment Report is an important source of information on recent advances in the evaluation of POP intercontinental transport. Key findings regarding the significance of POP intercontinental transport and recommendations for future work in this area can be essential for further activity of the EMEP directed to the evaluation of POP pollution and transboundary transport between the EMEP countries.

Evaluation of POP transport and pollution levels

Evaluation of POP pollution levels and transboundary transport in the EMEP domain for 2008 was carried out on the basis of EMEP measurements, emission data, and modelling of POP long-range transport.

Emission datasets for model assessment of POP long-range transport within the EMEP region were compiled on the basis of official data submitted by the EMEP Centre on Emission Inventories and Projections (CEIP) and unofficial expert estimates. For the evaluation of intercontinental transport emissions of PCDD/Fs, PCB-153 and γ-HCH within the Northern Hemisphere were constructed.

Several countries reported estimates of uncertainties in their emission inventories. These uncertainties range from several tens of per cent to several times. For better interpretation of the differences between measurements and modelling results the reporting of emission uncertainties by countries is strongly desirable.

Modelling results on spatial distribution of B[a]P concentrations along with regular measurements showed that the highest levels of B[a]P concentrations were characteristic of the central, eastern, and southern parts of Europe (about 0.08 – 1 ng/m^3, annual averages). The most significant concentrations (more than 1 ng/m^3) were obtained in the eastern part of the Ukraine. Lower concentrations were seen in the Northern and Western Europe.

Due to the strong seasonal and spatial variability of B[a]P air concentrations, their levels in the Central, Eastern and Southern Europe for particular periods of time can exceed the maximum permissible level of 1 ng/m^3, accepted in a number of countries. More detailed evaluation of spatial and temporal variability of B[a]P concentrations can be done in framework of case study, including the modelling with finer spatial resolution and specific monitoring campaigns. Reporting of emission seasonal variations by countries should essentially improve the quality of the assessment of environmental pollution.

Evaluation of transboundary transport of PCDD/Fs was carried out for the mixture of 17 toxic PCDD/F congeners. In addition to emission sources within the EMEP domain, the influence of PCDD/F emission sources of the USA and Canada was taken into account. Elevated levels of PCDD/F air concentrations were obtained in the central, eastern, and southern parts of Europe (about 2 – 5 fg TEQ/m^3). The most significant concentrations (more than 5 fg TEQ/m^3) can be seen in some regions of Southern and Eastern Europe. For further improvement of evaluation of pollution by PCDD/Fs the congener-specific emissions are needed.

Investigation of PCB-153 transport and pollution levels was carried out for the EMEP region with resolution 50×50 km^2. Since congener-specific data of PCBs had not been submitted by the countries, modelling was made on the basis of the global emission inventory of PCB-153 prepared by *K. Breivik et al.* [2007]. Elevated levels of PCB-153 concentrations (3 – 5 pg/m^3 and higher) were obtained for some regions of the Western and Southern Europe (western Germany, the Netherlands, northern France and northern Italy).

The environmental levels of lindane (γ-HCH) have been significantly decreased within the EMEP region during two recent decades due to the reduction of its application. Model predicted elevated γ-HCH air concentrations (50 pg/m^3 and above) in 2008 for the United Kingdom and Spain, while in other parts of the EMEP domain the level of concentrations was comparatively lower.

The country-to-country matrices for 2008 were calculated for B[a]P, PCDD/Fs, and PCB-153. It was found that the contribution of transboundary transport for the EMEP countries varied typically from 20 to

70% of deposition originated from the primary anthropogenic sources, depending on country size, geographical location and domestic emissions. Model calculations showed essential contribution of secondary emissions (re-emissions) to the contamination of EMEP countries by PCDD/Fs and PCB-153. For these pollutants the contribution of non-EMEP sources can also be essential for countries located near the boundaries of the EMEP domain.

In 2008 there were twenty EMEP monitoring sites measuring POPs, which were three more than in 2007. Twelve of these sites conducted parallel measurements of POPs in air and precipitation. According to the measurement data, elevated concentrations of POPs are often seen in central parts of Europe reflecting proximity to major emission source areas in Europe.

It is difficult to quantify the uncertainty in the POP measurements since it depends on several factors (methodology, sample handling and preparation etc). However, in the POP laboratory intercomparison performed in 2000-2002 large uncertainties were seen for some labs – a bias of factor two or three for some components for some laboratories, though several laboratories were also within 20% of the expected value for most of the species that were included in the intercomparison.

To reduce uncertainties in monitoring, emission inventories and modelling, an integrated monitoring/modelling/emission approach is now under the development at MSC-E. The main idea of this approach is to use complex analysis of the differences between the modelling results and measurement data in order to improve assessment of pollution levels in the EMEP region. In the framework of this work MSC-E started the development of a special modelling tool for the analysis of the agreement between measurements and calculations. The tool was tested at the comparison of model calculations and measurements of B[a]P in 2008.

The comparison of calculated and measured values of air concentrations for B[a]P showed that the agreement between measurements and calculated values was generally within a factor of two-three. Correlation coefficients between the measured and calculated monthly averages of air concentrations were 0.6 - 0.8 and higher. However, it was obtained that seasonal variations of B[a]P concentrations were strongly underestimated by the model. Thus, reporting the data on seasonal variations of emissions by countries is highly desirable. In addition, the analysis performed for B[a]P indicated the need in further refinement of model description of deposition processes.

The measurements of PCDD/F concentrations are stipulated by the EMEP on a voluntary basis at supersites only. However, the work on the comparison of calculated PCDD/F concentrations with measurements obtained on national basis is now ongoing.

Calculated values of annual means of air concentrations for PCB-153 agreed with measured ones within a factor of two. Correlation coefficients between seasonal variations of calculated and measured air concentrations were mainly in the range from 0.6 to 0.9. At the same time, there were considerable discrepancies between calculated and measured deposition values for PCB-153, presumably in winter months. This indicated the need to improve model description of deposition scheme for this pollutant in cold months (especially for snow scavenging).

The agreement between calculated and measured annual means of γ-HCH air concentrations was mainly within a factor of three. Calculated values of γ-HCH concentrations in precipitation were lower than measurements for almost monitoring sites and agreed with measured concentrations within a factor of three. On the opposite, the comparison of measured and calculated deposition fluxes at EMEP sites FI96, SE12 and SE14 showed that the model strongly overestimated the values of deposition fluxes. The reasons of such discrepancies can be a topic of consideration together with experts from monitoring community.

POP model development

The projected climate changes can essentially affect the POP transport. So, for evaluating climate change scenarios, investigation of the influence of meteorological parameters on POP fate in the environment is required.

For B[a]P, the most valuable factors determining its long-range transport are temperature and vegetation cover. The type of vegetation cover can be different depending on the geographical location of the source considered. For particular locations, precipitation intensity may essentially influence the ability to the long-range transport.

Similarly , the most valuable factors determining long-range transport of β-endosulfan are temperature and vegetation cover. However, in contrast to the B[a]P case, the dependence of the transport distance for β-endosulfan on temperature is different for different temperature ranges. Namely, for lower temperature range (taking place in Finland) transport distance increases with the increase of temperature.

To examine the influence of climate change on the POP fate in the environment, the projections both of meteorological parameters (temperature, precipitation) and geophysical parameters (particularly vegetation cover) are needed.

The evaluation of the feasibility of operational modelling of POPs atmospheric transport taking into account particle size distribution and estimation the sensitivity of modelling results to the diameter of carrier particles was continued. It was found that usage of size-segregated scheme of deposition led to better spatial correlation of annual mean modelled and measured B[a]P concentrations in air than mono-disperse one.

The investigation of long-range transport of perfluorooctane sulphonate anion (PFOS) and related substances was continued. Two different groups can be distinguished among these substances − PFOS in the ionic form and the so-called PFOS-related substances. For modelling of environmental transport of substances of the first group the model modification is required since classical models based on the partitioning in the system air/octanol/water are inapplicable for this purpose. On the opposite, modelling of long-range transport for the substances of the second group with the help of existing models is possible. However, for correct description of PFOS levels in the environment both their direct transport in the marine environment and atmospheric transport of their precursors should be investigated.

Co-operation

The work on evaluation of POP environmental contamination and model development was performed in close collaboration with CLRTAP subsidiary bodies (TFMM, TF HTAP), international organizations (AMAP, EU, HELCOM, OSPAR, UNEP, WMO, etc) and national experts. In the framework of this activity essential attention was paid to the assessment of pollution levels in EECCA countries and their information support.

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ANNEX A

COUNTRY-TO-COUNTRY DEPOSITION MATRICES FOR 2008

Table A.1. *Codes of countries*

Country/Region/Sea	Code	Country/Region/Sea	Code
Albania	AL	Monaco	MC
Armenia	AM	Montenegro	ME
Austria	AT	Netherlands	NL
Azerbaijan	AZ	Norway	NO
Belarus	BY	Poland	PL
Belgium	BE	Portugal	PT
Bosnia and Herzegovina	BA	Republic of Moldova	MD
Bulgaria	BG	Romania	RO
Croatia	HR	Russian Federation (European part)	RU
Cyprus	CY	Russian Federation (Asian part)	RUA
Czech Republic	CZ	Serbia	RS
Denmark	DK	Slovakia	SK
Estonia	EE	Slovenia	SI
Finland	FI	Spain	ES
France	FR	Sweden	SE
Georgia	GE	Switzerland	CH
Germany	DE	The Former Yugoslav Republic of Macedonia	MK
Greece	GR	Tajikistan	TJ
Hungary	HU	Turkey	TR
Iceland	IS	Turkmenistan	TM
Ireland	IE	Ukraine	UA
Italy	IT	United Kingdom	GB
Kazakhstan	KZ	Uzbekistan	UZ
Kyrgyzstan	KY	Baltic Sea	BAS
Latvia	LV	Black Sea	BLS
Lithuania	LT	Caspian Sea	CAS
Luxembourg	LU	North Sea	NOS
Malta	MT	Mediterranean Sea	MDT

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2008, kg/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CY	CZ	DE	DK	
AL	**377.3**	0.02	0.43	0.03	2.41	0.23	10.61	0.28	0.02	0.001	1.31	3.50	0.19	**AL**
AM	0.01	**333.5**	0.01	69.11	0.01	0.01	0.08	0.12	0.00	0.01	0.05	0.13	0.02	**AM**
AT	4.09	0.03	**494.9**	0.07	15.84	12.71	5.61	4.03	1.92	0.003	152.7	483.1	5.09	**AT**
AZ	0.02	42.24	0.03	**766.7**	0.04	0.04	0.11	0.67	0.00	0.01	0.13	0.40	0.08	**AZ**
BA	18.96	0.03	7.80	0.05	**669.7**	1.45	5.10	1.52	0.10	0.001	18.28	30.34	1.92	**BA**
BE	0.09	0.005	1.09	0.01	0.23	**782.2**	0.12	0.35	0.25	0.00	2.94	171.9	1.73	**BE**
BG	7.30	0.14	2.58	0.21	5.00	1.44	**1297**	3.77	0.05	0.02	10.97	21.42	1.55	**BG**
BY	2.36	0.21	8.82	0.35	6.20	11.75	8.95	**3252**	0.23	0.01	60.27	142.6	35.10	**BY**
CH	0.53	0.005	8.75	0.01	1.25	5.18	0.40	0.27	**21.94**	0.00	3.79	107.8	0.82	**CH**
CY	0.01	0.01	0.01	0.02	0.01	0.01	0.04	0.02	0.00	**5.13**	0.02	0.06	0.005	**CY**
CZ	2.84	0.03	76.89	0.05	8.64	16.74	6.79	5.72	0.97	0.001	**1562**	547.8	9.69	**CZ**
DE	3.37	0.05	106.6	0.12	8.44	396.7	3.22	8.09	14.83	0.002	262.1	**11562**	87.55	**DE**
DK	0.20	0.003	1.10	0.01	0.48	18.59	0.32	1.57	0.08	0.00	5.71	102.4	**697.9**	**DK**
EE	0.60	0.02	2.06	0.03	1.42	5.37	1.69	43.47	0.10	0.00	12.41	57.44	14.88	**EE**
ES	0.49	0.02	1.19	0.06	1.58	6.69	0.53	1.27	0.18	0.00	2.11	26.83	2.79	**ES**
FI	2.90	0.14	8.38	0.28	6.14	24.20	8.04	122.4	0.37	0.001	45.47	220.5	65.95	**FI**
FR	2.29	0.08	11.22	0.19	6.33	143.5	2.58	4.25	8.59	0.002	18.46	526.6	9.20	**FR**
GB	0.16	0.01	2.17	0.04	0.42	38.85	0.33	2.38	0.21	0.00	8.63	106.4	15.20	**GB**
GE	0.04	47.71	0.08	62.06	0.08	0.08	0.31	0.79	0.00	0.05	0.34	0.99	0.18	**GE**
GR	29.90	0.16	1.04	0.22	2.22	0.60	51.41	2.46	0.03	0.08	4.10	8.63	0.73	**GR**
HR	10.34	0.02	17.83	0.04	162.6	1.47	4.60	1.81	0.12	0.002	21.53	34.3	1.48	**HR**
HU	6.61	0.02	45.88	0.04	34.45	4.03	18.09	5.42	0.25	0.01	65.89	79.12	2.99	**HU**
IE	0.03	0.004	0.35	0.01	0.08	2.76	0.06	0.42	0.04	0.00	1.48	14.10	1.58	**IE**
IS	0.09	0.01	0.63	0.02	0.37	2.62	0.13	1.74	0.04	0.00	2.86	17.06	4.38	**IS**
IT	21.03	0.09	46.76	0.20	42.29	7.21	14.04	3.51	3.24	0.01	28.03	131.6	4.37	**IT**
KY	0.00	0.05	0.00	0.15	0.00	0.00	0.00	0.02	0.00	0.00	0.01	0.03	0.01	**KY**
KZ	0.26	0.73	0.51	4.41	0.47	1.32	1.04	18.50	0.02	0.004	2.85	11.66	3.44	**KZ**
LT	0.69	0.02	3.85	0.03	1.79	5.74	2.11	127.6	0.12	0.001	25.49	71.91	16.82	**LT**
LU	0.01	0.001	0.14	0.001	0.02	6.19	0.01	0.03	0.04	0.00	0.34	21.35	0.09	**LU**
LV	0.65	0.02	3.09	0.03	1.62	6.02	1.98	86.46	0.12	0.00	19.36	70.40	16.40	**LV**
MC	0.00	0.00	0.002	0.00	0.001	0.00	0.00	0.00	0.00	0.00	0.002	0.01	0.00	**MC**
MD	0.32	0.03	0.43	0.05	0.49	0.35	2.19	5.66	0.01	0.001	2.19	4.82	0.68	**MD**
ME	34.02	0.01	0.49	0.01	19.52	0.17	2.22	0.19	0.01	0.001	1.53	3.05	0.21	**ME**
MK	37.51	0.02	0.46	0.02	1.52	0.21	37.11	0.27	0.01	0.01	1.69	3.51	0.23	**MK**
MT	0.003	0.00	0.001	0.00	0.002	0.001	0.002	0.00	0.00	0.00	0.002	0.01	0.00	**MT**
NL	0.09	0.002	1.09	0.01	0.25	245.8	0.14	0.31	0.13	0.00	3.84	269.7	2.90	**NL**
NO	1.52	0.04	6.28	0.09	3.26	30.39	3.66	20.10	0.34	0.001	28.56	212.5	110.8	**NO**
PL	4.78	0.07	37.47	0.13	14.14	41.30	12.94	177.5	0.97	0.003	604.6	734.7	68.98	**PL**
PT	0.01	0.00	0.04	0.002	0.03	0.38	0.01	0.06	0.01	0.00	0.11	1.56	0.20	**PT**
RO	8.22	0.12	10.55	0.23	19.63	5.15	63.08	18.16	0.22	0.01	41.48	77.8	5.90	**RO**
RS	33.83	0.04	5.87	0.06	71.94	2.01	70.42	1.91	0.10	0.01	19.20	34.28	2.00	**RS**
RU	9.29	15.81	32.42	64.30	22.16	61.64	37.39	1235	1.23	0.08	187.8	655	157.5	**RU**
RUA	2.38	1.69	6.12	6.54	4.89	12.77	8.69	112.8	0.24	0.01	33.21	121.9	30.47	**RUA**
SE	2.68	0.07	9.95	0.17	5.84	51.07	6.57	54.77	0.46	0.001	53.88	376.8	301.5	**SE**
SI	2.25	0.01	32.86	0.03	14.08	0.87	1.48	1.20	0.08	0.001	10.67	22.13	0.62	**SI**
SK	2.55	0.02	23.70	0.04	9.69	3.80	7.62	5.39	0.18	0.001	169.7	71.47	3.74	**SK**
TJ	0.001	0.01	0.00	0.05	0.001	0.001	0.001	0.01	0.00	0.00	0.00	0.01	0.00	**TJ**
TM	0.01	0.33	0.01	2.38	0.01	0.03	0.03	0.34	0.00	0.001	0.07	0.26	0.06	**TM**
TR	1.86	96.58	1.58	21.03	1.95	1.23	22.63	9.43	0.04	0.61	6.46	15.62	1.62	**TR**
UA	4.36	1.07	11.64	1.96	9.33	12.20	20.31	321.7	0.28	0.02	77.90	154.1	21.35	**UA**
UZ	0.01	0.10	0.02	0.48	0.01	0.04	0.03	0.45	0.00	0.00	0.10	0.37	0.10	**UZ**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CY**	**CZ**	**DE**	**DK**	

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2008, kg/y (continued)*

Receptors ↓ Emitters →

	EE	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KY	
AL	0.05	0.34	0.05	0.57	0.15	0.01	42.08	0.86	1.31	0.05	0.00	70.75	0.001	**AL**
AM	0.02	0.00	0.02	0.01	0.01	3.13	0.09	0.01	0.04	0.002	0.00	0.08	0.002	**AM**
AT	0.57	2.63	0.41	10.69	4.39	0.01	4.89	23.52	61.62	1.00	0.005	192.5	0.005	**AT**
AZ	0.13	0.02	0.09	0.02	0.03	4.77	0.10	0.02	0.08	0.01	0.00	0.25	0.03	**AZ**
BA	0.25	1.44	0.21	1.90	0.85	0.01	10.31	84.29	29.86	0.24	0.002	161.5	0.002	**BA**
BE	0.24	7.04	0.19	36.42	15.11	0.001	0.08	0.23	0.32	2.10	0.01	6.58	0.00	**BE**
BG	0.64	0.26	0.55	0.91	0.73	0.04	57.52	2.90	12.48	0.20	0.002	23.40	0.01	**BG**
BY	33.23	2.28	14.29	4.67	7.67	0.10	4.50	4.99	23.42	1.81	0.02	31.97	0.01	**BY**
CH	0.08	4.69	0.07	30.52	2.49	0.001	0.51	1.60	1.15	0.69	0.002	173.1	0.001	**CH**
CY	0.004	0.001	0.003	0.002	0.003	0.001	0.12	0.01	0.02	0.00	0.00	0.06	0.00	**CY**
CZ	1.01	2.84	0.71	9.66	6.02	0.01	4.24	9.82	47.37	1.22	0.01	50.20	0.004	**CZ**
DE	4.89	38.66	2.84	162.9	75.05	0.01	3.56	7.88	16.53	14.74	0.06	195.9	0.01	**DE**
DK	1.06	3.08	0.74	5.20	15.03	0.001	0.17	0.34	1.35	3.05	0.02	4.65	0.00	**DK**
EE	**860.3**	1.42	34.37	2.44	3.81	0.01	0.99	1.07	3.60	0.89	0.01	9.55	0.001	**EE**
ES	0.32	**4144**	0.43	46.28	6.85	0.004	0.45	1.51	1.95	3.37	0.02	41.87	0.002	**ES**
FI	231.6	7.90	**1529**	10.52	18.85	0.06	4.66	4.59	15.00	4.63	0.07	43.60	0.01	**FI**
FR	1.37	242.8	1.23	**1461**	44.84	0.01	2.18	7.10	7.32	12.07	0.05	365.2	0.01	**FR**
GB	0.97	13.21	1.32	19.27	**819.3**	0.003	0.17	0.45	1.47	81.52	0.10	9.27	0.002	**GB**
GE	0.17	0.02	0.12	0.04	0.06	**58.58**	0.39	0.07	0.25	0.02	0.00	0.39	0.01	**GE**
GR	0.44	0.44	0.40	0.70	0.39	0.05	**976.8**	1.19	3.91	0.11	0.001	32.96	0.01	**GR**
HR	0.25	1.80	0.17	2.27	0.70	0.005	8.85	**373.6**	70.71	0.19	0.001	244.6	0.003	**HR**
HU	0.46	1.66	0.38	3.32	1.79	0.01	10.76	66.77	**1000**	0.42	0.003	89.07	0.005	**HU**
IE	0.23	3.63	0.30	2.32	30.28	0.001	0.04	0.08	0.20	**315.5**	0.04	1.85	0.001	**IE**
IS	0.58	1.30	0.80	1.64	4.25	0.002	0.06	0.31	1.12	1.79	**5.15**	4.17	0.003	**IS**
IT	0.68	29.32	0.57	38.91	3.52	0.02	24.67	47.71	28.01	1.05	0.01	**9057**	0.01	**IT**
KY	0.01	0.00	0.01	0.00	0.00	0.004	0.01	0.00	0.00	0.001	0.00	0.02	**710.2**	**KY**
KZ	6.82	0.40	5.52	0.53	1.14	0.21	0.51	0.33	1.25	0.29	0.01	3.34	191.2	**KZ**
LT	14.07	1.04	5.47	2.30	3.65	0.01	1.17	1.56	6.31	0.83	0.01	11.31	0.002	**LT**
LU	0.02	0.50	0.01	3.79	0.42	0.00	0.01	0.03	0.03	0.08	0.00	0.94	0.00	**LU**
LV	95.72	1.23	12.58	2.49	3.82	0.01	1.10	1.36	5.14	0.84	0.01	10.60	0.002	**LV**
MC	0.00	0.002	0.00	0.02	0.00	0.00	0.00	0.002	0.002	0.00	0.00	0.16	0.00	**MC**
MD	0.57	0.07	0.41	0.20	0.22	0.01	0.56	0.36	1.55	0.05	0.00	3.12	0.004	**MD**
ME	0.03	0.22	0.03	0.28	0.11	0.00	6.23	1.63	1.66	0.03	0.00	42.62	0.00	**ME**
MK	0.04	0.10	0.04	0.21	0.12	0.004	53.92	0.64	1.74	0.03	0.00	14.90	0.00	**MK**
MT	0.00	0.003	0.00	0.002	0.00	0.00	0.01	0.001	0.002	0.00	0.00	0.16	0.00	**MT**
NL	0.26	5.67	0.18	14.45	13.74	0.001	0.07	0.22	0.50	2.06	0.01	4.95	0.001	**NL**
NO	16.06	11.00	34.06	13.34	46.94	0.01	1.63	2.40	9.20	11.25	0.17	27.11	0.01	**NO**
PL	10.49	5.65	6.20	15.95	18.25	0.02	7.26	13.88	70.41	3.92	0.02	72.15	0.01	**PL**
PT	0.02	123	0.04	0.86	0.47	0.00	0.01	0.03	0.04	0.26	0.002	1.15	0.00	**PT**
RO	2.02	0.84	1.62	3.09	2.60	0.03	16.15	12.72	88.87	0.59	0.01	63.38	0.02	**RO**
RS	0.25	0.46	0.23	1.52	1.07	0.01	26.68	24.37	42.09	0.28	0.00	57.96	0.00	**RS**
RU	607.7	18.95	444.7	27.60	47.37	10.16	18.60	18.28	71.06	11.93	0.23	139.3	2.99	**RU**
RUA	56.65	4.75	59.54	5.92	11.45	0.49	4.72	3.72	14.17	3.15	0.12	32.95	15.91	**RUA**
SE	57.42	11.69	167.5	18.40	37.38	0.02	3.34	4.23	16.44	8.75	0.11	41.08	0.01	**SE**
SI	0.14	0.80	0.09	1.20	0.34	0.002	2.33	57.52	20.41	0.09	0.00	160.5	0.003	**SI**
SK	0.50	1.01	0.39	2.38	1.70	0.004	3.82	11.10	124.4	0.36	0.003	32.44	0.003	**SK**
TJ	0.00	0.00	0.00	0.00	0.001	0.002	0.002	0.001	0.002	0.00	0.00	0.01	25.29	**TJ**
TM	0.09	0.01	0.08	0.01	0.02	0.04	0.01	0.01	0.03	0.007	0.00	0.08	1.18	**TM**
TR	2.03	0.25	1.54	0.67	0.71	3.39	30.33	1.30	5.30	0.21	0.002	10.40	0.04	**TR**
UA	16.43	1.65	10.05	5.01	7.21	0.44	8.66	8.73	65.58	1.60	0.02	50.30	0.15	**UA**
UZ	0.13	0.01	0.12	0.02	0.04	0.02	0.01	0.01	0.04	0.01	0.00	0.10	104.7	**UZ**
	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KY**	

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2008, kg/y (continued)*
Receptors ↓ Emitters →

	KZ	LT	LU	LV	MC	MD	ME	MK	MT	NL	NO	PL	PT	
AL	0.08	0.10	0.04	0.17	0.00	0.19	8.93	24.52	0.01	0.09	0.05	5.08	0.02	**AL**
AM	0.15	0.01	0.00	0.03	0.00	0.01	0.00	0.002	0.00	0.00	0.00	0.13	0.00	**AM**
AT	0.16	1.00	1.57	1.48	0.00	0.11	1.27	1.51	0.01	3.53	0.65	94.59	0.13	**AT**
AZ	1.79	0.07	0.00	0.16	0.00	0.03	0.00	0.00	0.00	0.01	0.02	0.49	0.001	**AZ**
BA	0.11	0.41	0.18	0.66	0.00	0.25	17.70	2.67	0.01	0.58	0.28	36.30	0.06	**BA**
BE	0.03	0.18	17.89	0.44	0.00	0.01	0.02	0.02	0.00	26.69	0.35	4.07	0.34	**BE**
BG	0.94	1.49	0.17	2.59	0.00	4.58	1.48	24.97	0.005	0.55	0.37	54.81	0.02	**BG**
BY	1.44	178.1	0.84	173.7	0.00	9.76	0.82	1.65	0.002	5.50	4.11	924.8	0.16	**BY**
CH	0.03	0.07	0.82	0.12	0.00	0.02	0.11	0.14	0.003	0.95	0.13	3.93	0.18	**CH**
CY	0.00	0.00	0.00	0.01	0.00	0.003	0.00	0.01	0.00	0.00	0.00	0.07	0.00	**CY**
CZ	0.17	2.49	1.84	3.26	0.00	0.21	0.83	1.47	0.003	5.23	0.98	553.4	0.15	**CZ**
DE	0.32	5.35	54.97	10.57	0.00	0.23	0.55	0.71	0.01	170.4	5.15	431.2	1.49	**DE**
DK	0.03	0.69	0.46	1.65	0.00	0.05	0.04	0.06	0.00	8.29	2.94	28.06	0.10	**DK**
EE	0.13	29.96	0.48	240.9	0.00	0.81	0.16	0.34	0.00	2.75	2.42	134.6	0.14	**EE**
ES	0.11	0.22	0.37	0.51	0.00	0.04	0.17	0.20	0.001	1.56	0.51	5.89	106	**ES**
FI	1.27	57.17	1.66	207.7	0.00	3.32	0.80	1.71	0.004	10.91	20.19	401.1	0.54	**FI**
FR	0.46	1.27	42.45	2.56	0.01	0.23	0.63	0.83	0.01	17.15	1.88	38.32	6.91	**FR**
GB	0.14	0.98	1.27	1.78	0.00	0.06	0.04	0.07	0.00	13.43	3.76	39.59	1.26	**GB**
GE	0.42	0.18	0.01	0.37	0.00	0.13	0.01	0.02	0.00	0.04	0.03	2.26	0.002	**GE**
GR	0.57	0.67	0.07	1.20	0.00	1.28	0.70	34.02	0.02	0.21	0.17	18.00	0.02	**GR**
HR	0.16	0.41	0.26	0.70	0.00	0.20	3.44	2.05	0.02	0.66	0.25	38.48	0.10	**HR**
HU	0.27	1.29	0.56	1.64	0.00	0.55	3.36	5.75	0.01	1.52	0.50	170.5	0.14	**HU**
IE	0.04	0.16	0.15	0.31	0.00	0.01	0.01	0.02	0.00	0.90	0.43	4.16	0.33	**IE**
IS	0.28	0.80	0.20	1.61	0.00	0.08	0.04	0.04	0.00	1.30	2.04	20.65	0.13	**IS**
IT	0.38	0.71	0.86	1.39	0.003	0.45	3.33	3.98	0.13	1.78	0.59	43.55	0.59	**IT**
KY	118.8	0.01	0.00	0.03	0.00	0.00	0.00	0.001	0.00	0.003	0.004	0.09	0.002	**KY**
KZ	**4203**	6.38	0.13	16.87	0.00	0.97	0.12	0.25	0.00	1.07	1.94	46.11	0.14	**KZ**
LT	0.18	**830.3**	0.57	257.7	0.00	1.64	0.32	0.66	0.001	3.93	3.10	485.2	0.11	**LT**
LU	0.00	0.02	**28.27**	0.04	0.00	0.001	0.00	0.003	0.00	0.58	0.04	0.50	0.07	**LU**
LV	0.23	187.9	0.66	**1946**	0.00	1.65	0.30	0.62	0.001	4.54	4.43	313.6	0.16	**LV**
MC	0.00	0.00	0.00	0.00	**0.00**	0.00	0.00	0.00	0.00	0.00	0.00	0.004	0.00	**MC**
MD	0.31	1.25	0.06	1.94	0.00	**150.1**	0.15	0.55	0.001	0.28	0.20	32.15	0.01	**MD**
ME	0.04	0.08	0.04	0.12	0.00	0.08	**74.34**	3.83	0.01	0.10	0.06	5.68	0.04	**ME**
MK	0.10	0.12	0.05	0.19	0.00	0.21	1.04	**184**	0.01	0.12	0.07	7.33	0.02	**MK**
MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.001	**0.05**	0.00	0.00	0.01	0.00	**MT**
NL	0.04	0.28	2.31	0.76	0.00	0.01	0.02	0.03	0.00	**271.5**	0.84	9.12	0.75	**NL**
NO	1.09	12.14	2.23	31.81	0.00	0.98	0.54	0.94	0.002	17.27	**621.6**	190	1.18	**NO**
PL	0.87	69.50	6.01	56.16	0.00	2.96	3.22	6.31	0.01	33.44	10.14	**13614**	1.15	**PL**
PT	0.03	0.05	0.04	0.13	0.00	0.00	0.00	0.00	0.00	0.25	0.11	0.75	**654.5**	**PT**
RO	1.69	5.54	0.97	8.00	0.00	49.59	5.91	13.64	0.01	2.81	1.43	275.6	0.14	**RO**
RS	0.36	0.77	0.49	1.15	0.00	1.03	33.74	50.95	0.01	1.50	0.49	66.02	0.10	**RS**
RU	272.8	341.4	5.56	998.8	0.00	34.58	3.79	7.87	0.01	36.65	51.48	2288	2.44	**RU**
RUA	1302	45.68	1.26	124.6	0.00	6.53	0.91	1.90	0.003	8.37	16.66	400.2	1.00	**RUA**
SE	1.23	38.25	3.65	105.4	0.00	2.22	1.01	1.93	0.01	34.70	104.9	476.3	1.39	**SE**
SI	0.10	0.24	0.25	0.43	0.00	0.07	1.01	0.81	0.01	0.51	0.16	20.76	0.11	**SI**
SK	0.20	1.61	0.77	2.07	0.00	0.46	1.60	2.98	0.004	2.31	0.69	518.1	0.12	**SK**
TJ	7.95	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.001	0.001	0.03	0.001	**TJ**
TM	12.78	0.08	0.00	0.20	0.00	0.02	0.00	0.00	0.00	0.02	0.03	0.61	0.003	**TM**
TR	2.20	2.96	0.22	6.08	0.00	3.36	0.85	3.47	0.02	0.73	0.59	47.09	0.03	**TR**
UA	11.80	35.79	1.57	53.33	0.00	87.01	2.47	5.55	0.01	8.10	4.85	1320	0.23	**UA**
UZ	106.8	0.12	0.00	0.31	0.00	0.03	0.00	0.01	0.00	0.03	0.05	1.01	0.01	**UZ**
	KZ	**LT**	**LU**	**LV**	**MC**	**MD**	**ME**	**MK**	**MT**	**NL**	**NO**	**PL**	**PT**	

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2008, kg/y (continued)*

Receptors ↓ Emitters →

	RO	RS	RU	RUA	SE	SI	SK	TJ	TM	TR	UA	UZ	Total	
AL	18.65	50.66	0.30	0.01	0.17	0.51	1.20	0.00	0.001	6.68	11.10	0.001	**641.1**	**AL**
AM	0.26	0.02	0.38	0.01	0.01	0.00	0.02	0.001	0.01	15.49	1.82	0.004	**424.8**	**AM**
AT	23.92	20.84	1.12	0.03	0.97	131.8	33.18	0.001	0.002	4.60	11.38	0.002	**1816**	**AT**
AZ	0.85	0.05	5.76	0.50	0.05	0.01	0.05	0.01	0.16	6.71	8.80	0.06	**841.6**	**AZ**
BA	44.29	79.49	0.51	0.02	0.79	9.91	10.11	0.001	0.002	3.75	14.48	0.002	**1269**	**BA**
BE	0.49	0.17	0.18	0.01	0.30	0.37	0.25	0.00	0.00	0.10	0.98	0.00	**1082**	**BE**
BG	868.6	94.87	4.94	0.14	1.40	2.65	12.22	0.004	0.01	89.17	227.0	0.01	**2844**	**BG**
BY	223.8	16.41	48.02	0.68	17.65	6.97	47.09	0.01	0.01	16.85	986.5	0.01	**6323**	**BY**
CH	2.52	0.99	0.11	0.01	0.12	3.66	0.67	0.00	0.00	0.53	1.96	0.00	**382.8**	**CH**
CY	0.18	0.03	0.03	0.00	0.00	0.00	0.01	0.00	0.00	4.76	0.53	0.00	**11.2**	**CY**
CZ	37.31	19.10	1.65	0.05	2.02	16.39	119.8	0.001	0.002	3.74	21.27	0.002	**3162**	**CZ**
DE	20.16	7.62	2.99	0.12	7.35	8.44	15.75	0.002	0.003	2.83	23.93	0.004	**13746**	**DE**
DK	3.94	0.76	0.38	0.02	6.66	0.32	1.68	0.00	0.00	0.37	4.75	0.00	**924.3**	**DK**
EE	26.07	2.97	14.75	0.08	17.54	1.33	5.65	0.00	0.00	1.92	69.85	0.00	**1611**	**EE**
ES	4.39	1.67	0.55	0.03	0.48	2.15	1.30	0.001	0.002	0.34	4.83	0.002	**4422**	**ES**
FI	113.5	13.92	96.72	0.67	193.0	6.48	23.54	0.01	0.01	11.13	350.0	0.01	**3890**	**FI**
FR	21.52	6.54	1.86	0.10	1.98	14.12	5.52	0.003	0.01	2.03	24.14	0.01	**3069**	**FR**
GB	3.36	0.71	0.97	0.10	3.10	0.91	2.62	0.001	0.001	0.25	6.31	0.001	**1203**	**GB**
GE	4.71	0.23	9.77	0.03	0.14	0.06	0.25	0.004	0.02	46.19	20.27	0.01	**258.0**	**GE**
GR	99.16	24.63	2.01	0.07	0.67	0.94	3.59	0.003	0.01	114.0	72.13	0.01	**1493**	**GR**
HR	38.14	57.31	0.65	0.02	0.59	122.7	14.54	0.001	0.002	4.04	14.05	0.002	**1258**	**HR**
HU	338.7	130.1	1.74	0.07	1.35	57.10	243.4	0.002	0.002	10.84	66.75	0.002	**2474**	**HU**
IE	0.56	0.14	0.22	0.02	0.37	0.13	0.29	0.00	0.00	0.04	1.04	0.00	**384.8**	**IE**
IS	4.23	0.68	1.27	0.22	2.01	0.58	1.66	0.001	0.00	0.14	9.86	0.002	**98.9**	**IS**
IT	53.33	26.39	1.39	0.05	1.23	104.8	13.48	0.003	0.01	8.85	33.32	0.01	**9835**	**IT**
KY	0.13	0.01	0.22	0.03	0.01	0.00	0.01	80.88	0.20	0.54	0.76	34.41	**946.7**	**KY**
KZ	30.88	2.37	260.1	104.8	7.18	0.72	3.19	40.10	2.67	14.38	302.5	33.70	**5336**	**KZ**
LT	54.37	6.23	14.17	0.26	15.94	3.45	16.84	0.00	0.001	3.20	85.98	0.001	**2088**	**LT**
LU	0.06	0.02	0.02	0.00	0.03	0.06	0.04	0.00	0.00	0.01	0.13	0.00	**64.0**	**LU**
LV	53.83	5.93	17.04	0.17	27.37	2.95	13.69	0.001	0.001	3.68	121.9	0.002	**3048**	**LV**
MC	0.003	0.001	0.00	0.00	0.00	0.005	0.002	0.00	0.00	0.00	0.003	0.00	**0.2**	**MC**
MD	462.8	3.12	3.17	0.06	0.75	0.90	3.46	0.001	0.003	8.67	249.4	0.004	**943.7**	**MD**
ME	11.13	48.40	0.16	0.01	0.17	0.72	1.44	0.00	0.00	2.87	4.64	0.00	**268.2**	**ME**
MK	24.19	67.98	0.36	0.02	0.20	0.63	1.83	0.00	0.00	11.75	12.95	0.00	**467.5**	**MK**
MT	0.01	0.004	0.00	0.00	0.00	0.001	0.001	0.00	0.00	0.01	0.01	0.00	**0.3**	**MT**
NL	0.78	0.28	0.26	0.02	0.61	0.42	0.52	0.00	0.00	0.13	1.14	0.00	**856.2**	**NL**
NO	53.70	9.59	15.46	0.86	129.3	4.22	14.22	0.007	0.003	4.93	95.94	0.01	**1799**	**NO**
PL	305	62.43	21.42	1.63	33.54	35.85	355.4	0.004	0.01	18.05	312.1	0.01	**16871**	**PL**
PT	0.16	0.03	0.17	0.02	0.10	0.05	0.06	0.00	0.00	0.05	0.56	0.00	**785.4**	**PT**
RO	**12033**	235.9	12.38	0.38	4.96	22.03	68.27	0.01	0.01	92.44	659.1	0.02	**13936**	**RO**
RS	358.2	**1462**	1.59	0.09	1.23	12.28	22.47	0.00	0.00	19.07	52.20	0.00	**2482.1**	**RS**
RU	1006	79.88	**7312**	67.29	255.3	35.47	152.5	1.44	1.18	251.8	8760	1.80	**25867**	**RU**
RUA	214.5	18.34	1009	**2162**	59.34	7.35	31.63	4.35	0.69	46.00	1206	2.88	**7226**	**RUA**
SE	106.4	16.94	31.99	0.88	**1962**	7.04	29.60	0.01	0.01	6.64	194.7	0.01	**4357**	**SE**
SI	13.19	11.19	0.46	0.02	0.31	**598.5**	8.92	0.001	0.001	2.36	6.07	0.001	**995.2**	**SI**
SK	146.8	39.67	1.77	0.06	2.02	21.20	**947.8**	0.001	0.002	6.62	58.37	0.00	**2231**	**SK**
TJ	0.04	0.003	0.07	0.01	0.004	0.001	0.003	**317.1**	0.46	0.17	0.22	13.12	**364.6**	**TJ**
TM	0.59	0.03	2.35	0.34	0.09	0.01	0.05	7.64	**27.96**	0.65	6.81	16.03	**81.4**	**TM**
TR	221.6	17.76	19.52	0.23	2.30	2.59	7.80	0.02	0.06	**7853**	359.0	0.03	**8788**	**TR**
UA	1358	56.42	155.3	1.69	18.19	18.86	139.8	0.06	0.16	106.7	**29030**	0.18	**33227**	**UA**
UZ	0.83	0.05	4.37	0.78	0.15	0.02	0.08	138.6	5.79	0.91	8.92	**106.4**	**482.2**	**UZ**
	RO	**RS**	**RU**	**RUA**	**SE**	**SI**	**SK**	**TJ**	**TM**	**TR**	**UA**	**UZ**	**Total**	

***Table A.3**. Matrix of PCDD/F country-to-country deposition in 2008, g TEQ/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CY	CZ	DE	DK	
AL	**17.22**	0.000	0.03	0.01	0.19	0.01	0.21	0.003	0.01	0.00	0.09	0.02	0.004	**AL**
AM	0.01	**1.17**	0.003	2.67	0.005	0.002	0.01	0.002	0.001	0.00	0.01	0.002	0.001	**AM**
AT	0.09	0.00	**19.8**	0.01	0.32	0.30	0.03	0.02	0.46	0.00	2.63	1.08	0.05	**AT**
AZ	0.02	0.20	0.01	**47.4**	0.02	0.01	0.03	0.01	0.003	0.001	0.03	0.01	0.01	**AZ**
BA	0.58	0.00	0.25	0.01	**25.5**	0.05	0.08	0.01	0.03	0.00	0.60	0.09	0.02	**BA**
BE	0.002	0.00	0.02	0.001	0.004	**27.8**	0.001	0.002	0.03	0.00	0.03	0.98	0.02	**BE**
BG	0.39	0.002	0.12	0.06	0.30	0.05	**30.9**	0.03	0.02	0.001	0.45	0.09	0.02	**BG**
BY	0.06	0.001	0.15	0.04	0.12	0.15	0.11	**22.5**	0.03	0.00	1.44	0.26	0.33	**BY**
CH	0.03	0.00	0.34	0.004	0.05	0.15	0.01	0.002	**7.8**	0.00	0.08	0.31	0.01	**CH**
CY	0.01	0.00	0.002	0.01	0.01	0.001	0.01	0.00	0.001	**0.1**	0.01	0.001	0.00	**CY**
CZ	0.05	0.00	2.15	0.01	0.16	0.30	0.03	0.02	0.14	0.00	**53.7**	1.29	0.10	**CZ**
DE	0.06	0.00	2.75	0.02	0.12	7.57	0.02	0.04	2.73	0.00	3.10	**49.3**	1.12	**DE**
DK	0.004	0.00	0.02	0.002	0.01	0.26	0.003	0.005	0.01	0.00	0.12	0.25	**8.89**	**DK**
EE	0.01	0.00	0.02	0.003	0.02	0.06	0.01	0.09	0.01	0.00	0.16	0.09	0.12	**EE**
ES	0.12	0.001	0.10	0.03	0.17	0.27	0.05	0.01	0.09	0.00	0.15	0.17	0.05	**ES**
FI	0.05	0.001	0.08	0.02	0.08	0.26	0.06	0.19	0.03	0.00	0.58	0.32	0.42	**FI**
FR	0.14	0.001	0.36	0.05	0.26	5.13	0.05	0.02	2.17	0.00	0.47	2.89	0.14	**FR**
GB	0.01	0.00	0.07	0.01	0.02	1.72	0.01	0.01	0.06	0.00	0.22	0.75	0.30	**GB**
GE	0.03	0.21	0.01	4.81	0.03	0.01	0.06	0.01	0.004	0.001	0.05	0.01	0.01	**GE**
GR	1.78	0.003	0.08	0.08	0.26	0.04	2.84	0.02	0.03	0.002	0.26	0.06	0.02	**GR**
HR	0.30	0.00	0.60	0.01	5.29	0.05	0.06	0.01	0.04	0.00	0.76	0.11	0.02	**HR**
HU	0.12	0.00	1.50	0.01	0.90	0.10	0.13	0.03	0.04	0.00	2.78	0.22	0.04	**HU**
IE	0.003	0.00	0.01	0.003	0.01	0.16	0.002	0.002	0.01	0.00	0.03	0.10	0.04	**IE**
IS	0.01	0.00	0.02	0.01	0.01	0.08	0.005	0.01	0.01	0.00	0.08	0.06	0.07	**IS**
IT	1.20	0.001	1.50	0.05	1.67	0.26	0.25	0.02	0.89	0.00	0.94	0.40	0.06	**IT**
KY	0.01	0.01	0.01	0.24	0.01	0.01	0.02	0.004	0.004	0.00	0.03	0.01	0.003	**KY**
KZ	0.12	0.04	0.13	2.38	0.18	0.15	0.22	0.19	0.05	0.002	0.51	0.19	0.15	**KZ**
LT	0.02	0.00	0.06	0.004	0.03	0.09	0.02	0.55	0.02	0.00	0.63	0.16	0.26	**LT**
LU	0.00	0.00	0.004	0.00	0.001	0.22	0.00	0.00	0.01	0.00	0.005	0.13	0.001	**LU**
LV	0.02	0.00	0.05	0.004	0.03	0.09	0.02	0.32	0.01	0.00	0.38	0.14	0.22	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.02	0.001	0.02	0.02	0.03	0.01	0.09	0.03	0.004	0.00	0.12	0.02	0.01	**MD**
ME	0.65	0.00	0.02	0.01	0.82	0.01	0.04	0.002	0.01	0.00	0.08	0.01	0.004	**ME**
MK	1.59	0.00	0.03	0.01	0.11	0.01	0.66	0.003	0.01	0.00	0.09	0.02	0.004	**MK**
MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MT**
NL	0.002	0.00	0.02	0.001	0.00	6.92	0.001	0.002	0.02	0.00	0.05	1.81	0.04	**NL**
NO	0.04	0.001	0.10	0.02	0.07	0.51	0.03	0.06	0.05	0.00	0.48	0.44	0.94	**NO**
PL	0.12	0.00	0.89	0.02	0.35	0.74	0.11	0.72	0.15	0.00	37.78	2.15	1.06	**PL**
PT	0.01	0.00	0.01	0.002	0.01	0.03	0.004	0.00	0.01	0.00	0.01	0.02	0.01	**PT**
RO	0.36	0.003	0.34	0.08	0.81	0.13	1.64	0.10	0.06	0.001	1.93	0.25	0.08	**RO**
RS	1.26	0.00	0.23	0.02	2.65	0.06	0.71	0.01	0.03	0.00	0.86	0.11	0.03	**RS**
RU	0.49	0.12	0.65	7.28	0.73	1.04	1.08	4.38	0.22	0.004	4.10	1.39	1.54	**RU**
RUA	0.24	0.03	0.27	1.45	0.33	0.42	0.38	0.37	0.10	0.002	1.25	0.49	0.46	**RUA**
SE	0.06	0.00	0.14	0.03	0.10	0.69	0.05	0.15	0.05	0.00	0.92	0.75	4.13	**SE**
SI	0.04	0.00	1.09	0.01	0.30	0.02	0.01	0.005	0.02	0.00	0.30	0.06	0.01	**SI**
SK	0.07	0.00	0.74	0.01	0.27	0.08	0.07	0.03	0.03	0.00	13.34	0.18	0.05	**SK**
TJ	0.005	0.002	0.004	0.10	0.005	0.002	0.01	0.001	0.002	0.00	0.01	0.003	0.001	**TJ**
TM	0.02	0.02	0.02	1.59	0.02	0.01	0.04	0.01	0.01	0.001	0.06	0.02	0.01	**TM**
TR	0.57	0.36	0.18	1.81	0.38	0.12	2.39	0.11	0.06	0.03	0.61	0.16	0.07	**TR**
UA	0.23	0.01	0.34	0.37	0.42	0.26	0.85	1.64	0.07	0.001	3.13	0.44	0.30	**UA**
UZ	0.02	0.01	0.02	0.49	0.02	0.01	0.03	0.01	0.01	0.00	0.05	0.02	0.01	**UZ**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CY**	**CZ**	**DE**	**DK**	

***Table A.3**. Matrix of PCDD/F country-to-country deposition in 2008, g TEQ/y (continued)*

Receptors ↓ Emitters →

	EE	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KY	
AL	0.00	0.08	0.001	0.05	0.05	0.02	1.03	0.18	0.09	0.003	0.00	1.90	0.001	**AL**
AM	0.00	0.01	0.00	0.003	0.01	1.48	0.02	0.01	0.01	0.00	0.00	0.03	0.002	**AM**
AT	0.001	0.20	0.003	0.47	0.53	0.01	0.09	1.12	1.39	0.02	0.002	4.21	0.001	**AT**
AZ	0.001	0.02	0.001	0.01	0.03	3.83	0.07	0.02	0.02	0.002	0.00	0.11	0.02	**AZ**
BA	0.001	0.14	0.002	0.11	0.13	0.02	0.24	5.95	0.99	0.01	0.001	2.88	0.001	**BA**
BE	0.00	0.13	0.001	2.23	1.22	0.001	0.003	0.01	0.01	0.03	0.002	0.08	0.00	**BE**
BG	0.002	0.10	0.005	0.09	0.14	0.11	1.82	0.32	0.55	0.01	0.00	1.11	0.003	**BG**
BY	0.06	0.11	0.06	0.15	0.63	0.08	0.11	0.20	0.49	0.03	0.005	0.52	0.002	**BY**
CH	0.00	0.25	0.001	0.78	0.34	0.003	0.03	0.13	0.04	0.02	0.001	5.67	0.00	**CH**
CY	0.00	0.005	0.00	0.003	0.004	0.01	0.09	0.01	0.004	0.00	0.00	0.05	0.00	**CY**
CZ	0.002	0.13	0.005	0.36	0.54	0.01	0.06	0.41	1.14	0.02	0.002	0.75	0.00	**CZ**
DE	0.01	0.85	0.02	9.65	5.85	0.01	0.07	0.24	0.31	0.20	0.01	2.12	0.002	**DE**
DK	0.01	0.06	0.01	0.16	1.14	0.002	0.005	0.01	0.02	0.04	0.004	0.06	0.00	**DK**
EE	**2.00**	0.04	0.18	0.05	0.25	0.004	0.02	0.03	0.04	0.01	0.002	0.10	0.00	**EE**
ES	0.001	**94.8**	0.01	1.42	1.39	0.03	0.12	0.33	0.16	0.13	0.01	2.30	0.002	**ES**
FI	0.33	0.22	**7.89**	0.23	1.20	0.03	0.09	0.11	0.16	0.06	0.01	0.48	0.001	**FI**
FR	0.003	10.0	0.01	**58.9**	6.79	0.04	0.15	0.61	0.28	0.34	0.02	7.49	0.003	**FR**
GB	0.004	0.69	0.02	1.38	**127.5**	0.01	0.02	0.05	0.05	1.65	0.03	0.30	0.001	**GB**
GE	0.001	0.03	0.002	0.02	0.04	**46.3**	0.12	0.03	0.04	0.002	0.00	0.17	0.01	**GE**
GR	0.001	0.20	0.004	0.13	0.15	0.17	**32.2**	0.29	0.24	0.01	0.001	2.79	0.004	**GR**
HR	0.001	0.13	0.002	0.12	0.13	0.01	0.18	**32.8**	2.57	0.01	0.001	3.31	0.001	**HR**
HU	0.001	0.11	0.004	0.14	0.22	0.01	0.13	4.54	**48.7**	0.01	0.001	1.35	0.001	**HU**
IE	0.001	0.24	0.003	0.20	2.70	0.002	0.004	0.01	0.01	**7.96**	0.01	0.08	0.00	**IE**
IS	0.001	0.12	0.01	0.09	0.57	0.01	0.01	0.02	0.03	0.04	**0.89**	0.13	0.001	**IS**
IT	0.002	1.37	0.005	1.42	0.72	0.05	0.81	3.77	0.95	0.04	0.005	**159.1**	0.003	**IT**
KY	0.00	0.03	0.001	0.01	0.02	0.11	0.04	0.02	0.02	0.00	0.00	0.10	**33.7**	**KY**
KZ	0.03	0.31	0.07	0.22	0.69	1.07	0.33	0.23	0.27	0.03	0.008	1.09	7.97	**KZ**
LT	0.03	0.06	0.04	0.08	0.39	0.01	0.02	0.06	0.12	0.02	0.002	0.17	0.00	**LT**
LU	0.00	0.01	0.00	1.28	0.04	0.00	0.00	0.001	0.001	0.001	0.00	0.01	0.00	**LU**
LV	0.21	0.06	0.08	0.08	0.39	0.01	0.03	0.05	0.09	0.02	0.002	0.17	0.00	**LV**
MC	0.00	0.00	0.00	0.001	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.003	0.00	**MC**
MD	0.001	0.01	0.003	0.01	0.04	0.04	0.05	0.03	0.07	0.002	0.00	0.11	0.001	**MD**
ME	0.00	0.04	0.00	0.03	0.03	0.01	0.13	0.17	0.09	0.00	0.00	0.95	0.00	**ME**
MK	0.00	0.04	0.001	0.03	0.03	0.02	1.27	0.10	0.11	0.002	0.00	0.60	0.00	**MK**
MT	0.00	0.001	0.00	0.00	0.00	0.00	0.001	0.00	0.00	0.00	0.00	0.01	0.00	**MT**
NL	0.001	0.10	0.002	0.71	1.75	0.001	0.003	0.01	0.01	0.04	0.002	0.06	0.00	**NL**
NO	0.03	0.45	0.17	0.45	4.12	0.02	0.05	0.10	0.15	0.18	0.04	0.52	0.003	**NO**
PL	0.03	0.31	0.05	0.64	1.86	0.02	0.14	0.72	2.21	0.08	0.01	1.38	0.001	**PL**
PT	0.00	2.25	0.001	0.08	0.18	0.003	0.01	0.02	0.01	0.02	0.003	0.13	0.00	**PT**
RO	0.005	0.19	0.01	0.20	0.38	0.10	0.58	0.91	3.92	0.02	0.002	1.98	0.004	**RO**
RS	0.00	0.11	0.002	0.10	0.15	0.03	0.57	2.09	2.15	0.01	0.00	1.75	0.00	**RS**
RU	1.18	1.23	1.67	1.13	4.85	8.63	1.27	1.01	1.63	0.23	0.06	4.18	0.35	**RU**
RUA	0.08	0.72	0.26	0.52	2.10	0.96	0.57	0.44	0.57	0.11	0.04	2.18	1.30	**RUA**
SE	0.09	0.36	1.08	0.53	3.23	0.03	0.08	0.15	0.26	0.14	0.03	0.61	0.002	**SE**
SI	0.00	0.05	0.001	0.06	0.06	0.004	0.04	3.88	0.54	0.00	0.00	3.10	0.00	**SI**
SK	0.00	0.07	0.004	0.10	0.19	0.01	0.08	0.69	6.66	0.01	0.001	0.64	0.00	**SK**
TJ	0.00	0.011	0.00	0.005	0.01	0.04	0.01	0.01	0.01	0.00	0.00	0.04	1.59	**TJ**
TM	0.00	0.04	0.003	0.02	0.06	0.46	0.08	0.03	0.03	0.00	0.00	0.15	0.22	**TM**
TR	0.01	0.35	0.02	0.25	0.41	3.90	3.97	0.45	0.51	0.02	0.003	2.76	0.02	**TR**
UA	0.03	0.25	0.06	0.29	0.90	0.76	0.67	0.65	2.46	0.04	0.01	1.73	0.02	**UA**
UZ	0.00	0.04	0.003	0.02	0.06	0.22	0.05	0.03	0.03	0.00	0.001	0.13	4.80	**UZ**
	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KY**	

***Table A.3**. Matrix of PCDD/F country-to-country deposition in 2008, g TEQ/y (continued)*

Receptors ↓ Emitters →

	KZ	LT	LU	LV	MC	MD	ME	MK	MT	NL	NO	PL	
AL	0.004	0.001	0.00	0.002	0.004	0.02	0.43	7.96	0.02	0.005	0.002	0.13	**AL**
AM	0.03	0.00	0.00	0.001	0.00	0.004	0.001	0.02	0.001	0.00	0.00	0.01	**AM**
AT	0.01	0.004	0.01	0.01	0.01	0.01	0.03	0.17	0.01	0.08	0.02	1.22	**AT**
AZ	0.28	0.001	0.00	0.002	0.00	0.01	0.003	0.06	0.002	0.002	0.001	0.05	**AZ**
BA	0.01	0.002	0.001	0.004	0.01	0.02	0.90	0.70	0.02	0.01	0.01	0.52	**BA**
BE	0.001	0.001	0.15	0.004	0.00	0.00	0.00	0.004	0.001	0.86	0.01	0.07	**BE**
BG	0.03	0.01	0.002	0.02	0.01	0.26	0.12	8.74	0.02	0.02	0.01	0.80	**BG**
BY	0.04	0.95	0.003	0.88	0.004	0.31	0.03	0.34	0.01	0.08	0.07	10.76	**BY**
CH	0.002	0.001	0.01	0.002	0.03	0.002	0.01	0.05	0.01	0.03	0.01	0.08	**CH**
CY	0.002	0.00	0.00	0.00	0.00	0.002	0.002	0.04	0.001	0.00	0.00	0.01	**CY**
CZ	0.005	0.01	0.01	0.02	0.005	0.01	0.02	0.17	0.005	0.11	0.02	8.35	**CZ**
DE	0.01	0.04	0.35	0.14	0.02	0.01	0.01	0.11	0.01	4.80	0.16	6.71	**DE**
DK	0.002	0.01	0.002	0.03	0.00	0.002	0.001	0.01	0.001	0.17	0.09	0.53	**DK**
EE	0.003	0.10	0.001	1.19	0.001	0.01	0.003	0.04	0.001	0.03	0.04	0.93	**EE**
ES	0.01	0.002	0.005	0.01	0.03	0.01	0.03	0.22	0.03	0.08	0.03	0.24	**ES**
FI	0.02	0.13	0.004	0.64	0.003	0.04	0.01	0.18	0.01	0.14	0.29	2.45	**FI**
FR	0.02	0.01	0.44	0.03	1.09	0.02	0.04	0.27	0.03	0.50	0.08	0.78	**FR**
GB	0.01	0.01	0.01	0.03	0.003	0.004	0.003	0.03	0.002	0.66	0.20	0.58	**GB**
GE	0.06	0.002	0.00	0.004	0.001	0.03	0.01	0.11	0.004	0.003	0.002	0.11	**GE**
GR	0.03	0.005	0.001	0.01	0.01	0.12	0.11	15.03	0.06	0.01	0.01	0.46	**GR**
HR	0.01	0.002	0.002	0.005	0.01	0.01	0.13	0.42	0.01	0.02	0.01	0.60	**HR**
HU	0.01	0.01	0.003	0.01	0.01	0.03	0.08	0.67	0.01	0.03	0.01	2.79	**HU**
IE	0.002	0.001	0.002	0.01	0.001	0.001	0.001	0.01	0.00	0.06	0.03	0.07	**IE**
IS	0.01	0.003	0.001	0.01	0.001	0.003	0.002	0.02	0.001	0.04	0.08	0.24	**IS**
IT	0.02	0.01	0.01	0.01	0.50	0.03	0.27	1.68	0.25	0.06	0.02	0.91	**IT**
KY	6.05	0.001	0.00	0.002	0.001	0.004	0.002	0.04	0.002	0.001	0.001	0.03	**KY**
KZ	**194.0**	0.05	0.003	0.15	0.01	0.14	0.04	0.57	0.02	0.06	0.09	1.39	**KZ**
LT	0.01	**5.75**	0.002	1.50	0.001	0.04	0.01	0.08	0.002	0.05	0.04	5.07	**LT**
LU	0.00	0.00	**0.33**	0.00	0.00	0.00	0.00	0.001	0.00	0.01	0.001	0.01	**LU**
LV	0.01	1.01	0.002	**13.6**	0.001	0.03	0.01	0.07	0.002	0.05	0.06	2.39	**LV**
MC	0.00	0.00	0.00	0.00	**0.005**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.01	0.01	0.00	0.01	0.001	**10.0**	0.01	0.12	0.002	0.01	0.004	0.51	**MD**
ME	0.00	0.00	0.00	0.00	0.00	0.01	**3.88**	0.82	0.01	0.00	0.00	0.10	**ME**
MK	0.004	0.001	0.00	0.002	0.003	0.02	0.09	**73.3**	0.01	0.005	0.002	0.16	**MK**
MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.001	**0.06**	0.00	0.00	0.00	**MT**
NL	0.002	0.003	0.01	0.01	0.001	0.00	0.001	0.01	0.001	**10.1**	0.02	0.15	**NL**
NO	0.04	0.05	0.01	0.16	0.005	0.02	0.01	0.13	0.01	0.29	**12.3**	1.74	**NO**
PL	0.02	0.37	0.02	0.42	0.01	0.09	0.06	0.62	0.02	0.44	0.15	**256**	**PL**
PT	0.00	0.00	0.001	0.001	0.002	0.001	0.002	0.01	0.002	0.01	0.01	0.02	**PT**
RO	0.06	0.02	0.004	0.05	0.01	2.46	0.23	2.58	0.03	0.06	0.03	3.82	**RO**
RS	0.01	0.00	0.00	0.01	0.01	0.05	1.58	13.23	0.02	0.02	0.01	1.03	**RS**
RU	10.04	1.09	0.02	3.55	0.04	1.15	0.16	2.46	0.08	0.55	0.83	18.25	**RU**
RUA	34.18	0.13	0.01	0.43	0.02	0.23	0.07	0.97	0.04	0.20	0.34	3.83	**RUA**
SE	0.03	0.14	0.01	0.52	0.004	0.04	0.02	0.24	0.01	0.40	1.95	4.28	**SE**
SI	0.003	0.001	0.001	0.002	0.01	0.003	0.01	0.07	0.004	0.01	0.003	0.25	**SI**
SK	0.01	0.01	0.002	0.01	0.004	0.02	0.04	0.37	0.01	0.03	0.01	8.35	**SK**
TJ	0.61	0.00	0.00	0.001	0.00	0.002	0.001	0.02	0.001	0.001	0.00	0.02	**TJ**
TM	2.05	0.003	0.00	0.01	0.001	0.02	0.005	0.08	0.004	0.005	0.004	0.11	**TM**
TR	0.25	0.03	0.003	0.05	0.02	0.50	0.14	3.20	0.11	0.04	0.03	1.49	**TR**
UA	0.37	0.17	0.007	0.27	0.02	5.50	0.11	1.58	0.03	0.15	0.10	17.91	**UA**
UZ	7.53	0.003	0.00	0.01	0.001	0.01	0.004	0.08	0.003	0.005	0.01	0.12	**UZ**
	KZ	**LT**	**LU**	**LV**	**MC**	**MD**	**ME**	**MK**	**MT**	**NL**	**NO**	**PL**	

***Table A.3**. Matrix of PCDD/F country-to-country deposition in 2008, g TEQ/y (continued)*

Receptors ↓ Emitters →

	PL	PT	RO	RS	RU	RUA	SE	SI	SK	TJ	TM	TR	UA	UZ	Total	
AL	0.13	0.001	0.26	2.80	0.05	0.001	0.003	0.01	0.05	0.00	0.002	0.61	0.37	0.002	**33.94**	**AL**
AM	0.01	0.00	0.02	0.01	0.12	0.001	0.001	0.001	0.003	0.001	0.02	2.28	0.17	0.01	**8.17**	**AM**
AT	1.22	0.004	0.15	0.43	0.07	0.002	0.02	0.91	1.14	0.00	0.002	0.18	0.32	0.003	**37.59**	**AT**
AZ	0.05	0.00	0.07	0.04	1.24	0.08	0.003	0.002	0.01	0.01	0.21	1.99	0.59	0.13	**56.65**	**AZ**
BA	0.52	0.002	0.46	3.65	0.06	0.001	0.01	0.08	0.27	0.00	0.002	0.37	0.51	0.002	**45.28**	**BA**
BE	0.07	0.002	0.004	0.004	0.01	0.001	0.005	0.002	0.00	0.00	0.00	0.01	0.02	0.00	**33.77**	**BE**
BG	0.80	0.002	7.71	5.12	0.56	0.01	0.02	0.03	0.36	0.003	0.01	5.58	5.05	0.02	**71.19**	**BG**
BY	10.76	0.003	1.21	0.48	3.58	0.05	0.25	0.04	0.76	0.002	0.01	1.25	22.62	0.01	**71.40**	**BY**
CH	0.08	0.004	0.03	0.05	0.02	0.001	0.003	0.03	0.02	0.00	0.001	0.06	0.07	0.001	**16.53**	**CH**
CY	0.01	0.00	0.02	0.02	0.02	0.00	0.00	0.001	0.003	0.00	0.001	1.71	0.06	0.001	**2.19**	**CY**
CZ	8.35	0.003	0.19	0.42	0.10	0.003	0.03	0.09	2.12	0.00	0.001	0.14	0.42	0.002	**73.60**	**CZ**
DE	6.71	0.01	0.13	0.17	0.28	0.01	0.18	0.05	0.30	0.001	0.003	0.18	0.54	0.004	**100.45**	**DE**
DK	0.53	0.001	0.02	0.02	0.05	0.002	0.33	0.002	0.03	0.00	0.00	0.02	0.11	0.00	**12.54**	**DK**
EE	0.93	0.001	0.07	0.05	1.09	0.004	0.27	0.004	0.05	0.00	0.00	0.12	0.61	0.00	**7.93**	**EE**
ES	0.24	0.72	0.17	0.21	0.10	0.004	0.02	0.05	0.08	0.001	0.004	0.28	0.37	0.01	**104.69**	**ES**
FI	2.45	0.01	0.28	0.19	3.87	0.02	1.86	0.02	0.17	0.001	0.004	0.55	2.05	0.01	**25.84**	**FI**
FR	0.78	0.07	0.23	0.31	0.18	0.01	0.05	0.12	0.18	0.001	0.01	0.33	0.63	0.01	**101.71**	**FR**
GB	0.58	0.02	0.03	0.03	0.11	0.01	0.09	0.01	0.04	0.00	0.002	0.07	0.18	0.003	**137.06**	**GB**
GE	0.11	0.001	0.16	0.07	1.69	0.005	0.005	0.003	0.02	0.004	0.04	5.65	1.38	0.03	**61.40**	**GE**
GR	0.46	0.003	1.28	1.87	0.45	0.004	0.01	0.03	0.16	0.002	0.01	11.74	2.95	0.01	**76.08**	**GR**
HR	0.60	0.002	0.38	2.50	0.06	0.001	0.01	0.98	0.42	0.00	0.002	0.26	0.52	0.003	**52.90**	**HR**
HU	2.79	0.002	2.74	4.52	0.15	0.003	0.02	0.37	13.03	0.00	0.002	0.35	3.10	0.003	**89.10**	**HU**
IE	0.07	0.01	0.01	0.01	0.02	0.002	0.01	0.002	0.01	0.00	0.001	0.02	0.04	0.001	**11.92**	**IE**
IS	0.24	0.004	0.03	0.02	0.08	0.01	0.03	0.005	0.03	0.001	0.002	0.05	0.18	0.004	**3.15**	**IS**
IT	0.91	0.02	0.60	1.57	0.18	0.004	0.02	0.80	0.43	0.001	0.01	0.99	1.08	0.01	**184.97**	**IT**
KY	0.03	0.00	0.03	0.02	0.18	0.01	0.002	0.002	0.01	4.29	0.19	0.62	0.20	14.92	**60.98**	**KY**
KZ	1.39	0.01	0.93	0.50	27.10	10.43	0.18	0.03	0.23	2.22	1.56	5.81	9.92	14.48	**286.58**	**KZ**
LT	5.07	0.002	0.20	0.11	1.21	0.02	0.21	0.01	0.17	0.00	0.001	0.16	1.52	0.001	**18.98**	**LT**
LU	0.01	0.00	0.00	0.001	0.001	0.00	0.00	0.00	0.001	0.00	0.00	0.001	0.003	0.00	**2.09**	**LU**
LV	2.39	0.002	0.16	0.09	0.96	0.01	0.34	0.01	0.11	0.00	0.001	0.19	1.33	0.001	**22.87**	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**0.01**	**MC**
MD	0.51	0.00	3.82	0.13	0.30	0.003	0.01	0.01	0.09	0.001	0.003	0.98	9.46	0.004	**26.24**	**MD**
ME	0.10	0.00	0.14	2.18	0.02	0.00	0.00	0.01	0.05	0.00	0.001	0.20	0.15	0.00	**10.69**	**ME**
MK	0.16	0.001	0.34	3.96	0.05	0.001	0.004	0.01	0.07	0.00	0.001	0.71	0.41	0.002	**83.89**	**MK**
MT	0.00	0.00	0.00	0.001	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.001	0.001	0.00	**0.08**	**MT**
NL	0.15	0.002	0.005	0.01	0.02	0.001	0.01	0.002	0.01	0.00	0.00	0.01	0.03	0.00	**22.01**	**NL**
NO	1.74	0.01	0.19	0.18	0.75	0.04	1.08	0.02	0.14	0.002	0.01	0.33	1.03	0.01	**27.60**	**NO**
PL	**256**	0.01	1.28	1.16	1.61	0.15	0.49	0.15	5.39	0.001	0.004	0.65	10.87	0.01	**331.39**	**PL**
PT	0.02	**4.51**	0.02	0.02	0.01	0.001	0.00	0.003	0.01	0.00	0.00	0.02	0.03	0.00	**7.50**	**PT**
RO	3.82	0.01	**104**	8.87	1.07	0.02	0.06	0.11	2.02	0.004	0.02	4.69	20.19	0.02	**164.83**	**RO**
RS	1.03	0.00	3.28	**82.47**	0.16	0.003	0.02	0.07	0.64	0.00	0.00	0.99	1.39	0.005	**117.94**	**RS**
RU	18.25	0.04	5.83	2.48	**659**	4.75	2.38	0.16	1.89	0.29	0.85	24.06	131.12	1.53	**923.14**	**RU**
RUA	3.83	0.03	1.58	0.93	39.23	**182**	0.58	0.06	0.53	0.73	0.58	7.13	13.53	2.63	**305.15**	**RUA**
SE	4.28	0.01	0.34	0.29	1.28	0.03	**21.8**	0.02	0.28	0.001	0.01	0.44	1.94	0.01	**47.76**	**SE**
SI	0.25	0.001	0.09	0.22	0.02	0.001	0.003	**4.69**	0.18	0.00	0.001	0.07	0.16	0.001	**15.40**	**SI**
SK	8.35	0.002	0.83	0.97	0.12	0.003	0.03	0.12	**27.6**	0.00	0.001	0.26	2.30	0.002	**64.42**	**SK**
TJ	0.02	0.00	0.01	0.01	0.08	0.004	0.001	0.001	0.004	**19.3**	0.26	0.31	0.09	6.75	**29.34**	**TJ**
TM	0.11	0.001	0.10	0.06	0.99	0.06	0.01	0.004	0.02	0.91	**13.9**	1.60	0.90	9.86	**33.61**	**TM**
TR	1.49	0.01	3.64	1.71	4.89	0.03	0.06	0.06	0.41	0.02	0.11	**669**	19.06	0.11	**724.13**	**TR**
UA	17.91	0.008	11.10	2.07	15.41	0.09	0.25	0.11	3.40	0.02	0.07	12.65	**657**	0.10	**744.77**	**UA**
UZ	0.12	0.001	0.09	0.05	0.95	0.09	0.01	0.00	0.02	8.58	2.59	1.13	0.88	**52.1**	**80.39**	**UZ**
	PL	**PT**	**RO**	**RS**	**RU**	**RUA**	**SE**	**SI**	**SK**	**TJ**	**TM**	**TR**	**UA**	**UZ**	**Total**	

***Table A.4**. Matrix of PCB-153 country-to-country deposition in 2008, g/y*

Receptors ↓ Emitters →

	AL	AM	AT	BA	BE	BG	BY	CH	CY	CZ	DE	DK	
AL	**0.161**	0.001	0.012	0.005	0.020	0.017	0.003	0.007	0.000	0.030	0.069	0.001	**AL**
AM	0.000	**1.534**	0.002	0.000	0.004	0.001	0.002	0.001	0.001	0.005	0.015	0.000	**AM**
AT	0.001	0.001	**6.838**	0.011	0.358	0.003	0.014	0.264	0.000	1.249	1.873	0.008	**AT**
BA	0.007	0.001	0.084	**0.749**	0.055	0.006	0.007	0.018	0.000	0.184	0.253	0.003	**BA**
BE	0.000	0.000	0.009	0.000	**27.56**	0.000	0.002	0.023	0.000	0.024	3.877	0.003	**BE**
BG	0.005	0.005	0.043	0.011	0.069	**2.125**	0.024	0.016	0.001	0.146	0.292	0.004	**BG**
BY	0.001	0.003	0.057	0.006	0.212	0.010	**12.10**	0.021	0.000	0.386	0.976	0.046	**BY**
CH	0.000	0.000	0.149	0.001	0.172	0.001	0.002	**4.155**	0.000	0.042	0.564	0.002	**CH**
CY	0.000	0.001	0.001	0.000	0.001	0.001	0.000	0.000	**0.091**	0.002	0.005	0.000	**CY**
CZ	0.001	0.001	0.841	0.006	0.397	0.003	0.019	0.091	0.000	**17.87**	3.188	0.015	**CZ**
DE	0.001	0.001	0.871	0.004	9.675	0.003	0.031	1.514	0.000	1.827	**155.1**	0.160	**DE**
DK	0.000	0.000	0.006	0.000	0.291	0.000	0.002	0.005	0.000	0.036	0.720	**0.896**	**DK**
EE	0.000	0.000	0.007	0.000	0.069	0.001	0.041	0.004	0.000	0.041	0.287	0.015	**EE**
ES	0.002	0.002	0.046	0.006	0.377	0.005	0.007	0.062	0.000	0.077	0.762	0.010	**ES**
FI	0.000	0.001	0.025	0.002	0.290	0.003	0.095	0.016	0.000	0.140	1.001	0.051	**FI**
FR	0.002	0.003	0.157	0.008	5.834	0.006	0.019	1.451	0.000	0.252	11.85	0.027	**FR**
GB	0.000	0.001	0.025	0.001	1.834	0.001	0.008	0.032	0.000	0.084	2.907	0.043	**GB**
GE	0.000	0.374	0.006	0.001	0.016	0.006	0.010	0.003	0.002	0.018	0.058	0.001	**GE**
GR	0.038	0.006	0.032	0.008	0.059	0.234	0.018	0.019	0.003	0.085	0.210	0.003	**GR**
HR	0.003	0.001	0.182	0.136	0.061	0.005	0.008	0.021	0.000	0.234	0.287	0.003	**HR**
HU	0.002	0.001	0.500	0.032	0.118	0.010	0.022	0.026	0.000	0.765	0.619	0.006	**HU**
IE	0.000	0.000	0.005	0.000	0.189	0.000	0.002	0.008	0.000	0.015	0.375	0.006	**IE**
IS	0.000	0.000	0.004	0.000	0.067	0.000	0.003	0.004	0.000	0.015	0.173	0.007	**IS**
IT	0.014	0.003	0.552	0.047	0.372	0.027	0.021	0.535	0.000	0.398	1.313	0.010	**IT**
KY	0.000	0.012	0.006	0.001	0.012	0.002	0.004	0.004	0.001	0.014	0.047	0.001	**KY**
KZ	0.002	0.088	0.074	0.009	0.303	0.023	0.171	0.047	0.005	0.228	1.058	0.031	**KZ**
LT	0.000	0.000	0.022	0.002	0.112	0.001	0.160	0.010	0.000	0.159	0.528	0.032	**LT**
LU	0.000	0.000	0.001	0.000	0.332	0.000	0.000	0.004	0.000	0.003	0.552	0.000	**LU**
LV	0.000	0.000	0.014	0.001	0.110	0.001	0.116	0.008	0.000	0.095	0.490	0.028	**LV**
MD	0.000	0.001	0.007	0.001	0.017	0.008	0.023	0.002	0.000	0.034	0.079	0.002	**MD**
ME	0.015	0.000	0.010	0.016	0.013	0.004	0.002	0.004	0.000	0.026	0.050	0.001	**ME**
MK	0.027	0.001	0.011	0.004	0.016	0.044	0.003	0.005	0.000	0.032	0.061	0.001	**MK**
MT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**MT**
NL	0.000	0.000	0.009	0.000	6.743	0.000	0.002	0.012	0.000	0.034	7.377	0.007	**NL**
NO	0.000	0.001	0.026	0.002	0.456	0.002	0.031	0.020	0.000	0.112	1.206	0.099	**NO**
PL	0.002	0.001	0.339	0.017	1.006	0.009	0.242	0.097	0.001	8.113	7.839	0.148	**PL**
PT	0.000	0.000	0.004	0.000	0.046	0.000	0.001	0.005	0.000	0.008	0.103	0.002	**PT**
RO	0.005	0.005	0.122	0.037	0.172	0.119	0.078	0.036	0.002	0.541	0.818	0.012	**RO**
RS	0.012	0.001	0.074	0.094	0.070	0.049	0.012	0.017	0.001	0.246	0.324	0.004	**RS**
RU	0.006	0.207	0.229	0.033	1.266	0.080	2.692	0.124	0.012	1.078	4.795	0.199	**RU**
RUA	0.002	0.049	0.088	0.012	0.500	0.023	0.206	0.055	0.004	0.307	1.640	0.060	**RUA**
SE	0.001	0.001	0.043	0.003	0.774	0.003	0.073	0.026	0.000	0.228	2.430	0.440	**SE**
SI	0.000	0.000	0.384	0.010	0.035	0.001	0.004	0.015	0.000	0.119	0.160	0.001	**SI**
SK	0.001	0.000	0.276	0.012	0.098	0.005	0.021	0.018	0.000	2.830	0.570	0.007	**SK**
TJ	0.000	0.006	0.003	0.000	0.005	0.001	0.002	0.002	0.000	0.006	0.019	0.000	**TJ**
TR	0.007	0.665	0.073	0.016	0.167	0.187	0.094	0.042	0.067	0.205	0.615	0.011	**TR**
TU	0.000	0.041	0.011	0.001	0.034	0.004	0.015	0.007	0.002	0.031	0.126	0.003	**TU**
UA	0.003	0.025	0.134	0.027	0.360	0.070	1.015	0.046	0.004	0.820	1.693	0.047	**UA**
UZ	0.000	0.022	0.011	0.001	0.035	0.003	0.016	0.007	0.001	0.030	0.127	0.003	**UZ**
	AL	**AM**	**AT**	**BA**	**BE**	**BG**	**BY**	**CH**	**CY**	**CZ**	**DE**	**DK**	

***Table A.4**. Matrix of PCB-153 country-to-country deposition in 2008, g/y (continued)*

Receptors ↓ Emitters →

	EE	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	
AL	0.000	0.131	0.001	0.214	0.026	0.002	0.049	0.003	0.006	0.001	0.000	0.700	**AL**
AM	0.000	0.017	0.000	0.023	0.006	0.240	0.002	0.000	0.001	0.000	0.000	0.031	**AM**
AT	0.001	0.324	0.003	1.566	0.280	0.001	0.006	0.016	0.091	0.007	0.001	2.394	**AT**
BA	0.001	0.227	0.001	0.425	0.064	0.002	0.012	0.073	0.048	0.002	0.000	1.410	**BA**
BE	0.001	0.163	0.001	6.249	0.830	0.000	0.000	0.000	0.001	0.012	0.001	0.073	**BE**
BG	0.002	0.189	0.004	0.372	0.081	0.013	0.113	0.005	0.033	0.003	0.000	0.627	**BG**
BY	0.050	0.174	0.054	0.573	0.340	0.011	0.007	0.004	0.028	0.010	0.001	0.328	**BY**
CH	0.000	0.357	0.001	3.913	0.188	0.001	0.002	0.002	0.003	0.005	0.000	3.055	**CH**
CY	0.000	0.008	0.000	0.013	0.002	0.001	0.003	0.000	0.000	0.000	0.000	0.022	**CY**
CZ	0.002	0.204	0.004	1.204	0.319	0.001	0.004	0.006	0.068	0.008	0.001	0.487	**CZ**
DE	0.016	1.173	0.024	20.05	3.549	0.003	0.005	0.004	0.023	0.076	0.004	1.446	**DE**
DK	0.005	0.050	0.008	0.493	0.469	0.000	0.000	0.000	0.001	0.012	0.001	0.033	**DK**
EE	**1.548**	0.041	0.146	0.167	0.115	0.001	0.001	0.000	0.002	0.003	0.000	0.046	**EE**
ES	0.002	**118.6**	0.005	6.562	0.774	0.005	0.008	0.005	0.011	0.041	0.003	1.806	**ES**
FI	0.201	0.174	**4.831**	0.701	0.517	0.003	0.003	0.001	0.007	0.016	0.003	0.185	**FI**
FR	0.005	9.187	0.011	**227.1**	4.226	0.006	0.010	0.009	0.020	0.125	0.006	5.395	**FR**
GB	0.005	0.519	0.012	3.963	**52.91**	0.001	0.001	0.001	0.003	0.429	0.006	0.182	**GB**
GE	0.001	0.059	0.001	0.088	0.026	**4.049**	0.007	0.000	0.002	0.001	0.000	0.107	**GE**
GR	0.002	0.328	0.003	0.538	0.083	0.018	**1.205**	0.004	0.015	0.003	0.000	1.215	**GR**
HR	0.001	0.209	0.001	0.434	0.064	0.001	0.008	**0.323**	0.122	0.002	0.000	1.700	**HR**
HU	0.002	0.178	0.003	0.484	0.117	0.002	0.008	0.055	**2.030**	0.003	0.000	0.747	**HU**
IE	0.001	0.168	0.002	0.782	0.928	0.000	0.000	0.000	0.001	**1.662**	0.002	0.049	**IE**
IS	0.001	0.079	0.004	0.249	0.203	0.001	0.000	0.000	0.001	0.010	**0.195**	0.049	**IS**
IT	0.002	2.817	0.005	6.263	0.456	0.008	0.053	0.050	0.064	0.016	0.001	**89.12**	**IT**
KY	0.001	0.070	0.001	0.085	0.017	0.017	0.002	0.000	0.002	0.001	0.000	0.080	**KY**
KZ	0.033	0.663	0.066	1.264	0.528	0.163	0.021	0.005	0.023	0.018	0.003	0.891	**KZ**
LT	0.028	0.064	0.031	0.276	0.187	0.001	0.001	0.001	0.006	0.006	0.001	0.090	**LT**
LU	0.000	0.016	0.000	0.443	0.030	0.000	0.000	0.000	0.000	0.001	0.000	0.010	**LU**
LV	0.181	0.063	0.061	0.265	0.185	0.001	0.001	0.001	0.004	0.005	0.001	0.077	**LV**
MD	0.001	0.025	0.002	0.061	0.025	0.004	0.003	0.001	0.004	0.001	0.000	0.065	**MD**
ME	0.000	0.070	0.000	0.118	0.016	0.001	0.006	0.003	0.006	0.001	0.000	0.397	**ME**
MK	0.000	0.073	0.001	0.127	0.019	0.002	0.062	0.002	0.007	0.001	0.000	0.300	**MK**
MT	0.000	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.003	**MT**
NL	0.001	0.102	0.002	2.112	0.856	0.000	0.000	0.000	0.001	0.014	0.001	0.051	**NL**
NO	0.020	0.266	0.086	1.138	1.345	0.002	0.002	0.001	0.006	0.042	0.007	0.191	**NO**
PL	0.029	0.435	0.049	2.205	1.083	0.004	0.008	0.014	0.120	0.030	0.002	0.819	**PL**
PT	0.000	5.548	0.001	0.437	0.110	0.000	0.001	0.000	0.001	0.007	0.001	0.111	**PT**
RO	0.006	0.311	0.011	0.727	0.206	0.013	0.032	0.017	0.216	0.006	0.001	1.095	**RO**
RS	0.001	0.193	0.002	0.390	0.078	0.004	0.030	0.033	0.116	0.002	0.000	0.871	**RS**
RU	0.726	1.378	1.042	3.927	2.314	1.135	0.062	0.020	0.085	0.073	0.014	2.100	**RU**
RUA	0.065	0.743	0.167	1.771	0.971	0.102	0.022	0.007	0.026	0.034	0.010	1.003	**RUA**
SE	0.067	0.277	0.460	1.639	1.391	0.003	0.003	0.002	0.011	0.040	0.006	0.244	**SE**
SI	0.000	0.091	0.001	0.227	0.033	0.000	0.002	0.052	0.032	0.001	0.000	1.364	**SI**
SK	0.002	0.104	0.003	0.338	0.103	0.001	0.004	0.011	0.311	0.003	0.000	0.334	**SK**
TJ	0.000	0.033	0.000	0.038	0.007	0.008	0.001	0.000	0.001	0.000	0.000	0.034	**TJ**
TR	0.008	0.693	0.014	1.104	0.251	0.623	0.186	0.008	0.032	0.009	0.001	1.557	**TR**
TU	0.002	0.117	0.004	0.172	0.055	0.062	0.005	0.001	0.003	0.002	0.000	0.142	**TU**
UA	0.033	0.406	0.051	1.163	0.530	0.093	0.037	0.016	0.142	0.016	0.002	1.040	**UA**
UZ	0.003	0.110	0.005	0.167	0.057	0.036	0.004	0.001	0.003	0.002	0.000	0.129	**UZ**
	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	

***Table A.4**. Matrix of PCB-153 country-to-country deposition in 2008, g/y (continued)*

Receptors ↓ Emitters →

	KY	KZ	LT	LU	LV	MD	ME	MK	MT	NL	NO	PL	
AL	0.000	0.000	0.001	0.000	0.001	0.002	0.010	0.037	0.002	0.002	0.000	0.008	**AL**
AM	0.000	0.003	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.001	**AM**
AT	0.000	0.001	0.005	0.005	0.003	0.001	0.001	0.001	0.001	0.024	0.002	0.072	**AT**
BA	0.000	0.001	0.002	0.001	0.002	0.003	0.017	0.005	0.002	0.006	0.001	0.033	**BA**
BE	0.000	0.000	0.001	0.062	0.001	0.000	0.000	0.000	0.000	0.237	0.001	0.005	**BE**
BG	0.000	0.003	0.007	0.001	0.005	0.032	0.002	0.033	0.002	0.007	0.001	0.048	**BG**
BY	0.000	0.005	0.624	0.002	0.210	0.042	0.001	0.002	0.001	0.029	0.008	0.507	**BY**
CH	0.000	0.000	0.001	0.003	0.001	0.000	0.000	0.000	0.000	0.009	0.001	0.006	**CH**
CY	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	**CY**
CZ	0.000	0.001	0.011	0.005	0.006	0.002	0.000	0.001	0.001	0.034	0.002	0.468	**CZ**
DE	0.000	0.002	0.043	0.242	0.040	0.002	0.000	0.001	0.001	1.046	0.014	0.363	**DE**
DK	0.000	0.000	0.006	0.001	0.008	0.000	0.000	0.000	0.000	0.051	0.007	0.030	**DK**
EE	0.000	0.000	0.066	0.001	0.244	0.002	0.000	0.000	0.000	0.011	0.005	0.045	**EE**
ES	0.000	0.001	0.003	0.003	0.003	0.002	0.001	0.001	0.003	0.028	0.003	0.018	**ES**
FI	0.000	0.002	0.083	0.002	0.132	0.005	0.000	0.001	0.000	0.042	0.026	0.106	**FI**
FR	0.000	0.002	0.010	0.153	0.009	0.003	0.001	0.002	0.003	0.171	0.008	0.056	**FR**
GB	0.000	0.001	0.010	0.007	0.010	0.001	0.000	0.000	0.000	0.209	0.013	0.035	**GB**
GE	0.000	0.007	0.002	0.000	0.001	0.004	0.000	0.001	0.000	0.001	0.000	0.007	**GE**
GR	0.000	0.003	0.004	0.001	0.003	0.015	0.002	0.056	0.005	0.005	0.001	0.027	**GR**
HR	0.000	0.001	0.002	0.001	0.001	0.002	0.003	0.003	0.002	0.006	0.001	0.034	**HR**
HU	0.000	0.001	0.006	0.002	0.004	0.005	0.002	0.004	0.001	0.012	0.002	0.143	**HU**
IE	0.000	0.000	0.002	0.001	0.002	0.000	0.000	0.000	0.000	0.020	0.002	0.005	**IE**
IS	0.000	0.001	0.002	0.000	0.002	0.000	0.000	0.000	0.000	0.011	0.005	0.009	**IS**
IT	0.000	0.002	0.006	0.005	0.005	0.006	0.006	0.009	0.020	0.024	0.003	0.058	**IT**
KY	**1.816**	0.473	0.001	0.000	0.001	0.001	0.000	0.000	0.000	0.001	0.000	0.004	**KY**
KZ	0.556	**11.50**	0.054	0.003	0.054	0.025	0.001	0.004	0.002	0.036	0.015	0.113	**KZ**
LT	0.000	0.001	**3.522**	0.001	0.342	0.005	0.000	0.000	0.000	0.019	0.005	0.239	**LT**
LU	0.000	0.000	0.000	**0.127**	0.000	0.000	0.000	0.000	0.000	0.004	0.000	0.001	**LU**
LV	0.000	0.001	0.758	0.001	**2.739**	0.004	0.000	0.000	0.000	0.019	0.006	0.124	**LV**
MD	0.000	0.001	0.006	0.000	0.003	**0.926**	0.000	0.001	0.000	0.003	0.001	0.028	**MD**
ME	0.000	0.000	0.001	0.000	0.000	0.001	**0.074**	0.005	0.001	0.001	0.000	0.006	**ME**
MK	0.000	0.000	0.001	0.000	0.001	0.002	0.002	**0.283**	0.001	0.002	0.000	0.009	**MK**
MT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**0.002**	0.000	0.000	0.000	**MT**
NL	0.000	0.000	0.003	0.006	0.003	0.000	0.000	0.000	0.000	**2.177**	0.002	0.010	**NL**
NO	0.000	0.003	0.032	0.003	0.034	0.002	0.000	0.001	0.000	0.074	**1.109**	0.079	**NO**
PL	0.000	0.003	0.266	0.012	0.112	0.014	0.002	0.004	0.002	0.144	0.016	**11.49**	**PL**
PT	0.000	0.000	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.005	0.001	0.002	**PT**
RO	0.000	0.006	0.022	0.003	0.015	0.314	0.005	0.016	0.004	0.021	0.003	0.213	**RO**
RS	0.000	0.001	0.004	0.001	0.003	0.008	0.029	0.080	0.003	0.009	0.001	0.059	**RS**
RU	0.032	0.818	0.803	0.015	0.860	0.155	0.005	0.015	0.009	0.216	0.090	0.967	**RU**
RUA	0.116	2.592	0.114	0.006	0.124	0.030	0.002	0.005	0.004	0.087	0.039	0.225	**RUA**
SE	0.000	0.003	0.096	0.005	0.111	0.005	0.000	0.001	0.001	0.138	0.187	0.205	**SE**
SI	0.000	0.000	0.001	0.001	0.001	0.001	0.000	0.001	0.001	0.003	0.000	0.014	**SI**
SK	0.000	0.001	0.007	0.002	0.004	0.003	0.001	0.002	0.001	0.012	0.002	0.408	**SK**
TJ	0.101	0.057	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002	**TJ**
TR	0.001	0.025	0.024	0.002	0.016	0.057	0.003	0.016	0.010	0.018	0.004	0.088	**TR**
TU	0.019	0.176	0.004	0.000	0.003	0.003	0.000	0.001	0.000	0.003	0.001	0.011	**TU**
UA	0.002	0.041	0.144	0.005	0.084	0.610	0.003	0.011	0.005	0.059	0.012	0.894	**UA**
UZ	0.283	0.542	0.004	0.000	0.004	0.003	0.000	0.001	0.000	0.004	0.001	0.012	**UZ**
	KY	**KZ**	**LT**	**LU**	**LV**	**MD**	**ME**	**MK**	**MT**	**NL**	**NO**	**PL**	

***Table A.4**. Matrix of PCB-153 country-to-country deposition in 2008, g/y (continued)*

Receptors ↓ Emitters →

	PT	RO	RS	RU	RUA	SE	SI	SK	TJ	TR	UA	UZ	Total	
AL	0.001	0.017	0.076	0.021	0.000	0.002	0.001	0.009	0.000	0.010	0.045	0.000	**1.702**	**AL**
AM	0.000	0.002	0.001	0.056	0.001	0.001	0.000	0.001	0.000	0.032	0.025	0.003	**2.017**	**AM**
AT	0.002	0.011	0.020	0.036	0.001	0.009	0.045	0.236	0.000	0.003	0.049	0.000	**15.837**	**AT**
BA	0.001	0.032	0.146	0.032	0.001	0.006	0.006	0.057	0.000	0.008	0.072	0.000	**4.064**	**BA**
BE	0.001	0.000	0.000	0.006	0.000	0.003	0.000	0.001	0.000	0.000	0.004	0.000	**39.158**	**BE**
BG	0.001	0.403	0.182	0.210	0.003	0.010	0.002	0.055	0.000	0.085	0.570	0.002	**5.850**	**BG**
BY	0.002	0.076	0.021	1.203	0.008	0.117	0.002	0.098	0.000	0.021	2.499	0.002	**20.875**	**BY**
CH	0.001	0.002	0.002	0.008	0.000	0.002	0.002	0.005	0.000	0.001	0.010	0.000	**12.670**	**CH**
CY	0.000	0.001	0.001	0.007	0.000	0.000	0.000	0.001	0.000	0.019	0.007	0.000	**0.189**	**CY**
CZ	0.001	0.013	0.020	0.042	0.001	0.016	0.006	0.454	0.000	0.003	0.058	0.000	**25.881**	**CZ**
DE	0.007	0.010	0.008	0.129	0.005	0.100	0.003	0.063	0.000	0.003	0.079	0.001	**197.72**	**DE**
DK	0.000	0.001	0.001	0.020	0.001	0.098	0.000	0.005	0.000	0.000	0.012	0.000	**3.275**	**DK**
EE	0.000	0.004	0.002	0.279	0.001	0.111	0.000	0.006	0.000	0.001	0.058	0.000	**3.372**	**EE**
ES	0.347	0.011	0.009	0.046	0.002	0.014	0.003	0.016	0.000	0.005	0.050	0.001	**129.79**	**ES**
FI	0.002	0.013	0.006	1.041	0.009	0.525	0.001	0.021	0.000	0.006	0.180	0.001	**10.469**	**FI**
FR	0.028	0.016	0.014	0.081	0.005	0.031	0.006	0.033	0.000	0.006	0.087	0.001	**266.47**	**FR**
GB	0.006	0.002	0.002	0.052	0.004	0.048	0.000	0.008	0.000	0.001	0.025	0.000	**63.400**	**GB**
GE	0.000	0.010	0.003	0.583	0.002	0.003	0.000	0.004	0.000	0.084	0.161	0.006	**5.726**	**GE**
GR	0.001	0.074	0.057	0.161	0.002	0.007	0.001	0.025	0.000	0.145	0.333	0.002	**5.061**	**GR**
HR	0.001	0.023	0.083	0.027	0.001	0.005	0.061	0.075	0.000	0.006	0.064	0.000	**4.207**	**HR**
HU	0.001	0.186	0.164	0.061	0.001	0.012	0.023	0.960	0.000	0.007	0.286	0.000	**7.614**	**HU**
IE	0.003	0.001	0.000	0.011	0.001	0.008	0.000	0.001	0.000	0.000	0.005	0.000	**4.260**	**IE**
IS	0.001	0.001	0.001	0.025	0.005	0.015	0.000	0.003	0.000	0.001	0.014	0.001	**1.162**	**IS**
IT	0.008	0.042	0.065	0.086	0.002	0.014	0.036	0.082	0.000	0.018	0.149	0.001	**102.80**	**IT**
KY	0.000	0.004	0.002	0.093	0.010	0.002	0.000	0.003	0.360	0.014	0.037	2.223	**5.457**	**KY**
KZ	0.006	0.074	0.029	6.297	2.913	0.117	0.003	0.051	0.200	0.119	1.210	2.116	**31.418**	**KZ**
LT	0.001	0.012	0.005	0.320	0.002	0.102	0.001	0.028	0.000	0.003	0.174	0.000	**6.501**	**LT**
LU	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**1.528**	**LU**
LV	0.001	0.009	0.004	0.316	0.002	0.146	0.001	0.018	0.000	0.003	0.139	0.000	**6.001**	**LV**
MD	0.000	0.197	0.007	0.111	0.001	0.006	0.000	0.012	0.000	0.017	1.120	0.001	**2.810**	**MD**
ME	0.000	0.010	0.071	0.010	0.000	0.001	0.000	0.009	0.000	0.004	0.021	0.000	**0.975**	**ME**
MK	0.000	0.020	0.100	0.021	0.000	0.002	0.001	0.012	0.000	0.011	0.048	0.000	**1.314**	**MK**
MT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**0.008**	**MT**
NL	0.002	0.000	0.000	0.012	0.001	0.007	0.000	0.002	0.000	0.000	0.005	0.000	**19.556**	**NL**
NO	0.003	0.009	0.006	0.229	0.013	0.489	0.001	0.019	0.000	0.004	0.096	0.001	**7.268**	**NO**
PL	0.005	0.083	0.056	0.726	0.008	0.223	0.010	0.904	0.000	0.015	1.399	0.001	**38.109**	**PL**
PT	**1.566**	0.001	0.001	0.007	0.000	0.003	0.000	0.001	0.000	0.001	0.005	0.000	**7.982**	**PT**
RO	0.003	**4.815**	0.399	0.402	0.006	0.031	0.006	0.241	0.000	0.081	2.192	0.003	**13.393**	**RO**
RS	0.002	0.187	**2.681**	0.066	0.001	0.009	0.004	0.106	0.000	0.019	0.174	0.001	**6.073**	**RS**
RU	0.019	0.375	0.117	**160.5**	1.420	1.043	0.011	0.286	0.025	0.469	12.06	0.269	**204.28**	**RU**
RUA	0.013	0.094	0.038	9.059	**43.90**	0.293	0.004	0.084	0.063	0.114	1.312	0.418	**66.680**	**RUA**
SE	0.004	0.017	0.010	0.393	0.013	**7.441**	0.001	0.039	0.000	0.006	0.192	0.001	**17.034**	**SE**
SI	0.001	0.006	0.010	0.011	0.000	0.002	**0.223**	0.034	0.000	0.002	0.021	0.000	**2.867**	**SI**
SK	0.001	0.055	0.047	0.053	0.001	0.013	0.008	**3.822**	0.000	0.006	0.252	0.000	**9.752**	**SK**
TJ	0.000	0.001	0.001	0.037	0.003	0.001	0.000	0.001	**1.260**	0.007	0.015	0.939	**2.628**	**TJ**
TR	0.005	0.203	0.067	1.775	0.015	0.032	0.004	0.063	0.001	**7.052**	1.911	0.019	**18.064**	**TR**
UA	0.004	0.691	0.105	5.263	0.038	0.134	0.007	0.413	0.001	0.217	**51.26**	0.019	**67.81**	**UA**
UZ	0.001	0.009	0.004	0.410	0.047	0.009	0.000	0.007	0.614	0.025	0.135	**6.296**	**9.456**	**UZ**
	PT	**RO**	**RS**	**RU**	**RUA**	**SE**	**SI**	**SK**	**TJ**	**TR**	**UA**	**UZ**	**Total**	