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Persistent Organic Pollutants in the Environment

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EXECUTIVE SUMMARY

In accordance with the EMEP Work-plan for 2006 (EB.AIR/GE.1/2005/10/Rev.1) Meteorological Synthesizing Centre East (MSC-E) and Chemical Coordinating Centre (CCC) continued the investigations of the environmental pollution by polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), γ-hexachlorocyclohexane (γ-HCH or lindane) and hexachlorobenzene (HCB). The outcome of the studies is summarized in this Status Report.

At present measurements of POP concentrations in the atmosphere and precipitation are performed by 15 EMEP monitoring sites. Most of them are located in northern and central Europe, and there are no sites in southern and eastern parts of Europe. However there are some positive tendencies; i.e. Spain, Latvia and the United Kingdom have started to measure POPs. In addition, in 2006 a passive sampler campaign will cover the whole Europe as well as central Asia to better localize possible hotspots of various POPs in Europe.

The official data on POP emissions (PAHs, PCDD/Fs, HCB, PCBs and HCH) for at least one year within the period from 1990 to 2004 were submitted by 36 Parties to the Convention. It should be noted that the number of countries reporting official information on POP emissions and their spatial distribution is gradually increasing (for example, total PAH emissions were reported in this year by 35 countries compared with 32 in the previous year). According to official data and expert estimates, emissions of considered POPs tend to decrease from 1990 to 2004. In particular, emissions of four indicator PAHs (benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), indeno[1,2,3-cd]pyrene (I_P)) have decreased by 17 - 22%, and emissions of PCDD/Fs – by 35%.

One of the main conclusions of the TFMM Workshop on the Review of the EMEP Models on Heavy Metals and Persistent Organic Pollutants in Moscow in 2005 is “the MSCE-POP model represents the state-of-the-science and fits to the purpose of evaluating the contributions of long-range transport to the environment impacts caused by POPs”. Along with that, further improvement of the model is recommended. According with the recommendation of the Workshop, further development of the model is performed. In particular, this activity includes the refinement of POP physical-chemical properties, description of POP atmospheric degradation and scavenging, improvement of model description of air/soil exchange and further development of oceanic module. To take into account contributions of intercontinental transport of POPs to the pollution of the EMEP region, the combined hemispheric/regional approach to evaluation of POP pollution levels in Europe is under development. MSC-E continues the comparison of model approaches to the description of POP environmental behaviour in the framework of EMEP intercomparison study of POP models.

Model evaluation of environmental pollution levels was performed for the four indicator PAHs and PCDD/Fs (regional scale), PCBs, γ-HCH and HCB (hemispheric scale) for 2004. The evaluation of contamination of European region and of source-receptor relationships for PCDD/Fs was carried out on the basis of hemispheric/regional approach to take into account contributions of non-EMEP anthropogenic sources and of re-emissions from the environmental media. Pollution levels and source-receptor relationships for PAHs were evaluated at regional scale since these pollutants are mainly particle-bound. For PCBs, γ-HCH and HCB evaluation of transcontinental transport was performed.

Modelling of environmental pollution by PCDD/Fs was made with the use of physical-chemical properties of four toxic PCDD/F congeners. The levels of net deposition flux of PCDD/Fs in Europe differ from about 0.1 ng TEQ/m^2/y in northern Europe (Norway, northern parts of Sweden and Finland)

to 5 ng TEQ/m^2/y and higher in central and southern Europe (Germany, Poland, Czech Republic). Maximum pollution levels were calculated for Germany and neighbouring countries. The transboundary transport of PCDD/Fs was evaluated taking into account national anthropogenic emissions of European countries, non-EMEP anthropogenic sources (USA and Canada) and re-emissions. The contribution of transboundary transport within the EMEP region typically ranges from 30% to 70%. The contributions of non-EMEP anthropogenic sources to the pollution of European countries can be noticeable and reach up to 20% for countries located close to EMEP boundaries. The input of re-emissions to depositions in European countries is in the range from 20% to 40%.

Annual mean B[a]P concentrations in the surface atmospheric layer vary from 0.1 to 1 ng/m^3 and more. High levels of contamination are characteristic for Germany, Ukraine, Belgium and the Netherlands (up to 1 ng/m^3 and higher). Spatial distribution of the rest three indicator PAHs (B[b]F, B[k]F, and I_P) are similar to that of B[a]P. The levels of air concentrations of B[k]F in Europe are lower than for the rest three indicator compounds (from 0.01 – 0.05 ng/m^3 in northern Europe up to 0.3 ng/m^3 in eastern and southern European regions). The transboundary transport of B[a]P between European countries was evaluated by regional calculations (within the EMEP region). The contribution of the external sources to air concentrations and depositions of B[a]P is essential and varies typically from 20 to 80%.

Preliminary model estimates of pollution levels and contributions of intercontinental transport to depositions of γ-HCH, HCB and selected PCB congeners (PCB-153, PCB-118 and PCB-153) over selected receptor regions were obtained. These are the pollutants of significant long-range transport potential. These POPs are recommended by the EMEP Task Force on Hemispheric Transport of Air Pollution during its 2nd meeting in Moscow in June 2006 for the model evaluation of intercontinental transport due to availability of the global emission data and various measurements. The influence of intercontinental transport to the contamination of remote regions is exemplified by the Arctic.

Monitoring and model assessment of POP pollution in the EMEP region was carried out by the EMEP Centres in co-operation with the subsidiary bodies to the Convention (WGE, WGSR), international organizations (EU, HELCOM, OSPAR, SETAC, UNEP, WMO), as well as with national experts.

In 2006 MSC-E in co-operation with national experts continued the work on the intercomparison of POP models. Two EMEP expert meetings held by MSC-E in Moscow were attended by national experts from Canada, Germany, Japan, the Netherlands, Norway, Switzerland, the United Kingdom, and the USA. Stage II of the study, aimed at the comparison of POP mass balance estimates, calculated levels of depositions and concentrations in the main environmental compartments and their spatial distribution, is finalized. A scientific paper devoted to the results of Stage II is under preparation. Stage III of the intercomparison study being focused on ranking of 14 reference chemicals with respect to their long-range transport potential and overall environmental persistence obtained by different models is now ongoing.

CONTENTS

INTRODUCTION

This report summarizes the main results of 2006 activities of EMEP countries and EMEP Centres - Meteorological Synthesizing Centre-East (MSC-E), Meteorological Synthesizing Centre-West (MSC-W) and Chemical Coordinating Centre (CCC) - in evaluation of contamination of the European region by persistent organic pollutants (POPs). During 2006 the studies of POP long-range atmospheric transport and transboundary fluxes on the basis of measurements, emission data and modelling were continued. Modelling studies in accordance with the EMEP Work-plan for 2006 [*ECE/EB.AIR/GE.1/2005/10/Rev.1*] were performed for the following pollutants: polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), lindane (γ-HCH), and hexachlorobenzene (HCB).

In spite of the fact that spatial coverage of the EMEP monitoring network does not seem quite satisfactory, there are some positive tendencies in monitoring activities. In 2004 there were 6 sites measuring POPs in both compartments, and altogether 15 measurement sites, with an increase of 3 sites (in Spain, the United Kingdom and Latvia) since the previous year. Spain however has only reported data for December 2004. Most of the sites are connected to the OSPAR programme CAMP.

The MSCE-POP model has been reviewed at the EMEP/TFMM Workshop on the Review of the EMEP Models on Heavy Metals and Persistent Organic Pollutants in Moscow in 2005. The model is considered to fit for the purpose of evaluating the contributions of long-range transport to the environment impacts caused by POPs for the evaluation of the long-range transboundary transport and deposition of POPs in Europe and "within the limitations of current understanding of the sources and emissions of POPs and of their fate and behaviour in the environment, the MSC-E POP model represents the state-of-the-science". Along with that the TFMM Workshop in Moscow has recommended to continue further improvement of modelling approach for POPs.

Following these recommendations MSC-E has continued its work on further development of EMEP modelling approach for POPs. An essential attention was given to the refinement of data sets of physical-chemical properties and process parameterisations used in the MSCE-POP model.

Further refinement of physical-chemical properties of POPs considered in modelling activities (PCDD/Fs, PAHs, PCBs, γ-HCH and HCB) was performed by taking into consideration the temperature dependence of octanol/water partition coefficient as well as by harmonising three partition coefficients (octanol/water, air/water, and octanol/air) with the help of the least-squares adjustment procedure developed in [*Schenker et al.,* 2005].

A number of modifications were introduced into the description of processes included into the MSCE-POP model. The possibility of improving the agreement between model results and measurements for B[a]P by the refinement of degradation process description and usage of different scenarios of seasonal variations of emission has been considered.

To take into account the effect of temperature variations, the model description of POP absorption in soil and volatilisation to the atmosphere was modified using, in particular, the temperature dependence of octanol/water partition coefficient.

A simple description of POP snow scavenging was elaborated and implemented into the model. The analysis of the effect of this modification of MSCE-POP parameterisation of POP deposition with precipitation will be presented on the next TFMM Workshop on MSC-E models review and evaluation.

During this year the work on refining model description of POP behaviour in the marine environment was continued. In the framework of this task further modification of oceanic module in MSCE-POP model and sensitivity study with respect to main processes determining POP fate in the ocean were performed.

Further development of the MSCE-POP model has been continued by improving the consistency of hemispheric and regional modelling approaches. The nested modelling approach is developed to evaluate POP pollution levels for the European region and to provide estimates of transboundary fluxes between European countries accounting also for the contributions of non-European emission sources and of re-emission of POPs accumulated in environmental compartments. This modelling approach is based on the nesting of regional and hemispheric versions of EMEP/MSCE-POP model. A special attention is given to the compatibility of the input data used in hemispheric and regional modelling, and evaluation of transboundary transport of POPs between the European countries.

In accordance with the Work-plan for 2006, levels of POP pollution and transboundary transport for 2004 were evaluated for PAHs (4 indicator compounds: benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F) and indeno[1,2,3-cd]pyrene (I_P)), PCDD/Fs (toxic congeners), PCBs (PCB-28, PCB-118, PCB-153), γ-HCH and HCB. The evaluation of contamination of the EMEP region by PCDD/Fs was carried out with the help of combined hemispheric/regional approach in order to take into consideration the contributions of hemispheric emission sources located outside the EMEP region and the contribution of re-emission. Since the considered four indicator PAHs are present in the atmosphere mainly in particle-bound form, calculations for PAHs were performed at the regional scale. A preliminary comparison of obtained modelling results with monitoring data was carried out. In modelling of pollution by PCBs, γ-HCH and HCB the emphasis was put on evaluation of intercontinental transport and pollution of remote regions.

To evaluate POP pollution levels within the European region, the EMEP Centres closely co-operated with the subsidiary bodies to the Convention, international organizations, programmes and projects as well as with national experts.

In the framework of co-operation with the EMEP Task Force on Hemispheric Transport of Air Pollution (TF HTAP) MSC-E took part in the organizational workshop on Intercontinental Transport Modelling Intercomparison and presented the information on EMEP experience in the organization of POP model intercomparison study and evaluation of its results.

MSC-E has supported the organization of the second meeting of the TF HTAP convened in Moscow in June 2006. The meeting considered the hemispheric transport of POPs, mercury, and methane and discussed the outline and process of preparation of the TF HTAP assessment report. MSC-E chaired the POP session. During this session MSC-E and national experts from the USA, Canada, Norway, and the Czech Republic presented information on the state of the art in monitoring, emission inventories and modelling of POPs with emphasis on intercontinental transport and its contribution to POP pollution levels in different regions of the northern hemisphere. It is recognized that there is well-documented evidence of the intercontinental and hemispheric transport of POPs.

In 2006 MSC-E continued to contribute to the work of the Task Force on POPs on technical review of dossiers of new proposed substances [*ECE/EB.AIR/87*]. Modelling data as an additional information for the evaluation of hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), polychlorinated naphthalenes (PCN), and dicofol as candidates for POPs in accordance with the two criteria: potential for the long-range transboundary atmospheric transport (LRTP) and persistence were provided by the MSC-E through the UN ECE Secretariat to the Task Force on POPs [*Vulykh et al.*, 2005 a, b, c, d].

Besides, MSCE-POP model results on LRTP and P_{ov} of PentaBDE, endosulfan, dicofol, HCBD, PeCB and PCN are summarized in the MSC-E Technical Report [*Vulykh et al*., 2006].

In 2006 MSC-E in close co-operation with national experts continued the work on the intercomparison of POP models. Two EMEP expert meetings held by MSC-E in Moscow were attended by national experts from Canada, Germany, Japan, the Netherlands, Norway, Switzerland, the United Kingdom, and the USA. The main results of the Stage II of the study are presented in the revised Intermediate Technical Report [*Shatalov et al*., 2006]. The preparation of the scientific paper devoted to the main findings of the Stage II as well as activities within the Stage III of the intercomparison study is now ongoing.

The results of the work carried out during this year are presented in the Technical Reports of the EMEP Centres [*Aas and Breivik*, 2006; *Gusev et al*., 2006] as well as on the Internet *www.emep.int* and *www.msceast.org*.

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1. MONITORING OF POPs IN EMEP

1.1. Measurement network

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. 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 OSPARCOM.

The locations of the measurement sites, which have delivered POPs for 2004, are shown in Fig. 1.1. The measurement programs at the different sites are given in Table 1.1. Further details of the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [*Uggerud and Hjellbrekke*, 2005]. The sites are divided in those measuring both in air and precipitation, and those measuring only in one media. In 2004 it was 6 sites measuring POPs in both compartments, and altogether it was 15 measurement sites, an increase of 3 sites (in ES, GB, LV) since the previous year. Spain has however only reported data for December 2004. Most of the sites are connected to the OSPARCOM programme CAMP.

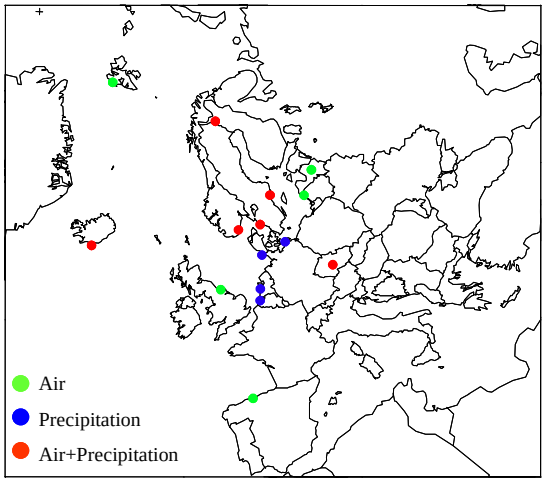


Fig. 1.1 *Measurement network of POPs in EMEP, 2004*

Table 1.1. *Measurements sites and programs for POPs in 2004*

Sites	POPs in air and aerosol	POPs in precipitation
BE04		Pesticides, HCHs
CZ03	PAHs, PCBs, pesticides, HCHs	PAHs, PCBs, pesticides, HCH
DE01		PAHs, PCBs, pesticides, HCB, HCHs
DE09		PAHs, PCBs, pesticides, HCB, HCHs
ES08	PAHs	
FI96	PAHs, PCBs, pesticides, HCHs	PAHs, PCBs, HCHs
GB14	PAHs, PCBs	
IS91	PCBs, pesticides, HCB, HCHs	PCBs, pesticides, HCB, HCHs
LV10	Benzo a-pyrene	
LV16	Benzo-a-pyrene	
NL91		γ–HCH
NO01	PCBs, HCB, HCHs	PCBs, HCB, HCHs
NO42	PAHs, PCBs, pesticides, HCHs, HCB	
SE12	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs
SE14	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs

It is quite evident from Fig. 1.1 that the spatial distribution in Europe is unsatisfactory, no sites in east of Europe, and only one site in south of Europe. However there are some positive tendencies; i.e. Spain, Latvia and the United Kingdom have started to measure POPs. In addition, CCC will coordinate a passive air sampling campaign in 2006 covering whole of Europe as well as central Asia to evaluate the spatial patterns in air. The results from that campaign will be available next year.

1.2. Measurement results of POPs in 2004

Details of the measurements results and methodology are found in the EMEP/CCC data report on heavy metals and POPs [*Aas and Breivik*, 2006]. In Tables 1.2 and 1.3 a summary of the results of the most common POP measurements in air and precipitation is presented. It can be difficult to compare the results, especially for precipitation, from the different sites as the measurement programs and methodologies may vary between sites.

Table 1.2. *Annual average concentrations of selected POPs in 2004 , pg/m^3; ng/m^3 for benzo-a-pyrene (B[a]P)*

	HCHs		PAHs	PCBs			Pesticides	
	α-HCH	γ-HCH	B[a]P	PCB-180	PCB-28	Ratio 28/180	pp_DDD	pp_DDT
CZ03	11.6	23.0	0.279	2.46	9.56	4	2.30	7.52
GB14			0.023	0.25	12.7	51		
LV10			0.163					
LV16			0.180					
NO01	16.5	10.1		0.20	1.64	8		
SE12	9.4	6.9	0.029	0.22	1.63	7		
SE14	8.2	7.4	0.068	0.63	1.57	2	0.25	0.90
FI96	9.4	3.1	0.033	0.06	1.93	32	0.08	0.18
IS91	5.0	7.6		0.17	4.52	27	0.19	0.24
NO42	17.1	2.8	0.003	0.04	1.97	50	0.03	0.10

In air, it is a quite clear south-north gradient with elevated concentration levels in the Czech republic for most of the POPs. The high levels at the stations in the Czech Republic and the United Kingdom is not surprising, considering the high historical usage of PCBs in central parts of Europe [*Breivik et al*., 2002]. PCBs consist of various individual chemical species (congeners). The Long-range Transport Potential (LRTP) of POPs is limited by the competing processes of atmospheric degradation and net atmospheric deposition. For PCBs, the atmospheric mobility of lighter congeners is believed limited by atmospheric reaction with OH-radicals, whereas the LRTP of heavier congeners is believed limited by atmospheric deposition. The atmospheric mobility of heavier PCBs (such as PCB-180) may thus be limited because they exhibit a greater affinity for atmospheric particles than lighter PCBs (e.g. PCB-28), and they thus deposit faster in comparison to the lighter counterparts. Stations experiencing elevated levels of PCB-180 may thus be indicative of areas that are under influence of contemporary PCB emission source regions. The relative contribution of individual PCB congeners is dependent on the site location. With the exception of the United Kingdom, the PCB-28/PCB-180 ratio tends to increase from south to north. This confirms that there are marked differences in the long-range transport potential (LRTP) within the group of PCBs [*Wania and Dugani*, 2003].

For the HCHs, we observe a general decline in concentrations of γ-HCH, going from south to north. This trend is in marked contrast to α-HCH, which even exhibit the highest concentrations at the northernmost site (NO42). The decline in γ-HCH may be attributed to the fact that it is the more water-soluble specie among the two HCHs. The LRTP of γ-HCH may thus be more limited in comparison to α-HCH because the former is more easily scavenged by rain.

Table 1.3. *Total deposition of selected POPs in 2004, ng/m^2/y*

	HCHs		PAHs	PCBs		Pesticides		mm
	α-HCH	γ-HCH	B[a]P	PCB-180	PCB-28	pp_DDD	pp_DDT	
Wet deposition:								
CZ03		2106	2937	57	47	202	211	635
BE04	399	3111				399		797
NL91		4967			23			927
DE01	195	943	806	73	72	40	146	554
DE09	221	1202	1361	77	37	59	192	562
NO01	537	1229		19	23			1399
IS91	67	39		4	32	4	6	791
Total deposition:								
SE12	170	300	1232	8	15			
SE14	124	305	1813	81	2			
FI96	86	85	2707	16	2			

For precipitation it is difficult to compare the results in Europe due to different methodology, e.g. Finland and Sweden are measuring deposition while others are measuring concentration in precipitation. In addition the concentration level in precipitation is very often below the detection limit. However the general picture is as for air components, that the level decreases from central Europe and north to Scandinavia and Iceland. The deposition is dependent on the precipitation amount; the relatively high level of HCHs at the Norwegian site can partly be explained by almost twice as much precipitation as many of the other sites.

2. EMISSIONS

Official data on the emission totals of PAHs, PCDD/Fs, HCB, PCBs and HCH for the period from 1990 to 2004 (for at least one year) were reported by 36 countries. Emission data reported by countries are available from WEBDAB: http://webdab.emep.int. The number of countries reporting the data on POP emissions increased considerably during recent years. Nevertheless, to estimate total emission values and their spatial distribution for European region expert estimates still have to be used.

2.1. Polycyclic aromatic hydrocarbons

Official data on the emission totals of polycyclic aromatic hydrocarbons (PAHs) were submitted by 35 countries for 1990-2004 (for at least one year).

According to the Protocol on POPs for the purposes of atmospheric emission inventories four indicator compounds of PAHs should be used. They are benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F) and indeno[1,2,3-cd]pyrene (I_P) [*ECE/EB.AIR/60, Annex III*). This year, model runs have been performed for all of them. The official information on total emission of these four PAHs is available for 26 European countries for 1990-2004 (for at least one year). 19 countries, namely, Belarus, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, France, Germany, Hungary, Iceland, Ireland, Latvia, Lithuania, Monaco, Poland, Republic of Moldova, Slovakia, Switzerland and the United Kingdom submitted emissions of each of four indicator compounds for the considered period (for at least one year). In 2006 Cyprus, Latvia and Switzerland reported the data on their national emissions for the first time. The Russian Federation submitted only B[a]P emissions. For the remaining countries, expert estimates were used [*Baart et al.*, 1995; *Denier van der Gon et al*., 2005].

The information on PAH spatial distributions was provided by 17 countries (Austria, Belgium, Bulgaria, Denmark, Finland, France, Hungary, Iceland, Latvia, Lithuania, Monaco, Netherlands, Norway, Poland, Slovakia, Spain, United Kingdom). For other countries, expert estimates were used [*van der Gon et al*., 2005].

According to the official data and expert estimates for the period from 1990 to 2004, European emissions of four indicator PAHs decreased by 17% - 22% depending on pollutant (Fig. 2.1, Table 2.1).

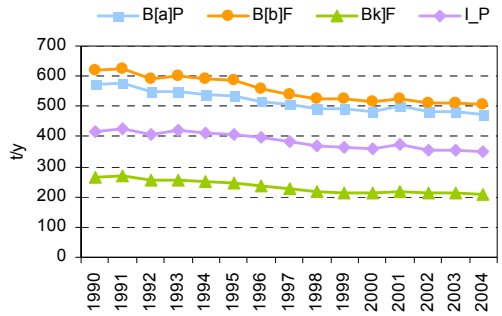


Fig. 2.1. *Time-series of PAH emissions*

Table 2.1. *Decrease of PAH emissions 1990-2004*

Pollutant	Decrease, %
B[a]P	18
B[b]F	18
B[k]F	22
I_P	17

Official information on emissions of four indicator PAHs by sectors in 2004 is available for 20 countries. The sector split for the national total four indicator PAH emissions is presented in Fig. 2.2. It can be seen that the Residential sector is the major contributor. On average its share amounts to 61%. The predominant source in the Residential sector is the combustion of wood. The second most important sector is the Metal Production. This sector is the largest source of PAHs for Iceland, Norway and Switzerland.

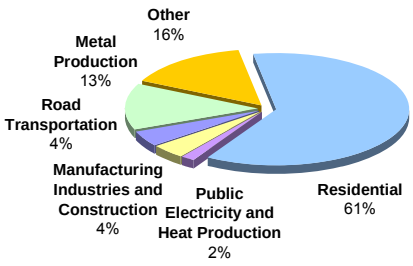


Fig. 2.2. *Sector split for PAHs in 2004*

The considerable contribution of the Residential sector to the total PAH emissions determines pronounced seasonal emission variation. At present, emission scenario taken from [*Baart et al.*, 1995], where the level of B[a]P emissions in winter is 10% higher than in summer, is used for calculations. The same seasonal distribution of PAH emissions is used for all the European countries. However, calculations using this scenario underestimate seasonal variations of B[a]P contamination levels (air concentrations and depositions). This shows that the uncertainties in emission seasonal variations cause additional uncertainties to the calculation results. The information on these variations from Parties to the Convention is strongly appreciated.

2.2. Dioxins and furans

Official data on total emission of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) (sum of toxicities of 17 toxic PCDD/F congeners) were reported by 33 European countries and Canada for the period from 1990 to 2004 (for at least one year). For the first time, Romania reported emission data. For the remaining European countries, expert estimates of PCDD/F total emission prepared by [*Denier van der Gon et al.*, 2005] were used. The value of PCDD/F emissions of the USA for 1995 was taken from the dioxin and furan inventories prepared by [*UNEP*, 1999].

The information on the spatial distribution of PCDD/F emissions was submitted by 17 countries (Austria, Belarus, Belgium, Bulgaria, Finland, France, Hungary, Iceland, Latvia, Lithuania, Monaco, Netherlands, Norway, Poland, Slovakia, Spain, Sweden). For the remaining European countries, expert estimates of spatial distribution of PCDD/F emissions were used [*Denier van der Gon et al.*, 2005]. For the evaluation of spatial distribution of the PCDD/F emissions in the North America over the 2.5^{o}x2.5^{o} calculation grid, data on the population density (1990) available in the Canadian Global Emissions Interpretation Centre (http://www.ortech.ca/cgeic) were used.

According to the available official data and expert estimates, the total annual emission of PCDD/Fs in the northern hemisphere and European region decreased by 30% and 35%, respectively between 1990 and 2004 (Fig. 2.3). The total emission of PCDD/Fs in the northern hemisphere amounted to 14.1 kg TEQ in 2004, including 11.2 kg TEQ/y in Europe and 2.9 kg TEQ/y in North America. The decrease of emissions of PCDD/Fs within the northern hemisphere was mainly due to reduction in emissions in Switzerland, France, the Czech Republic, the United Kingdom, the Netherlands, Belgium, Bulgaria and Canada.

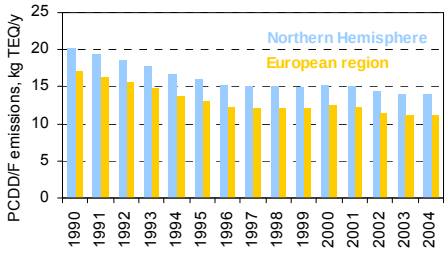


Fig. 2.3. *PCDD/F emissions in the northern hemisphere and European region for the period from 1990 to 2004*

According to the official data, PCDD/F emissions in most countries tend to decrease (1990-2004). The overall PCDD/F emissions in 22 countries reported data for both years 1990 and 2004 decreased by ~50%. The maximum decrease of the PCDD/F emissions was reported by the Netherlands (19 times), and the maximum increase - by Germany (2.8 times).

On the basis of the above data and using congener compositions of PCDD/F mixture in various European countries [*Pacyna et al*., 1999] spatial distributions for all 17 toxic congeners of PCDD/Fs for the period from 1990 to 2004 were prepared. At this stage, evaluation of overall toxicity of PCDD/F mixture was performed on the basis of simulations of group of four congeners (1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 2,3,4,7,8-PeCDF and 1,2,3,4,6,7,8-HpCDF) (feasibility of such evaluation and uncertainty analysis is described in [*Shatalov et al*., 2003]).

Official information on emissions of PCDD/Fs by sectors in 2004 is available for 25 countries. The sector split for the national total emission of PCDD/Fs is presented in Fig. 2.4. The largest contribution to the total PCDD/F emission is made by the Residential sector (24%). This sector is the largest source of PCDD/Fs for Austria, Belgium, Estonia, Finland, Lithuania and Slovenia. The next important sectors are the Manufacturing Industries and Construction, the Metal Production and the Waste Incineration.

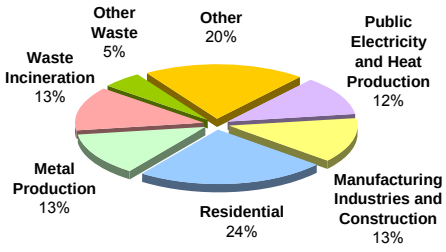


***Fig. 2.4**. Sector split for PCDD/Fs in 2004*

2.3. Hexachlorobenzene

Official information on total emission of hexachlorobenzene (HCB) was reported by 24 European countries along with Canada and the United States for the period from 1990 to 2004 (for at least one year). For the first time, Latvia, Romania, Slovakia, Slovenia and Sweden reported emission data. For the remaining European countries and the European part of Russia, the HCB expert estimates prepared by [*Pacyna et al*., 1999] were used. The HCB emission value in Japan for 2002 is taken from the emission inventory of Japan prepared by *E.Toda* [2005]. The HCB emission values in China, Pakistan, Republic of Korea and the Asian part of Russia were estimated on the basis of the relationship of the gross domestic product and official HCB emission data for 2004 taken for countries with similar economic indexes. The HCB emission value in India is taken from the EMEP Technical Report 7/2005 [*Shatalov et al*., 2005]. For the evaluation of HCB long-range transport on hemispheric scale available emission sources were aggregated into 5 groups shown in Fig. 2.5.

***Fig. 2.5**. Splitting of HCB emissions into groups of sources*

The information on the spatial distribution of HCB emissions was submitted by 7 countries (Austria, Belgium, France, Latvia, Poland, Slovakia and Spain). For the remaining European countries expert estimates of spatial distribution of HCB emissions were used [*Pacyna et al*., 1999]. For the evaluation of spatial distribution of the HCB emission in the northern hemisphere over the 2.5^{o}x2.5^{o} calculation grid, data on the population density (1990) available in the Canadian Global Emissions Interpretation Centre (http://www.ortech.ca/cgeic) were used.

According to the official data and expert estimates, the total emission of HCB in the northern hemisphere and European region decreased by 10% and 25%, respectively between 1990 and 2004 (Fig. 2.6). The total emission of HCB in the northern hemisphere amounted to 77 t in 2004, including 0.9 t in North America, 36.4 t in Central Asia, 14.2 t in Southeast Asia, 12.5 t in Europe and 12.8 t in Russia. The decrease of emission of HCB within the northern hemisphere is mainly due to reduction in emissions in the United Kingdom, Bulgaria, Spain and Canada.

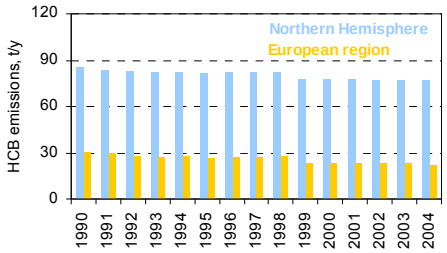


Fig. 2.6. *HCB emissions in the northern hemisphere and European region for the period from 1990 to 2004*

According to the official data, the total HCB emissions in 12 countries submitted data for both years 1990 and 2004 decreased by 75% (or 3.8 times). The maximum emission decrease was reported by the United Kingdom (118 times), and the maximum increase - by Belgium (3 times).

Official information on emissions of HCB by sectors in 2004 is available for 16 countries (Fig. 2.7). The maximum contribution to the total HCB emissions is made by the Manufacturing Industries and Construction sector (29%). The second most important sector is the Residential. This sector is the largest source of HCB for Austria, Estonia and Latvia.

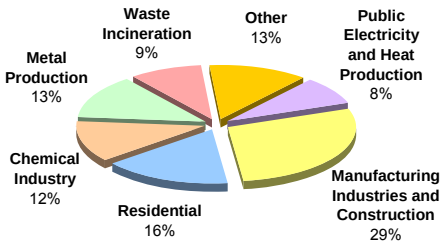


Fig. 2.7*. Sector split for HCB in 2004*

2.4. Polychlorinated biphenyls

Official information on total emission of polychlorinated biphenyls (PCBs) was submitted by 20 countries for the period from 1990 to 2004 (for at least one year). For the first time, Latvia, Romania, and Sweden reported emission data. According to the official data, the total PCB emissions in 12 countries submitted data for both years 1990 and 2004 decreased by 60% (or 2.7 times). The maximum emission decrease was reported by the Czech Republic (8.6 times), and the maximum increase - by Monaco (10%). The information on the spatial distribution of PCB emissions was submitted by Finland, France, Latvia and Lithuania.

Official information on emissions of PCB by sectors in 2004 is available for 15 countries. The maximum contribution to the total PCB emissions is made by the Public Electricity and Heat Production sector (20%). The next important sector is the Residential. This sector is the largest PCB sources for Bulgaria, France, Latvia and Slovakia.

The modelling of PCB long-range transport for the period from 1990 to 2004 was performed for the following individual PCB congeners: PCB-28, 118 and 153. The sources located over the whole northern hemisphere were taken into account in the computations. Emission data for selected PCB congeners were taken from the global emission inventory of 22 PCB congeners [*Breivik et al.*, 2002].

For the evaluation of intercontinental transport, PCB emission sources of the northern hemisphere were split into 5 groups (Fig. 2.8). For these groups gridded emission data for the period from 1990 to 2004 were prepared for selected PCB congeners.

For example, according to the data by [*Breivik et al*., 2002], PCB-153 emissions in the northern hemisphere amounted to 12 t, including 2.2 t in America, 5.9 t in Europe, 1.7 t in Africa and Central Asia, 0.7 t in Southeast Asia and 1.5 t in Russia in 2004.

Fig. 2.8*. Splitting of PCB emissions into groups of sources*

2.5. γ-Hexachlorocyclohexane

Official data on the emission totals of γ-hexachlorocyclohexane (γ-HCH) were submitted by 10 European countries for the period from 1990 to 2004 (for at least one year). Official information about usage of technical HCH and lindane was reported by 11 European countries for the considered period (for at least one year). For the remaining European countries, the compilation of the expert estimates prepared by [*Pacyna et al*., 1999 and *Denier van der Gon et al*., 2005] was used. The γ-HCH emission values in Canada, the USA, Mexico and Honduras for 1990 and China for 1990 and 1995 were estimated on the basis of the γ-HCH application in these countries and were described in [*Shatalov et al.,* 2003 and *Gusev et al*., 2005a]. For Canada, the USA and Mexico for 2000 and 2002, expert estimates of γ-HCH emission prepared by [*Li,* 2004] were used.

The information about the spatial distribution of γ-HCH emissions was submitted by Spain only. For the remaining European countries expert estimates of spatial distribution of γ-HCH emissions were used [*Pacyna et al*., 1999; *Denier van der Gon et al*., 2005]. The spatial distribution of γ-HCH emissions in North America was prepared by Dr.*Y.-F.Li* [2004]. For the evaluation of the γ-HCH emission spatial distribution in China over the 2.5^{o}x2.5^{o} calculation grid, data on using cropland area (1990) available in the Canadian Global Emissions Interpretation Centre [*http://www.ortech.ca/cgeic*] were used.

According to the available official data and expert estimates, the total annual emission of γ-HCH in the northern hemisphere and European region decreased by 75% and 93%, respectively between 1990 and 2004 (Fig. 2.9). The total emission of γ-HCH in the northern hemisphere amounted to 432 t in 2004, including 71 t/y in North America, 68 t/y in Central America, 200 t/y in Southeast Asia and 93 t/y in Europe.

Official information on emissions of γ-HCH by sectors is available for Belgium, Croatia, Germany, Spain and the United Kingdom for the period from 1990 to 2004 (for at least one year). In Belgium and the United Kingdom the maximum contribution to the total γ-HCH emissions is made by the Solvent and Other Product Use (Other sector). In Croatia, Germany, Spain γ-HCH emissions from the Agriculture (Other sector) contributed most of all to the total γ-HCH emissions.

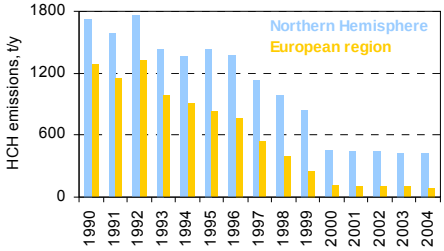


Fig. 2.9. *γ-HCH emissions in the northern hemisphere and European region for the period from 1990 to 2004*

For evaluation of the intercontinental transport emissions were split into three groups: sources of China, North America and Europe.

3. MODEL DEVELOPMENT

Following the recommendations of TFMM Workshop on the Review of the EMEP MSC-E Models on Heavy Metals and Persistent Organic Pollutants MSC-E has started its work on further improvement of the hemispheric and regional versions of MSCE-POP model. At current stage essential attention was given to the harmonization of physical-chemical properties of selected POPs and refinement of process parameterisations used in MSCE-POP model. In particular, the improvement of model description of POP degradation in the atmosphere, removal with precipitation (including scavenging with snow), and partitioning in soil was carried out. Particular attention was paid to further refinement of the description of POP fate in seawater compartment taking into account the transport of POPs with sea currents, partitioning of POPs in seawater, and the effect of ice cover.

The MSC-E hemispheric/regional modelling approach to the evaluation of POP pollution levels in the European region has been further developed. This approach is based on the nesting of hemispheric and regional versions of MSCE-POP models. Hemispheric version of the model permits to evaluate POP intercontinental transport and accumulation in environmental media on hemispheric/global scale with coarse spatial resolution during long periods of time. Regional version of the model is applied for the evaluation of POP distribution over the EMEP grid with finer spatial resolution. Thus the developed modelling approach can be applied for the evaluation of POP contamination levels in the northern hemisphere with emphasis to the European region and for estimation of POP intercontinental transport along with more detailed information on the transboundary fluxes between European countries.

3.1. Refinement of POP physical-chemical properties

One of the conclusions of the Workshop on the Review of the EMEP MSC-E Models on Heavy Metals and Persistent Organic Pollutants was “The parameterisations in the MSC-E POP model are based on up-to-date information and conform with those employed in most of the other models used to simulate the behaviour of POPs in the environment”.

Actually the selection of physical-chemical parameters and degradation rates for the MSCE-POP model parameterisation is usually based on the best available experimentally determined or theoretically derived parameter values taken from the literature. If it is possible this selection is relied on empirical values taking into account not only the accuracy of methods used for their determination but also existing scattering of values taken from the literature. An attention was paid to the internal consistency of such parameters as air/water, octanol/water and octanol/air partition coefficients and subcooled liquid vapour pressure.

To verify the MSCE-POP model parameterisation, a comparison of main pollutant-related parameters of PCBs, namely, physical-chemical properties and degradation rates was performed in the course of the POP model intercomparison study initiated under EMEP. It was found that the MSCE-POP model is close to other models in model parameterisation and description of main processes.

Last years several publications devoted to the selection of internally consistent data sets for modelling of POPs have been published. The works of *Cole and Mackay* [2000], *Beyer et al.* [2002], *Li et al.* [2003], *Xiao et al.* [2004], *Shen and Wania* [2005] and the most recent work of *Schenker et al.* [2005] included a description of different methods for evaluating and harmonizing data sets for many POPs including PCBs, PAHs, γ-HCH and HCB. In the latter paper the new least-squares adjustment

procedure was proposed for harmonizing physical-chemical properties including the energies of phase transitions.

This year to take into account the recent scientific findings in the field of selection and evaluation of physical-chemical properties for modelling and to improve model description of POP contamination levels in the environment, further refinement of physical-chemical properties of POPs included into modelling activities (PAHs, PCBs, PCDD/Fs, γ-HCH and HCB) is performed on the basis of up-to-date information. This refinement of MSCE-POP parameterisation includes the following main aspects:

- Implementation of temperature dependence of octanol/water partition coefficient;
- Harmonisation of octanol/water, air/water and octanol/air partition coefficients (K_{OW}, K_{AW}, K_{OA}) using, in particular, the least-squares adjustment procedure presented in [*Schenker et al.,* 2005].

Below the refinement of physical-chemical properties of selected POPs performed this year is briefly described. More detailed comparison of earlier accepted and refined values of physical-chemical parameters in the MSCE-POP model parameterisation can be found in Chapter 4 of the EMEP/MSC-E Technical Report 4/2006 [*Gusev et al*., 2006]. Besides, model parameterisations of all considered POPs used recently in calculations of the long-range atmospheric transport and depositions are given in detail in Annex A of [*Gusev et al*., 2005b].

Implementation of temperature dependence of K_{OW}. In the course of the POP model intercomparison study it was noted that in contrast to the MSCE-POP model most of participating POP models included octanol/water partition coefficient in the form of temperature dependence. Therefore, temperature dependence of K_{OW} partition coefficient available in the literature for all considered pollutants is introduced in the MSCE-POP model parameterisations this year. A comparison of renewed values of K_{OW} with the values used in the previous parameterisations for all considered pollutants can be found in Chapter 4 of the EMEP/MSC-E Technical Report 4/2006 [*Gusev et al.,* 2006].

This year temperature dependences of K_{OW} for four indicator compounds of **PAHs** - benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F) and indeno[1,2,3-cd]pyrene (I_P) - are implemented. To estimate the temperature dependences, the base values of K_{OW} at 25 °C are selected to be equal to those earlier used and taken from [*Mackay et al.*, 1992; *Beyer et al*., 2002] and value of internal energy of octanol-water phase transition (ΔU_{OW}) for B[a]P taken from *Lei et al*. [2000] cited by *A.Beyer et al*. [2002] are assumed to be equal to that of other considered PAHs. It should be noted that there are essential differences between earlier used value of K_{OW}, which is temperature independent, and new values of this coefficient for all considered PAHs at temperatures, which are much lower ord higher than 25 °C.

Selection of new temperature dependence of octanol/water partition coefficient for the following eight **PCB** congeners: PCB-28, PCB-52, PCB-101, PCB-105, PCB-118, PCB-138, PCB-153, and PCB-180 is performed on the basis of data from [*Schenker et al*., 2005]. It should be noted that for the refinement of the MSCE-POP model parameterisation K_{OW} values based on wet octanol are selected. A comparison of adjusted and previously used values of K_{OW} at 25 °C together with new temperature dependence coefficients demonstrates that the most essential difference between these values is characteristic of PCB-138. The minimum difference is found for PCB-180.

New temperature dependence of octanol/water partition coefficient of **γ-HCH** is also selected on the basis of data from [*Schenker et al*., 2005]. At that value of log K_{OW} = 3.76 at 25 °C based on dry octanol is recalculated to that based on wet octanol (K^*_{OW}) with the help of regression relation taken from *Beyer et al.* [2002]:

$$\log(K^*_{OW}) = \log(K_{OW}) - 0.117 \quad (3.1)$$

The comparison of K_{OW} value previously used in the MSCE-POP model parameterisation, which is temperature independent, and new adjusted values of selected temperature dependence for γ-HCH shows that the difference between previously used and adjusted values at 25°C is equal to 5%.

New temperature dependence of octanol/water partition coefficient of **HCB** is selected on the basis of data from [*Schenker et al*., 2005]. At that the final adjusted value of log K_{OW} = 5.61 at 25°C for dry octanol is recalculated to the wet octanol (K^*_{OW}) with the help of regression relation taken from *Beyer et al*. [2002]:

$$\log(K^*_{OW}) = 1.35 \cdot \log(K_{OW}) - 1.58 \quad (3.2)$$

New adjusted value of K_{OW} at 25°C is lower than the temperature independent value previously used in the MSCE-POP model approximately by 13%.

For **PCDD/Fs** no temperature dependence K_{OW} parameters is found in the literature.

Harmonisation of K_{OW}, K_{AW}, and K_{OA} partition coefficients. In the MSCE-POP model parameterisation of **PCDD/Fs** the refinement of the values of octanol/water, air/water and octanol/air partition coefficients and subcooled liquid vapour pressure (p_{OL}) is performed. The following four PCDD/F congeners are chosen for the assessment of general toxicity of PCDD/F mixture in the environment: 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 2,3,4,7,8-PeCDF and 1,2,3,4,6,7,8-HpCDF. In accordance with [*Dutchak et al.,* 2004] evaluation of PCDD/F total toxicity in main environmental media (atmosphere, soil, seawater and vegetation) with the help of modelling of the long-range transport and accumulation of these four congeners can be made within the accuracy about 20%.

An application of the least-squares adjustment procedure [*Schenker et al*., 2005] with minor modification[1] is performed for the harmonisation of $K_{AW,}$ K_{OW} and K_{OA} partition coefficients of the considered PCDD/F congeners. $K_{AW,}$ K_{OW} and K_{OA} are not independent being related to one another by the equations:

$$K_{OW} = K_{OA} \cdot K_{AW} \quad \text{or} \quad \log K_{OW} = \log K_{OA} + \log K_{AW} \quad (3.3)$$

The dimensionless coefficient K_{AW} can be expressed via the Henry' law constant K_H by:

$$K_{AW} = K_H / RT, \quad (3.4)$$

The full description of the adjustment procedure based on the least-squares method with constraints can be found in [*Gusev et al*., 2006]**.** The usage of adjustment procedure for the refinement of partition coefficients of PCDD/Fs resulted in the difference between previously used and adjusted values of the Henry's law constant within a factor of 2. At that there are no essential differences between earlier used and adjusted values of K_{OW}. The values of log K_{OA} obtained by the adjustment procedure are lower than previously used and agree better with available literature data.

[1] The authors applied the adjustment procedure for the following six parameters: saturated vapor pressure over subcooled liquid, solubility in water and octanol and the three mentioned partition coefficients. Since solubility of a POP is not included into the parameterization of MSCE-POP model and due to the absence of experimental data on temperature dependence of PCDD/F solubility we restricted ourselves by adjustment of three partition coefficients only.

Regarding subcooled liquid vapour pressure of PCDD/Fs it should be noted that in this year calculations coefficients of temperature dependence for 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, and 2,3,4,7,8-PeCDF obtained by [*Bulgakov and Ioannisian*, 1998], which were used earlier, are also utilised. The only exclusion is the congener 1,2,3,4,6,7,8-HpCDF. For this congener temperature dependence according to [*Mader and Pankow,* 2003] is applied.

For **PAHs** data on internal energy of octanol-air phase transition necessary for calculation of temperature coefficients of K_{OA} are not available in the recent literature. In this connection, the refinement of three partition coefficients for PAHs is carried out by defining temperature dependence of octanol/air partition coefficient from octanol/water and air/water partition coefficients using regression relation (3.3) and taking into account their temperature dependencies:

$$\Delta U_{OA} = \Delta U_{OW} - \Delta U_{AW} \qquad (3.5)$$

In this case obtained values of temperature dependences of these three coefficients are internally consistent. These calculations are performed on the basis of the temperature dependence of K_{AW} previously used in parameterisation of MSCE-POP model [*Ten Hulscher et al.,* 1992 cited by *Galarneau et al*., 2000] and above-mentioned new temperature dependence of K_{OW}.

The values of K_{OA} obtained for four indicator PAHs from the regression relations (3.3) and (3.5) exceed those earlier used at the considered temperatures within a range of factors from 0.8 to 5.3. The difference between these two values is declining with the temperature growth and ends near 20°C.

For **PCBs**, the three partition coefficients and subcooled liquid vapour pressure are renewed on the basis of the final adjusted values given in *Schenker et al*. [2005] and implemented in the MSCE-POP model parameterisations. It should be noted that, these final adjusted values were estimated by the authors with the use of the least-squares method on the basis of PCB data reported by *Li et al.* [2003]. The maximum difference between adjusted values of the Henry's law constant and previously used in MSCE-POP model parameterisation, not exceeding 2.4 times at the whole range of considered temperatures, is observed for PCB-138. For PCB-153 this difference is within a factor of 1.0 - 1.1 (for all considered temperatures). The values of log K_{OA} obtained by the adjustment procedure for all of the considered PCB congeners are lower than those previously used in the MSCE-POP model parameterisation. The difference between old and new values of K_{OA} for PCB-153 is within a factor of 2. The values of p_{OL} obtained in [*Schenker et al.,* 2005] are close enough to those used previously in the MSCE-POP model calculations.

For **γ-HCH**, the three partition coefficients and subcooled liquid vapour pressure are chosen to be equal to the final adjusted values given in *Schenker et al*. [2005]. The latter values were estimated by the authors with the use of the least-squares method on the basis of data reported by *Xiao et al*. [2004]. It should be noted that contrary to the new parameterisation of MSCE-POP model in the previous one two separate temperature dependencies of K_H were implemented: for seawater and fresh water. A comparison between adjusted and previously used temperature dependencies of Henry's law constant of γ-HCH demonstrates that the maximum differences between earlier used and adjusted values are within a factor of 3 (for all considered temperatures). The difference between old and new values of K_{OA} for this pollutant is within a factor of 0.8-3.6. The maximum differences are observed at the lowest temperatures. The values of p_{OL} obtained in [*Schenker et al.,* 2005] are close enough to those used earlier in the MSCE-POP model calculations. The maximum difference between earlier used and adjusted values does not exceed 10% at the whole range of considered temperatures.

For **HCB**, the three partition coefficients and subcooled liquid vapour pressure are chosen to be equal to the final adjusted values given in [*Schenker et al.*, 2005]. The latter values were estimated by the authors with the use of the least-squares method on the basis of data reported by *Snen and Wania* [2005]. The difference between adjusted values of the Henry's law constant and those previously used in MSCE-POP model parameterisation is not considerable (1.3 times for all considered temperatures). The adjusted values of K_{OA} exceed those earlier used by a factor of 2 for all considered temperatures. The difference between old and new values of p_{OL} for HCB is equal to 1.2 times for all considered temperatures.

All modifications described above are involved into the MSCE-POP parameterisation this year. More detailed comparison of renewed values of K_{OW}, K_{AW}, and K_{OA} partition coefficients and p_{OL} with their values used in the previous parameterisations for all considered pollutants can be found in Chapter 4 of the EMEP/MSC-E Technical Report 4/2006 [*Gusev et al.*, 2006]. The values of the rest parameters are not modified (see Annex B in the above cited report).

3.2. Refinement of process parameterisations in MSCE-POP model

Following the recommendations of EMEP/TFMM Workshop on the Review of the EMEP Models on Heavy Metals and Persistent Organic Pollutants MSC-E has started its work on further improvement of process parameterisation in MSCE-POP model. At current stage essential attention was given to the improvement of:

- model description of POP degradation processes in the atmosphere (photodegradation of particle-bound B[a]P);
- model description of POP partitioning in soil;
- model description of POP removal with precipitation (snow scavenging).

Below a brief presentation of these improvements is given. More detailed information on the topic can be found in [*Gusev et al.*, 2006].

Degradation of POPs in the atmosphere. In the course of the comparison of calculated B[a]P concentrations against measured ones carried out in the framework of MSCE-POP model review [*Gusev et al.*, 2006] it was found that the model underestimates seasonal variations of atmospheric concentrations of the pollutant. The example of this underestimation is shown in Fig. 3.1 where monthly mean B[a]P air concentrations measured at SE14 (Råö) in 2003 are compared with computed concentrations .

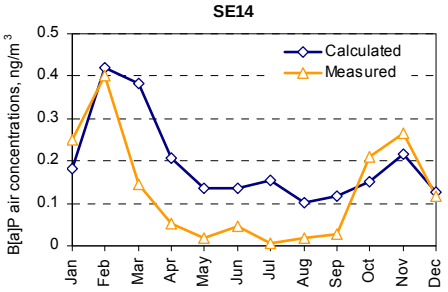


Fig. 3.1. *Calculated and measured B[a]P air concentrations at SE14 in 2003, ng/m^3*

One of the possible reasons for such underestimation can be neglecting of degradation of B[a]P sorbed on the atmospheric aerosol. B[a]P in gaseous phase is known to degrade rapidly in the atmosphere due to reaction with hydroxyl radical [OH]. Parameterization of this process is included in the MSCE-POP model. However, there is an evidence that its degradation can also take place on the surface of aerosol particles [*Kamens et al.*, 1988; *Valerio et al.*, 1984; *Behymer and Hites*, 1988; *Chen et al.*, 2001].

At present a preliminary description of B[a]P photodegradation on particles was elaborated and involved in the MSCE-POP model (see [*Gusev et al.*, 2006]). This allowed refining the agreement between calculated and measured values of B[a]P contamination levels in the environment. As an example the refinement of such an agreement due to modification of degradation process at SE14 is presented in Fig. 3.2. It is seen that the implemented description of B[a]P photodegradation allowed refining seasonal variations of B[a]P air concentrations.

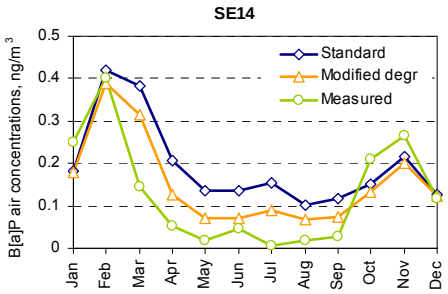


Fig. 3.2. *Modification of B[a]P air concentrations due to the modification of the description of degradation process at SE14 (Råö)*

Partitioning of POPs in soil. The implementation of temperature dependence of octanol/water partitioning coefficient K_{OW} for the considered POPs leads to essential changes in calculated atmospheric and soil concentrations. To exemplify these changes calculations of PCB-153 transport for one-year period were performed with and without temperature dependence of K_{OW} for PCB-153, and calculation results were compared.

The difference between monthly averages of PCB-153 concentrations in the EMEP domain calculated with and without temperature dependence of K_{OW} can reach – 64% for the atmosphere and 130% for soil in particular grid cells. The differences in monthly averages over all grid cells together with 95% confidence intervals (bars) are shown in Fig. 3.3 for all months.

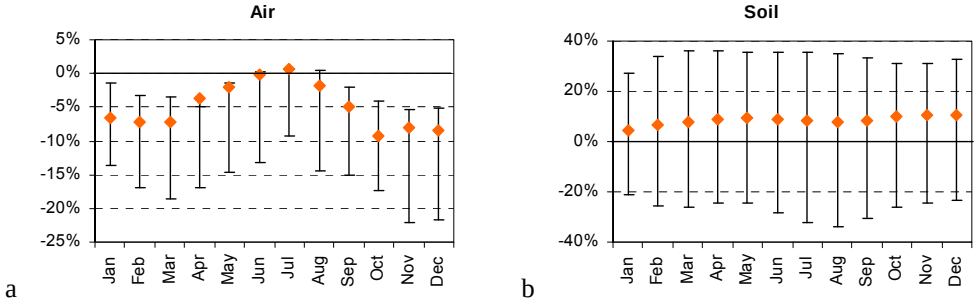


Fig. 3.3. *Differences between monthly averages of PCB-153 concentrations in the atmosphere (a) and in soil (b) over all EMEP grid cells calculated with and without K_{OW} temperature dependence, %. The bars show 95% confidence intervals*

It is seen that the implementation of K_{OW} temperature dependence leads to considerable differences in air and, particularly, soil concentrations. The differences in air concentrations reach up to 20%, and for soil concentrations – up to 30% and more in some grid cells. This leads in turn to the noticeable changes in re-emission flux from soil, see Fig. 3.4 where the differences in monthly averages in re-emission flux from soil within the EMEP region calculated with and without K_{OW} temperature dependence are shown.

More detailed analysis of sensitivity of model output with respect to K_{OW} temperature dependence can be found in [*Gusev et al.*, 2006].

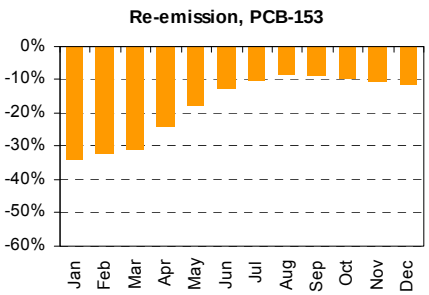


Fig. 3.4. *Differences in monthly means of values of re-emission flux calculated with and without taking into account K_{OW} temperature dependence for PCB-153, %*

Removal of POPs with precipitation. The importance of snow scavenging of POPs was confirmed by a lot of investigations. In particular, in *B.Commoner et al.* [2000] on the basis of evaluation of dioxin airborne transport from point sources in North America it was found that snow scavenging was more effective in removing organic pollutants than that by rain, due to significant surface area of snowflakes. Further, on the basis of measurements at a suburban site in Minnesota, in [*Franz and Eisenreich,* 1998] it was calculated that scavenging ratios by snow were normally several times higher than those by rainwater for PCBs and PAHs. Further, measurements of concentrations of various PCB congeners in air and precipitation within the Baltic region [*Agrell et al.,* 2001] have shown that scavenging ratios by snow are about twice higher than rain scavenging ratios at close values of temperature (about 0º C). Thus, the effect of snow scavenging should be taken into account while simulating long-range transport of persistence organic pollutants (POPs).

Theoretical description of snow scavenging based on the mechanism of sorption of gaseous POPs on the surface of snowflakes was proposed in [*Goss,* 1993; *Hoff et al.,* 1995; *Valsaraj et al.,* 1993; *Franz and Eisenreigh*, 1998; *Wania et al.,* 1999]. On the basis of these investigations a simple description of POP snow scavenging was elaborated and implemented into the model (for details see [*Gusev et al.,* 2006]).

The analysis of the effect of this modification of MSCE-POP parameterisation of POP deposition with precipitation will be presented on the next TFMM Workshop on MSC-E models review and evaluation.

3.3. Refinement of model description of POP fate in seawater

During this year the work on refining model description of POP behaviour in the marine environment presented in the previous Status Report was continued [*Gusev et al.,* 2005c]. In the framework of this task further modification of oceanic module in MSCE-POP model and sensitivity study with respect to main processes determining POP fate in the ocean was performed. Detailed description of these results can be found in the report [*Strukov,* 2006].

Main directions of the refinement of MSCE-POP oceanic module are:

1. POP partitioning in seawater.
2. Sedimentation.
3. Description of turbulent diffusion in open oceans.
4. Exchange between the atmosphere and snow pack and transport of POPs with sea ice.
5. Description of upper mixed layer.
6. Riverine input of POPs into oceanic waters.

An important part of the work on the refinement of oceanic module was sensitivity study with respect to recently introduced processes. The influence of POP partitioning in seawater and sedimentation on calculated pollution levels in the northern hemisphere was described in the previous Status Report [*Gusev et al.,* 2005c]. Furthermore, it was found that the introduction of sea ice module into the model leads to the change of concentrations in seawater in the Arctic region for PCB-153 up to 10 – 20%. The influence of changes in the description of upper mixed layer is rather strong. It can lead to the changes in seawater concentrations up to 100%. The riverine input of γ-HCH of four Siberian rivers (Ob, Yenisei, Lena and Pechora) leads to the increase of seawater concentrations in Kara Sea of about 10% and up to 50% near river estuaries. All these investigations show considerable role of the above listed processes in calculations of contamination of seawater. The refinement of seawater

concentrations of POPs leads in turn to the refinement of re-volatilisation of these pollutants from the marine environment.

The pathways of the POP long-range transport in the marine environment can be illustrated by the maps of dispersion of POPs entering to the ocean at certain locations. An example of the results of such calculations is presented in Fig. 3.5, where the dispersion of PCB-153 entering the ocean from the atmosphere in mid latitudes in the North-eastern Atlantic (55º-60º N, 10º-20º W) after 1-year simulations (Fig. 3.5a) and 10-year simulations (Fig. 3.5b) is shown.

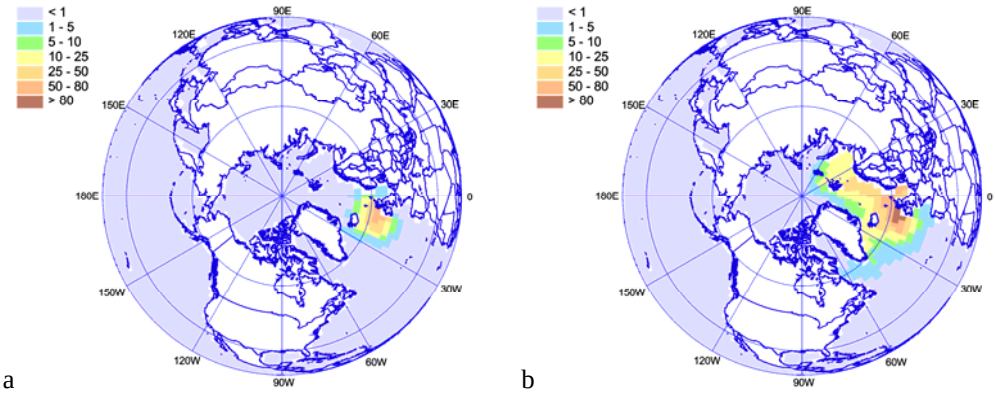


Fig. 3.5. *Spatial distribution of PCB-153 water concentrations resulting from uniform flux from the atmosphere in the Northern Atlantic after 1-year (a) and 10-year (b) simulations, % of maximum concentration*

It is seen that after 1 year the pollutant from the point of release reaches north boundaries of the Scandinavian Peninsula, and after 10 years noticeable concentrations of PCB-153 reach New Land boundaries. This testifies that marine transport can contribute noticeably to the long-range transport of POPs at least at long-range time scale.

It should be noticed that the pattern of POP dispersion due to marine transport essentially differs with the location of the entering point. To exemplify this, the results of simulation of marine transport of PCB-153 entering the ocean near the west coast of North America (near the region with intensive emissions) are shown in Fig. 3.6 for one-year (Fig. 3.6 a) and 10-year (Fig. 3.6 b) simulations.

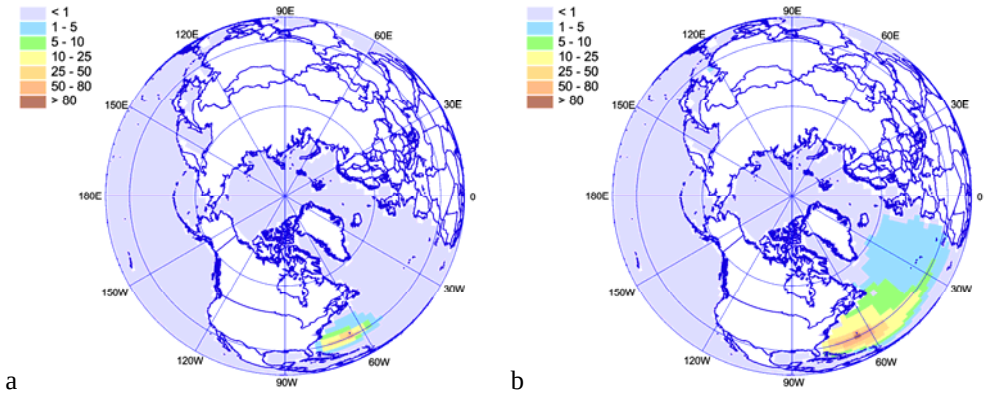
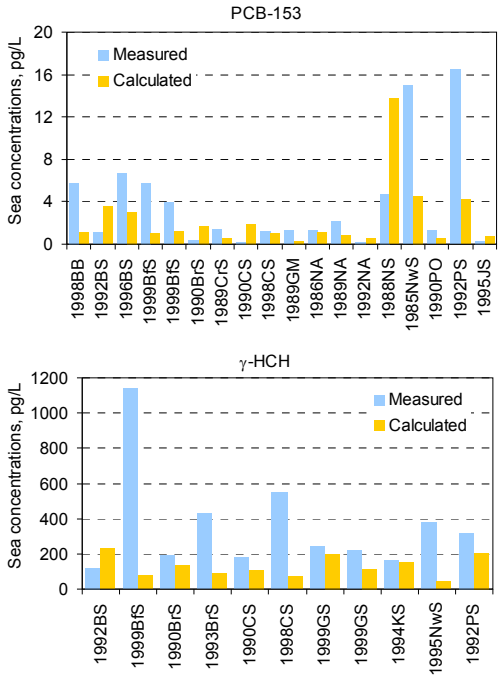


Fig. 3.6. *Spatial distribution of PCB-153 water concentrations resulting from uniform flux from the atmosphere in near the west coast of North America after 1-year (a) and 10-year (b) simulations, % of maximum concentration*

It is seen that under the influence of the Gulf Stream and further of the North-Atlantic Current the pollutant spreads from the source to the north-east reaching European borders.

The refined oceanic module was incorporated into the multicompartment MSCE-POP model. To illustrate the comparison between measured and calculated POP concentrations in seawater simulations of long-range transport from emission sources of the northern hemisphere (see subsection 2.1) was undertaken for PCB-153 (for the period from 1970 to 2000) and for γ-HCH (for the period from 1985 to 2002). Calculated seawater concentrations of these pollutants were compared with available measurements. The results of the comparison are presented in plots in Fig. 3.7.



Abbreviations:

BB	Baffin Bay
BS	Barents Sea
BfS	Beaufort Sea
BrS	Bering Sea
CrS	Caribbean Sea
CS	Chukchee Sea
GM	Gulf of Mexico
GS	Greenland Sea
KS	Kara Sea
NA	North Atlantic
NS	North Sea
NwS	Norwegian Sea
PO	Pacific Ocean
PS	Pechora Sea
JS	Sea of Japan

Fig. 3.7. *Comparison of measured seawater concentrations for PCB-153 and γ-HCH with that calculated by modified MSCE-POP model*

The analysis of the plots shows that for PCB-153 about 50% of the calculated values agree with measurements within a factor of 3 and more than 70% – within a factor of 4. For γ-HCH over 60% of calculated values agree with measurements within a factor of 2. It should be mentioned, however, that the basis of the comparison is far from being sufficient for validation of the model. For more reliable validation more measurements of POP concentrations in seawater are required. Such measurements could be obtained from measurement campaigns, oceanic cruises, etc.

The deviations of calculated data from measurements can be explained by the sufficiently high degree of averaging in the model calculations (whereas measurements normally are performed locally under specific hydrometeorological conditions), shortcomings of the model and measurement errors. E.g. it is well known [*AMAP Assessment 2002,* 2004], that PCB-153 concentrations in seawater can be distorted by the lab equipment and floating craft used in measurements. In view of these remarks the model adequately enough reflects measurements of PCB-153 and γ-HCH in seawaters.

In the case of γ-HCH, there is general underestimation of seawater concentrations. This underestimation could be explained by the fact that the arrival of γ-HCH with river flow was not taken into account under the experiment.

3.4. Development of hemispheric/regional POP modelling approach

Here the description of modelling approach to the evaluation of POP contamination levels in Europe based on combination of hemispheric and regional simulations is given. The necessity of combining regional and hemispheric simulations is conditioned by the peculiarities of POP behaviour in the environment followed by POP physical-chemical properties. Namely, a lot of POPs are characterized by significant long-range transport potential that leads to the necessity of consideration of their spreading from emission sources on the hemispheric/global scale. In addition, essential residence time in the environmental media characteristic of certain POPs leads to their ability to be accumulated there during long periods of time. As a result the re-emission of such POPs to the atmosphere can be essential and modelling approach should take into account their historical emission.

Complex modelling approach to the assessment of POP pollution levels taking into account the above peculiarities of POP fate is being developed at MSC-E. It is based on the nesting of hemispheric and regional versions of MSCE-POP model [*Gusev et al*., 2005b]. Hemispheric version of the model is used for the evaluation of POP intercontinental transport and accumulation in environmental media on hemispheric/global scale with coarse spatial resolution and for long periods of time. Regional version of the model is applied for the evaluation of POP distribution over the EMEP grid with finer spatial resolution taking into account initial and boundary conditions obtained by hemispheric calculations.

3.4.1. Meteorological and geophysical information

Hemispheric/regional multicompartment modelling approach developed for the evaluation of POP pollution levels both on the hemispheric scale and for the European region with finer resolution requires a set of input data including meteorological and geophysical information. For modelling of POP long-range transport and accumulation in environmental media the geophysical information includes data on land use/land cover, leaf area index data, organic matter content in soil, sea currents data, ice cover, and organic matter content in seawater. Available meteorological and geophysical information for model input, compatibility of data for hemispheric and regional scales, and further directions of its refinement are discussed in this section.

Meteorological data

Meteorological input for MSCE-POP model for regional and hemispheric scales is prepared by means of meteorological pre-processing systems. The set of meteorological data for the hemispheric scale with resolution 2.5˚x2.5˚ is supplied by System of Diagnosis of the Lower Atmosphere (SDA) developed by Hydrometeorological Centre of Russia [*Frolov et al.,* 1994; *Rubinstein et al*., 1997,1998; *Frolov et al.,* 1997 a,b,c]. The pre-processing of meteorological data for regional scale with resolution 50x50 km^2 is based on the PSU/NCAR mesoscale model MM5 [*Grell et al*., 1995; http://www.mmm.ucar.edu/mm5/overview.html]. These systems utilize input meteorological data with rough spatial resolution and perform short-term weather forecasts for the POP transport model grid, which are stored for the subsequent use in POP transport modelling. Preparation of regional and hemispheric meteorological information is based on one and the same set of initial meteorological information, namely, on NCEP/DOE re-analysis data [*Kanamitsu et al.,* 2002].

To improve the consistency between hemispheric and regional meteorological data it would be reasonable to use one meteorological pre-processing system based on the nested modelling approach. Such system will permit to obtain necessary meteorological input data with coarse and fine

spatial resolution for subsequent modelling of POP fate on the hemispheric and regional scales. As input information for meteorological data preprocessing it is planned to consider application of reanalysis data of ECMWF.

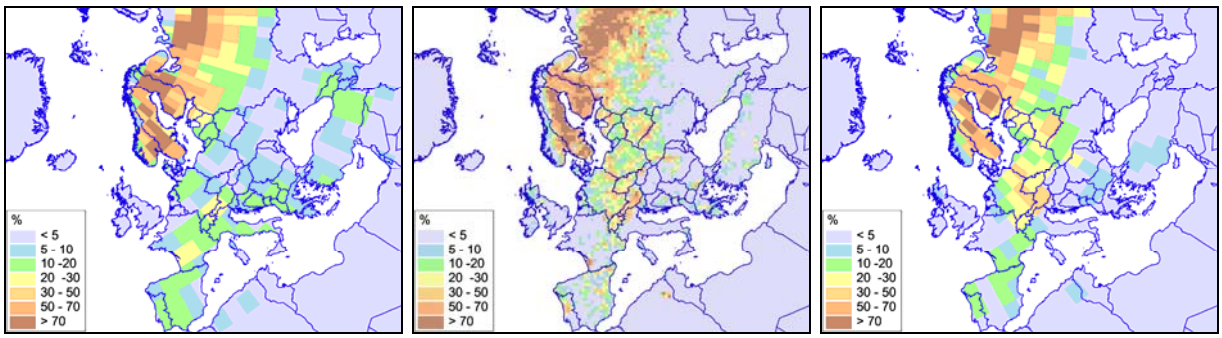
Land cover data

Land cover data is an important information for the evaluation of POP ecosystem-specific depositions and gaseous exchange between the atmosphere and underlying surface of different types.

For regional scale modelling, 17-category land use/land cover dataset developed by the Coordinating Centre for Effects (CCE) is used. The dataset is partly based on the database developed in the framework of EC Programme on Coordination of Information on the Environment (CORINE) [*dataservice.eea.eu.int*]. Since the CORINE Land Cover data do not cover entire EMEP area, Stockholm Environment Institute (SEI) database was used to fill the gaps [http://www.york.ac.uk/inst/sei/APS/projects.html]. In order to unify the CORINE and SEI inventories an ecosystem classification EUNIS (European Nature Information System) was adopted. For hemispheric scale modelling 24-category USGS Land Use/Land Cover dataset obtained from NCAR Mesoscale Modelling System (MM5) [*Guo and Chen*, 1994] is used. Both sets provide information on fractions of the cell area covered by particular types of underlying surface.

Regional and hemispheric versions of MSCE-POP model require usage of seven general categories of land cover, namely, deciduous forests, coniferous forests, grassland, bare land, urban and built-up land, glaciers, and water bodies. The comparison of spatial distributions of land use data for regional and hemispheric scales shows that, in general, these two sets of data are compatible. For the numerical evaluation of the compatibility, spatial distribution of land use types at regional scale was interpolated to the hemispheric grid and compared with spatial distribution of USGS dataset. Only the grid cells with more than 90% of intersection with the EMEP grid were taken into account. Significant correlation (about 0.9) was obtained between the major part of land use categories used in hemispheric and regional scale datasets. The comparison of spatial patterns of land use categories is exemplified by spatial distribution of coniferous forest in Fig. 3.8.

It can be seen that spatial patterns of coniferous forests distribution in the hemispheric (Fig. 3.8a) and regional (Fig. 3.8b) scale datasets are quite close to each other. For the comparison Fig. 3.8c presents the regional scale distribution of coniferous forest interpolated into hemispheric MSCE-POP model grid.



a b c

Fig. 3.8. *Fractions of coniferous forest according to hemispheric (a) and regional (b) model versions; (c) - recalculation of the fraction used in the regional model to the hemispheric grid, %*

It should be mentioned that currently the compatibility of land cover data used in hemispheric and regional versions of the MSCE-POP model can be considered as satisfactory. However it is evident that for the combined usage of hemispheric and regional modelling it is important to use the same set of land cover/land use information.

Leaf area data

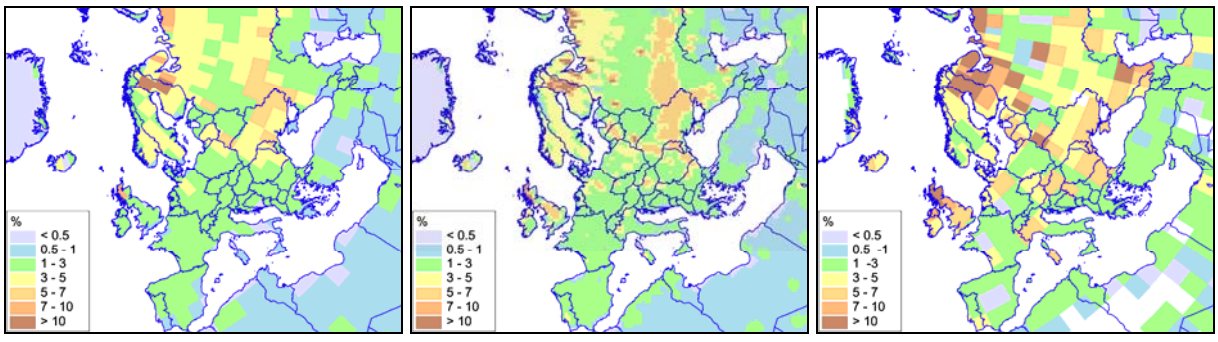
Leaf area data for hemispheric and regional MSCE-POP model grids were obtained on the basis of geographically resolved leaf area index data with monthly resolution of NASA Goddard Space Flight Center [*Sellers et al.,* 1994, 1995]. These data were interpolated from initial 1°×1° grid to 50×50 km and 2.5°×2.5° model grids used in regional and hemispheric versions of MSCE-POP model. Consistency of these data in relation to the land cover information was investigated by correlation analysis.

Organic carbon content in soil

For both regional and hemispheric model versions, the spatial distribution of organic carbon content in soil was constructed on the basis of spatial distribution of soil types and their specific organic matter content. The spatial distribution of soil types was adopted from the GIS dataset "Global Distribution of FAO Soil Units at 1°×1° Resolution", which distinguishes 27 soil types [*Zobler*, 1986; *http://www.giss.nasa.gov/data/landuse*].

The organic carbon content for individual soil types was obtained on the basis of the study of Fraters, 1993. Following these data the fraction of organic carbon in soils for all grid cells both for regional and hemispheric model grids was calculated.

The spatial patterns of organic carbon content in soils for hemispheric scale with resolution 2.5°×2.5°and for regional scale with resolution 50x50 km^2 is presented in Fig. 3.9a and b, respectively. Hemispheric scale data provide smoother distribution of organic carbon content. It should be taken into account that the values of content can essentially vary within one and the same cell of the hemispheric grid. The difference between the values for 50×50 km^2 grid within one and the same hemispheric grid cell can reach 7 – 10% with typical value in the EMEP region about 3-5%. This shows that for more accurate evaluation of soil contamination and, as a consequence, of re-emission flux from soil to the atmosphere it is reasonable to use regional version of the model with finer spatial resolution. The ranges of variation of organic carbon content in soil within one and the same hemispheric cell (the difference between maximum and minimum values of organic carbon content) are shown in Fig. 3.9c.



a b c

Fig. 3.9. *Fractions of organic carbon content in soil for hemispheric model grid (a) and regional model grid (b) and ranges of variation of organic carbon content in hemispheric model grid cells (c), %*

3.4.2. Evaluation of POP pollution levels on hemispheric scale and for European region

Developed hemispheric/regional modelling approach is based on one-way nesting of hemispheric and regional versions of MSCE-POP model. This means that initial and boundary data for regional model with finer resolution are obtained by calculations with hemispheric version of the model taking into account emission sources of the entire northern hemisphere. Nesting of hemispheric and regional scale modelling allows describing spatial distribution of POP contamination within the European region with finer spatial resolution using fine resolution input data (meteorology, land use and land cover).

To evaluate POP pollution levels for a particular year, there is a need to consider in model computations some preceding period, during which a chemical is accumulated in and re-emitted from environmental media (soil, seawater, vegetation, etc). Thus model simulations should be carried out for two periods of time, which can be referred as the evaluation period and the preceding period. The length of the preceding period can be defined taking into account physical-chemical properties of the considered POP, in particular, rate of accumulation and degradation in environmental media.

To obtain initial conditions for regional MSCE-POP model, POP content in environmental media is calculated by simulations of transport and accumulation of a pollutant within the preceding period. Then the result is interpolated from the coarse hemispheric grid into the fine resolution EMEP grid. For obtaining boundary conditions for regional scale model the daily mean POP concentrations along the lateral and upper boundaries of the EMEP grid are derived from hemispheric model run. This distribution of concentrations is interpolated into the finer resolution EMEP grid and stored for subsequent use in regional scale model run. The procedure used for interpolation of concentrations from coarse hemispheric grid into finer regional grid takes into account the differences in the co-ordinate systems applied in hemispheric and regional model versions.

The typical example of the computation scheme for the evaluation of pollution levels for 2004 is shown in Fig. 3.10. To obtain annual and seasonal levels of PCDD/F concentrations and depositions for 2004 the preceding period is conventionally chosen to be 1990-2003.

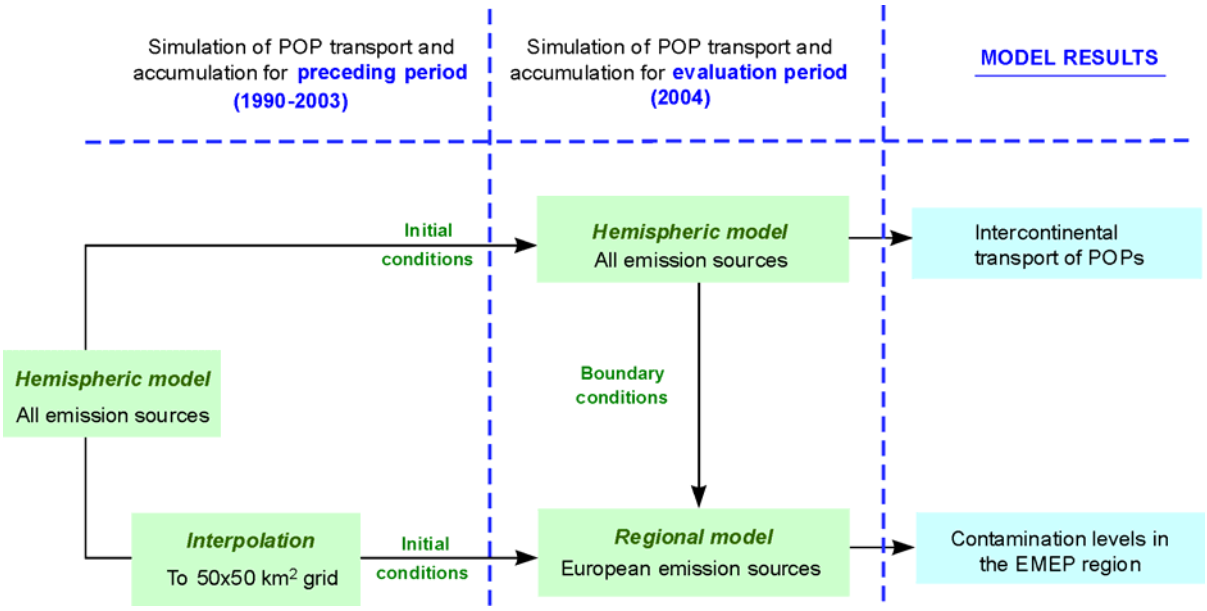


Fig. 3.10. *The computation scheme of complex hemispheric/regional evaluation of POP contamination levels based on one-way nesting of hemispheric and regional versions of MSCE-POP model*

The evaluation of pollution levels in 2004 begins with the simulation of POP transport on hemispheric scale from all emission sources within the northern hemisphere for the period 1990 – 2003. Concentrations in main environmental compartments obtained to the end of this run are interpolated to 50×50 km^2 grid for subsequent use in regional modelling. The hemispheric model run for 2004 is performed with the initial conditions generated by the previous model run for the period 1990-2003. During this model run boundary conditions at the borders of the EMEP grid for the subsequent regional scale model are generated. Using generated initial and boundary conditions for 2004 regional modelling of POP transport within the EMEP region is performed for the evaluation of contamination levels in the EMEP region with finer spatial resolution (50×50 km^2). To evaluate intercontinental transport of a particular POP from the emission sources to the receptors, separate hemispheric model runs can be performed for each of the selected emission source. Application of this approach can lead to some uncertainties in modelling results due to the model non-linearity. Evaluation of these uncertainties on the basis of MSCE-POP model results presented in [*Gusev et al*., 2006] showed that the differences obtained were within the range of several percents.

To check the applicability of the developed hemispheric/regional modelling approach, evaluation of contamination levels for one of 17 toxic congeners of PCDD/Fs was undertaken. For simulations the congener 2,3,4,7,8-PeCDF with maximum fraction in emission toxicity (about 40%) was chosen. For these illustrative calculations the length of the preceding period is set up to 13 years (1990-2002). This period is also characterized by availability of official information on PCDD/F emission submitted by European countries. Since the half-life of selected PCDD/F congener in soil is about 60 years, for more realistic computations this period should actually be longer. Modelling was based on the PCDD/F emission data for the European and North American regions described in subsection 2.1 above. For both regions official data on emissions of PCDD/Fs were applied. In case when officially reported data were missing, expert estimates of emission were used. Though available emission data cover only part of the northern hemisphere the results obtained can be used to analyse the importance of atmospheric transport of POPs across the Northern Atlantic and to evaluate the contribution of North American emission sources to the pollution of the European region. Spatial distribution of emissions used in calculations is shown in Fig. 3.11a.

The results of calculations are displayed in Figs. 3.11 b – d. Namely, Fig. 3.11b shows spatial distribution of annually averaged air concentrations of the selected congener obtained by the hemispheric model.

The comparison of the map in Fig. 3.11b with the map of spatial distribution of air concentrations obtained by the regional model run for 2003 with initial and boundary conditions (Fig. 3.11c) shows that this spatial distribution is compatible with that obtained by the hemispheric model. From the other hand, regional run without initial and boundary conditions and the same emission data (Fig. 3.11d) shows considerable underestimation of 2,3,4,7,8-PeCDF air concentrations. This illustrates the importance of including to the evaluation process both the sources outside the EMEP region, and from the re-emission of pollutant accumulated in the environmental media of the northern hemisphere earlier.

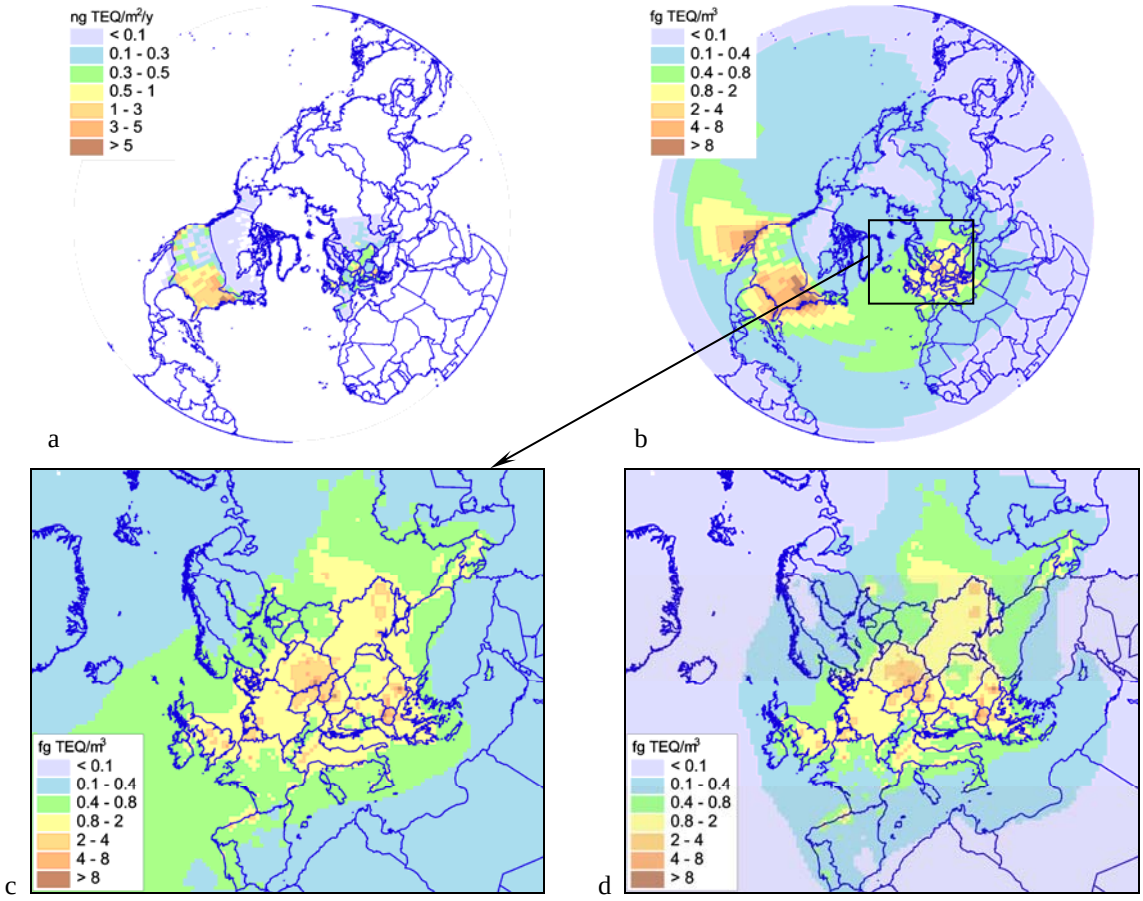


Fig. 3.11. *Spatial distribution of annual total emission of 2,3,4,7,8-PeCDF for 2003, ng TEQ/m^2/y (a) and annual mean 2,3,4,7,8-PeCDF air concentrations obtained in different model runs: results of hemispheric model run for the northern hemisphere (b), regional model run with initial and boundary conditions provided by hemispheric model (c), regional model run with European emission sources only (d), fg TEQ/m^3*

3.4.3. Evaluation of transboundary transport for European region

To evaluate transboundary transport of POPs the following peculiarities of their dispersion in the environment should be taken into account. Many of POPs are pollutants of global concern. Due to their physical-chemical properties the dispersion of POPs in the environment has a number of peculiarities. In the atmosphere POPs can present in gaseous and particle-bound phases. Being emitted to the atmosphere from different sources, POPs are transported to the receptors by atmospheric transport and deposited to the environmental media (soil, seawater, vegetation). Later on, POPs accumulated in soil, seawater and vegetation are re-emitted to the atmosphere and undergo further cycles of atmospheric transport, deposition and re-emission.

Thus, for the evaluation of contributions to the pollution of any particular receptor (European country) there is a need to take into account the influence of several groups of POP emission sources. These are:

- European anthropogenic emission sources (annual anthropogenic emissions of each European country);
- Remote anthropogenic emission sources (annual anthropogenic emissions within the northern hemisphere, i.e. in North America, Asia, etc.);

- Re-emission of POPs from environmental media to the atmosphere (re-volatilisation of POPs accumulated in environmental media within the northern hemisphere in the preceding and evaluation periods of time). Re-suspension of POPs with soil particles is not currently taken into account and is supposed to be considered in the nearest future.

To describe transboundary transport of POPs between the European countries with rather fine spatial resolution (50×50 km^2) and to evaluate contributions of remote sources including re-emission outside the European region a combined hemispheric/regional scale modelling is required.

Distinguishing between contributions created by anthropogenic emission sources and re-emission is important since anthropogenic emissions can be regulated in contrast to re-emission being a result of POP releases, which took place earlier. However re-emission can support the levels of air pollution for a long periods depending on the residence time of a particular POP in environmental media.

Taking all this into account the evaluation of transboundary transport of POPs for the European region is made on the basis of hemispheric/regional modelling approach described in subsection 3.2 above. This modelling approach permits to describe POP contamination levels in the northern hemisphere with emphasis to the European region. Also it allows estimating contributions caused by transboundary fluxes from European countries, intercontinental transport from remote sources, and re-emission to the pollution of particular receptor. The scheme of the approach developed for the evaluation of POP transboundary transport within the European region is presented in Fig. 3.12.

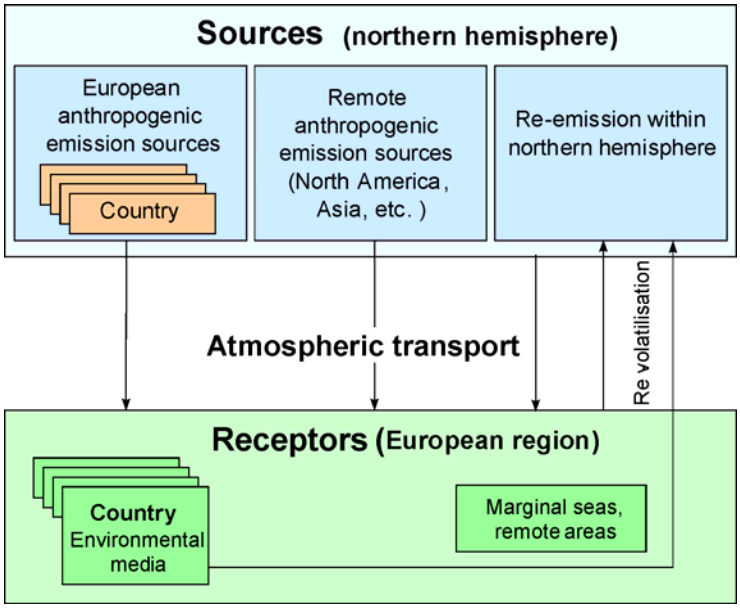


Fig. 3.12. *Evaluation of transboundary transport of POPs within the European region*

Currently within this modelling approach European countries are used as the receptors. However, it is possible to consider more detailed splitting of the emission sources and receptors, for example, the administrative units of countries, and to include additional receptors like marginal seas.

For the evaluation of source-receptor relationships, POP mass emitted by each emission source to the atmosphere (each European country, remote emission sources, re-emission) is considered separately. The contributions of annual emissions of each European country to each receptor are tracked during the period of computations. These contributions are presented in the form of country-to-country matrices for annual air concentrations and depositions. Thus using these matrices it is possible to characterize annual transboundary fluxes of POPs due to anthropogenic emissions of European countries. Along with that the matrices contain the contributions of two additional sources: remote emission sources and re-emission.

Remote emission sources include annual anthropogenic emissions from sources of other continents, i. e. North America, Asia, etc. Currently their effect is considered as aggregated contribution of all annual anthropogenic emissions outside the EMEP grid. However developed hemispheric/regional modelling approach permits to consider contributions of particular continents or regions within the northern hemisphere.

The re-emission is represented by annual POP flux from the environmental media resulted from POP accumulation in, for example, soil or seawater, during the whole simulation period. It depends on different factors like physical-chemical properties of particular POP, properties of environmental media, and temporal variations of emissions. Within the model the re-emission flux is dynamically computed at each model time step and for each grid cell for particular environment compartment. This allows to obtain the contribution of re-emission to the pollution of each European country. At current stage the evaluation of POP transboundary transport does not consider contributions of particular European country to overall re-emission flux. At the same time it is possible to trace the fate of POP emission of each European country or the particular emission sources within long period of time in case when historical emission of countries is known.

At present the contributions of all the emissions outside the EMEP grid are aggregated to compare the influence of annual anthropogenic emissions of European countries with contributions of re-emission within the European region and emission sources of the northern hemisphere beyond the EMEP grid. However developed modelling approach is capable to distinguish contributions of anthropogenic emissions of various remote sources to the pollution levels in European countries.

Technical aspects of calculations of source-receptor relationships within the EMEP domain are considered in more detail in [*Gusev et al.,* 2006].

4. EVALUATION OF POP TRANSPORT AND POLLUTION LEVELS IN THE ENVIRONMENT

In accordance with the Work-plan of MSC-E, this year the environmental contamination and transboundary transport of PAHs (4 indicator congeners) – benzo[a]pyrene (B[a]P), benzo[k]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F) and indeno[1,2,3-cd]pyrene (I_P), PCDD/Fs (toxic congeners), PCBs, γ-HCH and HCB in 2004 were evaluated. The calculations were performed by means of the improved version of MSCE-POP model. The improvements concern the refinement of physical-chemical properties of the pollutants, improvement of model description of POP degradation in the atmosphere, removal with precipitation (including scavenging with snow) and partitioning in soil.

Modelling of EMEP region pollution by PCDD/Fs was carried out with the help of hemispheric/regional approach to take into account the contributions of hemispheric sources located outside the EMEP region and the contribution of re-emission. Since the considered four indicator PAH congeners are present in the atmosphere mainly in particle-bound form, calculations for PAHs were carried out at regional scale.

In the examination of PCBs, γ-HCH and HCB the emphasis was put on evaluation of intercontinental transport and pollution of remote regions. The comparison of calculated results with available measurements is presented in detail in [*Shatalov et al.,* 2005].

4.1. Pollution levels and source-receptor relationships in the EMEP domain

This section presents the results of the evaluation of pollution levels of PCDD/Fs and four indicator PAHs (B[a]P, B[b]F, B[k]F, and I_P) within the EMEP region in 2004. For B[a]P and the "indicator congener" of PCDD/Fs (2,3,4,7,8-PeCDF, see [*Dutchak et al.,* 2004]) source-receptor relationships in 2004 were investigated.

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

Pollution levels in the EMEP region

Calculations of pollution levels of polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs) in the EMEP region were performed for four congeners: 2,3,4,7,8-PeCDF, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD and 1,2,3,4,6,7,8-HpCDF. Usage of the above four congeners allows evaluating spatial distributions of the total PCDD/F toxicity with difference not more than 20% for concentrations in the main environmental compartments (the atmosphere, soil, seawater, vegetation) compared with the results of simulations of all 17 toxic congeners, see EMEP Status Report [*Dutchak et al.,* 2004]. The emission data used in the calculations are based on official data or on expert estimates in case when official information is not available (subsection 2.1).

Evaluation of pollution levels of PCDD/Fs in Europe was carried out with the help of hemispheric/regional approach (subsection 3.2). Namely, first PCDD/F transport for the period from 1990 to 2003 was simulated by hemispheric model version to obtain initial conditions for regional simulations. Spatial distribution of depositions to the northern hemisphere in 2003 in comparison with emissions used in modelling for one of the above congeners (2,3,4,7,8-PeCDF) is shown in Fig. 4.1.

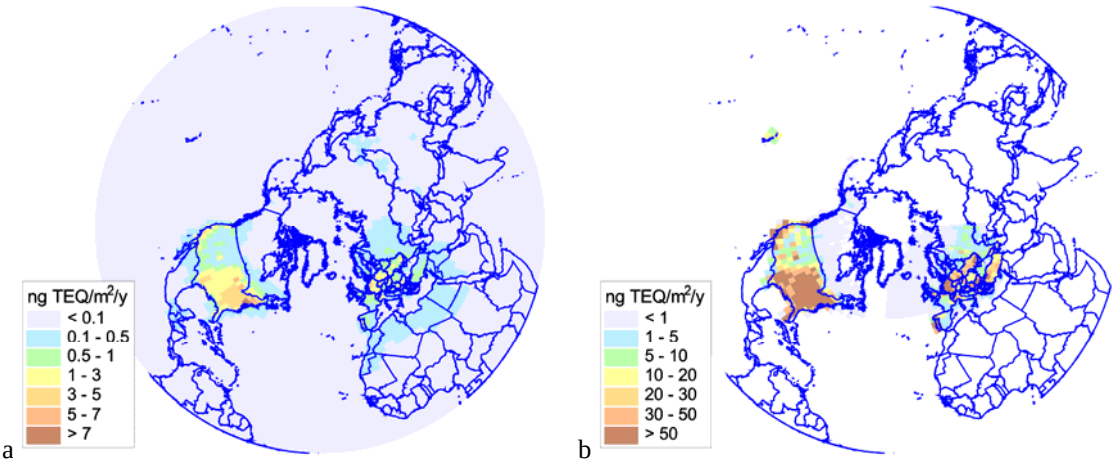


Fig. 4.1. *Deposition flux of 2,3,4,7,8-PeCDF in 2003 within the northern hemisphere (a) in comparison with emissions (b), ng TEQ/m^2/y*

Then, simulations for 2004 for the above four PCDD/F congeners were carried out by the regional model with resolution 50x50 km^2 with obtained boundary and initial conditions. As an example, calculated values of air concentrations in the surface atmospheric layer for four selected congeners are shown in Fig. 4.2 (in toxicity units).

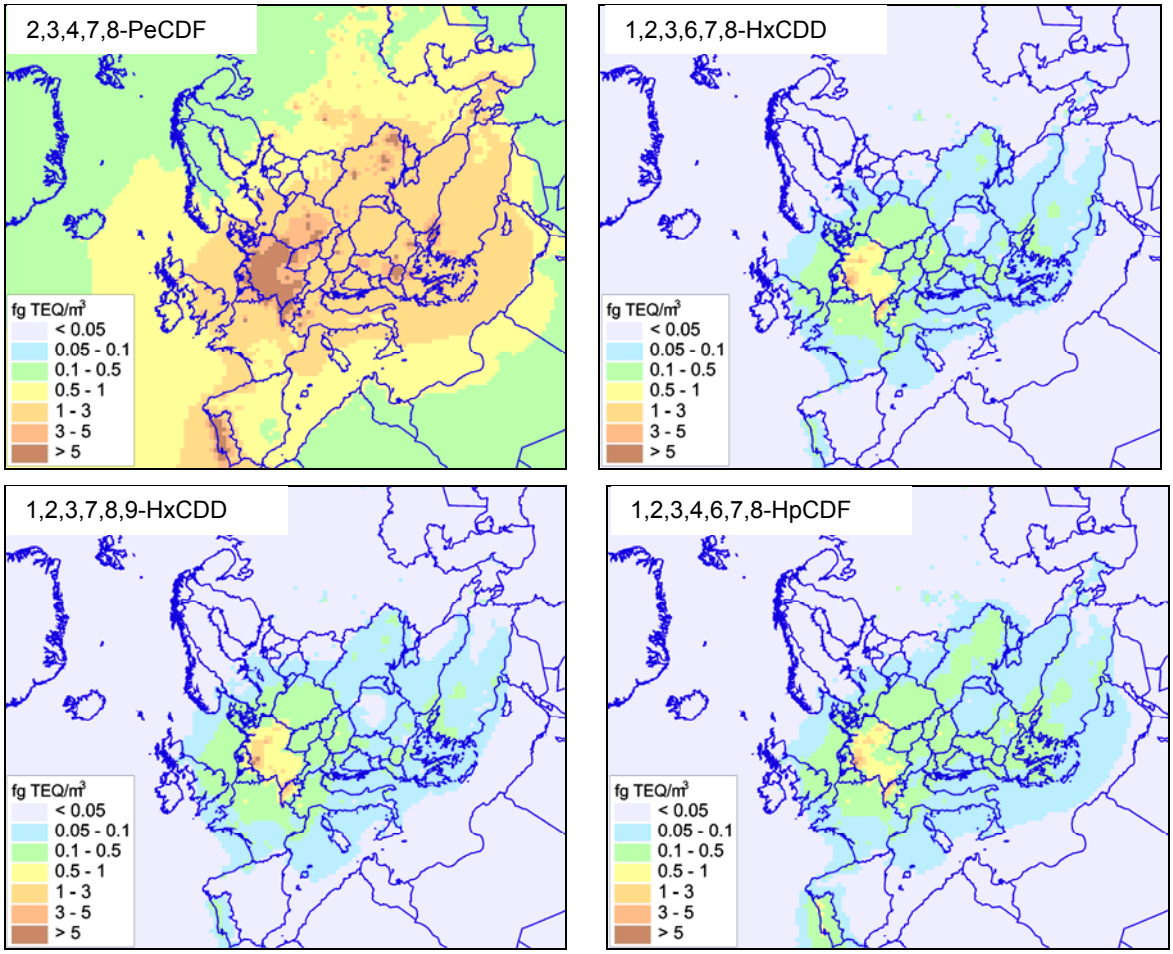


Fig. 4.2. *Concentrations of four selected PCDD/F congeners in the surface atmospheric layer in 2004 calculated on the basis of hemispheric/regional approach, fg TEQ/m^3*

It is clear that spatial patterns of depositions for selected congeners are different. Highest levels of contamination are calculated for 2,3,4,7,8-PeCDF contributing about 40% of the overall toxicity in emissions (totally about 4200 g TEQ/y in Europe). Emissions of the three rest congeners are about the same (200 g TEQ/y in Europe). In spite of similarity in emission totals, spatial distributions of depositions are different. For example, contamination levels in Ukraine are higher for 1,2,3,4,6,7,8-HpCDF than for two HxCDD congeners.

Usage of the selected four congeners allows taking into account differences in physical-chemical properties of various toxic PCDD/F congeners while evaluating pollution levels in the EMEP domain. To estimate spatial distributions of air concentrations and depositions for the sum of all 17 toxic PCDD/F congeners it is assumed that the whole mixture consists of the four selected congeners with emissions proportional to their emissions estimated in subsection 2.1. The proportionality coefficients are chosen in so that the sum of emission totals of these four congeners coincides with emission total for the overall PCDD/F mixture.

As an example of the results of these calculations, the maps of spatial distribution of PCDD/F toxicity in depositions in the EMEP region calculated with the help of this approach in comparison of emission data are presented in Fig. 4.3.

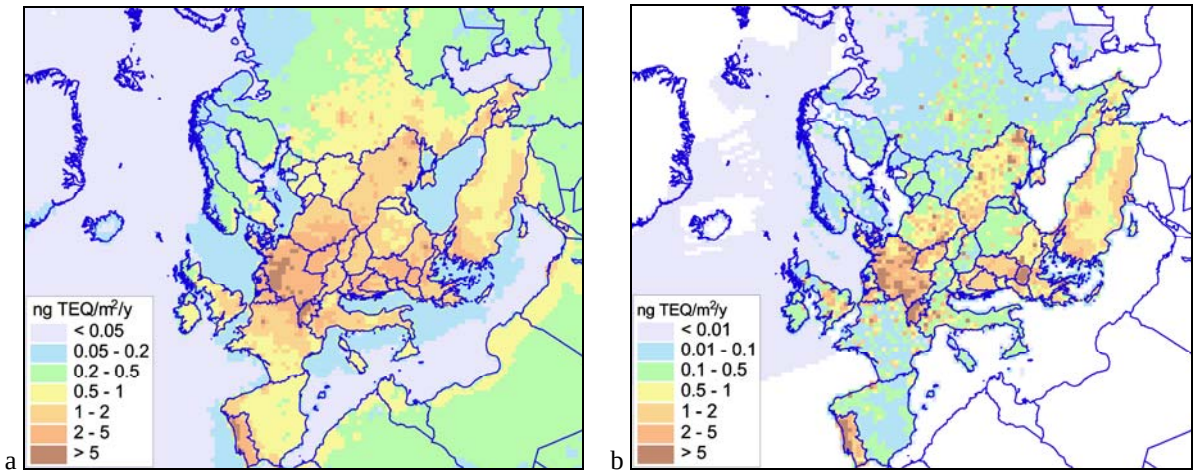


Fig. 4.3. *Calculated deposition fluxes (a) of PCDD/F mixture in 2004 in comparison with emissions (b), ng TEQ/m^2/y*

The levels of net deposition flux in Europe differ from about 0.1 ng TEQ/m^2/y in northern Europe (Norway, northern parts of Sweden and Finland) to 5 ng TEQ/m^2/y and higher in central and southern Europe (Germany, Poland, Czech Republic). Maximum pollution levels are calculated for Germany and neighbouring countries. The reason for that is high values of officially reported emission levels in Germany (about 30% of total PCDD/F toxicity in Europe). High values of deposition levels (0.5 – 2 ng TEQ/m^2) due to high emission densities are calculated for Turkey and Ukraine.

The comparison of spatial distributions of depositions and emissions in Europe (Figs. 4.3.a and b, respectively) shows that spatial distribution of European emissions strongly influences pollution levels in Europe. However, as will be shown further, the inputs of other emission sources (hemispheric anthropogenic sources located outside the EMEP region and re-emissions due to long-term accumulation) also contribute noticeably to the contamination of some European countries.

It should be taken into account that pollution levels calculated in the current year are essentially higher than the results obtained in 2005. First, the total value of refined emissions of PCDD/Fs in Europe is about 2.4 times higher than that used in 2005. Second, the contributions of non-EMEP sources (USA and Canada) and of re-emissions were taken into account in calculations of the current year. Emission data for PCDD/F require further refinement for sources both of European countries and of the entire northern hemisphere.

Comparison with measurements

Calculated atmospheric concentrations were compared with available measurements. Since measurements of PCDD/Fs are not presently included in EMEP monitoring programme, measurements obtained from various campaigns and national activities were used. Measurements at various rural/background locations within the period from 1990 to 2004 (totally 59 measured values) were included into the comparison. It was found that for the congener 2,3,4,7,8-PeCDF (with 40% contribution to the overall PCDD/F toxicity) the median of frequency distribution of calculated-to-measured ratio is about 0.8. About 55% of calculated values agree with measurements within a factor of two and about 75% – within a factor of three. So, taking into account possible uncertainties in emission and measurement data, the agreement between measurements and calculations for this congener can be concluded to be reasonable. The model slightly underestimates obtained values of PCDD/F air concentrations. In addition values of calculated air concentrations were compared with measurements at urban sites (totally 44 measurements). Here the model shows concentrations about 5 times lower than measurements. This corresponds to the difference between PCDD/F contamination levels at urban and background locations. Similar results were obtained for 1,2,3,4,6,7,8-HpCDF. For the congeners 1,2,3,6,7,8-HxCDD and 1,2,3,7,8,9-HxCDD the model underestimates air concentrations about three times. This may be conditioned by the fact that the congener composition of emissions was quite rough (Section 2). For the refinement of the results on evaluation of air contamination by PCDD/Fs further work on emission inventories including seasonal variations is needed.

For the evaluation of reliability of hemispheric calculations, additional comparison of these calculations with measurements for monitoring sites located outside the European region was performed. Calculated air concentrations at Canadian sites appeared to be underestimated several times by the model whereas air concentrations at the US measurement sites are overestimated. This is conditioned by the fact that for Canada official emission data was used in contrast with the USA where expert estimates of PCDD/F emissions for 1995 are only available (see Fig. 4.1b). Refinement of emission data in North America should further improve calculation results.

Transboundary transport

Here we present evaluation of contributions of different source categories to the pollution levels of each European country.

To take into account contributions of emission sources located outside the EMEP region, the above-described hemispheric/regional approach was used. To evaluate the influence of re-emission process due to accumulation of PCDD/Fs in the environmental media prior to 2004, preceding computations were carried out for the period 1990-2003 by hemispheric model with anthropogenic emission sources of the northern hemisphere. So, the following sources are considered for evaluation of source-receptor relationship for PCDD/Fs:

- Anthropogenic sources of each European country.
- Anthropogenic source including emissions outside the EMEP region (further referred as non-EMEP sources).
- Re-emission due to accumulation of PCDD/Fs in the environmental media from anthropogenic emission sources of the entire northern hemisphere during the period from 1990 to 2003.

Each European country is considered as a separate receptor.

Source-receptor matrices for PCDD/Fs in 2004 were calculated both for deposition and for air concentrations. The calculations of source-receptor matrices for PCDD/Fs were made with the use of physical-chemical properties of the "indicator" congener 2,3,4,7,8-PeCDF. The possibility to use the properties of this congener in calculations of source-receptor relationships for PCDD/Fs was substantiated earlier (see [*Dutchak et al*., 2004]). The assumption that all PCDD/F mixture emitted in the northern hemisphere consists of this congener leads to the uncertainty in source-receptor matrix of several per cent. The data on source-receptor matrix for air concentrations are available in the Internet (www.msceast.org).

Below we illustrate source-receptor relationships by the example of deposition matrix.

The main output of the model calculations, which allows obtaining more detailed source-receptor information, is the set of spatial distributions of pollution caused by each particular source (anthropogenic emissions of each European country, anthropogenic emissions of all non-EMEP sources and re-emissions). Spatial distributions of 2,3,4,7,8-PeCDF annual depositions originated from anthropogenic emission sources are exemplified by some of European countries (Austria, Finland and Italy) (Fig. 4.4).

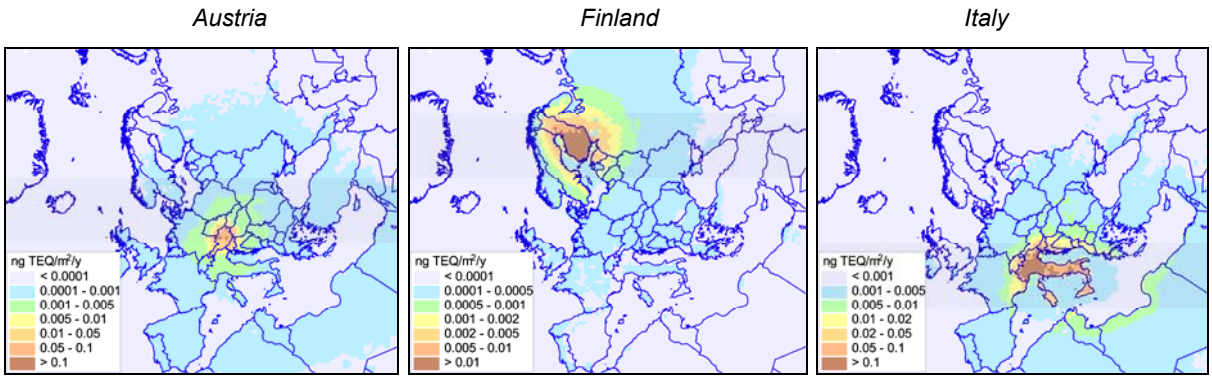


Fig. 4.4. *Annual total depositions of 2,3,4,7,8-PeCDF originated from national emissions of Austria, Finland and Italy in 2004, ng TEQ/m^2/y*

Spatial distributions of depositions caused by anthropogenic emissions of non-EMEP sources and re-emissions are displayed in Fig. 4.5.

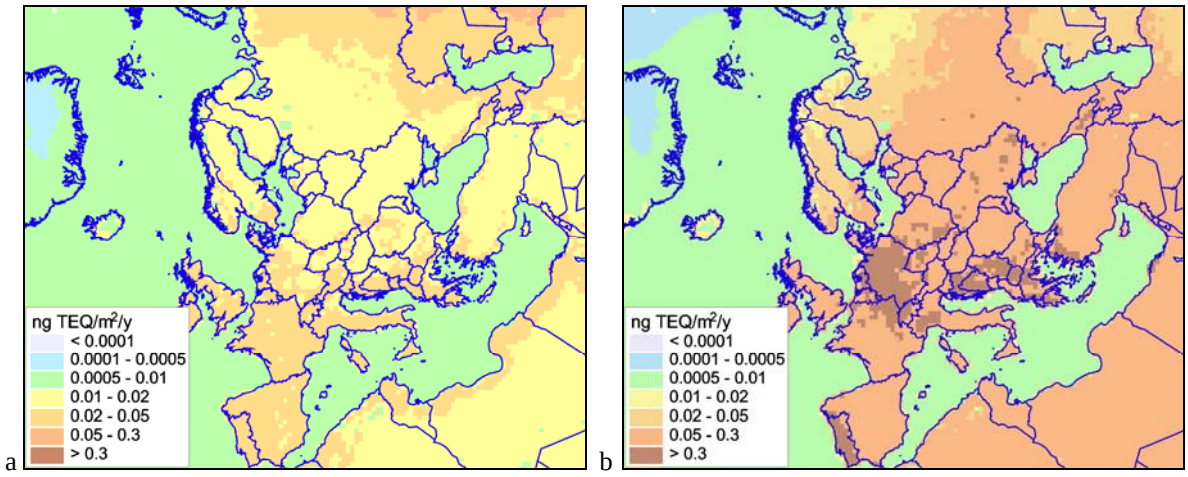


Fig. 4.5. *Annual total depositions of 2,3,4,7,8-PeCDF originated from non-EMEP (a) sources and re-emissions (b) in 2004, ng TEQ/m^2/y*

On the basis of spatial distribution of depositions caused by each source, source-receptor relationships in the EMEP region were evaluated. Further we distinguish export of pollution from and import to each European country.

Export means the transboundary transport from anthropogenic emission sources of each particular country to the territories of other countries. It is characterized by

- spatial distribution of pollution levels originated by annual anthropogenic emissions of each particular country in 2004 with resolution 50×50 km (Fig. 4.3);
- fractions of depositions from emission sources of a particular country to the entire EMEP region deposited to the territories of all other countries.

Import is the set of contributions of particular sources (including non-EMEP sources and re-emissions) to deposition levels in a given country (receptor). It is characterized by

- contributions of anthropogenic emissions of each European country and of non-EMEP sources to deposition levels in a given country;
- contribution of re-emission to the pollution of each considered receptor;
- spatial distribution of contributions of external sources to the pollution levels for each considered receptor (50×50 km).

Below the results of calculations of source-receptor relationships are presented.

Export. Export can be first characterized by the fraction of depositions to the entire EMEP region due to emissions of the given country deposited to the territory of all the rest countries (export fraction). The plot of these fractions for all European countries is displayed in Fig. 4.6.

These fractions depend significantly on the geographic location of a given country, size of its territory, and spatial distribution of emission sources within the country. Typically, about 40 – 60% of depositions caused by national emission sources of a country are deposited outside this country. Significant export fractions (about 80% and more) are characteristic of Iceland, Luxembourg, Monaco, and Denmark.

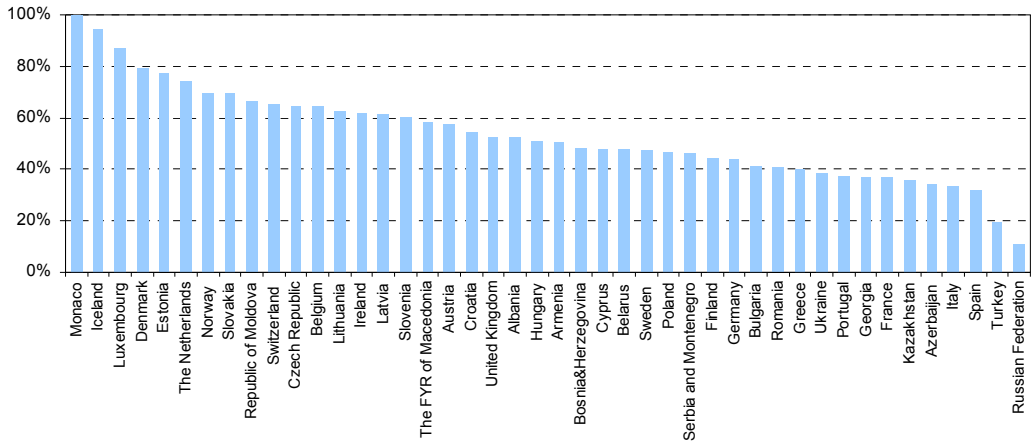


Fig. 4.6. *Export fractions of 2,3,4,7,8-PeCDF for European countries, %*

The information on the export can be presented in a more detailed way representing the distribution of depositions due to the country's sources between surrounding countries (Fig. 4.7).

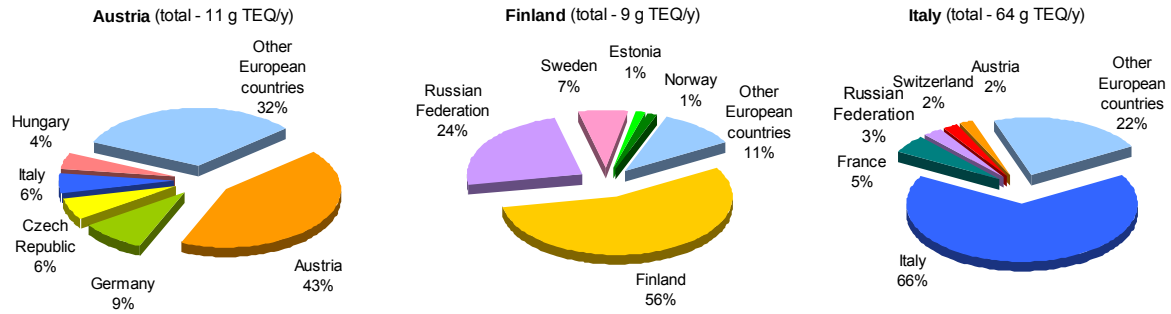


Fig. 4.7*. Transboundary transport from some countries, % of depositions*

For example, for Austria over 50% of depositions originated from national sources (totally 11 g TEQ) are deposited outside the country (transboundary depositions). These transboundary depositions are distributed between Germany, the Czech Republic, Italy and Hungary (25% altogether) and other European countries (32%).

Import. Spatial distributions of pollution originated from European countries, exemplified in Fig. 4.4, can be summarised with the aim to obtain total annual depositions from European emission sources for each of considered receptors. Thus, it is possible to compare value of pollution originated from national emission sources with the value of pollution caused by transboundary transport from other European countries (import). In Fig. 4.8 the contribution of transboundary transport to total annual depositions of 2,3,4,7,8-PeCDF from European emission sources over particular country is presented (blue bars). The plot also shows contributions of atmospheric transport from non-EMEP sources (see Fig. 4.1b above) located outside the EMEP region within the northern hemisphere (yellow bars). To reveal the magnitude of the atmospheric transboundary transport of anthropogenic emissions (from European countries and non-EMEP sources), the contributions of re-emission are not taken into account in this diagram.

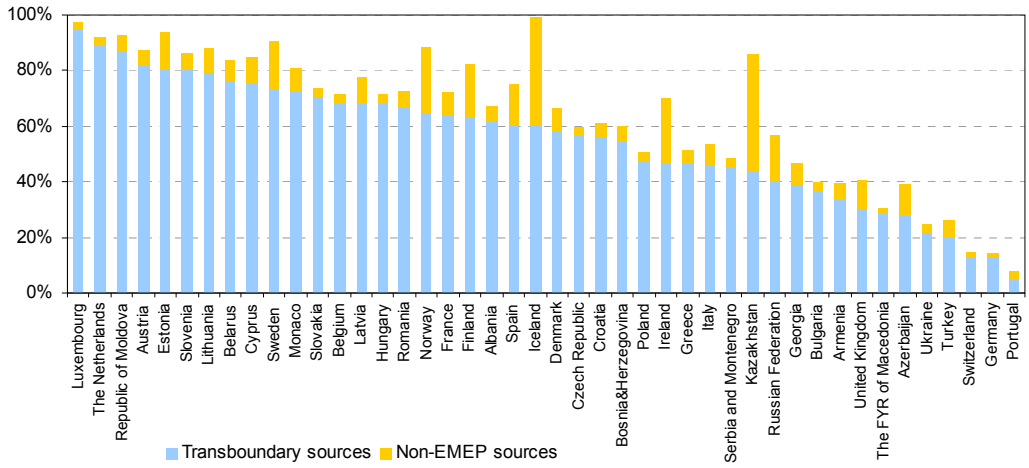


Fig. 4.8. *Contributions of transboundary transport to total annual depositions of 2,3,4,7,8-PeCDF from European emission sources for each European country, %*

Contributions of other European countries to the deposition levels of a particular country are significant for the majority of European countries. Typically they vary from 20% to 80% (Fig. 4.8). Besides, for some countries the contribution of atmospheric transport from non-EMEP sources (hemispheric transport) is also noticeable. For countries located near EMEP boundaries such as Iceland, Ireland, Norway and Finland this contribution could be about 20% or higher.

Similarly to the export, the information on import of 2,3,4,7,8-PeCDF depositions to European countries due to atmospheric transport can be presented in a more detailed way. Namely, for each country the fractions of total annual deposition determined by emissions of other European countries are calculated. This information is exemplified below by import diagrams for Austria, Finland and Italy (Fig. 4.9).

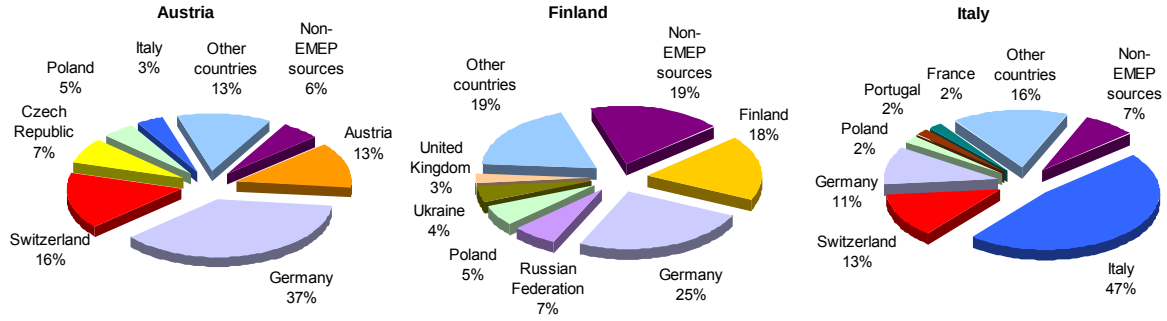


Fig. 4.9. *Import of 2,3,4,7,8-PeCDF depositions originated from atmospheric transport for Austria, Finland, and Italy*

These countries are located in different parts of the European region, namely, in northern, central, and southern Europe. For Austria and Finland the contribution of atmospheric transboundary transport is significant while national emissions contribute not more than 20%. In case of Italy the contribution of transboundary transport from other European countries is about 50%. Besides, calculations show significant contribution of non-EMEP sources to the depositions to Finland (about 20%).

Contribution of re-emission. As it was mentioned above, the process of re-volatilisation of POPs earlier accumulated in the environmental media is considered at the current stage as re-emission. The contributions of this process to annual total depositions over European countries for 2004 are presented in Fig. 4.10. Re-emission contributions for all European countries are close to one another varying from 20% to 45%.

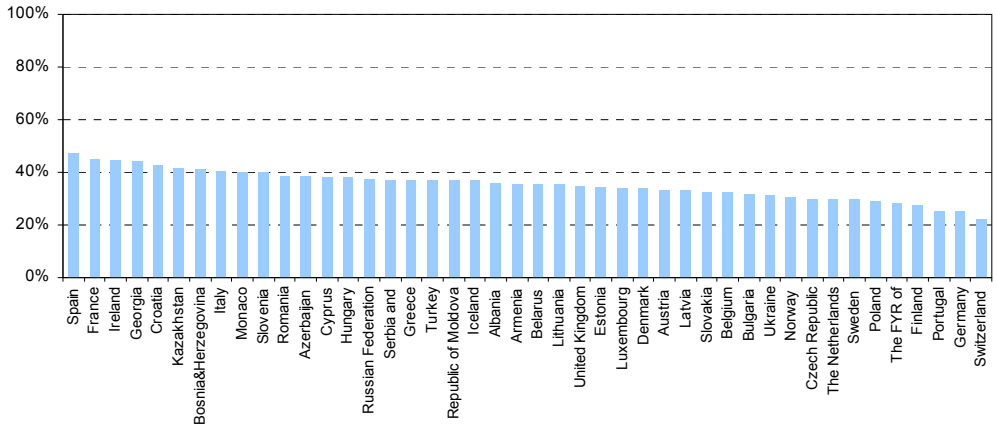


Fig. 4.10. *Contributions of re-emission to 2,3,4,7,8-PeCDF depositions over the territories of European countries, %*

Country-specific information. Putting together contributions of the atmospheric transport and re-emissions it is possible to evaluate total depositions for each European country in 2004 and contributions of all above categories of sources (European anthropogenic sources, non-EMEP sources within the northern hemisphere and re-emissions) to these depositions.

The calculated values of total depositions of 2,3,4,7,8-PeCDF to the territories of European countries in 2004 are displayed in Fig. 4.11.

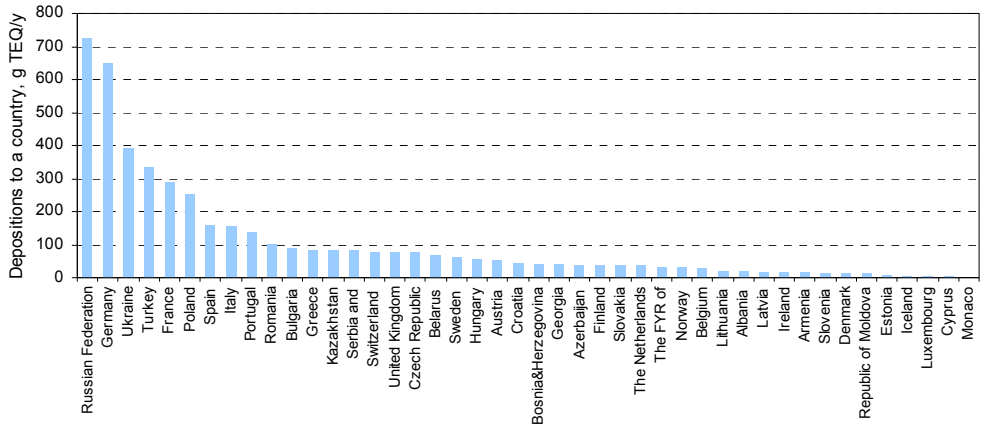


Fig. 4.11. *Total depositions of 2,3,4,7,8-PeCDF to the territories of European countries, g TEQ/y*

These depositions strongly depend on the country area, its locations and emission distributions. As it was mentioned above, high deposition values (more than 300 g TEQ) for Germany, Ukraine and Turkey are explained by high emission levels in these countries. The contributions of different sources to depositions are exemplified by Austria, Finland and Italy are presented in Fig. 4.12.

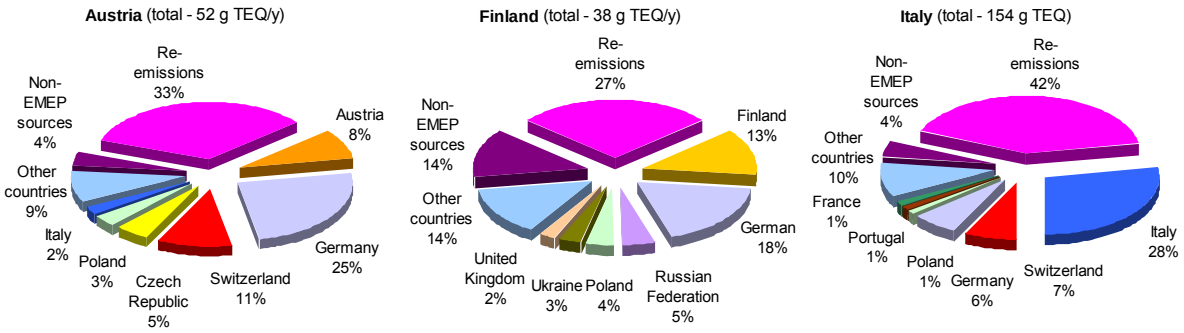
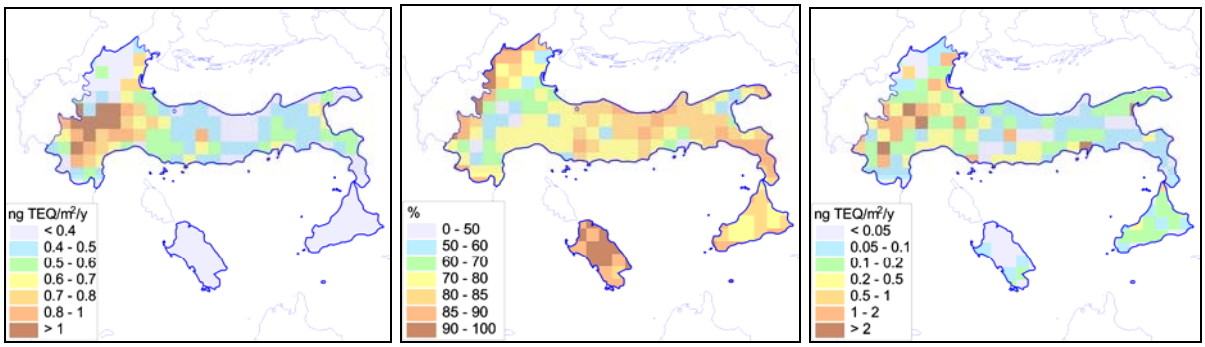


Fig. 4.12. *Contributions of all emission sources to 2,3,4,7,8-PeCDF depositions over the territories of Austria, Finland and Italy*

At the same plots values of total deposition to a country due to all considered emission sources are put. So, the information of source-receptor relationships in the European region includes values of total depositions to the European countries together with the fractions of these depositions originated by various emission sources.

Peculiarities of spatial distribution of total annual 2,3,4,7,8-PeCDF depositions and contributions of transboundary transport for a particular European country are exemplified in Fig. 4.13. Distribution of total annual depositions over Italy together with distribution of deposition fractions due to transboundary transport from European and non-EMEP anthropogenic sources and re-emissions is presented. Along with that spatial distribution of PCDD/F emissions of Italy in 2004 is shown in the figure.



a b c

Fig. 4.13*. Spatial distribution of annual total depositions of 2,3,4,7,8-PeCDF to Italy, ng TEQ/m^2/y, (a); contribution of transboundary transport to depositions over Italy, % (b); annual emission of 2,3,4,7,8-PeCDF in Italy, ng TEQ/m^2/y, (c)*

On the average, the joint contribution of transboundary transport (including non-EMEP sources) and re-emission for Italy is about 56%. However, for different locations inside the country this contribution can be quite different. Highest contribution of non-EMEP sources together with re-emission is characteristic of regions with low levels of national emissions and for regions located close to the country borders.

The full information on source-receptor relationships for 2,3,4,7,8-PeCDF depositions presented in the form of country-to-country matrix can be found in Annex B.

4.1.2. Polyaromatic hydrocarbons (PAHs)

Pollution levels in the EMEP region

Calculations of pollution levels of the four indicator PAH congeners (B[a]P, B[b]F, B[k]F and I_P) in the EMEP region were performed with the help of regional model with resolution 50x50 km^2 for 2004. For evaluation of atmospheric pollution levels (air concentrations and depositions) calculations were done for one-year period. The latter simplification is feasible for the assessment of the selected four PAHs since they exist in the atmosphere mainly in particle-bound phase and re-emission processes seem not to be essential for evaluation of atmospheric pollution levels. The emission data used in the calculations are based on official data and include expert estimates when official information is not available (subsection 2.1).

Benzo[a]pyrene (B[a]P). The maps of spatial distributions of B[a]P concentrations in surface atmospheric layer and depositions in comparison with emissions are shown in Fig. 4.14.

Atmospheric concentration levels in some European regions (Germany, Ukraine, Belgium and the Netherlands) are close to or even higher than the level of 1 ng/m^3 accepted in some countries as the limit value of atmospheric concentrations. Comparatively high levels of B[a]P air concentrations (0.1 – 0.3 ng/m^3) are calculated for such countries as France and Belarus.

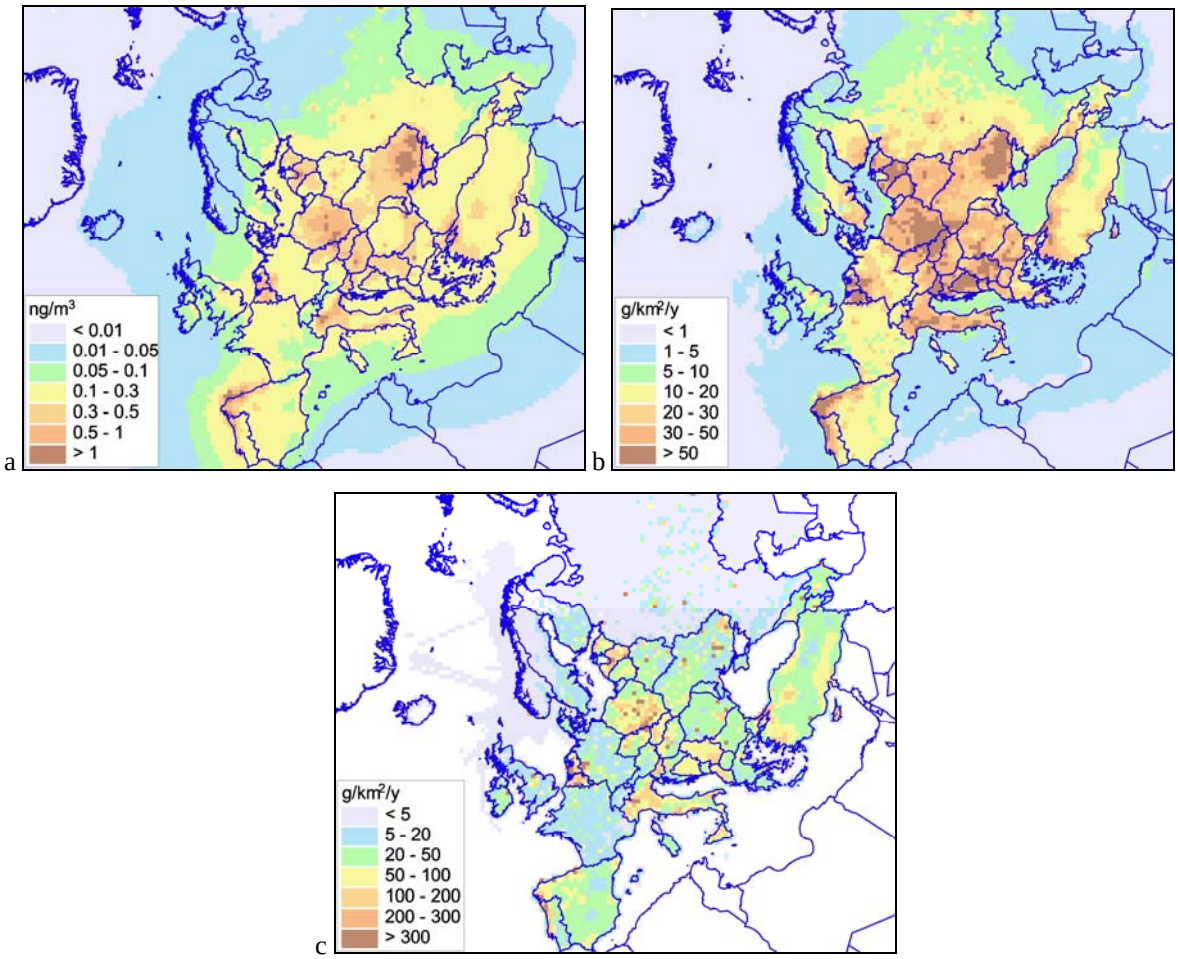


Fig. 4.14. *Calculated air concentrations (a) and deposition fluxes (b) of B[a]P in 2004 in comparison with emissions (c)*

For B[a]P, calculated values were compared with available measurement data for 2004. The base of comparison includes EMEP measurement sites CZ3, FI96, LV10, LV16, SE12, SE14 and NO42 for atmospheric concentrations, CZ3, DE1 and DE9 for concentrations in precipitation and FI96, SE12 and SE14 for deposition fluxes. In general, most annual averages of calculated air concentrations agree with measurements within a factor of three. Annual averages of calculated and measured air concentrations are in a good agreement at CZ3 and FI96 (calculation-to-measurement ratio is 0.7 and 0.9, respectively). Some overestimation of air concentrations (about factor of two) takes place at sites LV10 and LV16. Calculated values of annual averaged concentrations in precipitation at three above sites agree with measurements within a factor of two. The comparison of measured and calculated values of deposition flux is hampered by the fact that measured bulk deposition include wet deposition and some unknown part of dry deposition. However, with regard to this fact calculated values of deposition flux are in a reasonable agreement with available measurements.

Detailed analysis of the agreement between calculations and measurements for B[a]P for a larger time period is given in detail in [*Gusev et al*., 2005].

The rest indicator congeners. For the rest three indicator congeners (B[b]F, B[k]F and I_P) the maps of spatial distributions of depositions in comparison with their emissions are shown in Fig. 4.15 a, b and c, respectively.

Air concentrations of B[b]F and I_P in Europe vary from very low (0.01 – 0.05 ng/m^3) in northern Europe to rather high (up to 1 ng/m^3 and even higher) in eastern and southern European regions. In general, spatial pattern of air pollution for these compounds is close to that for B[a]P. Higher emission densities in the northern part of Spain lead to increase of their air concentrations and deposition levels in northern Spain and southern France compared with B[a]P.

The levels of air concentrations of B[k]F in Europe are lower (from 0.01 – 0.05 ng/m^3 in northern Europe up to 0.3 ng/m^3 in eastern and southern European regions). Absolute values of air concentrations and deposition fluxes are smaller than for the two above considered pollutants since emission levels for B[k]F are considerably lower according to emission data used in modelling. Spatial pattern of air contamination for B[k]F is similar to that for B[a]P and B[b]F and I_P.

In view of similarities between spatial patterns of emissions and atmospheric contamination evaluating of source-receptor relationships for PAHs at present stage of investigation can be performed on the example of B[a]P. This choice is conditioned by larger amount of measurement data and more reliable emission inventories for B[a]P. In future, for more detailed evaluation of source-receptor relationships for the rest three PAHs more information on substance-specific emissions is needed.

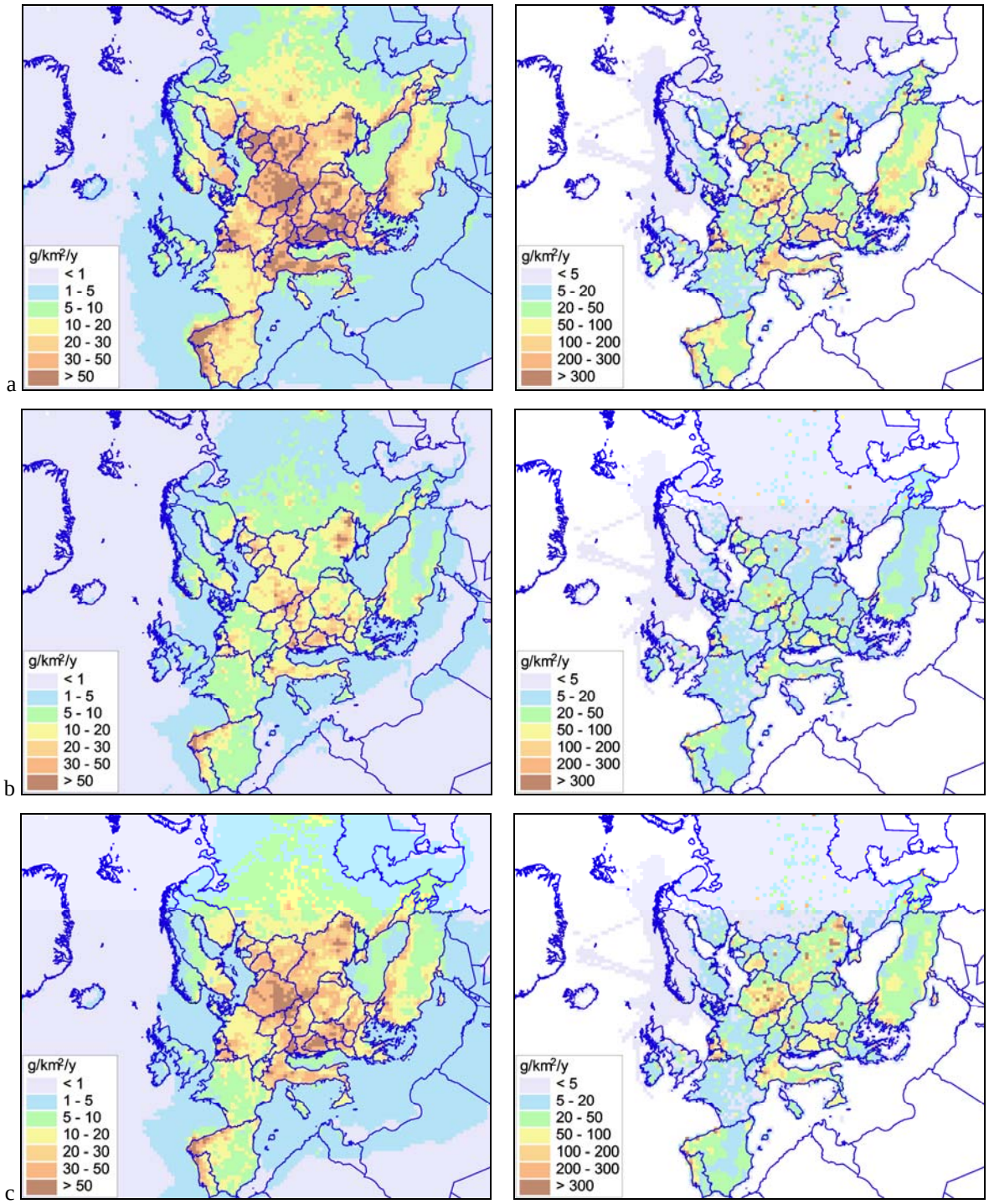


Fig. 4.15. *Calculated deposition fluxes (on the left) and emissions (on the right) of B[b]F (a), B[k]F (b) and I_P (c) in 2004*

Transboundary transport of B[a]P

Here the calculated source-receptor relationships in the EMEP region for PAHs in 2004 are exemplified by B[a]P. In calculations the above-described emission data were used. Taking into account the fact that the considered pollutant exists in the atmosphere predominantly in particulate form, evaluation of source-receptor relationships was restricted to evaluation of atmospheric transport from European anthropogenic sources.

Similar to the case of PCDD/Fs, source-receptor matrices for B[a]P in 2004 were calculated both for deposition and for air concentrations. Below we illustrate source-receptor relationships by the example of deposition matrix.

Spatial distribution of B[a]P annual depositions originated from national emission sources of Austria, Finland and Italy is presented in Fig. 4.16.

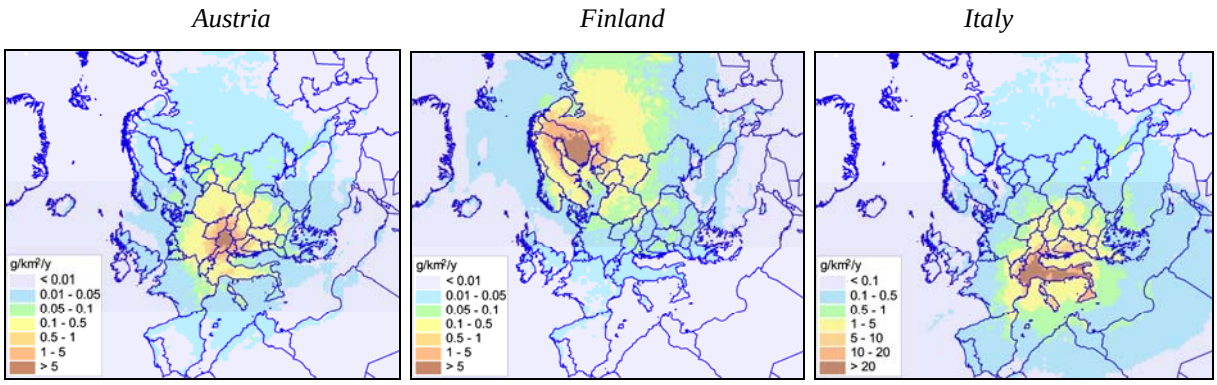


Fig. 4.16. *Annual total depositions of B[a]P originated from national emissions of Austria, Finland and Italy in 2004, g/km^2/y*

On the basis of spatial distribution of depositions caused by each source region source-receptor relationships in the EMEP region was evaluated. As stated above, along with atmospheric transport contributions from re-emissions should be evaluated.

Export. The plot of export fractions for all European countries (that is, the fractions of depositions to the entire EMEP region due to emissions of the given country) is displayed in Fig. 4.17.

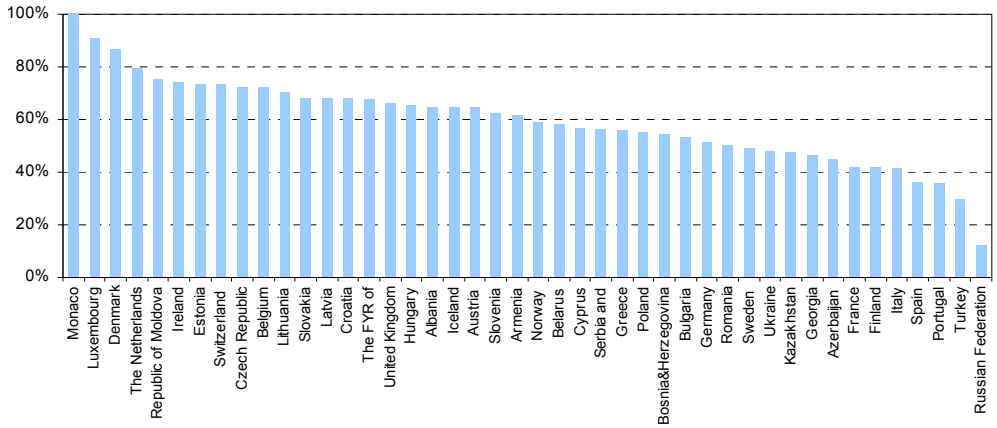


Fig. 4.17. *Export fractions of B[a]P for European countries, %*

These fractions depend significantly on the geographic location of a given country, size of its territory, and spatial distribution of emission sources within the country. Typically, about 30 – 70% of depositions caused by national emission sources of a country are deposited outside this country. Significant export fractions (about 80% and more) are characteristic of Luxembourg, Monaco, Denmark and the Netherlands.

The distribution of total depositions caused by the country's sources to the EMEP region between surrounding countries is shown in Fig. 4.18.

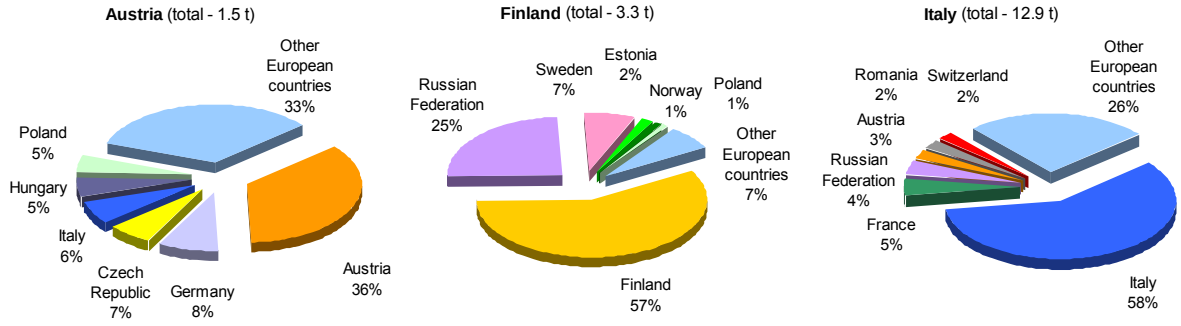


Fig. 4.18*. Distribution of depositions of B[a]P due to national emissions of Austria, Finland and Italy between surrounding European countries, %*

For example, for Austria over 60% of depositions originated from national sources (totally 1.5 tonnes) are deposited outside the country (transboundary depositions). These transboundary depositions are distributed between Germany, the Czech Republic, Italy, Hungary and Poland (about 30% altogether) and other European countries (33%).

Import. The calculated values of total depositions of B[a]P to the territories of European countries in 2004 are displayed in Fig. 4.19.

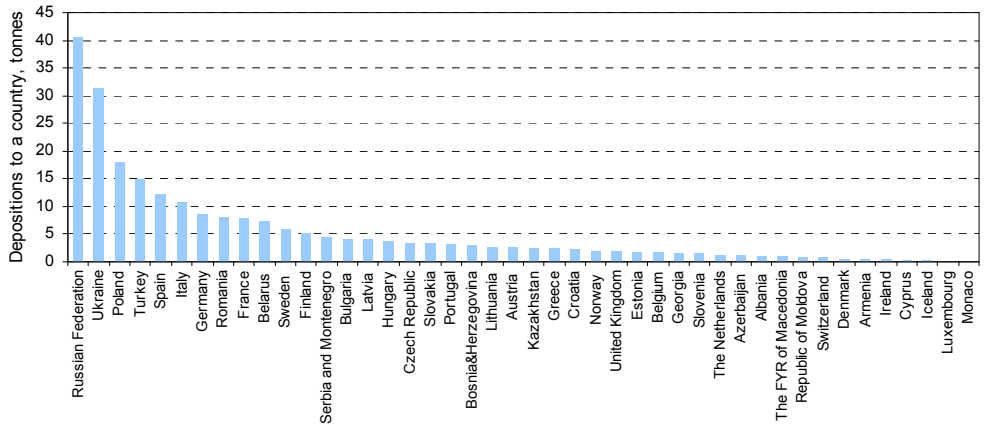


Fig. 4.19. *Total depositions of B[a]P to the territories of European countries, t*

Again, these depositions strongly depend on the country area, its locations and emission distributions. Comparatively high deposition in e.g. Germany, France and Belarus is mainly determined by transboundary transport. The contributions of transboundary transport to depositions in European countries are shown in the plot in Fig. 4.20.

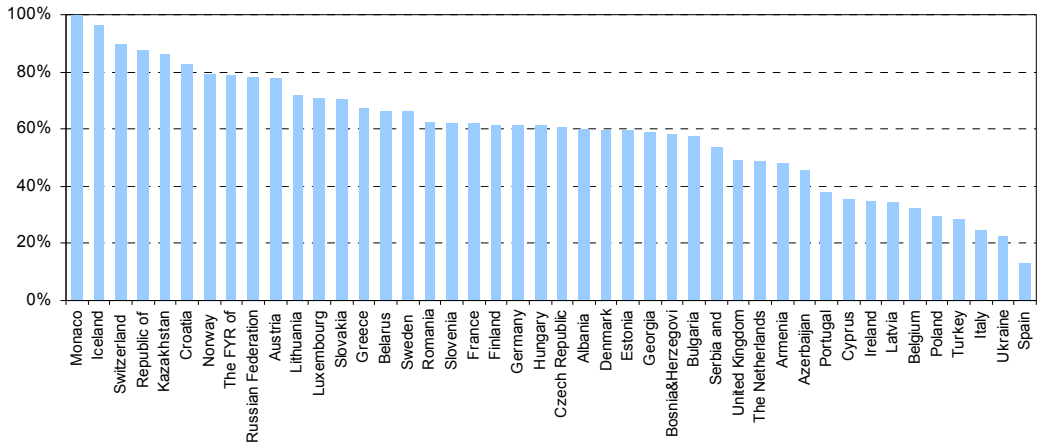


Fig. 4.20. *Contributions of transboundary transport to total annual depositions of B[a]P from European emission sources for each European country, %*

Typically contributions of other European countries to the deposition levels of a particular country vary from 20% to 80%.

The information of contributions of various European countries to deposition levels of a particular country is presented in Fig. 4.21 for Austria, Finland and Italy.

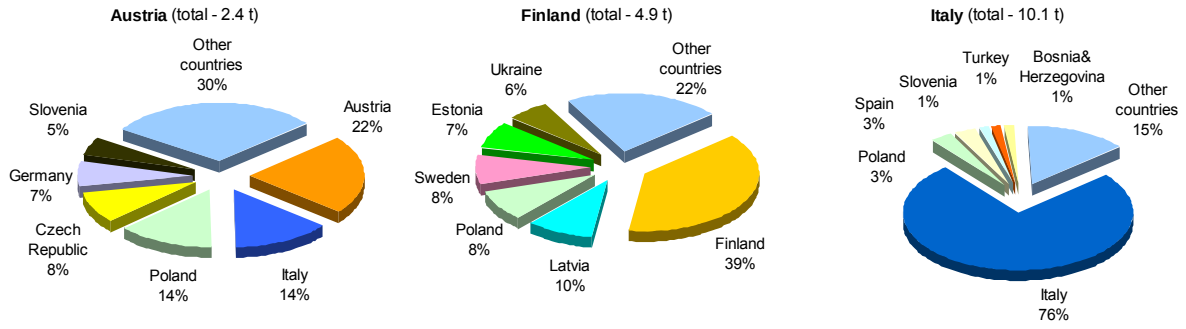
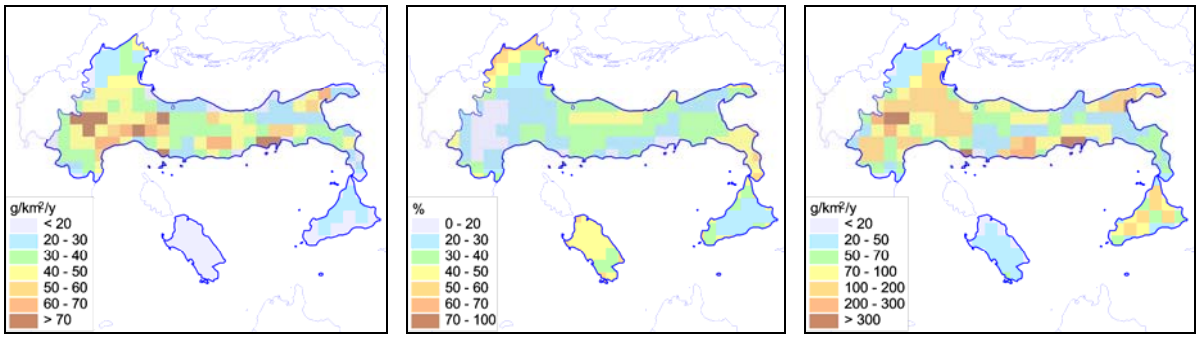


Fig. 4.21. *Import of B[a]P depositions originated from atmospheric transport for Austria, Finland, and Italy, %*

For Austria the contribution of atmospheric transboundary transport to total depositions (2.4 tonnes) is significant (about 80%) while national emissions contribute slightly less than 20%. In Finland the contribution of the transboundary transport to depositions amounts to about 60%. In case of Italy the contribution of transboundary transport from other European countries is only 24%.

Peculiarities of spatial distribution of total annual B[a]P depositions and contributions of transboundary transport for a particular European country are exemplified in Fig. 4.22. Distribution of total annual depositions over Italy together with distribution of deposition fractions due to transboundary transport is presented. Along with that spatial distribution of B[a]P emissions of Italy in 2004 is shown in the figure.



a b c

Fig. 4.22*. Spatial distribution of annual total depositions of B[a]P to Italy, g/km^2/y, (a); contribution of transboundary transport to depositions over Italy, %, (b); annual emission of B[a]P in Italy, g/km^2/y, (c)*

On the average, the joint contribution of transboundary transport for Italy is about 25%. However, for different locations inside the country this contribution can be quite different reaching 50% and higher. Highest contribution of transboundary transport is characteristic of regions with low levels of national emissions and for regions located close to the country borders.

Detailed information on source-receptor relationships for B[a]P depositions can be found in the Annex B.

4.2. Pollution levels in the Northern Hemisphere

The section presents the results of the evaluation of pollution levels and intercontinental transport of PCBs, HCB, and γ-HCH for 2004. These are the pollutants of significant long-range transport potential. Therefore the evaluation of environmental pollution within the European region requires taking into account the contribution of intercontinental transport from the remote emission sources. At the moment global emission inventories are available for PCBs, HCB, and HCHs. However, the inventories on PCBs and HCB provide information on total annual emissions of countries. Spatial distributions of these emissions were obtained on the basis of population density. For HCHs global emission data with spatial distribution are available for α-HCH and β-HCH. For γ-HCH (lindane) there is available information on its emission for the European region, North America, and China. Due to availability of the global emission data and various measurements EMEP Task Force on Hemispheric Transport of Air Pollution during its 2nd meeting in Moscow in June 2006 recommended to currently focus at the model evaluation of intercontinental transport on these POPs. In this section preliminary model estimates of pollution levels and contributions of intercontinental transport to depositions of these POPs over selected receptor regions are presented.

4.2.1. Pollution levels of PCBs

Modelling of long-range transport and depositions of PCBs was carried out for three PCB congeners PCB-28, PCB-118, and PCB-153 using updated version of hemispheric MSCE-POP model. In the current year the development of the model was continued including the improvement of model description of POP degradation in the atmosphere, removal with precipitation (including scavenging with snow), partitioning in soil, and the refinement of model description of POP fate in seawater compartment (see subsections 3.2 and 3.3).

Computations were performed for the period 1970-2004 using the global emission inventory of PCBs [*Breivik et al.*, 2002]. Since this inventory was prepared for the period 1930-2000 it was assumed that the level of PCB emission in 2001-2004 was not changed since 2000. Several groups of emission sources were considered, namely: Europe, Russia, Africa and Central Asia, Southeast Asia, and America (subsection 2.4).

For the evaluation of PCB intercontinental transport separate model runs were made for each of the selected groups of emission sources. On the basis of obtained modelling results distribution of PCB congeners depositions over selected receptor regions (Fig. 4.23) were described and contributions of distinguished groups of emission sources to depositions over the Arctic region were estimated.

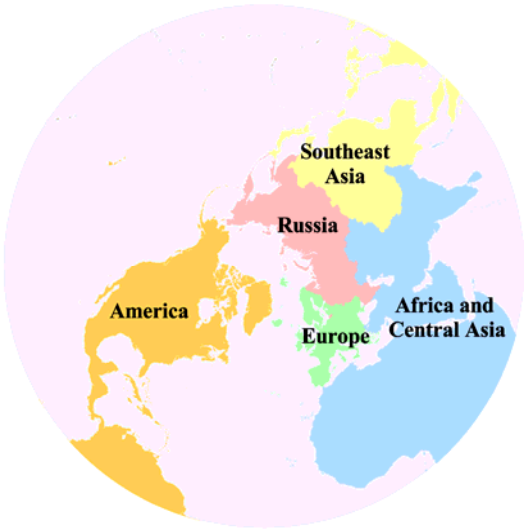


***Fig. 4.23**. Selected receptor regions*

Intercontinental transport

In Figs. 4.24 – 4.26 spatial distribution of PCB-28, PCB-118, and PCB-153 annual emission fluxes for 2004 is illustrated. According to the inventory of [*Breivik et al.*, 2002] total annual emission of lighter congener PCB-28 is comparatively higher than that of heavier congeners PCB-118 and PCB-153. In particular, total annual emission of PCB-28 within the northern hemisphere used for computations for 2004 accounts for 53 tonnes while annual emissions of PCB-118 and PCB-153 are about 18 and 11 tonnes, respectively.

Spatial distribution of annual mean air concentrations and net annual deposition fluxes of selected PCBs for 2004 resulted from all considered emission sources is shown in Figs. 4.24 – 4.26 b and c.

Significant levels of calculated PCB-28 air concentrations (about 30 pg/m^3 and higher) and net annual depositions (about 20 g/km^2/y) in Europe can be noted for Germany and European part of Russia. Relatively lower levels of pollution of European countries are obtained for PCB-118. In case of PCB-153, due to different character of spatial distribution of emissions, higher levels of PCB-153 air concentrations (about 15 pg/m^3 and higher) and net annual depositions (about 10 g/km^2/y) are obtained for France, Belgium, the Netherlands, and Germany.

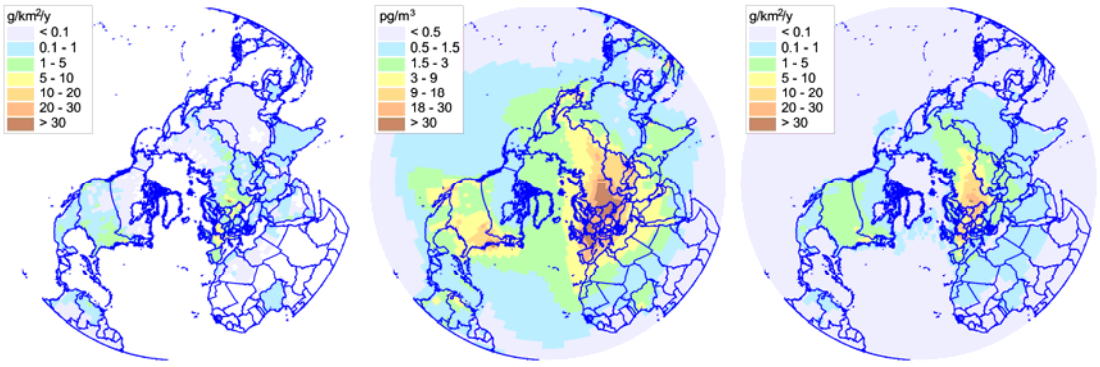


Fig. 4.24. *Spatial distribution of PCB-28 annual emissions, g/km²/y (a), annual mean air concentrations, pg/m³ (b), and net depositions, g/km²/y (c) for 2004*

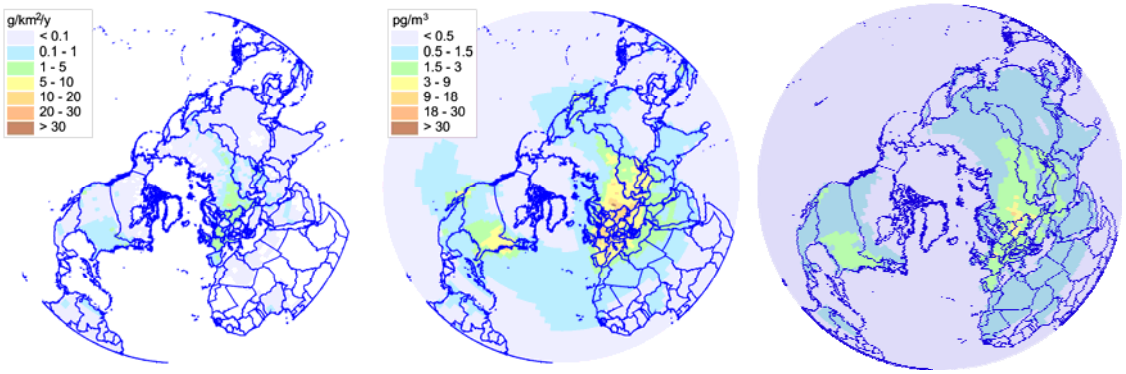


Fig. 4.25. *Spatial distribution of PCB-118 annual emissions, g/km²/y (a), annual mean air concentrations, pg/m³ (b), and net depositions, g/km²/y (c) for 2004*

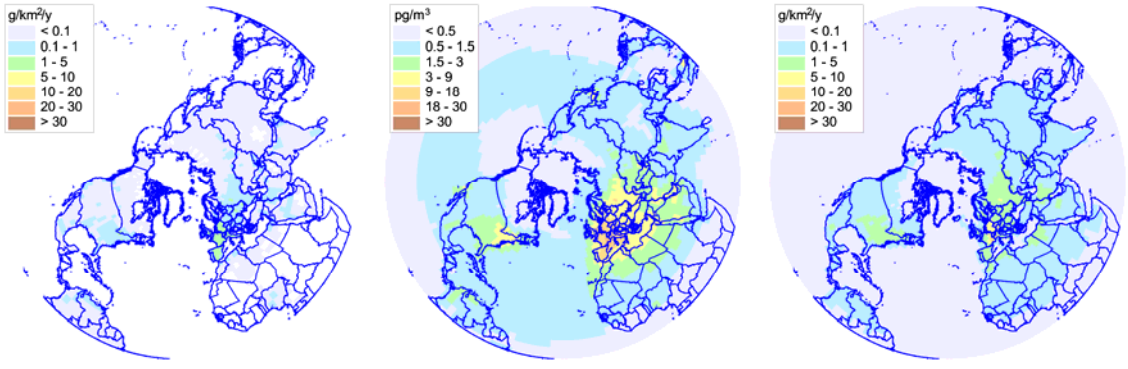


Fig. 4.26*. Spatial distribution of PCB-153 annual emissions, g/km²/y (a), annual mean air concentrations, pg/m³ (b), and net depositions, g/km²/y (c) for 2004*

The calculations of PCB long-range transport permit to evaluate the distribution of pollution from emission sources of various regions of the northern hemisphere. Intercontinental transport of selected PCB congeners was evaluated for the following groups of emission sources: Europe, America, Russia, Southeast Asia, and Africa and Central Asia. To characterize the influence of intercontinental transport on the pollution of different regions of the northern hemisphere total depositions over land territories of the selected receptors (Fig. 4.23) were considered. Depositions over marine regions

were not taken into account in calculation of contribution of intercontinental transport. In Figs. 4.27 – 4.28 the distribution of depositions of three selected PCB congeners over selected receptors originated from emission sources of Europe and Southeast Asia is presented. More volatile PCB congeners have higher contributions of intercontinental transport to the depositions. In particular, 25%, 17%, and 10% of PCB-28, PCB-118, and PCB-153, respectively, emitted from European emission sources are deposited over the territory of Russia (Fig. 4.27). Similarly, emissions of PCB-28, PCB-118, and PCB-153 originated from Southeast Asia contribute 24%, 11%, and 10% to depositions over America, respectively (Fig. 4.28).

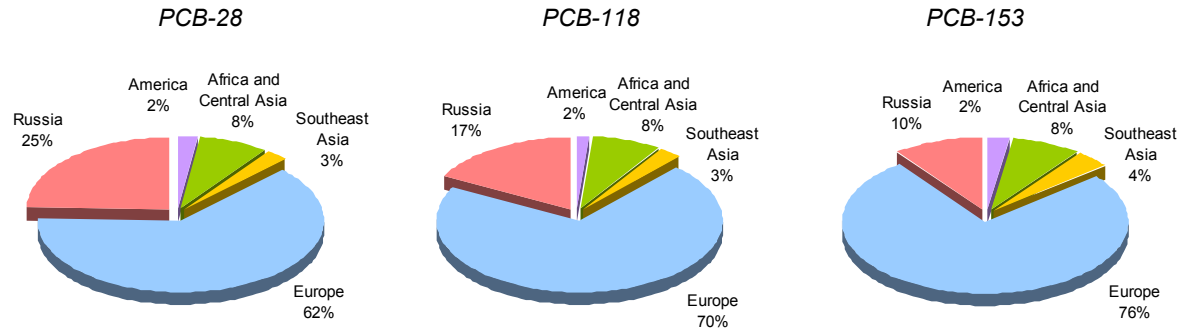


Fig. 4.27. *Distribution of three PCB congeners (PCB-28, PCB-118, PCB-153) depositions originated from emission sources of Europe over land territories of selected receptor regions of the northern hemisphere for 2004*

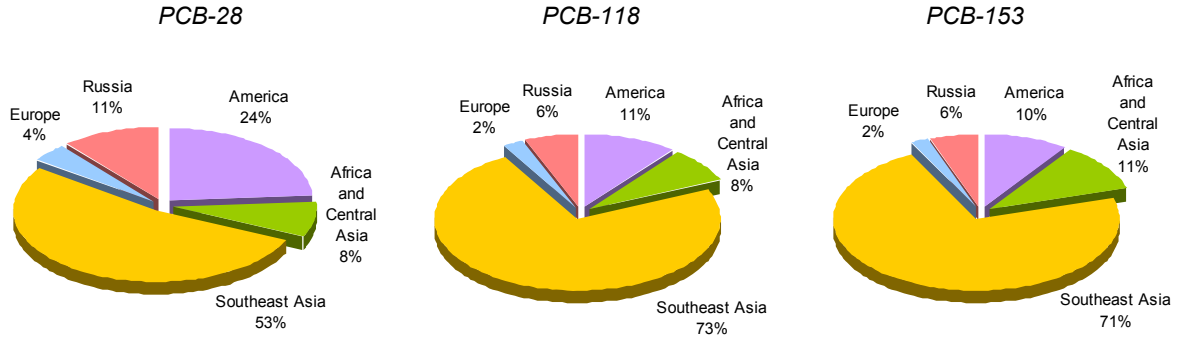


Fig.4.28. *Distribution of three PCB congeners (PCB-28, PCB-118, PCB-153) depositions originated from emission sources of Southeast Asia over land territories of selected receptor regions of the northern hemisphere for 2004*

Pollution of remote regions

The influence of intercontinental transport to the contamination of remote regions is exemplified by the Arctic. In Fig. 4.29 the contributions of selected groups of emission sources to annual depositions of PCB-28, PCB-118, and PCB-153 over the Arctic region for 2004 are presented. In accordance with the modelling results for 2004 about 9%, 5%, and 4% of total hemispheric emissions of PCB-28, PCB-118, and PCB-153, respectively, is deposited over the Arctic region. In case of PCB-28 the most significant contribution to the depositions belongs to Russia (47%) followed by Europe (35%), and America (11%). Close pattern of contributions is obtained for depositions of PCB-118. In particular, 53% of depositions is originated from the emission sources of Russia, 28% from European emission sources, and 9% from American emission sources. Different pattern of spatial distribution of PCB-153 emission resulted in different contributions of selected groups of emission sources to the depositions over the Arctic region. Thus, contributions of Europe, Russia, and America are accounted for 54%, 23%, and 12%, respectively.

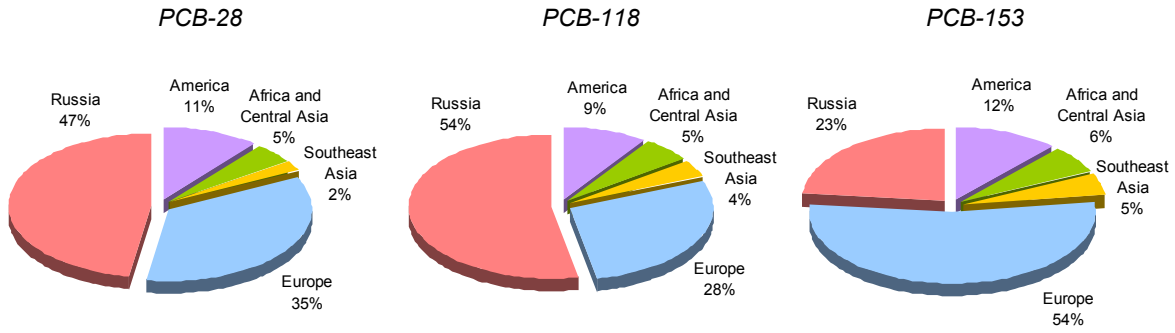


Fig. 4.29. *Contributions of selected groups of emission sources of PCB-28, PCB-118, and PCB-153 to depositions over the Arctic region in 2004*

4.2.2. Pollution levels of HCB

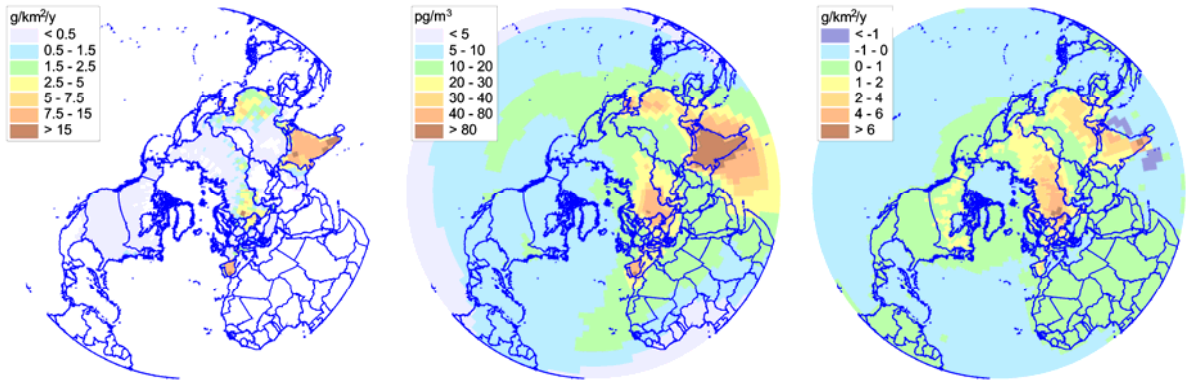
Modelling of intercontinental transport of HCB within the northern hemisphere was carried out for the period 1990-2004 using the hemispheric version of MSCE-POP model. The emission scenario used in computations for this period is described in section 2.3 of this report. This year the expert estimates of HCB emissions were complemented by the available official emission data submitted by countries. It should be noted that most of included official emissions were essentially lower than expert estimates of HCB emissions [*Pacyna et al.*, 1999; *Bailey*, 2001] used in calculations made in the previous year. The major changes in emissions in comparison to the scenario of the previous year are characteristic of the USA and Japan, for which official data used are about 90% lower than expert estimates.

Intercontinental transport

The evaluation of HCB intercontinental transport was carried out on the basis of emission scenario described in subsection 2.3. The following groups of emission sources within the northern hemisphere were distinguished: Europe, Russia, Central Asia, Southeast Asia, and North America. Separate model runs were made for each of the selected groups of emission sources. On the basis of calculations performed pathways of HCB atmospheric transport were evaluated and contributions of selected emission sources to depositions over the receptor regions (Fig. 4.23). Evaluation of remote regions pollution by HCB was performed on the example of the Arctic region.

Spatial distribution of annual emissions, annual mean air concentrations and net annual deposition fluxes of HCB for 2004 is shown in Fig. 4.30. Elevated levels of HCB air concentrations for 2004 about 40-80 pg/m^3 and higher are obtained for countries of Southeast Asia, European part of Russia, Spain, and Portugal. Significant level of HCB air concentrations (higher than 80 pg/m^3) is characteristic of India. In comparison to the results for 2003 presented in the previous Status report [*Gusev et al.,* 2005c] essentially lower values of air concentrations are obtained for North America. This can be explained by the differences in emissions used for computations for 2003 and 2004. It can be noted also that spreading of air concentrations obtained for 2004 is comparatively wider than for 2003. This can be explained by the refinement of physical-chemical properties of HCB (subsection 3.1), in particular, partition coefficients and scavenging ratio used for the description of HCB removal with precipitation.

Elevated levels of net deposition fluxes can be noted over the territory of Russia, countries of Eastern Europe, and in Southeast Asia (about 10 g/km^2/y). Over the seawater re-volatilization fluxes of HCB are obtained for most of the regions within the northern hemisphere, in particular, in the vicinity of India.



a b c

Fig. 4.30. *Spatial distribution of HCB annual emissions, g/km^2/y (a), annual mean air concentrations, pg/m^3 (b), and net depositions, g/km^2/y (c) for 2004*

The distribution of HCB depositions over the land territories of selected receptors (Fig. 4.23) from emission sources of Europe, Southeast Asia and North America for 2004 is presented in Fig. 4.31. 58% of total HCB depositions over land originated from European emission sources are deposited over Russia, 16% over Europe, 13% over Central Asia. In case of emission sources of Southeast Asia 41% and 36% are deposited over Southeast Asia and Central Asia, respectively, and 12% reach the territory of North America. Major part of total HCB depositions over land originated from North American emission sources (about 70%) is deposited over its own territory. In comparison to the modelling results for 2003 it can be noted that the calculated fractions of depositions due to intercontinental transport of HCB are increased. This is explained by the changes in physical-chemical properties of HCB and inclusion of official emission data into the modelling as it was mentioned above.

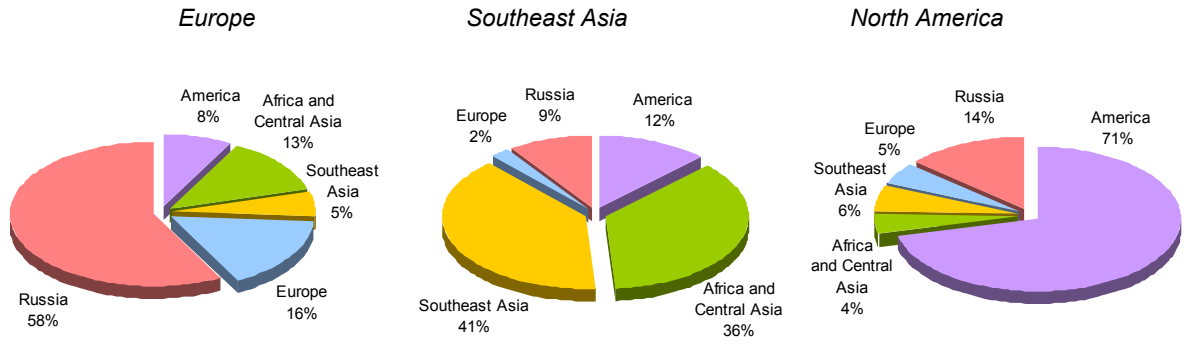


Fig. 4.31. *Distribution of HCB depositions originated from emission sources of Europe, Southeast Asia, and North America over land territories of selected receptor regions within the northern hemisphere in 2004*

Pollution of remote regions

The contributions to total annual depositions over the Arctic region of HCB emitted by selected groups of emission sources for 2004 are presented in Fig. 4.32. Due to inclusion of official data on emissions for the USA and some European countries the pattern of contributions to total annual depositions for 2004, as compared to the results of the previous Status report, is noticeably changed. European emission sources contribute 36% to the total HCB depositions from the selected emission sources, 30% is originated from Russian emission sources, 22% from Southeast Asia and 11 % from Central Asia. Relative contribution of North American emission sources is essentially lower (1%) due to the usage of significantly lower annual total emission of HCB for North America.

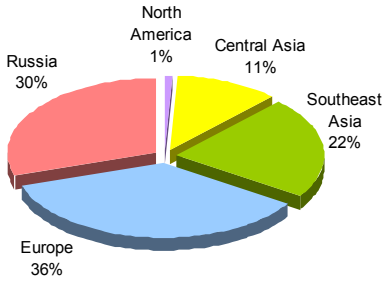


Fig. 4.32. *Contributions of selected groups of emission sources of HCB to depositions over the Arctic region in 2004*

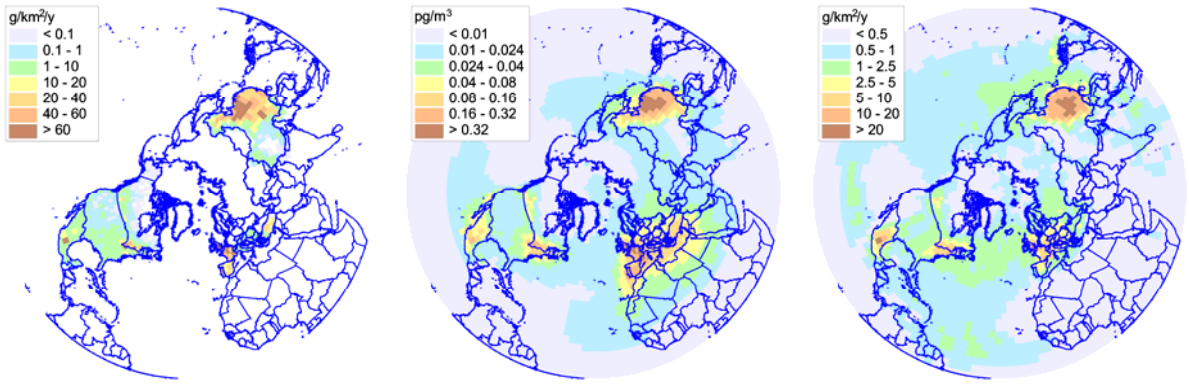
4.2.3. Pollution levels of γ-HCH

Modelling of intercontinental transport of γ-HCH (lindane) within the northern hemisphere was performed using the hemispheric version of the MSCE-POP model for the period 1990-2004. Annual emissions of γ-HCH and their spatial distribution used for computations are described in subsection 2.5 of this report. It should be noted that currently the complete global emission inventory for γ-HCH is not available. Data on emissions or usage of lindane are compiled from several available sources. Obtained estimates of emissions cover only part of the northern hemisphere. However, this information permits to carry out preliminary estimates of the intercontinental transport of γ-HCH from European, North American, and Chinese emission sources.

Intercontinental transport

For the evaluation of γ-HCH intercontinental transport separate model runs were made for each of the selected groups of emission sources. On the basis of obtained modelling results distribution of γ-HCH depositions to the selected receptor regions (Fig. 4.23) was described and contributions of selected emission sources to depositions over the Arctic region were estimated.

Spatial distribution of annual emissions, annual mean air concentrations and net annual deposition fluxes of γ-HCH for 2004 is shown in Fig. 4.33. It can be seen that emission sources of North America and Europe can essentially contribute to the contamination of the Northern Atlantic and the Arctic region. Emission sources of Southeast Asia under favorable conditions can contribute to the pollution of North America.



a b c

Fig. 4.33. *Spatial distribution of γ-HCH annual emissions, g/km²/y (a), annual mean air concentrations, pg/m³ (b), and net depositions, g/km²/y (c) for 2004*

The contributions to γ-HCH depositions over land from the emission sources of Europe, China and North America for 2004 are presented in Fig. 4.34. 63% of total γ-HCH depositions over land originated from European emission sources is deposited over Europe, 16% over Russia, 13% over Africa and Central Asia, and 4% over America. In case of emission sources of China 81% is deposited over Southeast Asia, 9% over America, and 6% over Russia. Like for HCB major part of total γ-HCH depositions over land originated from North American emission sources (90%) is deposited over its own territory.

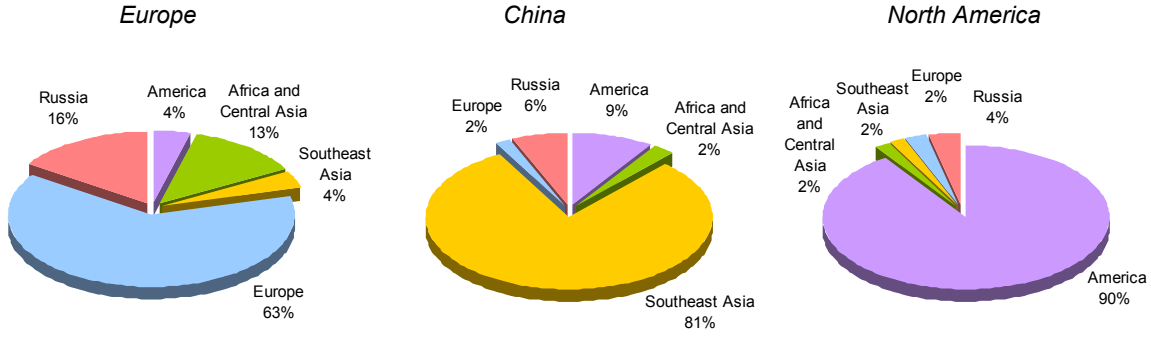


Fig. 4.34. *Distribution of γ-HCH depositions originated from emission sources of Europe, China, and North America over land territories of selected receptors within the northern hemisphere in 2004*

Pollution of remote regions

The contributions to total annual γ-HCH depositions over the Arctic region for 2004 for the selected groups of emission sources are presented in Fig. 4.35. Significant contribution belongs to emission sources of China (44%). Contribution of European emission sources accounts for 33% and North American emission sources for 23%. This illustrates the relative significance of the intercontinental transport from the selected three groups of emission sources.

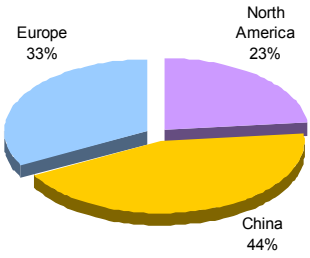


Fig. 4.35. *Contributions of selected groups of emission sources of γ-HCH to depositions over the Arctic region in 2004*

5. CO-OPERATION

During this year the EMEP Centres continued to work in close co-operation with the subsidiary bodies to the Convention, international organizations, programmes and projects as well as with national experts. In particular, the MSCE-POP model was reviewed by the EMEP Task Force on Measurements and Modelling. MSC-E continued to support the work of the Task Force on POPs on technical review of dossiers of new proposed substances, and to carry out the POP model intercomparison study. MSC-E took part in the activity of the EMEP Task Force on Hemispheric Transport of Air Pollution attending the organizational workshop in Washington in January 2006 and supporting the organization of the Task Force meeting in Moscow in June 2006. Experts of MSC-E participated in Summer School "The Advances and Trends in Environmental Chemistry and Ecotoxicology of Persistent, Toxic Substances", organized by EU DG Research – Centre of Excellence for Environmental Chemistry and Ecotoxicology and took part in the 16th SETAC Europe Annual Meeting.

5.1. Task Force on Measurements and Modelling

The MSCE-POP model was evaluated at the EMEP Task Force on Measurements and Modelling meeting in Zagreb and the EMEP/TFMM Workshop on the Review of the EMEP Models on Heavy Metals and Persistent Organic Pollutants in Moscow in 2005. The Workshop in Moscow has recommended continuing further improvement of modelling approaches for POPs. In particular, the Task Force pointed out to the following issues:

- degradation of POPs in particle-bound and gaseous phase in the atmosphere including photodegradation;
- seasonal dependence of soil volatilisation;
- evaluation of concentrations at boundaries of the EMEP grid;
- seasonal variations of emissions;
- application of the MSCE-POP model to screening a wider range of POPs for their potential environmental significance.

Following these recommendations MSC-E has started its work on further improvement of POP modelling. The results of this work are presented in the intermediate EMEP/MSC-E Technical Report [*Gusev et al*., 2006].

To improve the MSCE-POP model, an essential attention was given to the refinement of data sets of POP physical-chemical properties and process parameterisations used in the MSCE-POP model.

Further improvement of physical-chemical properties of POPs considered in modelling activities (PCDD/Fs, PAHs, PCBs, γ-HCH and HCB) was performed by taking into account the temperature dependence of octanol/water partition coefficient as well as by harmonising all three partition coefficients (octanol/water, air/water, and octanol/air) with the help of the least-squares adjustment procedure developed in [*Schenker et al*., 2005] (subsection 3.1).

In the course of the comparison of model results with measurements it was found that the model considerably underestimated seasonal variations of B[a]P air concentrations for a number of monitoring sites. Possible reasons of this disagreement could be connected with the neglecting of

B[a]P degradation in particle-bound phase and underestimation of seasonal variations of B[a]P emissions. The possibility of improving the agreement between model results and measurements for B[a]P by the refinement of degradation process description and usage of different scenarios of seasonal variations of emissions is considered (subsection 3.2).

To improve model description of POP volatilisation from soils, an additional modification of MSCE-POP model parameterisation is performed. The processes of POP absorption in soil and volatilisation to the atmosphere are described in the model taking into account the effect of temperature variations. However, the parameterisation of POP partitioning in soil previously used in the model did not take into account temperature dependence of octanol/water partition coefficient K_{OW}. At present this temperature dependence is included to the model description of POP behaviour in soils. The sensitivity analysis of POP soil contamination with respect to K_{OW} is presented (subection 3.2).

Further development of the MSCE-POP model has been continued by improving the consistency of hemispheric and regional modelling approaches. The combined modelling approach based on the nesting of regional and hemispheric versions of MSCE-POP model is developed to evaluate POP pollution levels for the European region and to provide estimates of transboundary fluxes between European countries accounting also for the contributions of non-European emission sources and of re-emission of POPs accumulated in environmental compartments (Section 3.3).

More detailed description of the refinement of the sets of POP physical-chemical properties and of model description of main environmental processes is given in the EMEP/MSC-E Technical Report [*Gusev et al.,* 2006].

The results of this work were discussed at the 7th Meeting of the Task Force on Measurements and Modelling in Helsinki in 2006. The Task Force agreed with the conclusions of the workshop that within the limitations of current understanding of the POPs fate in the environment, the MSCE-POP model represents the state of the science and fits for the purpose of evaluating the contribution of long-range transport to the environmental impacts caused by the POPs. The Task Force agreed also that further scientific investigations on MSCE-POP model are required. The results of these investigations will be published in the nearest future in the final EMEP/MSC-E Technical Report.

5.2. Task Force on POPs

In 2006 MSC-E continued to contribute to the work of the Task Force on POPs requested by the Executive Body to conduct a technical review of dossiers of new proposed substances [*ECE/EB.AIR/87*]. MSC-E attended two meetings of the Task Force (Dessau, Germany, February 2006; Tallinn, Estonia, May 2006).

Modelling data as an additional information for the evaluation of hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), polychlorinated naphthalenes (PCN), and dicofol as potential new POPs in accordance with the two criteria: potential for long-range transboundary atmospheric transport (LRTP) and overall persistence (P_{ov}) were provided by MSC-E to the Task Force on POPs through the UN ECE Secretariat. This information has been submitted in a number of information notes [*Vulykh et al*, 2005 a,b,c,d], posted on Convention's website and summarized in the MSC-E technical report [*Vulykh et al*., 2006]. Below a brief description of the model approach to the evaluation of LRTP and P_{ov} for the new substances is presented. To characterize the LRTP, model estimates of half-life of the considered chemicals in the atmosphere ($T_{1/2}^{air}$, days) calculated with allowance of all processes removing them from the atmosphere are used. LRTP is also illustrated by the Transport Distance (*TD*, km) that is the distance from the source at which annual mean atmospheric concentration of a

chemical in question drops 1000 times compared with the concentration near the point source. Overall persistence is enumerated by half-life in the environment ($T_{1/2}^{env}$, days) estimated for the considered substances on the basis of the model calculations of their atmospheric transport taking into account deposition processes, degradation and exchange of a pollutant between main environmental media. The results of calculations for HCBD, PeCB, BDE-47 and BDE-99, PCN-47, dicofol, pentachlorophenol (PCP) and two isomers of endosulfan (α- and β-) are illustrated by the plots in Fig. 5.1. Evaluated values of the long-range transport potential and overall persistence of the above listed substances are compared with those of two well-known substances already included in the Protocol on POPs – benzo(a)pyrene (B[a]P) and hexachlorobenzene (HCB) – used as benchmark substances of regional and global concern, respectively.

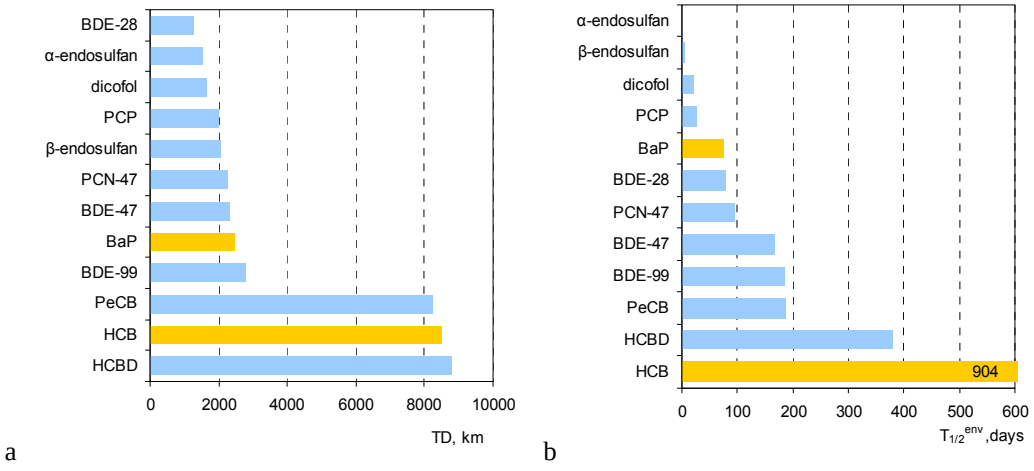


Fig. 5.1. *Transport distance (a) and overall persistence (b) for selected substances*

In addition to the calculations of *TD* and $T_{1/2}^{env}$, model simulations allow evaluating spatial distributions of the pollution by new substances originated from a conventional emission point source. As an example, Fig. 5.2 shows the maps of air concentrations of PCN-47 originated from the conventional point sources of identical power (1 t/y) located at three different points of the northern hemisphere (in Europe, North America and Asia).

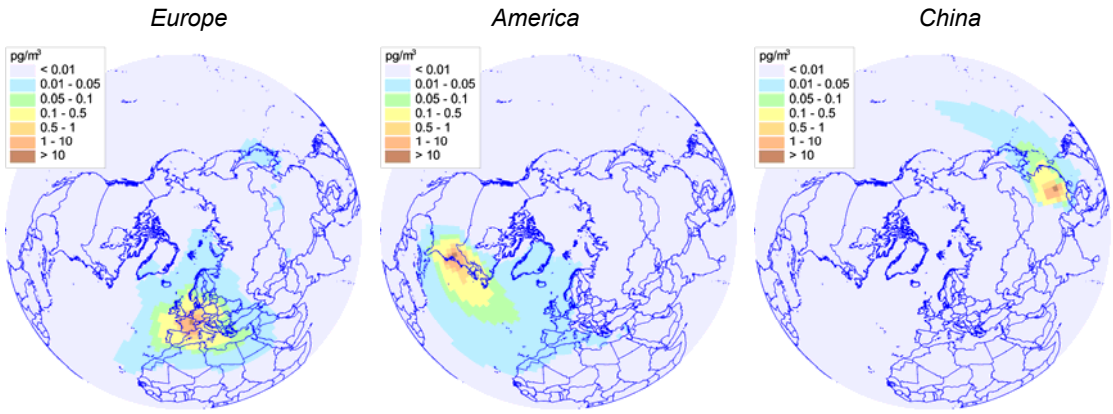


Fig. 5.2. *The spatial distribution of air pollution by PCN-47 from sources (1 t/y) located in Europe, America, and China*

At present, the activities on evaluation of LRTP and Pov of potential new POPs are continued. In particular, within the framework of the third stage of the POP model intercomparison study initiated under EMEP the comparison of different POP models from the viewpoint of evaluation of LRTP and Pov on the basis of 14 reference pollutants with wide variety of physical-chemical properties is carried out. Besides, it is planned to proceed with model evaluation of LRTP and Pov for such pollutants as octaBDE, PFOS, and SCCP.

5.3. Task Force on Hemispheric Transport of Air Pollution

In the framework of co-operation with the EMEP Task Force on Hemispheric Transport of Air Pollution (TF HTAP) MSC-E took part in the organizational workshop on Intercontinental Transport Modelling Intercomparison, which was held in Washington in January 2006. The aim of the workshop was to organize the evaluation and intercomparison of global and regional models of intercontinental transport of pollutants. MSC-E presented the information on EMEP experience in the organization of POP model intercomparison study and evaluation of its results.

MSC-E has supported the organization of the second meeting of the TF HTAP convened in Moscow in June 2006. The meeting considered the hemispheric transport of POPs, mercury, and methane and discussed the outline and process of preparation of the TF HTAP assessment report. MSC-E has chaired the session devoted to the discussion of the scientific issues relevant to intercontinental transport of POPs. During this session MSC-E and national experts from the USA, Canada, Norway, and the Czech Republic presented information on the state of the art in monitoring, emission inventories and modelling of POPs with emphasis to intercontinental transport and its contribution to POP pollution levels in different regions of the northern hemisphere. It is recognized that there is a well-documented evidence for the intercontinental and hemispheric transport of POPs.

The following issues were discussed as the recommendation for the future work in the evaluation of POP intercontinental transport and its contribution to the pollution levels on the regional scale:

- modelling studies of POP intercontinental transport can be carried out for PCBs, HCB, and HCHs due to the availability of global emission inventories and monitoring data for these pollutants;
- while modelling POP intercontinental transport a special attention should be given to such processes as gas-particle partitioning, degradation, and gaseous exchange with underlying surface;
- evaluation of POP source-receptor relationships at hemispheric scale should take into account contributions from direct transport from anthropogenic sources to receptor regions, multi-hop transport and historical emissions;
- further development of nested hemispheric and regional scale models can improve estimates of intercontinental transport contribution to POP pollution levels on the regional scale (EMEP);
- further development of EMEP monitoring strategy including passive sampling to meet the requirements of evaluation of POP intercontinental transport;
- further work on the evaluation of uncertainties in emission inventories and measurement data is required;

- development of tools for the evaluation of new POPs that may be distributed on hemispherical scale;
- development of global POP emission inventories based on existing EMEP inventories in close co-operation with Stockholm Convention, UNEP Chemicals and other international and national programmes.

5.4. Co-operation with national experts

In 2006 MSC-E continued the work on the intercomparison of POP models. Two EMEP expert meetings held by MSC-E in Moscow were attended by national experts from Canada, Germany, Japan, the Netherlands, Norway, Switzerland, the United Kingdom, and the USA. The Stage II of the study aimed at the comparison of POP mass balance estimates, calculated levels of depositions and concentrations in the main environmental compartments and their spatial distribution as well as at the performance of a number of sensitivity studies has been finalized. The main results of the stage are presented in the revised Intermediate Technical Report 5/2006 [*Shatalov et al*., 2006]. The scientific paper devoted to the main outcome of the Stage II is under preparation. The Stage III of the intercomparison study focused on the comparison of relative order in ranking of 14 reference chemicals with respect to their long-range transport potential and overall environmental persistence obtained by different models is ongoing.

The work on the review of MSCE-POP models under the Task Force on Measurement and Modelling was carried out in the collaboration with national experts in monitoring. The report [*Shatalov et al.,* 2005] describing the comparison of calculation results with monitoring data was written in close co-operation with Dr. A.Sweetman and Prof. I.Holoubek.

MSC-East takes part in annual Summer School on Environmental Chemistry and Ecotoxicology organized by EU-DG Research Centre of Excellence for Environmental Chemistry and Ecotoxicology together with the EC DG Research and DG Environment. The representatives of MSC-E are involved in the work of the School as invited lecturers on modelling POP fate in the environment.

This year MSC-E attended the 16th Annual Meeting of SETAC Europe (Society of Environmental Toxicology and Chemistry) held in The Hague in May 2006. The meeting was focused on Controversies and Solutions in Environmental Sciences. MSC-E made a presentation “POP fate modelling: Assessment of environmental pollution with the help of MSCE-POP multicompartment transport model” prepared in cooperation with the national experts from the Czech Republic and the UK (Prof. I Holoubek and Dr. A. Sweetman). The participants were informed of the recent progress in the development of EMEP modelling approach to the assessment of long-range atmospheric transport and levels of environmental concentrations and depositions for a wide range of POPs: PCDD/Fs, PAHs, PCBs, HCB, and γ-HCH. The comparison between the model results and measurement data taken from EMEP stations and national monitoring networks were also presented.

6. FUTURE ACTIVITIES

In order to further improve the quality of estimates of the levels of POP pollution in the EMEP region, the following activities are proposed:

- Further develop of regional and hemispheric versions of the MSCE-POP model in accordance with the recommendations of the EMEP/TFMM Workshop on model review:
  - refine datasets of physical-chemical properties used for modelling;
  - develop the model parameterization for POP resuspension and volatilization from soils;
  - improve the model description of degradation in the atmosphere and deposition processes;
  - prepare meteorological and geophysical data for hemispherical/global modelling.
- Prepare input data for the model application. Employ the ECMWF reanalysis for the data preprocessing; prepare mapped emission data for regional modelling based both on official and expert estimates.
- Continue the development of hemispheric/regional approach to evaluation of pollution levels in the EMEP domain. Calculate PCDD/Fs and PAHs dispersion at the hemispheric scale for evaluation of the European pollution from global sources and boundary conditions for the EMEP regional modelling.
- Start the development of inverse modelling for selected POPs on the basis of measurement data including passive sampling.
- On the basis of the improved model prepare the information on PAHs and toxic congeners of PCDD/Fs for 2005:
  - air concentrations and depositions over Europe;
  - evaluation of source-receptor relationships;
  - estimates of depositions on marginal seas (Mediterranean, Baltic, Black and North Seas);
  - evaluation of media response to possible emission reduction scenario for PCDD/Fs.
- Further investigate intercontinental transport of air pollution for selected POPs (PCBs, HCB and γ-HCH) at the hemispheric scale and evaluate its impact on contamination levels of the EMEP region, Kazakhstan and Kyrgyzstan.
- Compare model results (concentration in air and precipitation, deposition fluxes) against available monitoring data.
- Complete Stage III of the POP model intercomparison study (comparison of different model approaches to ranking a number of reference chemicals with respect to long-range transport potential and overall persistence) and the analysis of agreement and discrepancies between model simulations of previous stages; cooperate with national experts on POP modelling issues.
- Contribute to the activities of the TF on POPs and WGE, co-operate with AMAP, HELCOM, EC, UNEP.
- Cooperate with the TF HTAP on evaluation of the intercontinental transport of POPs.

CONCLUSIONS

In this Status Report the progress in the evaluation of POP pollution levels and source-receptor relationships within the European region and at the hemispheric scale is discussed.

The main conclusions of the work carried out are summarized below.

Monitoring of POPs

- Measurements of POP concentrations in air and in precipitation, and of deposition fluxes for 2004 were performed at 15 monitoring sites of EMEP and submitted to CCC. Compared to the previous year the number of sites measuring POPs is increased by 3 sites (in Spain, the United Kingdom, and Latvia).
- 6 monitoring sites submitted data on simultaneous measurements of POP concentrations in air and in precipitation. The spatial distribution of monitoring sites across Europe is non-uniform and requires further improvement.
- Measurements of air concentrations indicate a clear south-north gradient with elevated concentration levels in Central Europe for most of the POPs. The ratio of measured concentrations PCB-28/PCB-180 increases towards north indicating marked differences in the long-range transport potential of different PCB congeners.

Emission data

- The official data on POP emissions (PAHs, PCDD/Fs, HCB, PCBs and HCH) for the period from 1990 to 2004 (at least for one year) were submitted by 36 Parties to the Convention. It should be noted that the number of countries reporting official information on POP emissions and their spatial distribution is gradually increasing. Nevertheless, to estimate total emission of POPs and their spatial distribution over the European region expert estimates still have to be used.
- According to the official data and expert estimates the emissions of all the pollutants of concern tend to decrease from 1990 to 2004. In particular, emissions in the European region of four indicator PAHs (benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), indeno[1,2,3-cd]pyrene (I_P)) have decreased by 17 - 22% depending on particular PAH, European emissions of PCDD/Fs by 35%, of HCB by 25% and of γ-HCH by 93%.

Model review and evaluation

- According with the conclusions of the EMEP/TFMM Workshop on the Review of the EMEP Models on Heavy Metals and Persistent Organic Pollutants (Moscow, Russia, Ocober, 2005) “The model is considered to represent the state-of-the-science and to fit for the purpose of evaluating the contributions of long-range transport to the environment impacts caused by POPs for the evaluation of the long-range transboundary transport and deposition of POPs in Europe”. Along with that the TFMM Workshop recommended to continue further improvement of modelling approach for POPs.
- Key physical-chemical properties of PCDD/Fs, PCBs, PAHs, HCHs, and HCB harmonised in accordance with recently developed adjustment procedure were applied in modelling.

- Improvement of the agreement between MSCE-POP model results and measurements for B[a]P due to the refinement of model description of degradation process and usage of different scenarios of seasonal variations of emission was analysed.
- Model description of POP absorption in soil and volatilisation was modified taking into account temperature dependence of the octanol/water partitioning coefficient K_{OW}. Modification of model parameterisation of POP partitioning in soil resulted in the refinement of calculated POP re-emission flux on the seasonal level.
- The refined description of POP partitioning between gaseous, dissolved, and sorbed on particles phases in seawater has been incorporated into the MSCE-POP model together with the description of sedimentation process. This resulted in the improvement of the evaluation of re-volatilisation fluxes from marine surface.
- Combined hemispheric/regional modelling approach to evaluation of POP intercontinental transport within the northern hemisphere with emphasis to the European region is under development.
- The compatibility of model input data for hemispheric and regional scale modelling is required for further development of hemispheric/regional modelling approach for POPs.

Model assessment of POP pollution levels

- Model evaluation of the environmental pollution levels within the European region was carried out for the four indicator PAHs, PCDD/Fs, PCBs, γ-HCH and HCB for 2004. Transboundary transport of B[a]P and PCDD/Fs in 2004 was evaluated. For PCBs, γ-HCH and HCB the emphasis was put to the evaluation of intercontinental transport and contamination of remote regions (the Arctic).
- The levels of net deposition flux of PCDD/Fs in Europe differ from about 0.1 ng TEQ/m^2/y in northern Europe (Norway, northern parts of Sweden and Finland) to 5 ng TEQ/m^2/y and higher in central and southern Europe (Germany, Poland, Czech Republic). Maximum pollution levels are calculated for Germany and neighbouring countries. The reason for that is high values of officially reported emission levels in Germany (about 30% of total PCDD/F toxicity in Europe). High values of deposition fluxes (0.5 – 2 ng TEQ/m^2) are calculated for Turkey and Ukraine.
- The transboundary transport of PCDD/Fs was evaluated taking into account national anthropogenic emission sources of European countries, non-EMEP anthropogenic sources (USA and Canada) and re-emission. The contribution of transboundary transport within the EMEP region typically ranges from 30% to 70%. The contributions of non-EMEP anthropogenic sources to the pollution of European countries can be noticeable and reach up to 20% for countries located close to EMEP boundaries. The input of re-emission to depositions in European countries is in the range from 20% to 40%.
- Annual mean B[a]P concentrations in the surface atmospheric layer vary from 0.1 to 1 ng/m^3 and more. High levels of contamination are characteristic for Germany, Ukraine, Belgium and the Netherlands (up to 1 ng/m^3 and higher). Spatial distribution of the rest three indicator PAHs (B[b]F, B[k]F, and I_P) is similar to that of B[a]P. The levels of air concentrations of B[k]F in Europe are lower than for the rest three indicator compounds (from 0.01 – 0.05 ng/m^3 in the northern Europe up to 0.3 ng/m^3 in eastern and southern European regions).
- The transboundary transport of B[a]P between European countries was evaluated. The contribution of the external sources to air concentrations and depositions of B[a]P is essential and varies typically from 20 to 80%.

- Preliminary estimates of intercontinental transport of selected PCB congeners, HCB, and γ-HCH were carried out using hemispheric version of the MSCE-POP model. Intercontinental transport of these POPs makes a noticeable contribution to the contamination of different regions of the northern hemisphere including remote regions like the Arctic.

Co-operation

- MSC-E contributed to the work of the Task Force on POPs on technical review of dossiers of new proposed substances providing model estimates of long-range transport potential and persistence for hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), polychlorinated naphthalenes (PCN), and dicofol.
- MSC-E supported the organization of the second meeting of the EMEP TFHTAP convened in Moscow in June 2006. The meeting considered availability of information on POP emissions and measurements and discussed modelling of source-receptor relationships for POPs. Further directions for the improvement of modelling of POP intercontinental transport were formulated.
- Collaborative work on the EMEP intercomparison study of POP models was continued by MSC-E and national experts. Two expert meetings held by MSC-E in Moscow were attended by the experts in modelling and monitoring of POPs from Canada, Germany, Japan, the Netherlands, Norway, Switzerland, the United Kingdom, and the USA.
- In co-operation with national experts MSC-E continued the studies of POP pollution levels in Europe on the basis of joined interpretation of modelling results and available measurements.

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Annex A

EMEP work-plan for POPs in 2006

2.2. Atmospheric measurements and modelling

Description/objectives

To support the implementation of protocols to the Convention; provide the measurement and modelling tools necessary for further abatement policies; compile and evaluate information on transboundary air pollution and implement the EMEP monitoring strategy adopted in 2004. The Task Force on Measurements and Modelling, led by the United Kingdom and co-chaired by WMO, reviews and assesses the scientific and operational activities of EMEP related to monitoring and modelling, evaluates their contribution to the effective implementation and further development of the protocols and reviews national activities on measurements, modelling and data validation.

Main activities and time schedule for monitoring:

(b) Review, store and make available the 2005 monitoring data (CCC, MSC-W, MSC-E);

Main activities and time schedule for atmospheric modelling for POPs:

(a) Prepare information using 2004 data to evaluate concentration and deposition fields of PAHs and toxic congeners of PCDD/Fs and to assess source-receptor relationships. Compare model results for concentrations in air and precipitation and deposition fluxes with measurements; analyse contributions from northern hemisphere sources to the European region and the contribution of European sources to other regions for PCBs, HCB, and γ-HCH (MSC-E);

(b) Further develop the MSC-E model and its input databases of geophysical and meteorological data for regional and hemispherical modelling, paying special attention to POP transport in the marine environment at the hemispherical scale (MSC-E);

(c) Work on the third stage of the POP model intercomparison study; ranking a number of chemicals with respect to long-range transport potential and overall persistence (MSC-E, Parties);

(d) Work on evaluation of long-range transport potential and persistence of new substances (MSC-E).

2.4. Hemispheric transport of air pollution

Description/objectives:

To develop a fuller scientific understanding of the hemispheric transport of air pollution and estimate such transport for specific air pollutants. The Task Force on the Hemispheric Transport of Air Pollution, led by the United States and the European Community, will coordinate activities, including collaboration with other international bodies and networks with related interests within and outside the UNECE region.

Main activities and time schedule:

(c) Link measurements at regional and hemispheric scales; investigate further the intercontinental transport of air pollution and assess its impact on European surface pollution levels, using the EMEP monitoring data (CCC, MSC-E, MSC-W).

Annex B

COUNTRY-TO-COUNTRY DEPOSITION MATRICES FOR 2004

Table B.1*. Country codes*

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

***Table B.2**. Matrix of B[a]P country-to-country depositions in 2004, kg/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BE	BG	BA	BY	CH	CS	CY	CZ	DE	DK	EE	
AL	346	0.1	2.2	0.2	2.4	26.1	13.4	2.5	0.3	102	0.5	6.2	4.5	0.9	0.7	**AL**
AM	0.4	225	0.2	51.5	0.3	1.3	0.4	1.2	0.03	0.9	1.3	0.6	0.6	0.2	0.2	**AM**
AT	8.1	0.1	532	0.2	47.4	13.0	28.9	8.8	12.7	39.1	0.1	205	176.6	13.7	4.3	**AT**
AZ	1.0	70	0.5	551	1.0	4.0	1.3	4.6	0.1	2.9	2.7	1.8	1.8	0.9	1.2	**AZ**
BE	0.3	0.02	2.3	0.03	1132	0.6	0.7	1.1	1.3	0.9	0.03	5.9	86.3	4.8	0.8	**BE**
BG	33.3	1.1	10.6	1.9	10.4	1677	34.8	27.6	1.0	274	3.7	36.0	22.9	6.7	5.7	**BG**
BA	39.0	0.2	22.2	0.3	11.3	27.9	1084	8.6	1.3	226	0.8	55.7	27.4	4.8	2.9	**BA**
BY	6.3	1.8	16.9	3.4	39.3	27.6	17.4	2336	2.0	35.4	0.9	103	84.2	43.8	66.4	**BY**
CH	1.7	0.02	11.1	0.0	15.8	1.6	4.0	1.1	71.1	4.3	0.1	8.6	46.8	2.4	0.6	**CH**
CS	119	0.5	20.9	0.9	12.5	220	223	16.6	1.4	1908	2.1	62.1	30.3	6.2	5.0	**CS**
CY	0.5	0.2	0.2	0.2	0.3	1.3	0.5	0.6	0.04	1.1	166	0.6	0.5	0.1	0.1	**CY**
CZ	4.6	0.2	97.6	0.3	49.1	11.6	15.9	11.4	5.7	30.6	0.2	1325	207	18.2	5.6	**CZ**
DE	5.8	0.2	122	0.4	845	10.9	14.5	26.2	53.5	22.0	0.4	323	3105	109	16.8	**DE**
DK	0.4	0.04	1.6	0.1	18.8	0.9	0.9	3.4	0.3	1.4	0.0	8.4	32.6	204	2.5	**DK**
EE	0.7	0.2	2.1	0.4	11.1	2.2	1.7	37.6	0.3	3.0	0.1	12.5	22.1	13.8	686	**EE**
ES	4.4	0.1	6.4	0.2	70.6	5.5	9.6	4.2	3.1	9.2	0.8	13.0	52.9	10.7	3.2	**ES**
FI	2.4	1.4	7.6	2.5	56.6	9.0	5.6	105	1.3	10.3	0.8	45.0	94.4	69.3	357	**FI**
FR	8.3	0.2	26.9	0.3	541	9.7	21.5	9.5	46.0	21.6	0.9	52.1	390	30.2	6.9	**FR**
GB	0.9	0.1	3.8	0.1	89.6	2.1	2.0	4.1	1.3	2.9	0.2	13.3	61.4	12.8	3.2	**GB**
GE	2.1	76.4	1.0	81.0	1.9	8.5	2.7	7.9	0.2	5.7	4.0	3.5	3.4	1.6	1.4	**GE**
GR	66.0	0.8	5.5	1.3	6.7	221	20.3	12.8	0.7	93.7	6.4	17.6	13.0	3.5	3.0	**GR**
HR	20.0	0.1	41.0	0.2	10.4	20.2	245	7.1	1.5	116	0.5	65.2	27.2	5.0	2.3	**HR**
HU	10.6	0.3	77.5	0.5	15.8	33.1	64.7	16.4	2.0	168	0.6	137	44.2	8.2	4.4	**HU**
IE	0.2	0.01	0.5	0.03	7.4	0.3	0.4	0.6	0.2	0.6	0.1	1.7	6.0	2.0	0.6	**IE**
IS	0.2	0.05	0.8	0.1	6.0	0.5	0.6	1.4	0.2	0.7	0.03	2.7	6.7	2.4	0.7	**IS**
IT	60.2	0.4	89.4	0.6	43.0	50.6	137.4	13.9	20.8	116	5.2	97.6	97.7	13.6	5.7	**IT**
KZ	2.9	10.0	3.7	34.8	9.5	16.6	5.7	46.1	0.5	12.0	1.7	14.9	16.5	8.6	14.5	**KZ**
LT	1.6	0.3	6.4	0.6	19.5	4.7	4.3	162	0.8	7.9	0.1	42.0	42.9	26.3	33.1	**LT**
LU	0.03	0.001	0.3	0.002	24.9	0.1	0.1	0.1	0.2	0.1	0.003	0.7	11.5	0.4	0.1	**LU**
LV	1.5	0.4	5.0	0.7	20.4	4.6	3.7	118.2	0.7	6.9	0.2	30.5	42.1	29.8	117.2	**LV**
MD	1.7	0.4	1.6	0.7	2.7	18.7	3.4	16.6	0.2	8.8	0.5	7.1	5.6	2.1	2.2	**MD**
MC	0.001	0.0	0.003	0.0	0.002	0.001	0.004	0.001	0.001	0.003	0.0	0.004	0.005	0.001	0.0	**MC**
MK	63.0	0.2	2.3	0.3	2.1	81.2	10.4	3.2	0.2	127	1.0	7.7	4.6	1.0	0.9	**MK**
NL	0.2	0.0	1.9	0.02	255	0.5	0.5	1.2	0.6	0.7	0.03	6.3	115.8	5.4	0.9	**NL**
NO	2.1	0.3	6.2	0.6	62.6	6.1	4.8	23.6	1.3	8.8	0.4	31.3	85.3	84.9	23.5	**NO**
PL	12.8	1.2	75.1	1.9	158	36.8	40.6	207	7.8	78.3	0.8	1000	493	132	52.4	**PL**
PT	0.7	0.0	0.8	0.0	9.0	0.8	1.2	0.7	0.3	1.3	0.1	1.7	7.1	1.8	0.6	**PT**
RO	37.7	2.4	36.5	4.1	31.8	399	111	87.9	3.2	416	5.9	113.2	69.9	19.6	17.2	**RO**
RU	46.3	87.8	66.9	208	219	239	91.8	1570	9.4	192	24.8	321	391	233	947	**RU**
SK	7.2	0.3	47.8	0.5	17.2	20.9	27.3	20.4	1.9	61.4	0.4	281	48.0	10.3	5.8	**SK**
SI	5.1	0.04	67.6	0.1	6.6	7.5	27.2	3.2	1.4	25.9	0.1	36.4	19.2	2.9	1.3	**SI**
SE	4.0	1.0	11.7	2.0	126	13.9	9.9	88.5	2.0	18.9	0.7	74.9	196	337	111.3	**SE**
TR	29.0	87.7	12.9	33.5	19.8	195	30.5	67.3	1.7	77.5	139	42.8	37.8	13.9	11.9	**TR**
UA	24.1	10.1	43.4	17.8	67.5	163	64.0	544	4.6	144	10.1	206	144	52.4	52.4	**UA**
Total, t/y	0.98	0.58	1.49	1.00	4.10	3.60	2.39	5.63	0.27	4.38	0.38	4.82	6.38	1.55	2.58	**Total, t/y**
	AL	**AM**	**AT**	**AZ**	**BE**	**BG**	**BA**	**BY**	**CH**	**CS**	**CY**	**CZ**	**DE**	**DK**	**EE**	

***Table B.2**. Matrix of B[a]P country-to-country depositions in 2004, kg/y (continued)*
Receptors ↓ Emitters →

	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	MD	
AL	25.0	0.6	5.8	2.2	0.3	41.6	3.6	10.1	1.1	0.01	106	0.1	1.0	0.2	2.9	0.6	**AL**
AM	1.7	0.2	0.4	0.4	24.4	1.2	0.2	0.6	0.1	0.002	3.3	0.4	0.3	0.0	0.9	0.2	**AM**
AT	67.0	3.4	54.2	27.0	0.3	8.9	31.9	109	8.9	0.1	335	0.1	5.5	4.4	17.7	0.8	**AT**
AZ	5.3	0.8	1.3	1.3	68.4	3.2	0.5	1.8	0.5	0.01	9.1	3.4	1.2	0.1	4.0	0.7	**AZ**
BE	57.4	0.8	114	46.9	0.04	0.5	0.4	1.2	11.1	0.1	12.1	0.0	0.9	26.9	3.2	0.0	**BE**
BG	42.5	3.9	13.6	9.0	2.4	117	11.7	63.8	3.6	0.04	130	0.5	8.6	0.8	22.6	9.5	**BG**
BA	51.8	2.6	17.0	8.6	0.3	22.5	107	118	3.2	0.04	290	0.1	4.6	0.9	12.1	0.9	**BA**
BY	58.3	31.0	29.2	36.9	3.9	11.3	9.3	57.8	13.1	0.2	74.0	1.2	273	2.4	485	12.4	**BY**
CH	52.8	0.6	77.0	12.4	0.04	2.2	3.2	4.4	4.6	0.04	291	0.01	0.6	1.6	2.2	0.1	**CH**
CS	56.3	4.0	17.7	9.8	0.9	66.8	46.0	170	3.9	0.04	251	0.2	7.4	1.0	19.8	2.8	**CS**
CY	2.8	0.1	0.7	0.4	0.2	2.2	0.2	0.6	0.1	0.002	5.6	0.02	0.2	0.0	0.5	0.1	**CY**
CZ	51.3	4.4	44.9	26.9	0.4	6.0	12.5	94.8	9.4	0.1	82.0	0.1	7.4	4.5	23.2	1.0	**CZ**
DE	332	14.1	450	227	0.6	7.9	9.4	45.0	64.1	0.5	224	0.2	17.8	91.0	64.4	1.2	**DE**
DK	20.6	2.4	10.2	24.1	0.1	0.5	0.5	2.7	6.6	0.1	7.3	0.01	2.8	0.7	9.3	0.1	**DK**
EE	14.8	49.6	7.0	11.3	0.4	1.0	0.9	4.4	4.0	0.1	9.7	0.1	40.7	0.6	460	0.5	**EE**
ES	9855	3.4	207	66.3	0.2	7.7	6.0	9.1	32.8	0.3	187	0.1	3.0	4.7	10.5	0.3	**ES**
FI	77.2	1886	35.4	62.4	2.8	4.3	3.0	15.3	22.1	0.5	37.9	1.3	82.0	2.9	475	2.2	**FI**
FR	1614	6.7	2779	221	0.4	12.7	14.6	23.6	87.2	0.6	613	0.1	7.2	72.9	25.7	0.5	**FR**
GB	191	3.2	74.2	918	0.1	1.6	1.1	4.0	164	0.5	23.0	0.03	3.1	3.8	11.8	0.2	**GB**
GE	9.8	1.0	2.6	2.3	571	6.8	1.0	3.5	1.0	0.01	19.6	0.9	2.2	0.1	5.9	1.5	**GE**
GR	45.4	2.1	11.8	6.2	1.7	749	6.7	26.9	2.7	0.03	139	0.4	4.3	0.5	11.7	3.6	**GR**
HR	42.6	1.9	15.6	7.5	0.3	15.7	335	172	2.8	0.03	273	0.05	3.8	0.8	10.3	0.7	**HR**
HU	37.2	3.4	18.4	10.9	0.6	12.9	78.9	1287	4.0	0.04	152	0.1	7.5	1.3	19.5	1.5	**HU**
IE	38.4	0.7	10.4	35.5	0.0	0.4	0.2	0.7	265	0.2	4.6	0.01	0.5	0.4	2.0	0.03	**IE**
IS	22.7	0.9	5.5	11.0	0.1	0.3	0.3	1.2	5.8	4.3	5.5	0.03	0.8	0.3	2.7	0.1	**IS**
IT	297	5.0	131	36.2	0.8	79.1	92.7	106	14.4	0.2	7563	0.2	7.3	3.7	22.0	1.8	**IT**
KZ	29.3	11.1	9.1	11.3	19.0	7.5	2.6	11.2	4.4	0.1	31.9	310	14.3	0.6	50.3	4.0	**KZ**
LT	26.0	14.6	13.1	18.4	0.7	2.5	2.6	14.6	6.3	0.1	22.9	0.2	691	1.1	475	1.5	**LT**
LU	4.8	0.1	14.0	2.3	0.003	0.1	0.1	0.1	0.6	0.0	1.5	0.001	0.1	28.6	0.3	0.004	**LU**
LV	30.1	28.0	13.6	20.7	0.8	2.2	2.1	11.1	7.1	0.1	20.6	0.2	220	1.2	2622	1.3	**LV**
MD	6.5	1.3	2.5	2.4	0.9	3.3	1.4	7.3	0.9	0.01	13.5	0.2	3.7	0.2	9.2	97.8	**MD**
MC	0.015	0.0	0.021	0.002	0.0	0.002	0.003	0.004	0.001	0.0	0.2	0.0	0.0	0.0	0.0	0.0	**MC**
MK	13.5	0.7	3.7	1.9	0.3	68.4	3.1	13.7	0.8	0.01	52.0	0.1	1.3	0.2	3.6	0.7	**MK**
NL	43.1	0.8	40.0	46.7	0.0	0.4	0.3	1.2	10.5	0.1	7.8	0.01	1.0	3.5	3.6	0.1	**NL**
NO	101	37.9	40.3	117	0.7	3.4	2.7	13.7	39.7	0.7	33.4	0.2	18.5	2.8	73.7	1.0	**NO**
PL	175	33.5	108	108	2.9	19.3	27.8	196	35.0	0.4	188	0.5	123	10.7	256	6.5	**PL**
PT	1013	0.6	21.0	10.8	0.04	1.2	0.7	1.1	6.0	0.1	19.3	0.01	0.5	0.6	1.7	0.05	**PT**
RO	97.3	11.4	37.5	25.4	5.9	64.5	42.9	287	9.7	0.1	293	0.9	27.2	2.4	71.1	49.1	**RO**
RU	478	800	174	262	277	109	42.5	198	101.1	1.8	482	262	514	12.4	2288	59.4	**RU**
SK	31.1	4.3	18.0	12.2	0.6	10.0	20.3	271	4.4	0.05	84.5	0.1	10.1	1.4	25.8	1.6	**SK**
SI	22.8	1.0	10.5	4.5	0.1	5.3	69.8	59.3	1.6	0.02	222	0.02	1.8	0.6	5.3	0.3	**SI**
SE	147.9	234.2	70.7	147.0	2.1	6.8	5.4	29.5	46.5	0.7	56.0	0.8	76.7	5.4	326	2.8	**SE**
TR	113.4	8.6	30.3	20.9	56.6	163	12.4	48.1	8.9	0.1	249	2.3	17.2	1.3	47.2	20.6	**TR**
UA	142	32.8	61.0	60.1	26.8	54.7	33.3	251	22.2	0.3	258	6.5	97.3	4.6	233	104	**UA**
Total, t/y	15.55	3.25	4.80	2.69	1.07	1.71	1.05	3.75	1.04	0.01	12.88	0.59	2.31	0.30	8.22	0.39	**Total, t/y**
	ES	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KZ**	**LT**	**LU**	**LV**	**MD**	

***Table B.2**. Matrix of B[a]P country-to-country depositions in 2004, kg/y (continued)*
Receptors ↓ Emitters →

	MC	MK	NL	NO	PL	PT	RO	RU	SK	SI	SE	TR	UA	ReEmis	Total, t/y	
AL	0.0	31.2	2.0	0.4	28.2	2.9	15.1	1.6	5.5	2.0	1.2	24.1	39.7	60	0.9	**AL**
AM	0.0	0.2	0.3	0.1	3.9	0.2	1.9	2.1	0.4	0.2	0.3	78.1	27.9	29	0.5	**AM**
AT	0.0	2.6	37.8	4.6	332	6.0	19.5	3.4	55.9	121.1	11.1	14.6	49.4	149	2.6	**AT**
AZ	0.0	0.5	1.0	0.4	13.2	0.7	6.1	16.8	1.4	0.5	1.4	112	118	81	1.1	**AZ**
BE	0.0	0.1	121.2	2.1	18.0	6.2	0.8	0.6	1.1	0.7	3.6	1.6	4.3	60	1.7	**BE**
BG	0.0	50.9	9.6	2.6	195.1	4.8	368	14.7	36.5	8.4	7.9	264	401	277	4.2	**BG**
BA	0.0	7.3	10.6	1.8	201	4.7	37.4	3.0	47.6	22.5	5.7	31.0	61.8	213	2.8	**BA**
BY	0.0	3.0	41.5	14.0	1334	7.4	84.3	92.6	67.1	10.5	63.4	60.2	1298	433	7.4	**BY**
CH	0.0	0.4	9.8	1.1	22.2	4.1	2.0	0.5	2.4	5.8	1.8	4.3	5.8	43	0.7	**CH**
CS	0.0	69.8	11.8	2.4	253	5.9	161	6.9	64.3	15.9	7.8	81.5	159	313	4.4	**CS**
CY	0.0	0.2	0.3	0.1	3.0	0.2	1.5	0.5	0.4	0.2	0.2	52.8	11.4	15	0.3	**CY**
CZ	0.0	1.9	45.8	5.4	881	5.0	20.5	4.4	134	18.8	14.9	13.6	64.1	147	3.5	**CZ**
DE	0.0	1.9	753	26.1	751	34.5	19.3	10.3	37.7	16.0	66.4	24.6	101	424	8.5	**DE**
DK	0.0	0.1	28.4	6.5	59.4	2.0	1.7	1.5	2.5	0.7	23.7	1.8	14.9	26	0.5	**DK**
EE	0.0	0.3	12.7	6.3	126	1.6	5.1	23.2	4.9	1.1	42.6	5.5	67.1	82	1.8	**EE**
ES	0.0	1.2	46.2	6.4	53.9	596	6.0	2.6	5.1	7.6	9.8	22.3	27.9	783	12.2	**ES**
FI	0.0	1.1	64.7	48.6	413	8.5	20.9	127	17.9	3.7	410	33.2	303	219	5.2	**FI**
FR	0.01	2.4	206	13.8	162	103	12.4	5.2	14.7	21.8	23.9	29.2	46.4	590	7.9	**FR**
GB	0.0	0.3	75.8	8.4	53.4	23.1	3.1	2.3	3.6	1.6	10.9	4.3	20.7	85	1.9	**GB**
GE	0.0	1.1	1.9	0.7	25.9	1.3	13.3	19.8	2.7	0.9	2.2	265	226	135	1.5	**GE**
GR	0.0	58.7	5.9	1.5	97.8	5.3	76.3	9.0	16.3	4.6	4.4	309	227	195	2.5	**GR**
HR	0.0	4.3	9.5	1.7	205	3.6	26.3	2.4	50.3	120	5.1	22.9	45.2	160	2.1	**HR**
HU	0.0	5.0	15.0	2.6	438	3.4	94.7	4.9	329	61.6	8.3	31.0	123	198	3.5	**HU**
IE	0.0	0.1	6.2	1.5	7.4	4.8	0.6	0.4	0.5	0.3	1.9	0.8	3.8	15	0.4	**IE**
IS	0.0	0.1	6.1	2.5	14.1	3.5	1.1	0.7	1.1	0.4	2.5	1.5	8.1	4.3	0.1	**IS**
IT	0.01	13.9	33.4	5.6	318	25.9	49.6	6.2	55.1	145	13.3	144	122	669	10.7	**IT**
KZ	0.0	1.5	9.8	4.5	120	4.1	36.1	204	9.7	2.8	15.7	107	1027	285	2.5	**KZ**
LT	0.0	0.7	21.4	7.4	533	3.0	12.5	22.6	16.9	3.4	39.9	9.0	147	115	2.6	**LT**
LU	0.0	0.01	3.8	0.2	1.7	0.4	0.1	0.0	0.1	0.1	0.3	0.2	0.3	5.1	0.1	**LU**
LV	0.0	0.6	22.2	10.4	332	3.2	11.5	26.3	12.3	2.7	62.4	9.9	137	144	4.1	**LV**
MD	0.0	1.0	2.8	0.8	70.9	0.8	147	7.4	5.7	1.3	2.6	29.3	287	53	0.8	**MD**
MC	0.0	0.0	0.002	0.0	0.01	0.001	0.001	0.0	0.002	0.01	0.0	0.003	0.0	0.02	0.0	**MC**
MK	0.0	169	1.9	0.4	35.7	1.4	19.7	1.9	7.4	1.8	1.3	38.9	43.6	61	0.9	**MK**
NL	0.0	0.1	618	2.5	20.4	5.8	0.7	0.6	1.2	0.5	3.8	1.4	4.6	40	1.2	**NL**
NO	0.0	0.9	76.5	387	243	11.6	12.7	15.6	13.1	3.3	150	10.9	105	84	1.9	**NO**
PL	0.0	5.5	167	26.2	12256	21.9	90.9	42.0	373	36.8	109	57.1	577	667	18.0	**PL**
PT	0.0	0.2	6.5	1.1	7.2	1851	0.9	0.4	0.6	0.9	1.7	3.4	4.4	208	3.2	**PT**
RO	0.0	24.0	29.5	7.0	693	11.0	2778	35.0	145	31.8	22.7	269	1012	530	8.0	**RO**
RU	0.0	24.0	239	136	2908	56.1	508	8007	188	45.8	571	1440	12468	3305	40.6	**RU**
SK	0.0	3.3	16.5	3.1	948	3.0	55.4	5.7	939	24.9	10.8	22.2	121	173	3.4	**SK**
SI	0.0	1.6	5.8	1.0	102	1.9	10.1	1.2	22.4	478	2.7	7.7	18.1	81	1.3	**SI**
SE	0.0	1.8	159	169	729	15.1	31.4	47.3	32.0	6.4	1878	30.0	310	277	5.8	**SE**
TR	0.0	14.8	19.0	6.5	285	13.2	221	71.6	33.7	11.2	18.2	9834	1630	1083	14.8	**TR**
UA	0.0	13.3	68.5	18.3	2055	16.8	577	271	214	33.3	64.9	500	23043	1667	31.5	**UA**
Total, t/y	0.0	0.52	3.02	0.95	27.35	2.89	5.56	9.12	2.97	1.28	3.70	14.00	44.51			
	MC	**MK**	**NL**	**NO**	**PL**	**PT**	**RO**	**RU**	**SK**	**SI**	**SE**	**TR**	**UA**			

***Table B.3**. Matrix of 2,3,4,7,8-PeCDF country-to-country depositions in 2004, mg TEQ/y*

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BE	BG	BA	BY	CH	CS	CY	CZ	DE	DK	EE	
AL	4028	1.3	15	2.2	11	391	111	5.2	182	1141	0.7	74	523	11	0.5	**AL**
AM	3.2	5617	2.4	953	3.4	23	4.2	2.8	38	11	1.7	12	136	4.5	0.2	**AM**
AT	37	1.2	4448	2.5	141	114	130	23	5518	245	0.1	2465	12984	106	3.1	**AT**
AZ	9.5	1267	7.0	14309	11	74	14	12	108	37	3.4	38	431	16	0.8	**AZ**
BE	2.0	0.3	15	0.5	5793	7.6	4.4	2.9	494	8.1	0.02	70	9906	42	1.1	**BE**
BG	221	10	59	17	40	37209	209	51	517	2739	3.2	372	2188	60	2.8	**BG**
BA	234	1.4	107	2.7	36	290	9510	17	551	2123	0.4	473	2189	38	1.6	**BA**
BY	27	12	84	25	114	248	75	7304	787	212	0.6	965	7310	358	31	**BY**
CH	9.5	0.4	78	0.9	55	21	23	3.9	52398	34	0.1	130	4090	28	0.8	**CH**
CS	898	4.0	110	7.4	45	2772	1707	32	647	26523	1.1	621	2738	56	2.4	**CS**
CY	3.1	2.3	1.3	1.7	1.4	23	3.2	1.6	18	10	226	6.6	62	1.7	0.1	**CY**
CZ	21	1.4	643	2.8	138	104	73	22	2312	202	0.1	21854	17316	150	3.4	**CZ**
DE	38	3.7	978	8.3	2547	144	92	68	27536	196	0.3	5299	418679	1166	15	**DE**
DK	1.9	0.4	9.3	0.9	67	9.8	4.5	7.0	140	11	0.02	100	3250	2982	1.6	**DK**
ES	40	4.2	113	9.0	326	129	93	27	2691	139	0.6	465	8995	211	5.9	**ES**
EE	2.3	1.0	8.0	2.1	24	16	5.6	48	109	14	0.1	93	1463	93	308	**EE**
FI	11	8.2	35	17	140	83	25	138	520	64	0.8	351	6853	450	136	**FI**
FR	62	5.2	250	11	2269	176	162	39	24982	240	0.6	949	49194	393	10	**FR**
GB	8.7	1.8	42	4.1	405	42	20	14	841	41	0.1	247	7911	168	3.6	**GB**
GE	12	1224	8.6	1484	12	102	16	14	126	46	3.4	45	487	18	0.8	**GE**
GR	693	11	48	16	40	4525	181	30	586	1080	6.2	251	1905	48	2.1	**GR**
HR	149	1.2	252	2.4	40	234	2204	17	719	1262	0.3	721	2700	46	1.7	**HR**
HU	53	2.2	464	4.4	58	343	410	36	861	1737	0.2	1737	4440	80	2.6	**HU**
IE	3.4	0.7	13	1.6	51	14	7.9	4.6	282	15	0.1	64	1368	41	1.0	**IE**
IS	1.2	0.4	4.9	0.9	17	6.5	3.1	2.8	90	5.9	0.02	31	579	21	0.4	**IS**
IT	434	5.0	585	8.8	180	674	957	41	11395	1079	3.0	1231	9759	157	5.0	**IT**
KZ	26	157	39	703	63	258	52	97	550	137	2.1	242	2771	121	8.9	**KZ**
LT	5.9	1.8	26	3.7	49	38	16	318	283	41	0.1	346	3303	225	14	**LT**
LU	0.2	0.02	2.4	0.05	63	0.8	0.5	0.3	93	1.0	0.003	9.6	1617	3.6	0.1	**LU**
LV	4.8	1.7	19	3.7	44	32	13	175	223	33	0.1	227	2767	195	50	**LV**
MC	0.006	0.0	0.02	0.0	0.01	0.01	0.02	0.001	0.5	0.02	0.0	0.05	0.5	0.006	0.0	**MC**
MD	9.7	3.0	12	5.5	10	228	22	35	108	77	0.2	88	604	21	1.2	**MD**
MK	517	1.6	15	2.6	10	1276	71	6.3	145	1468	0.7	84	507	11	0.5	**MK**
NL	2.0	0.3	15	0.6	1396	7.9	4.4	3.6	289	8.3	0.0	94	19038	62	1.2	**NL**
NO	10	2.6	32	6.1	167	55	23	36	589	56	0.2	275	6602	573	12	**NO**
PL	57	6.8	358	13	442	328	183	488	2857	504	0.4	11294	51730	1137	26	**PL**
PT	6.4	0.8	18	1.8	54	21	14	5.6	415	23	0.1	81	1536	42	1.2	**PT**
RO	204	17	178	30	99	5004	621	151	1283	3954	2.4	1159	5911	162	7.8	**RO**
RU	257	1009	386	3396	708	2729	487	2943	4872	1401	22	3046	34769	1833	603	**RU**
SK	27	1.8	235	3.5	44	156	112	32	608	371	0.2	2920	3583	74	2.6	**SK**
SI	19	0.3	361	0.7	19	58	121	7.6	460	153	0.05	361	1418	22	0.8	**SI**
SE	19	7.1	59	16	313	121	46	124	837	119	0.7	634	14385	2546	41	**SE**
TR	206	1818	118	461	135	3444	233	146	1633	754	145	637	5975	185	8.2	**TR**
UA	122	85	234	170	224	1874	326	1396	2067	1048	5.6	2104	13454	473	26	**UA**
Total, g TEQ/y	8.5	11	10	22	16	63	18	14	152	49	0.4	62	747	14	1.3	**Total, g TEQ/y**
	AL	**AM**	**AT**	**AZ**	**BE**	**BG**	**BA**	**BY**	**CH**	**CS**	**CY**	**CZ**	**DE**	**DK**	**EE**	

***Table B.3**. Matrix of 2,3,4,7,8-PeCDF country-to-country depositions in 2004, mg TEQ/y (continued)*
Receptors ↓ Emitters →

	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	MD	
AL	36	2.9	70	66	3.0	883	72	45	8.6	1.4	374	0.8	2.1	0.3	2.6	1.8	**AL**
AM	12	1.2	19	27	436	29	4.4	3.9	3.6	0.7	22	8.0	0.7	0.1	1.0	0.8	**AM**
AT	109	13	566	457	2.9	119	390	292	45	4.6	1223	1.1	12	5.1	17	2.2	**AT**
AZ	33	4.7	55	87	1233	83	14	13	11	2.2	64	107	2.9	0.2	4.2	2.9	**AZ**
BE	64	3.7	1571	706	0.6	9.3	6.9	5.4	38	2.0	54	0.2	2.2	30	4.3	0.2	**BE**
BG	89	14	190	235	25	2265	166	271	29	4.4	431	6.4	15	1.0	16	29	**BG**
BA	70	8.1	172	176	3.2	334	2113	388	21	2.9	780	1.0	9.2	1.0	10	3	**BA**
BY	106	95	324	608	28	129	104	218	63	6.7	245	13	501	2.3	381	36	**BY**
CH	93	4.2	928	251	1.1	36	47	19	27	2.9	1250	0.4	2.2	2.1	3.6	0.3	**CH**
CS	94	14	215	238	8.7	1081	859	716	29	4.2	760	2.7	13	1.2	15	8.6	**CS**
CY	5.2	0.5	8.7	10.6	2.5	69	3.2	2.7	1.4	0.3	17	0.3	0.4	0.0	0.5	0.5	**CY**
CZ	85	15	456	420	3.4	80	149	390	41	3.7	297	1.2	13	5.1	20	2.5	**CZ**
DE	521	63	5503	3780	9.4	152	156	200	285	20	1193	3.9	40	119	69	4.1	**DE**
DK	35	12	136	414	1.0	8.3	6.8	11	29	2.3	30	0.4	5.2	0.7	8	0.3	**DK**
ES	21093	45	2923	2128	11	181	156	78	275	42	1168	4.5	15	7.1	26	2.2	**ES**
EE	19	129	60	129	2.2	10.3	7.6	10.6	13	1.3	27	0.9	49	0.5	307	0.9	**EE**
FI	112	4925	359	777	18	59	34	48	80	9.5	139	9.3	92	2.4	254	4.5	**FI**
FR	3111	58	44306	5612	13	249	289	136	524	44	3213	5.2	25	138	44	3.0	**FR**
GB	306	25	1175	30097	4.6	40	30	28	928	19	190	1.8	9.0	4.4	16	1.1	**GB**
GE	39	4.4	61	87	11694	109	16	16	12	2.2	78	18	3.4	0.2	4.5	4.9	**GE**
GR	126	12	236	253	24	25314	144	143	33	5.4	725	6.0	9.2	1.0	11	14	**GR**
HR	67	7.8	184	182	3.0	270	9746	608	21	2.8	1139	0.9	9.0	1.1	10.8	2.5	**HR**
HU	68	14	216	251	5.0	178	1471	10036	28	3.4	509	1.8	15	1.7	17	5.4	**HU**
IE	107	8.5	246	794	1.8	15	11	9.2	2600	8.1	73	0.7	2.6	0.9	4.7	0.3	**IE**
IS	25	3.4	60	148	0.9	5.2	4.3	4.7	21	23	23	0.4	1.4	0.3	2.1	0.2	**IS**
IT	537	29	1714	952	12	1332	1710	451	114	16	42750	4.0	20	5.2	28	5.9	**IT**
KZ	138	52	269	492	268	181	58	63	61	10	224	6876	26	1.3	43	14	**KZ**
LT	38	41	126	258	4.4	26	24	41	25	2.3	67	1.5	1538	1.0	429	3.3	**LT**
LU	5.7	0.3	342	34	0.1	1.1	0.9	0.7	2.4	0.2	8.0	0.02	0.2	58	0.4	0.02	**LU**
LV	35	74	113	236	3.9	21	18	27	23	2.3	54	1.5	367	0.9	2490	2.3	**LV**
MC	0.03	0.001	0.4	0.04	0.0	0.02	0.04	0.01	0.005	0.0	0.9	0.0	0.001	0.0	0.001	0.0	**MC**
MD	17	5.1	40	61	8.5	52	23	38	7.0	1.0	55	2.1	6.5	0.2	7.1	545	**MD**
MK	28	3.0	55	59	3.4	1416	49	59	7.6	1.2	185	0.9	2.4	0.2	2.9	2.2	**MK**
NL	53	4.7	554	723	0.7	8.8	7.0	6.7	40	2.1	44	0.3	2.3	3.5	4.5	0.2	**NL**
NO	135	121	424	1372	6.0	46	33	45	130	13	133	2.7	24	2.5	46	2.1	**NO**
PL	257	105	1064	1611	18	243	311	850	147	13	614	5.4	216	10	190	16	**PL**
PT	1206	10	353	402	2.2	29	22	13	55	8.3	153	0.9	3.0	1.1	5.2	0.4	**PT**
RO	168	37	419	533	47	837	558	1160	63	8.7	852	11	41	2.5	47	190	**RO**
RU	1039	2124	2446	4780	3418	1780	556	713	559	75	1929	3509	710	14	1500	152	**RU**
SK	44	12	156	187	4.1	105	223	1781	20	2.2	223	1.3	16	1.3	18	3.8	**SK**
SI	26	3.1	87	77	0.9	54	1167	150	8.4	1.1	937	0.3	4.0	0.6	5.0	0.8	**SI**
SE	210	619	721	1830	16	89	64	101	169	17	216	6.9	98	4.4	191	5.9	**SE**
TR	464	50	745	987	1009	3769	230	243	131	25	1124	45	34	3.0	44	74	**TR**
UA	296	113	779	1236	263	772	417	1007	139	18	876	87	159	5.1	167	476	**UA**
Total, g TEQ/y	31	8.9	70	64	19	42	21	20	6.9	0.4	64	11	4.1	0.4	6.5	1.6	**Total, g TEQ/y**
	ES	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KZ**	**LT**	**LU**	**LV**	**MD**	

***Table B.3**. Matrix of 2,3,4,7,8-PeCDF country-to-country depositions in 2004, mg TEQ/y (continued)*
Receptors ↓ Emitters →

	MC	MK	NL	NO	PL	PT	RO	RU	SK	SI	SE	TR	UA	ReEmis	Back	g TEQ/y	
AL	1.3	2562	6.7	11	156	149	64	33	37	4.4	3.9	219	216	6751	652	18.9	**AL**
AM	0.2	14	2.2	6.9	32	58	10	48	4.3	0.6	1.6	1034	152	5157	586	14.5	**AM**
AT	5.8	111	81	54	1694	417	59	87	334	255	23	108	324	17581	1938	52.5	**AT**
AZ	0.6	42	7.0	20	112	158	35	432	15	1.8	5.9	1323	644	14641	2570	38.1	**AZ**
BE	0.7	6	341	20	101	208	3.4	21	5.9	1.4	8.3	15	36	9594	656	29.9	**BE**
BG	2.6	3469	25	44	919	396	1792	270	244	15	19	2852	2480	29199	2198	91.4	**BG**
BA	3.8	386	24	27	832	267	144	67	239	39	12	220	416	16565	1310	40.2	**BA**
BY	2.1	121	85	117	6918	501	273	1672	389	16	120	354	10454	24623	3270	69.3	**BY**
CH	7.7	24	26	25	154	289	8.9	27	17	11	6.7	39	64	17601	1179	79.0	**CH**
CS	3.6	4965	30	39	1176	393	760	136	408	28	18	580	1064	30606	1947	82.4	**CS**
CY	0.1	14	0.9	3.9	17	19	7.2	13	2.8	0.4	0.6	717	70	917	138	2.4	**CY**
CZ	2.5	82	93	41	5879	338	67	95	538	32	31	93	402	22974	1497	77.0	**CZ**
DE	13	121	1937	241	4966	1907	82	337	209	37	160	234	801	163212	7505	650.6	**DE**
DK	0.4	6.9	76	49	348	148	6.1	38	12	1.2	76	16	82	4508	733	13.4	**DK**
ES	17	116	175	312	853	27944	52	280	75	30	65	295	532	75644	12302	160.1	**ES**
EE	0.3	9.3	20	37	449	82	12	380	16	1.2	68	28	262	2579	648	7.5	**EE**
FI	1.6	48	111	325	1494	506	60	1788	71	5.6	660	210	1214	10519	5157	37.9	**FI**
FR	202	177	635	297	1436	5544	78	351	128	58	96	328	680	131781	13471	291.8	**FR**
GB	2.6	31	284	169	456	1485	21	133	30	6.3	41	76	250	27178	5237	78.0	**GB**
GE	0.7	55	7.6	22	137	171	51	503	18	2.2	6.3	2485	1053	17237	1601	39.1	**GE**
GR	3.9	5551	25	52	608	524	376	209	132	13	16	3973	1586	30498	2575	82.6	**GR**
HR	4.6	261	27	23	1108	258	110	65	304	315	13	190	378	18429	1229	43.3	**HR**
HU	3.0	250	40	32	2764	295	564	112	5008	126	21	193	1670	21729	1407	57.3	**HU**
IE	1.0	11	32	42	127	541	7.1	49	9.4	2.4	12	28	88	7082	2042	15.8	**IE**
IS	0.3	4.3	12	29	75	131	4.1	19	5.6	0.8	5.4	14	50	1371	914	3.7	**IS**
IT	125	850	105	116	1912	1817	211	219	350	322	45	1089	894	62547	6820	153.6	**IT**
KZ	2.4	124	44	72	802	720	162	5661	80	8.7	52	1188	4880	34454	20192	82.4	**KZ**
LT	0.6	24	39	53	2451	173	36	458	66	4.0	74	54	810	6878	1053	19.5	**LT**
LU	0.1	0.8	9.2	1.7	11.2	17.3	0.4	2.1	0.7	0.2	0.7	1.6	3.4	1209	57	3.6	**LU**
LV	0.5	20	34	61	1271	156	28	424	42	2.9	98	49	613	5583	1084	16.7	**LV**
MC	0.7	0.01	0.005	0.004	0.1	0.1	0.005	0.01	0.01	0.01	0.002	0.02	0.02	2.5	0.3	0.0	**MC**
MD	0.4	53	7.4	9.4	397	79	761	136	49	2.8	6.2	245	3433	4504	461	12.2	**MD**
MK	0.9	17256	6.2	11	181	117	82	37	48	3.7	3.8	310	247	9809	576	34.7	**MK**
NL	0.6	6.4	2082	22	134	202	3.8	24	7.9	1.5	11	15	45	10803	683	36.4	**NL**
NO	1.6	39	140	2530	968	669	38	273	55	5.5	301	85	474	9696	5186	31.4	**NO**
PL	5.4	235	324	202	88641	1141	322	816	2010	54	216	355	5037	74415	5477	254.3	**PL**
PT	2.3	19	31	66	155	95824	9.2	59	13	4.2	14	50	106	34667	2675	138.2	**PT**
RO	5.8	1227	65	87	3316	751	17368	593	1130	47	48	1928	9452	40031	4143	103.9	**RO**
RU	18	1236	525	1440	12881	4928	1703	196843	996	80	1085	12282	64197	270977	74830	727.8	**RU**
SK	1.4	122	30	29	4936	183	188	93	6723	35	19	117	1156	12238	970	37.8	**SK**
SI	2.4	57	13	9.2	490	95	28	26	103	1107	5.4	42	108	5343	481	13.4	**SI**
SE	2.5	78	269	1160	2974	934	93	695	139	10	4136	208	1359	18131	7407	61.2	**SE**
TR	9.4	1029	86	215	1760	2046	1008	1736	269	30	65	156451	9854	123596	12534	335.6	**TR**
UA	7.3	639	158	194	11085	1401	2591	5916	1750	53	143	4018	201341	123245	8757	391.7	**UA**
Total, g TEQ/y	0.5	41	8.1	8.3	167	154	29	221	22	2.8	7.8	194	329				
	MC	**MK**	**NL**	**NO**	**PL**	**PT**	**RO**	**RU**	**SK**	**SI**	**SE**	**TR**	**UA**				