

Convention on Long-Range Transboundary Air Pollution

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*Co-operative programme for monitoring
and evaluation of the long-range
transmission of air pollutants in Europe*

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EXECUTIVE SUMMARY

In accordance with the 2005 Work-plan for Implementation of the Convention (*ECE/EB.AIR/83/add2, item 2.7*) 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) and hexachlorobenzene (HCB). The outcome of the studies is summarized in this Status Report.

At present the measurements of POP concentrations in the atmosphere and precipitation are performed by 12 EMEP monitoring sites. Most of the sites are located in Northern and Central Europe, and no sites in southern and eastern parts of Europe. The adopted EMEP monitoring strategy for 2004-2009 (EB.AIR/GE.1/2004/5) aims to improve the distribution of monitoring sites over European region.

The official data on POP emissions (PAHs, PCDD/Fs, PCBs, HCB) for the period from 1990 to 2003 (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. According to the official data and expert estimates the emissions of all the pollutants of concern tend to decrease from 1990 to 2003. 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 18 - 33% depending on particular PAH, and emissions of PCDD/Fs by 63%.

In course of preparation for the review of MSCE-POP regional model MSC-E has summarized the information on model sensitivity to variations of pollutant-specific and environmental parameters in the EMEP/MSCE Technical Report 5/2005 [*Gusev et al., 2005a*]. The report provides also a detailed description of MSCE-POP regional model along with the input data required for modeling of POPs. This information was presented at the sixth TFMM meeting in Zagreb, Croatia in April 2005. During the meeting it was recommended that the continuation of MSCE-POP model review at the forthcoming Workshop in Moscow in October 2005 will be focused on the comparison of model results with measurements and POP model intercomparison study.

Model evaluation of the environmental pollution levels in the current year was performed out for the four indicator PAHs, PCDD/Fs, PCBs, and HCB for the period 1990-2003. The evaluation of trends in European region contamination by PAHs and PCDD/Fs is carried out at the regional scale. Transboundary transport was estimated for B[a]P and PCDD/Fs. For PCBs and HCB evaluation of pollution levels was performed on the hemispheric scale.

Annual mean B[a]P deposition flux over Europe has decreased by 20% on average during the period 1990-2003 and its value for 2003 is amounted to 16 g/km²/y. Average value of B[a]P annual air concentrations over Europe amounts to 0.2 ng/m³. At the same time in central and eastern parts of the Europe contamination levels exceed 1 ng/m³. Total annual depositions of B[b]F, B[k]F, and I_P in European countries in period 1990-2003 have decreased by 21%, 28%, and 15%, correspondingly. The most pronounced changes in deposition levels can be noted for countries of western and southeastern parts of Europe.

Evaluation of environmental pollution by dioxins and furans (PCDD/Fs) was made for the indicator congener 2,3,4,7,8-PeCDF. Following the reduction of emission PCDD/F annual total depositions over Europe have decreased by 47% in period 1990-2003. The decrease of annual mean air concentration of PCDD/F is accounted for 41% during this period. Mean value of PCDD/F annual air concentration

over Europe in 2003 is accounted for 2.6 fg TEQ/m³ varying from 0.02 to 15 fg TEQ/m³ for most of European countries. Mean level of annual PCDD/F deposition fluxes is about 0.6 ng TEQ/m²/y.

The transboundary transport of B[a]P and PCDD/Fs between European countries was assessed. The contribution of the external sources to air concentrations of B[a]P and PCDD/Fs is essential and varies typically from 30 to 70%. In case of PCDD/Fs noticeable contribution to air concentrations and depositions over Europe belongs also to the re-emission from underlying surface.

The POP intercontinental transport was investigated by the example of HCB and PCB-153 using the hemispheric version of MSCE-POP model. HCB and PCBs are the pollutants with significant long-range transport potential therefore evaluation of pollution of European region and remote regions like the Arctic by these POPs requires consideration of the influence of remote emission sources. In the framework of co-operation with national experts MSC-E participated in German Canadian Project "Quality of measuring data on atmospheric inputs of POPs" where it contributed with the study of pollution levels of B[a]P and γ -HCH within the OSPAR region, over Europe, and on the hemispheric scale.

The development of the hemispheric MSCE-POP model was continued during this year with the refinement of its ocean block, in particular, the descriptions of the processes of diffusion, partitioning to particulate organic matter, and sedimentation within the seawater for POPs were refined.

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

In the framework of POP model intercomparison study the third expert meeting was organized in Moscow in February 2005. Fulfillment of the II and III Stages of intercomparison programme were discussed. Results of Stage II are presented in the EMEP/MSCE-POP Technical Report 4/2005 [Shatalov *et al.*, 2005a].

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INTRODUCTION

In the current year the investigations of the environmental pollution by persistent organic pollutants (POPs) on the basis of measurements, emission data and modeling results were continued. The investigations were performed in accordance with the Work-plan for 2005 (*ECE/EB.AIR/83/Add.2*, item 2.7). The following pollutants were under consideration: polycyclic aromatic hydrocarbons (B[a]P, B[b]F, B[k]F, I_P), polychlorinated dibenzo-p-dioxins and dibenzofurans (2,3,4,7,8-PeCDF congener), polychlorinated biphenyls (PCB-153) and HCB.

To prepare for the review of the regional MSCE-POP multicompartiment transport model MSC-E has further studied the model sensitivity to variations of pollutant-specific and environmental parameters. The emphasis was given to the processes affecting POP dispersion in the atmosphere and to the influence of POP re-emission from the underlying surface. The results of the work were summarised in the EMEP/MSCE-POP Technical Report 5/2005 [*Gusev et al.*, 2005a] along with a detailed description of MSCE-POP model and were presented at the sixth TFMM meeting in Zagreb, Croatia in April 2005.

In the framework of the evaluation of pollution levels within the European region measurement data from the EMEP monitoring network were collected and analyzed (Section 1), emission data were prepared for modeling purposes on the basis of the officially submitted information and expert estimates of emissions (Section 2), sensitivity study of MSCE-POP model was performed and detailed description of the model was prepared (Section 3). The following model calculations were made (Sections 4.1 and 4.2):

- long-term trends of pollution in period 1990-2003 and pollution levels in 2003 for the European region by B[a]P, B[b]F, B[k]F, and I_P (Section 4.1.1) and PCDD/Fs (Section 4.1.2);
- transboundary transport between European countries for B[a]P (Section 4.1.1) and PCDD/Fs (Section 4.1.2);
- contribution of the intercontinental transport to the pollution of the European region and contribution of European emission sources to the pollution of remote regions (by the example of the Arctic) for PCB-153 and HCB (Section 4.2);

To carry out the model assessment of the environmental pollution on the hemispheric scale the ocean block of MSCE-POP model was modified. In particular, the descriptions of the processes of diffusion, partitioning to particulate organic matter, and sedimentation within the seawater for POPs were refined (Annex B).

For the purposes of the evaluation of POP pollution levels within the European region EMEP Centres closely co-operated with the subsidiary bodies to the Convention, international organizations, programmes and projects as well as with national experts. The work on the support of the Task Force on POPs in preparation for the review of the Protocol on POPs was continued.

In the framework of POP model intercomparison study MSC-E has organized the third expert meeting aimed at the discussion of intercomparison results obtained during the Stage II. During the meeting the experts have discussed the results on computational experiments of the Stage II and elaborated an extended work-programme and time-schedule of the Stage III.

MSC-E has participated in international projects such as German Canadian Project “Quality of measuring data on atmospheric inputs of POPs” and 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.

The results of the work done are presented in the Technical Reports of the EMEP Centres as well as on the Internet www.emep.int and www.msceast.org.

Acknowledgements

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1. MONITORING OF POPs

1.1. Measurement network

POP_s 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 POP_s 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 POP_s for 2003, are shown in Fig.1. The different measurements programs are given in Table 1. Further details of the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POP_s [Aas and Breivik, 2005]. The sites are divided in those measuring both in air and precipitation, and those measuring only in one media. In 2003 it was 6 sites measuring POP_s in both compartments, and altogether it was 12 measurement sites.

Table 1. Measurements sites and programs for POP_s in 2003

Sites	POP _s in air and aerosol	POP _s in precipitation
BE04		Pesticides, HCHs
CZ03	PAHs, PCBs, pesticides, HCHs	PAHs, pesticides
FI96	PAHs, PCBs, pesticides, HCHs, HCB	PAHs, PCBs, HCHs
DE01		PAHs, PCBs, pesticides, HCB, HCHs
DE09		PAHs, PCBs, pesticides, HCB, HCHs
IS91	PCBs, pesticides, HCB, HCHs	PCBs, pesticides, HCB, HCHs
IE02		PCBs, pesticides, HCB, HCHs
NL91		γ-HCH
NO99	HCB, HCHs	HCB, HCHs
NO42	PAHs, pesticides, HCHs, HCB, PCBs	
SE12	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs
SE14	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs

It is quite evident from Fig.1 that the spatial distribution in Europe is unsatisfactory, no sites in south and east of Europe. Most of the sites are connected to the OSPARCOM programme CAMP. The adopted EMEP monitoring strategy for 2004-2009 (EB.AIR/GE.1/2004/5) aims to improve the current unsatisfactorily site distribution. At "Core sites level 2" (supersites), POP_s in both air and precipitation should be measured at around 20 stations distributed over the domain, this is a big challenge and it is necessary to get closer cooperation with research programmes etc that to campaign measurements of POP_s.

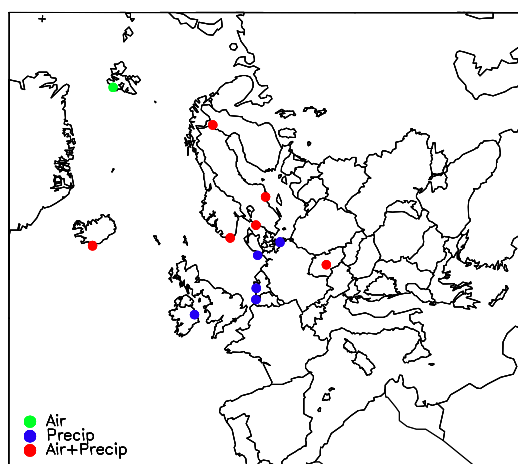


Fig.1. Measurement network of POP_s in EMEP, 2003

1.2. Measurement results of POPs in 2003

Details of the measurements results and methodology are found in the EMEP/CCC data report on heavy metals and POPs. It is difficult to compare the results from the different sites since the measurement programs can vary and the methodology differs. E.g. much of the data from Ireland and Belgium, and to some extent from Iceland, are mainly below the detection limits and here one can only say something about the upper concentration limits. 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.

1.3. PCBs in air and precipitation

PCBs consist of various individual chemical species (congeners). Altogether, 209 different congeners are possible, although only a few are regularly monitored in environmental samples. The data presented herein includes seven individual PCB congeners (PCBs 28, 52, 101, 118, 138, 153, 180). During 2003 these PCBs were monitored in air at stations in the Czech Republic (CZ03), Sweden (SE12 and SE02/SE14), Finland (FI96), Iceland (IS91) and Norway (NO42). These sites have also measured since before 1999 and Fig.2 shows the trend in the annual average air concentrations of the sum of the seven PCBs (Σ_7 PCB). In precipitation there have also been measurements of PCB in Germany (DE01 and DE09) and Ireland (IE02). At IE02 the PCB-28 was not measured in precipitation so here the sum of the six others are given.

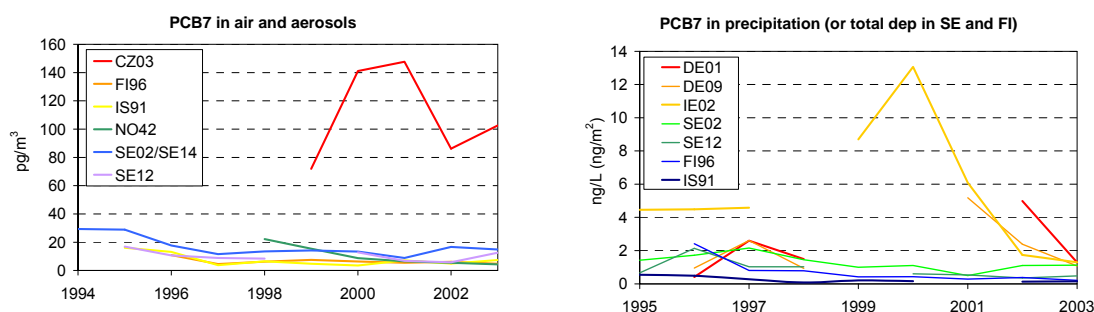


Fig.2. Trends of Σ_7 PCB in air and precipitation

Elevated levels of PCBs in air are observed at the station in the Czech Republic (higher than 80 pg/m^3) as compared to the other stations, which are all below 20 pg/m^3 . For precipitation the level is varying from year to year, especially in Ireland that observed high concentrations in 1999-2001. It is very difficult to measure POPs in precipitation because of low concentrations, much of these measurements are below the detection limits. The trends are not significantly changing, there might be a tendency to lower concentrations in Scandinavia but this can also be due to more sensitive instrumentation.

The high air concentrations of levels in the stations in the Czech Republic is not surprising, considering that the high historical usage of PCBs in central Europe [Breivik et al, 2002]. It is also known that former Czechoslovakia were among the European countries where PCBs were produced in significant amounts until 1984 [Taniyasu et al, 2003]. Large differences in atmospheric PCB levels across Europe were also noted by F.M.Jaward et al. [2004]. They carried out a campaign during the

summer of 2002, deploying 71 passive air samplers throughout Europe, and found that the atmospheric levels of PCBs were found to vary by as much as two orders of magnitude. Elevated levels were found in urban areas, suggesting that densely populated regions tend to be key contemporary source regions of PCBs to the atmosphere.

The relative contribution of individual PCB congeners is dependent on the site location. Fig.3 conveys the message that there is a tendency that the heavier congeners (PCBs 138, 153 and 180) are enriched in the southernmost stations (Czech Republic), whilst the lighter congeners (PCBs 28 and 52) are enriched at the northernmost sites (Iceland, Finland). This clearly confirms that there are marked differences in the long-range transport potential (LRTP) within the group of PCBs [Wania and Dugani, 2003]. LRTP is controlled by the competing processes of atmospheric degradation and net atmospheric deposition. The data in Fig.3 suggest that it is net atmospheric deposition that tends to reduce the LRTP of the heavier PCBs as compared to the lighter PCBs. This is caused by a change in phase partitioning in the atmosphere for which the heavier PCBs get increasingly sorbed to atmospheric particles at decreasing temperatures. This again contributes to a depletion of heavier congeners due to atmospheric deposition, and thus an increased prevalence of lighter congeners in the northernmost air samples [Wania and Su, 2004].

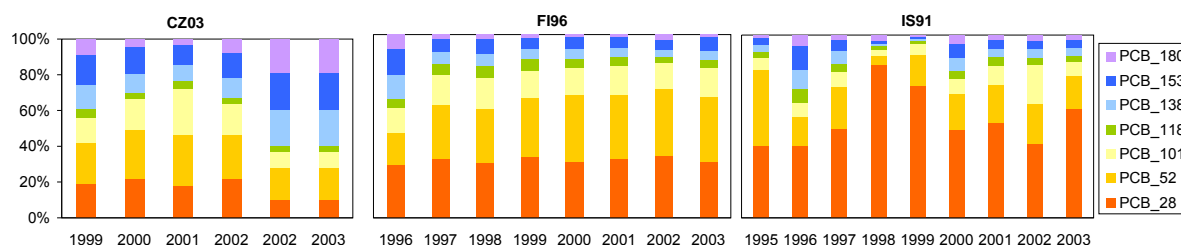


Fig.3. Trends in relative contribution of individual PCB congeners to $\Sigma_7\text{PCB}$ in air at three sites, notice the difference in time periods

The trend of the relative importance of the different congeners is varying. In CZ03 the lighter PCB gets of less importance while there is a tendency for increased importance of these at the Finnish site. At Island there is high inter-annual variability. These changes can have different reason, e.g. meteorological variability, possible influences of nearby emission sources and changes in methodology. A more thorough analysis's is necessary to explain the different tendencies.

2. EMISSIONS

Official data on the emission totals of PAHs, PCDD/Fs, HCB and PCBs were reported by 36 countries for the period from 1990 to 2003 (for at least one year). The officially reported emission data are available from WEBDAB: <http://webdab.emep.int>. The number of countries reporting 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 32 countries for 1990-2003 (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 22 European countries for 1990-2003 (for at least one year). 17 countries, namely, Belarus, Croatia, the Czech Republic, Denmark, Estonia, France, Germany, Hungary, Iceland, Ireland, Lithuania, Luxembourg, Monaco, Poland, Republic of Moldova, Slovakia and the United Kingdom submitted emissions of each from four indicator compounds for the considered period (for at least one year). In 2005 Croatia, France, Germany and Iceland 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; Berdowski et al., 1997; Pacyna et al., 1999; Tsibulsky et al., 2001].

The information about PAH spatial distributions was provided by 13 countries (Austria, Belgium, Bulgaria, Denmark, Finland, France, Hungary, Iceland, Netherlands, Norway, Poland, Spain, United Kingdom). For other countries, expert estimates were used [Pacyna et al., 1999; Berdowski et al., 1997].

According to the official data and expert estimates for the period from 1990 to 2003, European emissions of four indicator PAHs decreased by 18% - 33% depending on pollutant (Fig.4, Table 2).

Official information on emissions of four indicator PAHs by sectors in 2003 is available for 15 countries: Austria, Belarus, the Czech Republic, Denmark, Estonia, Finland, France, Hungary, Iceland, Ireland, Lithuania, Republic of Moldova, Norway, Sweden and the United Kingdom.

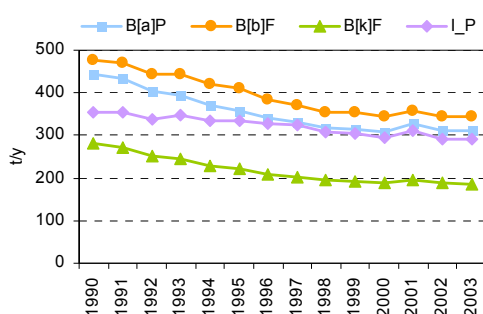


Fig. 4 Time-series of PAH emissions

Table 2. Decrease of PAH emissions 1990-2003

Pollutant	Decrease, %
B[a]P	30
B[b]F	28
B[k]F	33
I_P	18

The sector split for the national total four indicator PAH emissions is presented in Fig.5. Graph shows that the Residential sector is the major contributor. On average its share amounts to 58%. The predominant source in the Residential sector is combustion of wood. The second most important sector is the Metal Production. This sector is the largest source of PAHs for Iceland, Republic of Moldova and Norway.

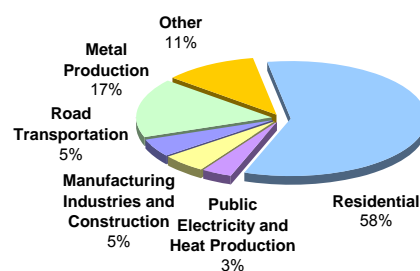


Fig. 5. Sector split for PAHs in Europe (15 countries) in 2003

The considerable contribution of the Residential sector to the total PAH emissions determines pronounced seasonal emission variation. In the model calculations data on the seasonal distribution of PAH emissions taken from [Baart et al., 1995] were used. According to [Baart et al., 1995] the level of PAH emissions in winter is higher than in summer. In the model calculations the same seasonal distribution of PAH emissions was used for all the countries. It causes additional uncertainties to the calculation results.

2.2. Dioxins and furans

Official data on total emissions of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) were reported by 34 countries for the period from 1990 to 2003 (for at least one year). For the first time, Iceland, Switzerland and the FYR of Macedonia reported emission data.

The information about the spatial distribution of PCDD/F emissions was submitted by 13 countries (Austria, Belarus, Belgium, Bulgaria, Finland, France, Hungary, Iceland, Netherlands, Norway, Poland, Spain, Sweden). For the remaining countries expert estimates of PCDD/F total emissions and their spatial distribution [Pacyna et al., 1999] were used.

According to the official data and expert estimates, the European emissions of PCDD/Fs dropped by 63% (or 2.7 times) between 1990 and 2003 (Fig. 6). The total PCDD/F emissions in the EMEP region amounted to 4.7 kg TEQ in 2003.

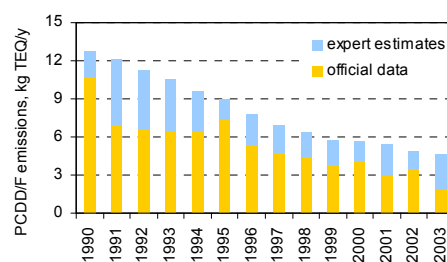


Fig. 6. PCDD/F emissions in Europe for the period from 1990 to 2003

According to the official data, PCDD/F emissions in most countries tend to decrease (1990-2003). The overall PCDD/F emissions in eighteen countries reported data for both years 1990 and 2003 decreased by 78% (or 4.6 times). The maximum decrease of the PCDD/F emissions was reported by the Netherlands (19 times), and the maximum increase - by Monaco (1.2 times) (Fig. 7).

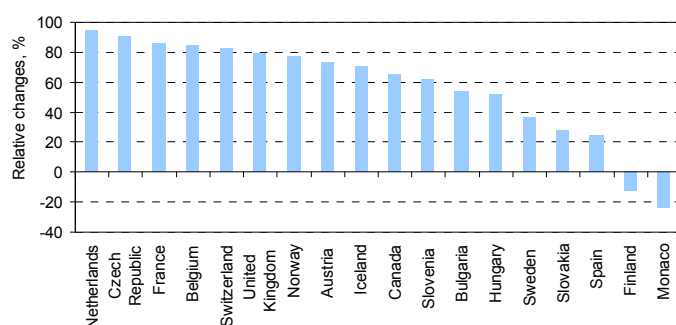


Fig. 7. Relative changes of PCDD/F emissions for 1990-2003 in 18 countries

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 2003 were prepared. At this stage, evaluation of overall toxicity of PCDD/F mixture was performed on the basis of simulations of 2,3,4,7,8-PeCDF congener (feasibility of such evaluation and uncertainty analysis see [Shatalov et al., 2003]).

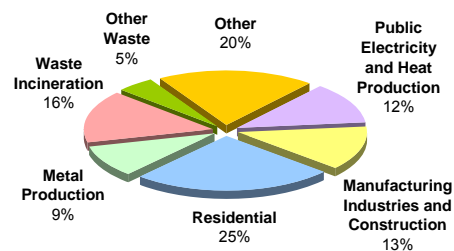


Fig. 8. Sector split for PCDD/Fs in Europe (23 countries) in 2003

Official information on emissions of PCDD/Fs by sectors in 2003 is available for 23 countries. The sector split for the national total emissions of PCDD/Fs is presented in Fig. 8. The largest contribution to the total PCDD/F emissions is made by the Residential sector (25%). The next most important sector is the Waste Incineration. This sector is the largest source of PCDD/Fs for Belarus, Cyprus, Iceland, Republic of Moldova, Switzerland and the United Kingdom.

2.3. Hexachlorobenzene

Official information about total emissions of hexachlorobenzene (HCB) was reported by 24 countries for the period from 1990 to 2003 (for at least one year). For the first time, Estonia and Finland reported emission data. The information about the spatial distribution of HCB emissions was submitted by Austria, Belgium, France, Poland and Spain.

According to the official data, the total HCB emissions in 7 countries submitted data for both years 1990 and 2003 decreased by 19% (or 1.2 times). The maximum emission decrease was reported by Bulgaria (12 times), and the maximum increase - by Belgium (3.4 times) (Fig. 9).

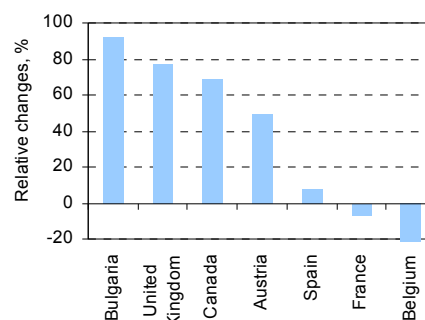


Fig. 9. Relative changes of HCB emissions for 1990-2003 in 7 countries

Official information on emissions of HCB by sectors in 2003 is available for 14 countries. The maximum contribution to the total HCB emissions is made by the Manufacturing Industries and Construction sector (27%). The second most important sector is the Metal Production.

For modeling of HCB long-range transport within the Northern Hemisphere an emission scenario was prepared last year with the use of different expert estimates (EMEP Status Report 3/2004 [Dutchak et al., 2004]).

For the evaluation of intercontinental transport HCB emission sources of the Northern Hemisphere were split into 5 groups. The spatial distribution of emissions for each group was prepared (Fig. 10).



Fig. 10. Splitting of HCB emissions into groups of sources

In accordance with the emission scenario, HCB emissions in the Northern Hemisphere amounted to 92 t, including 12.5 t in North America, 36.4 t in Central Asia, 16.7 t in South-eastern Asia, 12.5 t in Europe and 14 t in Russia in 2003.

2.4. Polychlorinated biphenyls

Official information about total emissions of polychlorinated biphenyls (PCBs) was submitted by 19 countries for the period from 1990 to 2003 (for at least one year). According to the official data submitted for the both years, the total PCB emissions of Bulgaria, the Czech Republic, France, Hungary, Monaco, Slovakia, Slovenia, Spain and the United Kingdom decreased by 4.3 times. The information about the spatial distribution of PCB emissions was submitted by Finland and France.

Official information on emissions of PCB by sectors in 2003 is available for 12 countries. The maximum contribution to the total PCB emissions is made by the Public Electricity and Heat Production sector (39%). The next important sector is the Residential. This sector is the largest PCB sources for Bulgaria and Lithuania.

The modeling of PCB long-range transport for the period from 1990 to 2003 was performed for individual PCB congener (PCB-153). The sources located over the whole Northern Hemisphere were taken into account in the computations. Emission data of PCB-153 were taken from the global emission inventory of 22 PCB congeners [Breivik *et al.*, 2002].

For the evaluation of intercontinental transport PCB sources of the Northern Hemisphere were split into 6 groups (Fig. 11). For these groups gridded emission data for the period from 1990 to 2003 were prepared.

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, 3.3 t in North-western Europe, 2.6 t in South-eastern Europe, 1.7 t in Africa and Central Asia, 0.7 t in South-eastern Asia and 1.5 t in Russia in 2003.



Fig. 11. Splitting of PCB-153 emissions into groups of sources

3. MODEL REVIEW

According to 2005 work-plan on POPs, regional version of MCSE-POP model is to be reviewed at the TFMM Workshop to be held in Moscow in October 2005. As a first step for the review MSC-E prepared the Technical Report [Gusev *et al.*, 2005a] with a detailed model description and the results of sensitivity study of the model to variation of pollutant-specific and environmental parameters. These results were presented at the sixth TFMM meeting in Zagreb, Croatia in April 4 – 7, 2005. Following TFMM recommendation forthcoming Workshop will be focused at comparison of modelling results with available measurement data and at the intercomparison of POP models.

Here a brief description of MSCE-POP model and results of sensitivity study are presented. The information on the comparison of model results with measurements and on model intercomparison is given in Sections 4.1, 4.2, and 5.2, respectively.

An unique feature of POPs is their ability for re-emission. Thus, to evaluate long-range transport and deposition of POPs multi-compartment approach is used. For the description of POP exchange with and accumulation in main environmental compartments the atmospheric module is complemented with additional modules for soil, seawater, and vegetation.

At present there exist two versions of MSCE-POP model – regional and global. One is operating within the EMEP region with resolution 50×50 km, and another is designed for calculations within the whole northern hemisphere with resolution $2.5 \times 2.5^\circ$. Regional version of the model was recently modified to enlarge vertical coverage in the atmosphere. The development of hemispheric version is currently underway. In particular, the approach to the description of POP behaviour in the marine environment is refined and tested on the basis of this version (Annex B).

MSCE-POP model is being developed for several purposes:

- evaluation of atmospheric transport and deposition of POPs on regional (EMEP) and hemispheric scales;
- evaluation of spatial distribution of POPs in the atmosphere, soil, vegetation, and seawater;
- evaluation of transboundary and intercontinental transport of POPs;
- evaluation of POP partitioning between main environmental compartments;
- assessment of temporal and spatial trends;
- projection of future levels of POP contamination and trends under various emission scenarios;
- estimation of long-range transport potential and overall persistence of new potential POPs;
- study of environment pollution by POPs on the basis of monitoring/modeling approach.

3.1. Brief description of MSCE-POP model

Model structure. Both regional and hemispheric versions of MSCE-POP model are three-dimensional Eulerian multi-compartment models, which consider main environmental compartments and include basic processes describing POP emission, long-range transport, deposition, degradation, and gaseous exchange between the atmosphere and the underlying surface (Fig. 12). MSCE-POP model domain covers practically the whole troposphere, upper soil layer of 20 cm, and seawater compartment within the model grid.

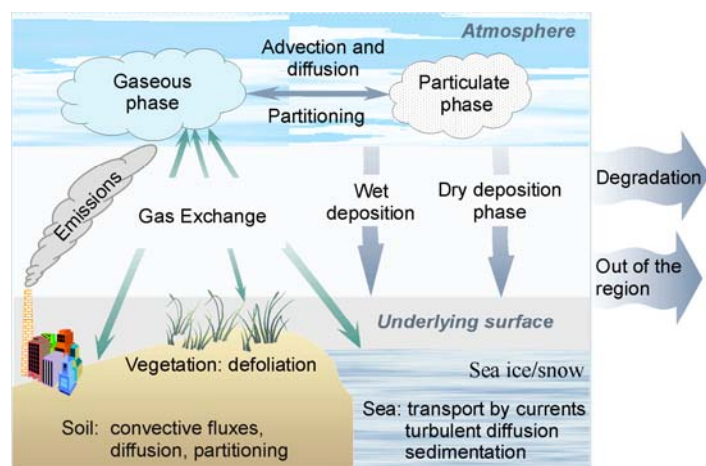


Fig. 12. The scheme of processes included into the MSCE-POP model

Current version of the model considers partitioning of POPs between the following environmental compartments: the atmosphere, soil, seawater, vegetation, and forest litter (Fig. 12). Selection of compartments and processes is based on current understanding of their importance with regard to the description of POP dispersion and accumulation in the environment.

Processes included. The following processes affecting the long-range transport of POPs are included in the model:

Atmosphere:

- advective transport and turbulent diffusion;
- partitioning between the gaseous and particulate phase;
- wet and dry deposition of POP in particulate and gaseous phase to the underlying surface;
- degradation.

Vegetation:

- gaseous exchange with the atmosphere;
- degradation;
- defoliation and transfer to upper soil layer.

Soil:

- gaseous exchange with the atmosphere;
- partitioning in soil between the gaseous, solid and liquid phases;

- vertical transport due to convective water fluxes, diffusion, and bioturbation;
- degradation.

Seawater:

- gaseous exchange with the atmosphere;
- advective transport by sea currents and turbulent diffusion;
- partitioning between the dissolved and particulate phase;
- sedimentation;
- degradation.

Input and output information. An inclusion of additional environmental compartments, comparing to MSCE-HM model, increases the amount of input data required for modelling. Along with meteorological and emission data the MSCE-POP model uses information on soil and vegetation properties, data on sea currents, and physical-chemical properties of considered POPs.

MSCE-POP model provide the following output information:

- spatial distribution of deposition and concentrations of POPs in environmental media with specification for different types of underlying surface;
- source-receptor relationships;
- long-term trends of POP content in main environment compartments;
- distribution of POPs between environmental media;
- long-range transport potential and overall persistence.

The details on model description of the above processes can be found in MSC-E Technical Report [Gusev *et al.*, 2005a] prepared for the purposes of model review.

3.2. Sensitivity study

In course of preparation to the model review MSC-E has carried out a study of MSCE-POP model sensitivity aimed at the evaluation of the effect of uncertainties of individual parameters related to the POP properties and the analysis of the influence of environment parameters on the model result, in particular, air concentrations and deposition fluxes of the selected POPs.

For the description of the long-range transport of POPs in the environment several media should be considered in the model, namely, atmosphere, soil, seawater, vegetation, etc. Among these media the atmosphere provides the most dynamic pathway of a pollutant dispersion and therefore is the most important from point of view of the long-range transport. Taking this into account the analysis of MSCE-POP model sensitivity at current stage was mostly focused on the processes which POP undergo in the atmosphere. To evaluate model sensitivity the values of computed POP atmospheric concentrations and deposition fluxes were considered. The analysis was performed for the following processes affecting POPs in the atmosphere: gas/particle partitioning, degradation, wet deposition of POP in particulate and gaseous phases, absorption by and re-volatilisation from the underlying surface. For the analysis of model sensitivity individual congener PCB-153 was chosen as a pollutant with essential fractions of both gaseous and particulate phases in the atmosphere.

The set of input parameters used in the sensitivity analysis is shown in Table 3. These are pollutant-specific parameters used for model description of POP behavior in the environment and some environmental parameters.

To analyze the sensitivity conventional ranges of parameter variations are defined as “base”, “high” and “low” values for all input parameters. “Base” values of parameters represent those ones selected in model parameterization and used for the evaluation of environment pollution. “High” and “low” values of pollutant-specific parameters were selected on the basis of the literature data [Shatalov *et al.*, 2003]. For the analysis it was assumed that “high” and “low” values differ by an order of magnitude. For each parameter “base” value is a geometrical mean between “high” and “low” ones. For environmental parameters mean values and characteristic ranges over Europe are used which are obtained from the geophysical and meteorological input data of the model.

Table 3. Input parameters used for evaluation of model sensitivity

Parameter	Notation	Base value	Low value	High value
Pollutant-specific parameters of PCB-153				
Henry's law constant at 10 °C, Pa·m ³ /mol	K_H	3.78	1.20	12.0
Subcooled liquid vapor pressure at 10 °C, Pa	p_{OL}	$9.69 \cdot 10^{-5}$	$3.06 \cdot 10^{-5}$	$3.06 \cdot 10^{-4}$
Octanol/air partitioning coefficient at 10 °C, dimensionless	K_{OA}	$3.64 \cdot 10^{10}$	$1.15 \cdot 10^{10}$	$1.15 \cdot 10^{11}$
Octanol/water partitioning coefficient, dimensionless	K_{OW}	$7.94 \cdot 10^6$	$2.51 \cdot 10^6$	$2.51 \cdot 10^7$
Washout ratio for particulate phase, dimensionless	W_p	$1.5 \cdot 10^5$	$4.7 \cdot 10^4$	$4.7 \cdot 10^5$
Mass transfer coefficient to vegetation (deciduous forest), 1/s	$K_{avdecid}$	30	9.49	94.9
Mass transfer coefficient to vegetation (coniferous forest), 1/s	$K_{avconif}$	4.6	1.45	14.5
Degradation coefficient, 1/s	K_d	$1.2 \cdot 10^{-7}$	$3.7 \cdot 10^{-8}$	$3.7 \cdot 10^{-7}$
Environmental parameters				
Ambient air temperature	T	10 °C	0 °C	20 °C
Atmospheric aerosol specific surface, m ² /m ³	θ	$1.5 \cdot 10^{-4}$	$1.5 \cdot 10^{-5}$	$1.5 \cdot 10^{-3}$
Concentration of OH radicals in the atmosphere, molecules/cm ³	C_{OH}	$1.0 \cdot 10^6$	$1.0 \cdot 10^5$	$5.0 \cdot 10^6$
Precipitation amount, mm/hour	$Prec$	0.1	0.01	0.5
Organic carbon fraction in soil, dimensionless	f_{oc}	0.05	0.01	0.1

The sensitivity of air concentrations (S_a) and depositions (S_d) to the variations of considered parameter A are estimated as:

$$S_a = \frac{\Delta C_a}{\Delta A} \quad (\text{for air concentrations}), \quad S_d = \frac{\Delta D}{\Delta A} \quad (\text{for depositions}) \quad (1)$$

respectively, where:

$$\Delta C_a = \frac{C_a^{high} - C_a^{low}}{C_a^{base}} \cdot 100\%, \quad \Delta D = \frac{D^{high} - D^{low}}{D^{base}} \cdot 100\%, \quad \Delta A = \frac{A^{high} - A^{low}}{A^{base}} \cdot 100\%. \quad (2)$$

Here C_a^{high} , C_a^{low} , C_a^{base} are the values of air concentrations and D^{high} , D^{low} and D^{base} are the values of depositions calculated for “high”, “low” and “base” values of the considered parameter A (A^{high} , A^{low} and A^{base}). The rest of the parameters are set to the base values.

Evaluation of the sensitivity of air concentrations and depositions was carried out on the basis of one-year period calculations with meteorology of 2000 using a single conventional point source located in France. To analyze the role of re-emission process the values of sensitivities were calculated for different parts of the model domain, namely, for the region close to the emission source (within a

distance of 1000 km from the source) and for the remote regions beyond this limit. Detailed results of sensitivity analysis are given in the MSC-E Technical Report [Gusev *et al.*, 2005a].

Values of sensitivity of calculated air concentrations and depositions to variations of the selected parameters for the region close to the emission source are demonstrated in Fig. 13.

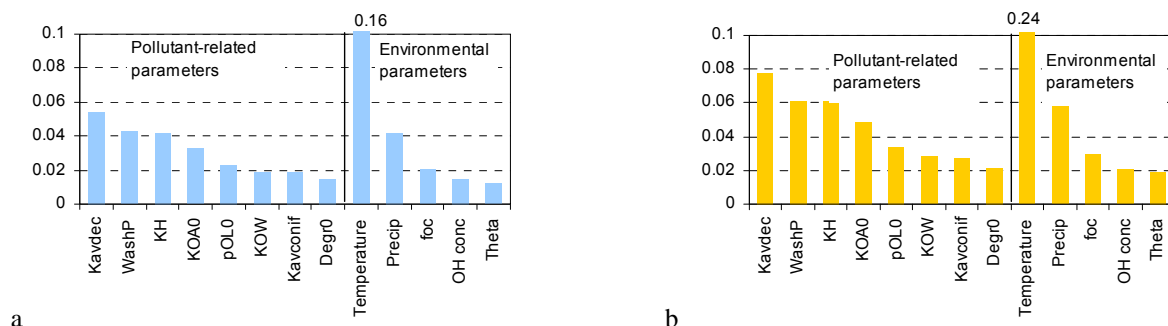


Fig. 13. Sensitivity of air concentrations (a) and depositions (b) with respect to PCB-153 pollutant-related and environmental parameters

Among environmental parameters both air concentrations and depositions are mostly sensitive to the changes of the ambient temperature and of precipitation amount. Relatively high values of sensitivities were obtained for mass transfer coefficient for deciduous forests, washout ratio, and Henry’s law constant. Relatively low sensitivity of model output parameters to variation of degradation rate is explained by significant persistence of PCB-153 in the atmosphere. In comparison to other processes the removal of PCB-153 from the atmosphere due to degradation is less effective within the region close to emission source.

With the increasing distance from emission source the relative importance of particular processes is changing as well as sensitivity to their parameters. Thus the role of deposition and the exchange with underlying surface becomes more important and sensitivities to variations of selected parameters increase (Fig. 14).

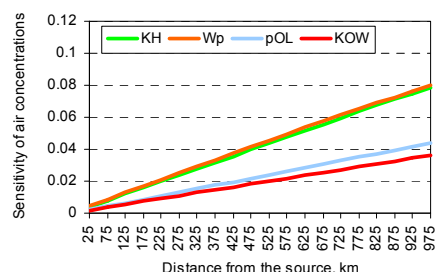


Fig. 14. The dependence of sensitivities of air concentrations with respect to a number of input parameters on the distance from the point source

As an example the spatial distribution of PCB-153 air concentrations calculated with the different values of Henry’s law constant and resulted sensitivity of model output to variation of this parameter can be considered (Fig. 15 and 16).

The increase of the Henry’s law constant leads to the decrease of wet deposition of a pollutant in gaseous phase as well as the decrease of its absorption by the underlying surface (soil, seawater). Comparing two maps in Fig. 15 it can be seen that higher value of Henry’s law constant resulted in more significant dispersion of a pollutant air concentrations in horizontal direction.

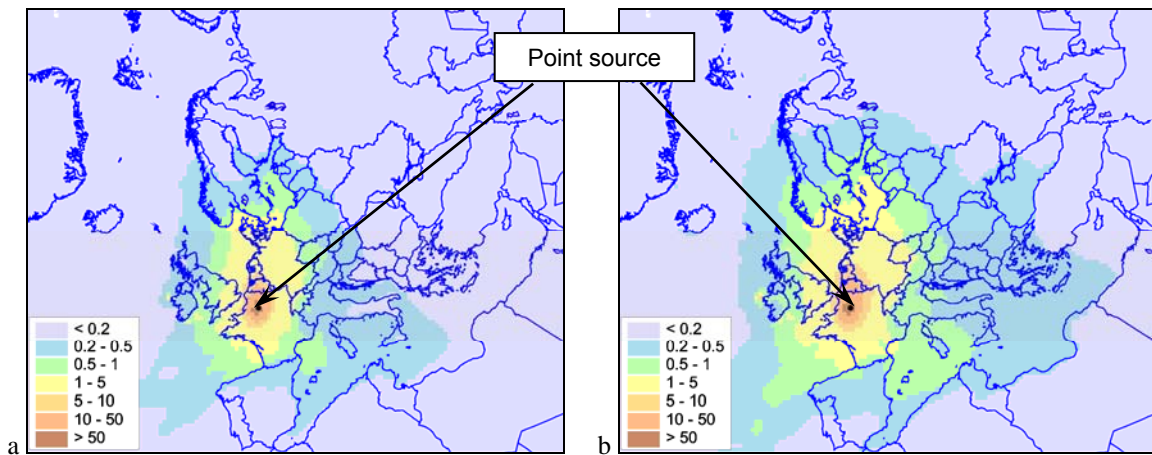


Fig.15. Calculated values of air concentrations of PCB-153 (pg/m^3) originated from a conventional point source of the power 2.8 t/y located in France (Paris) for “low” (a) and “high” (b) values of Henry’s law coefficient. Arrows show the location of the point source

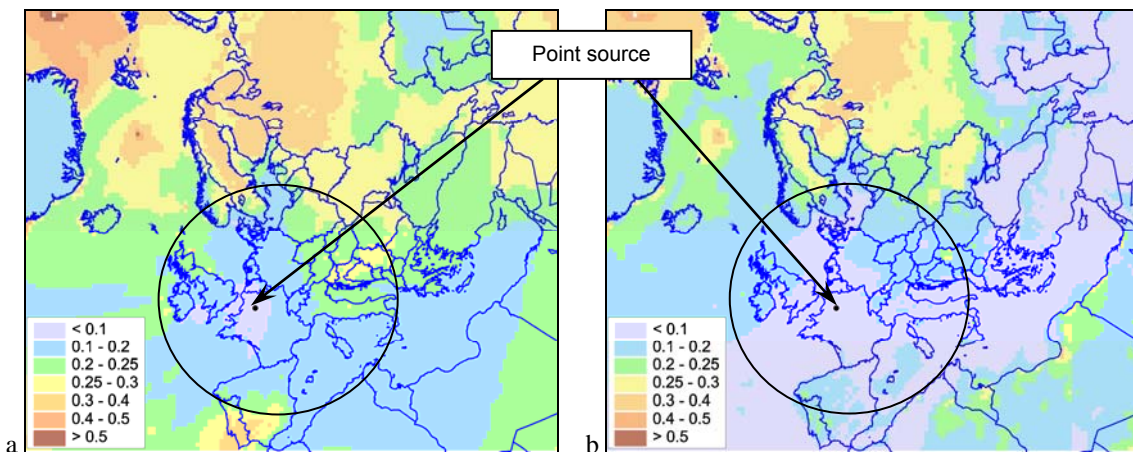


Fig. 16. Spatial distributions of sensitivity of air concentrations (a) and depositions (b) with respect to Henry’s law constant coefficient. Arrows show the location of the point source (1000 km – zone is marked)

Spatial distribution of sensitivities of air concentrations and depositions to variation of Henry’s law constant are presented in Fig. 16. It can be seen that sensitivities are growing with the distance from the point source. As it was mentioned above, with the increase of the distance from the emission source its influence becomes lower and thus contribution of removal processes to changing of concentrations in the atmosphere increases.

The values of calculated sensitivities allow estimating uncertainties of model output if the uncertainties of the input parameters are known. To do that we suppose that variations of these parameters are independent and that the uncertainty of each parameter is chosen in accordance to Table 1. Under these assumptions the uncertainty of output values *caused by uncertainties in pollutant-specific data* is estimated as 30% for air concentrations and 40% for depositions. The uncertainties calculated *taking into account both pollutant-specific and environmental parameters* can reach 50% for air concentrations and 70% for depositions. However, it should be taken into account that rather wide range of variation of the pollutant-specific parameters is defined in this study for the evaluation of extent of model response. It is supposed that the uncertainty introduced by the base values of pollutant-specific parameters to the model results is actually lower.

Results of model sensitivity study indicate that near emission sources the influence of re-emission process is not rather significant and can be neglected. However, for the evaluation of pollution levels in remote regions and for sufficiently long periods of time the influence of re-emission process for some of POPs can be essential. To evaluate the influence of re-emission on air concentrations modeling of PCB-153 long-range transport was carried out with and without taking re-emission into account for 50-years period. The contributions of PCB-153 re-emission from soil to its atmospheric concentrations at different distances from the emission source after 1, 10, 20, 30, 40 and 50 years from the beginning of simulations are presented in Fig. 17.

The contribution of re-emission process is increasing with distance from the emission source and with time. Even for one-year period its contribution at 5000 km from the considered point source equals to about 9% and for 50-year period reaches 65%. So, for PCB-153 the influence of re-emission from soil can be considerable especially for regions located far from main emission sources.

More detailed description of the results of the sensitivity study can be found in EMEP/MSC-E Technical Report [Gusev et al., 2005a].

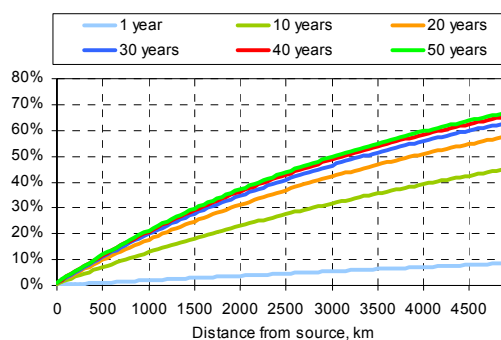


Fig. 17. The influence of re-emissions for various calculation periods

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

In accordance with the Work-plan on POPs, this year the environmental contamination by PAHs (4 indicator congeners – B[a]P, B[b]F, B[k]F and I_P), PCDD/Fs (toxic congeners), PCBs and HCB was evaluated. The evaluation of contamination of the EMEP region by PAHs and PCDD/Fs is carried out at the regional scale. It includes spatial distribution of atmospheric contamination (air concentrations and deposition for 2003), long-term trends (for the period from 1990 to 2003) and source-receptor matrices for 2003.

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

4.1. Pollution levels in the EMEP domain

This section presents the results of the evaluation of pollution levels of four indicator PAHs (benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-cd]pyrene) and PCDD/Fs within the EMEP region for the period 1990-2003. For B[a]P and PCDD/Fs the evaluation of transboundary transport of pollution was carried out.

4.1.1. Benzo[a]pyrene

Calculations of the benzo[a]pyrene (B[a]P) pollution levels in the EMEP region were performed with the 50x50 km² model for the period from 1990 to 2003. The emission data used in the calculations are based on official data and include expert estimates if official information is not available [Pacyna *et al.*, 1999] (Section 2.1).

The assessment of long-term trends of air pollution by B[a]P, its spatial distribution and transboundary transport in 2003 is given below.

Pollution trends in period 1990 – 2003. Emission of B[a]P to the atmosphere over Europe have decreased by 30% in period 1990-2003. Following the reduction of emissions annual mean atmospheric concentrations and depositions have decreased by 20% on average. Temporal variation of annual total B[a]P emissions and total depositions over Europe are given in Fig. 18.

Changes in emissions and depositions for individual European countries in period 1990-2003 are given in Fig. 19. The most significant decrease of national B[a]P emissions (more than 50%) is a characteristic of Luxembourg, Monaco, United Kingdom, Czech Republic, Slovakia, Republic of Moldova, Croatia and the Netherlands (Fig. 19a). It can be seen that level of pollution in these countries has changed differently. The most significant decrease of B[a]P depositions

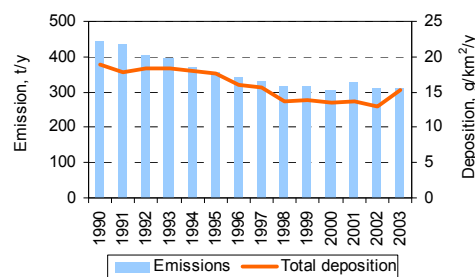


Fig. 18. Decrease of B[a]P total annual depositions over Europe in comparison with variation emissions in period 1990-2003

took place in the United Kingdom, the Netherlands, Slovakia, Republic of Moldova, and Czech Republic. The changes of depositions in other countries were less essential that can be explained by the influence of the transboundary transport.

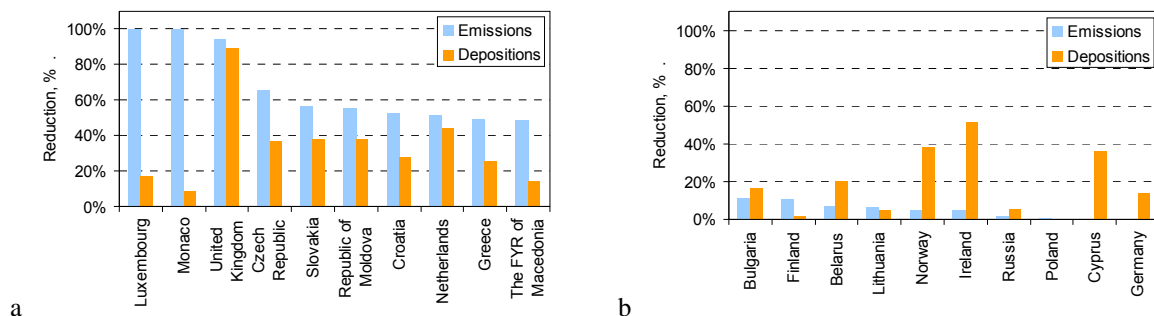


Fig. 19. Countries with highest (a) and lowest (b) reduction of B[a]P emission in 1990-2003 and corresponding reduction of total depositions

The influence of changes in transboundary transport is illustrated in Fig. 19b where countries with lowest changes of B[a]P emissions in period 1990-2003 are shown. It can be seen that significant decrease of depositions took place in Ireland, Norway, and Germany which is the result of the reduction of emissions in the United Kingdom. Changes of depositions in Cyprus, Belarus, and Bulgaria are caused by the emission decrease in countries of Central Europe.

Pollution levels in 2003. According to modelling results bulk of B[a]P emitted to the atmosphere in 2003 is deposited to the underlying surface (over 60%). About 13% of annual emission is degraded in the atmosphere and 26% is transported outside the EMEP domain (Fig. 20).

Significant amount of B[a]P depositions to soil lead to essential accumulation (over 95%) of the pollutant in this media (half-life of B[a]P in soil is about 2 years) as it is shown in Fig. 21.

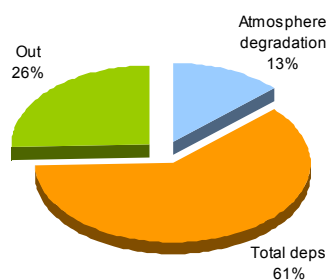


Fig.20. Total depositions, degradation, and atmospheric transport out of model domain of annual B[a]P emission in 2003

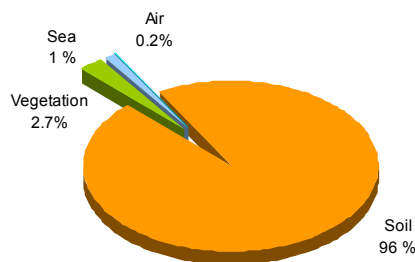


Fig. 21. Distribution of B[a]P between main environmental compartments

The mean value of B[a]P deposition flux over European countries in 2003 amounts to 16 g/km²/y. Spatial distribution of total B[a]P depositions in comparison with the emission density within the EMEP region is shown in Fig.22. High values of deposition fluxes (exceeding 50 g/km²/y) are obtained for the southern and eastern parts of Europe, in particular, Estonia, Latvia, Lithuania, Poland, Slovakia, Bosnia and Herzegovina, Serbia and Montenegro.

Average value of air concentrations in surface layer over the EMEP region amounts to about 0.2 ng/m^3 . (Fig. 23) However, in some regions of Central and South-Eastern Europe contamination levels exceed 1 ng/m^3 .

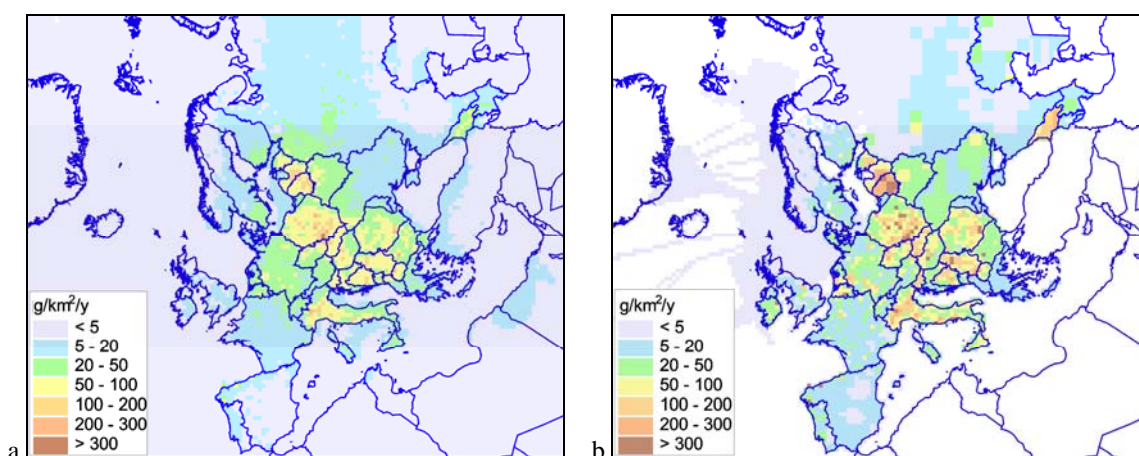


Fig. 22. Spatial distribution of B[a]P total depositions (a) and emissions (b) for 2003

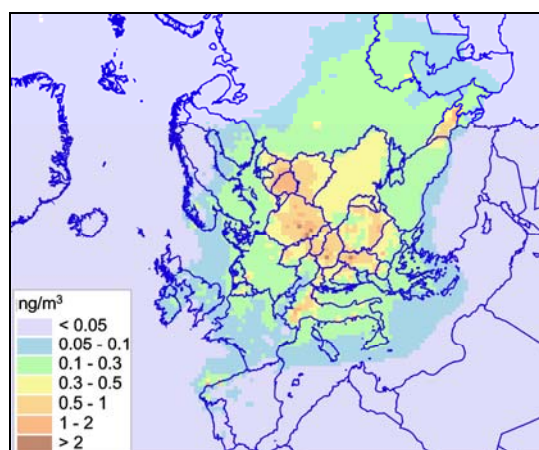


Fig. 23. Spatial distribution of B[a]P concentrations in surface air layer, 2003

Transboundary transport. The model allows evaluating both the contribution of each particular country to B[a]P air pollution in different European regions and the contribution of the transboundary transport to pollution in each particular European country.

In most of the European countries the contribution of external sources to B[a]P air pollution varies from 30 to 70% (Fig. 24). However the contribution of external sources depends significantly on the levels of internal B[a]P emissions in the country under consideration as well as its geographic position. For instance in such countries as Monaco and Luxembourg external sources are almost entirely responsible for the air contamination by B[a]P. The effect of transboundary transport on air concentrations is more significant than that of domestic emission flux in such countries as Cyprus, Albania, Iceland and Republic of Moldova (about 90% of the total concentration), Armenia (80%) and Croatia (75%). At the same time, the contribution of transboundary transport to the air concentrations in Spain does not exceed 20%.

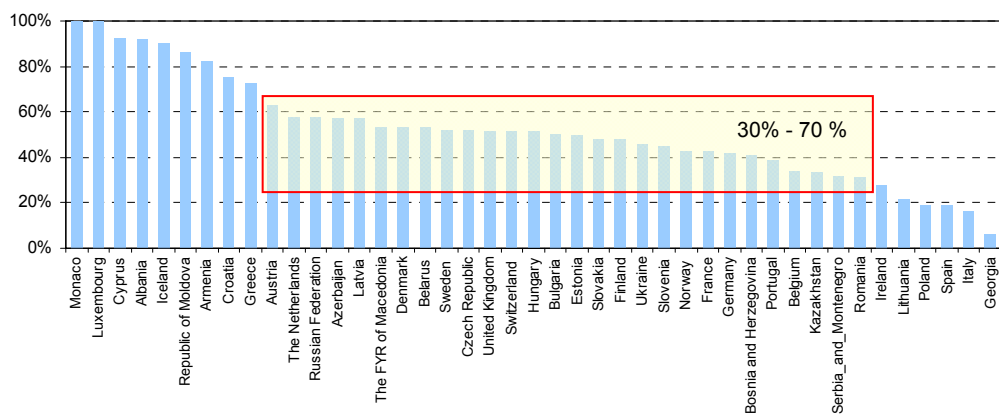


Fig. 24. Relative contributions of external emission sources to the B[a]P annual mean air concentrations in EMEP countries

The absolute contribution of external sources to the B[a]P mean annual air concentrations over the territory of some countries is also of interest. The countries, where air concentrations exceeded the average annual European value (0.2 ng/m^3), are shown in Fig. 25 together with transboundary contributions (export). The most significant effect of transboundary transport is observed for Republic of Moldova, Croatia, the FYR of Macedonia, the Netherlands, and Latvia.

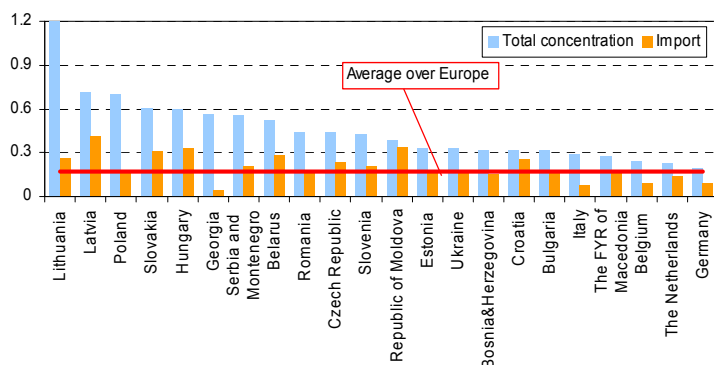


Fig. 25. Contributions of external emission sources to the B[a]P mean annual air concentrations in some European countries. Annual mean value of air concentration of B[a]P over Europe in 2003 (0.2 ng/m^3) is shown as a red line on the diagram

On the basis of model results obtained it is possible to identify the countries with the most significant contribution to B[a]P transboundary transport. Fig. 26 shows a diagram illustrating fractions of national emissions transported outside the borders of countries and deposited to the territories of other countries. For some countries this fraction can exceed 40%.

Next diagram shows the contribution of transboundary transport to B[a]P total depositions for European countries in 2003 (Fig. 27). The highest contributions of transboundary transport are typical for the countries with small territories (Monaco, Luxembourg, Albania, Iceland, Cyprus, etc.). For more than a half of European countries the fraction of imported depositions is about 60% and even higher.

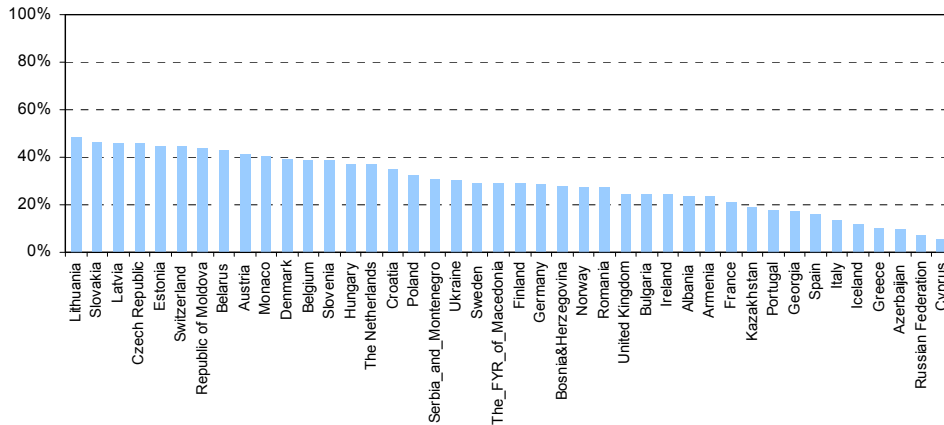


Fig. 26. Fractions of B[a]P national emissions deposited to the territory of other countries

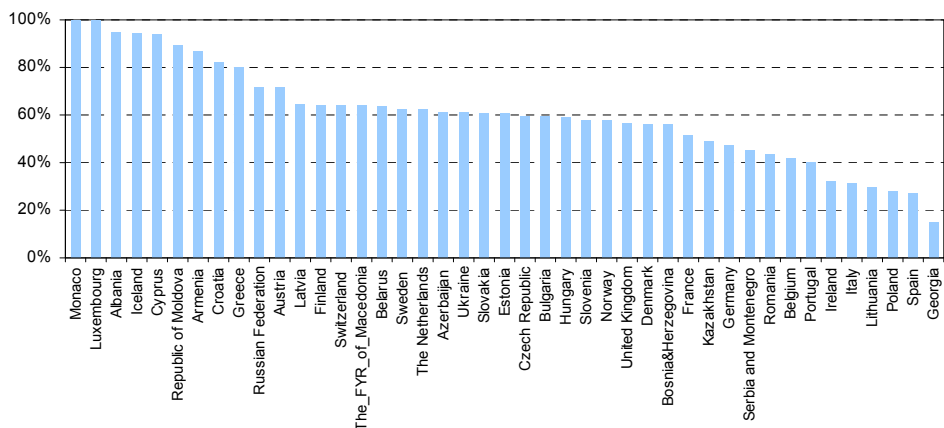


Fig. 27. Contributions of external emission sources to B[a]P depositions to the territories of European countries

Country-specific information. Model calculations can be used for the analysis of transboundary transport for each European country. The contribution of transboundary transport to the pollution of a particular country strongly depends on its location and area. As an example we shall consider three European countries - Albania, Czech Republic and Spain. The contributions of emission sources of various European countries to air concentrations in their territories are shown in Fig. 28.

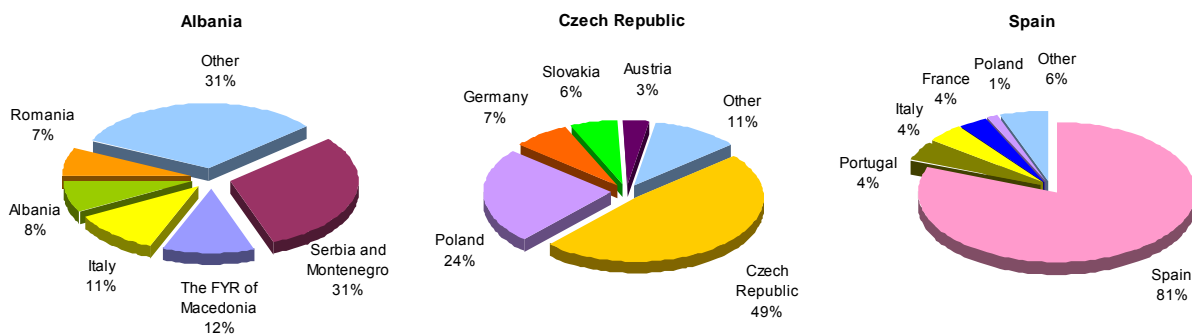


Fig. 28. Import diagrams for air concentrations in three European countries – Albania, Czech Republic, and Spain

In case of Albania the major contribution to air concentrations over its territory belongs to emission sources of neighboring countries. In particular, Serbia and Montenegro contributed 31%, the FYR of Macedonia – 12%, Italy – 11%, Romania – 7 %, and other countries contribute 31%. Due to rather small national emission (221 kg/y) the contribution of own emission sources of Albania to air concentration is about 8% only. For Czech Republic the contribution of its own emissions to air pollution of its territory is accounted for 49%. Emissions of neighboring countries (Poland, Germany, Slovakia, Austria and others) contribute 51%. And in case of Spain air pollution is mainly determined by its own emission sources (about 80%). This is conditioned by the geographical location of Spain and its rather significant area.

Different pattern can be seen in export diagrams for these countries, which represent contributions of their emission sources to depositions on other European countries (Fig. 29). The fraction of national emission deposited to the territory of these three countries varies from 36% to 55%, with lowest value in case of Czech Republic and highest in case of Spain. It means that Czech Republic exported about two thirds of its national emission of B[a]P in 2003.

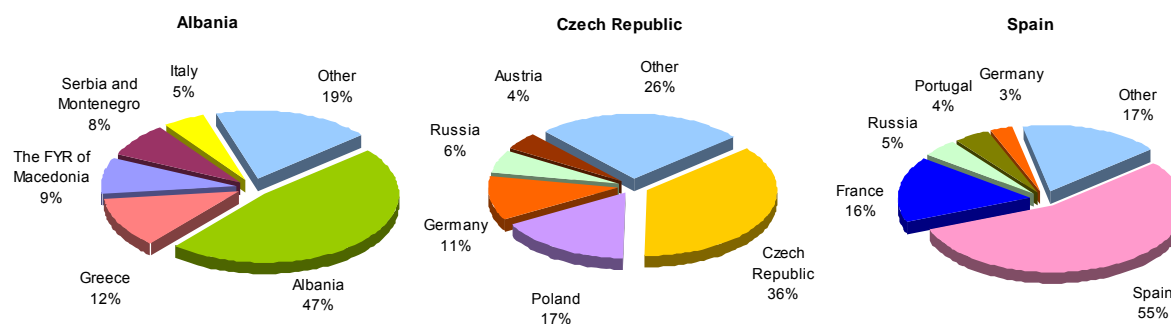


Fig. 29. Export diagrams for depositions in three European countries – Albania, Czech Republic, and Spain

Model results on transboundary transport of B[a]P for each European country are available in the Annex C of this report.

Comparison against measurements. Modeling results on B[a]P concentrations in air and in precipitation and deposition fluxes were compared with available measurements for the period 1990-2003. On the average the model slightly underestimates air concentrations of B[a]P. About 60% of calculated values of B[a]P air concentrations agree within a factor of two with observed concentrations and over 80% within a factor of three. The comparison of calculated values of B[a]P concentrations in precipitation and deposition fluxes with measured ones has shown that about 70% of measured concentrations in precipitation agree with calculations within a factor of two and over 80% – within a factor of three. More detailed analysis of the comparison results will be given in the MSC-E Technical Report [Shatalov et al., 2005b].

4.1.2. Benzo[b]fluoranthene

Calculations of pollution levels by benzo[b]fluoranthene in the EMEP region with resolution 50x50 km² for 1990 and 2003 were carried out using regional MSCE-POP model. The emission data used in the calculations are based on official data and expert estimates where official information is not available (Section 2.1). As it was shown in Section 2.1 B[b]F emission of European countries has reduced in period 1990-2003 by 28%. Pollution levels in this period have decreased by 17% for annual mean air concentrations and by 21% for total annual depositions.

At the same time on the level of individual countries the changes are different. In Fig. 30 the spatial distribution of computed B[b]F annual mean air concentrations for 1990 and 2003 is presented. Essential decrease in annual B[b]F concentrations in air can be noted for the United Kingdom, the Netherlands, Germany, Sweden, France, Spain, Serbia and Montenegro, and Romania. Some decrease can also be seen in Czech Republic, Slovakia, and Hungary. Levels of B[b]F annual mean air concentrations in Baltic countries, Belarus, and the Ukraine did not change significantly.

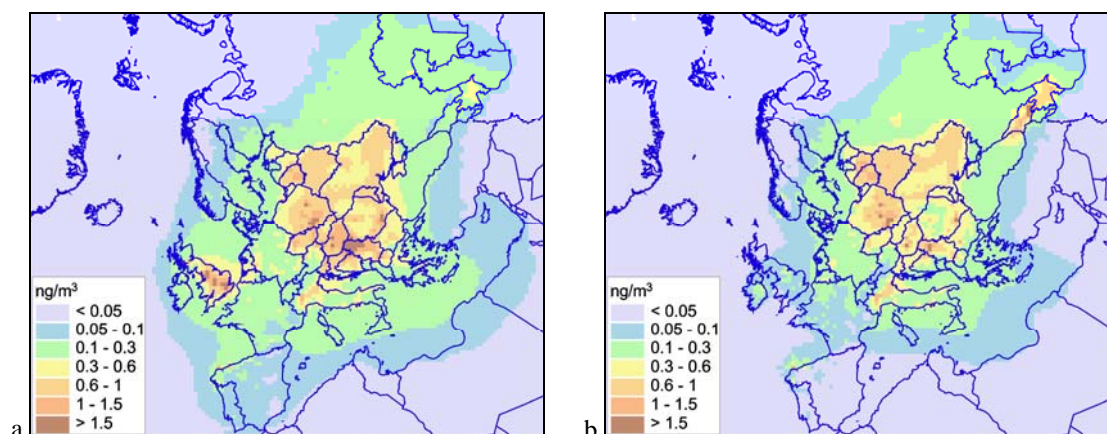


Fig. 30. Spatial distribution of B[b]F annual mean air concentrations for 1990 (a) and 2003 (b), ng/m^3

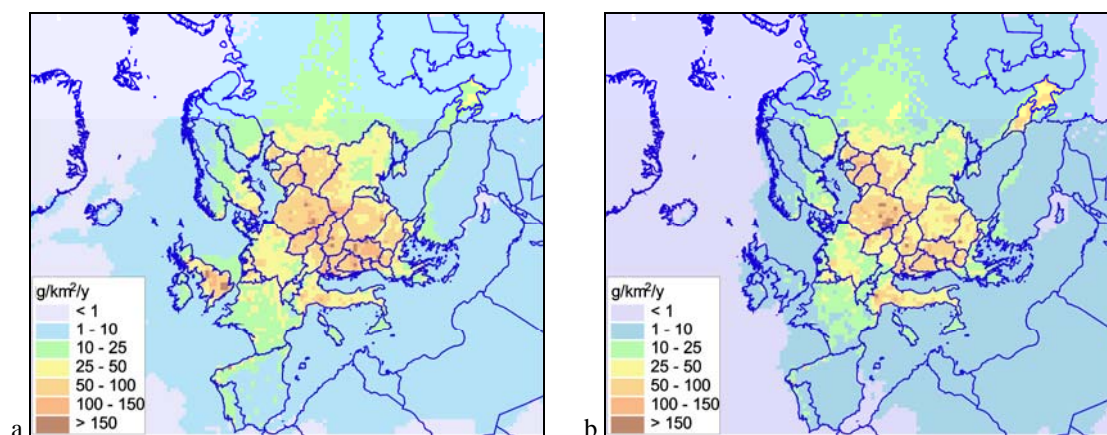


Fig. 31. Spatial distribution of B[b]F total annual depositions for 1990 (a) and 2003 (b), $\text{g}/\text{km}^2/\text{y}$

Similar pattern of changes from 1990 to 2003 has been obtained for B[b]F total annual depositions (Fig. 31). Most essential decrease of B[b]F depositions can be noted for the United Kingdom, the Netherlands, France, Spain, Sweden, Germany, Czech Republic, Slovakia, and Hungary.

4.1.3. Benzo[k]fluoranthene

Evaluation of pollution levels by benzo[k]fluoranthene in the EMEP region for 1990 and 2003 was performed with spatial resolution $50 \times 50 \text{ km}^2$. The emission data for B[k]F used in the calculations are based on official data and expert estimates (Section 2.1). Similar to B[b]F annual emissions of B[k]F within the European region have reduced by 33%. According to modeling results annual mean air concentrations of B[k]F over Europe have decreased by 27% and annual total depositions by 28%.

Calculated levels of annual mean air concentrations are given in Fig. 32. As for B[b]F following the reduction of emission significant decrease in air concentrations can be noted for the United Kingdom,

France, Spain, the Netherlands, Germany, Czech Republic, Slovakia, Hungary, and Serbia and Montenegro.

The spatial distribution of computed B[k]F annual total deposition for 1990 and 2003 is presented in Fig. 33. Like in the case of B[b]F the most pronounced changes in deposition levels took place in countries of western and central parts of Europe, namely, United Kingdom, the Netherlands, France, Spain, Sweden, Germany, Czech Republic, Slovakia, and Hungary.

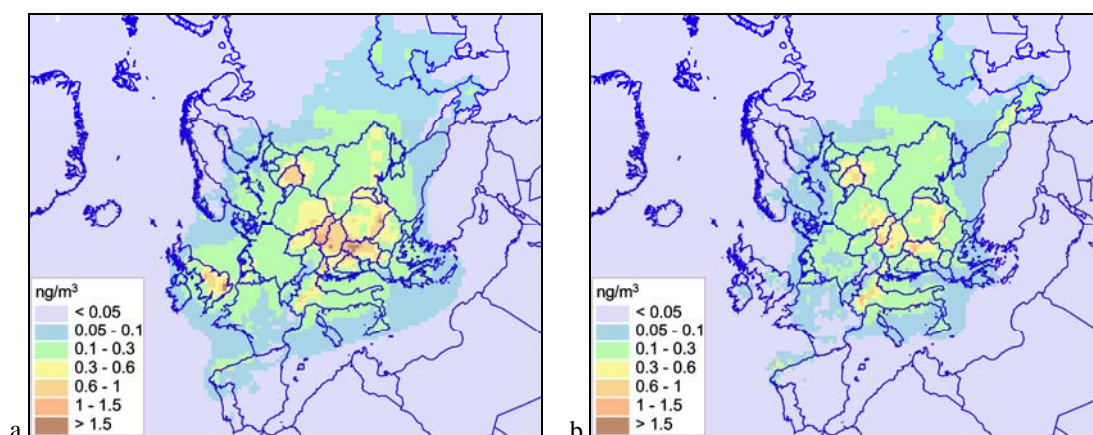


Fig. 32. Spatial distribution of B[k]F annual mean air concentrations for 1990 (a) and 2003 (b), ng/m^3

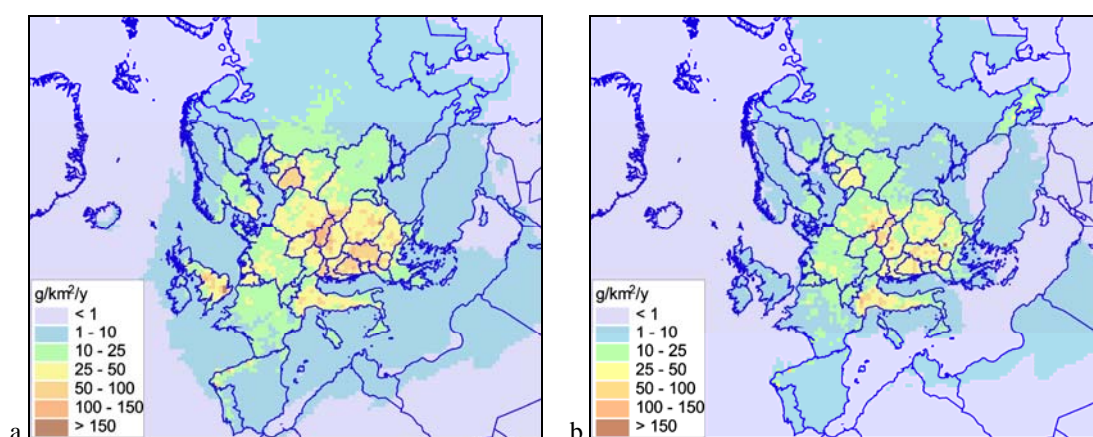


Fig. 33. Spatial distribution of B[k]F total annual depositions for 1990 (a) and 2003 (b), $\text{g/km}^2/\text{y}$

4.1.4. Indeno[1,2,3-cd]pyrene

Pollution levels of indeno[1,2,3-cd]pyrene within the EMEP region were obtained with the $50 \times 50 \text{ km}^2$ resolution for 1990 and 2003. The emission data used in the calculations are based on official data and expert estimates where official information is not available (Section 2.1). I_P emissions of European countries have reduced in period 1990-2003 by 18%. Following to reduction of emission pollution levels in this period have decreased by 11% for annual mean air concentrations and by 21% for annual total depositions.

Spatial distribution of I_P annual mean air concentrations (Fig. 34) and total annual depositions (Fig. 35) and their temporal changes in period 1990-2003 in general follow B[b]F concentrations and depositions. Notable changes in pollution levels by I_P are the characteristic of countries in western and central parts of European region.

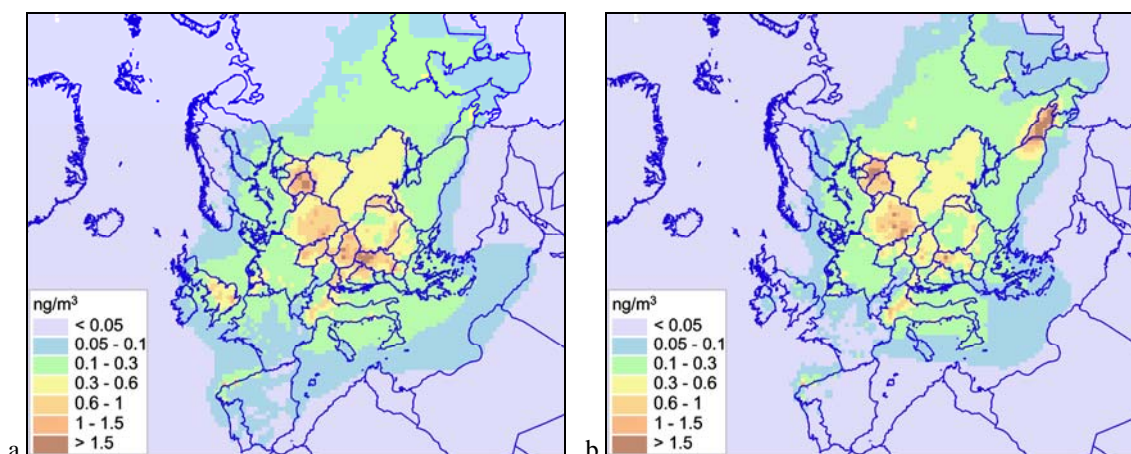


Fig. 34. Spatial distribution of I_P annual mean air concentrations for 1990 (a) and 2003 (b), ng/m^3

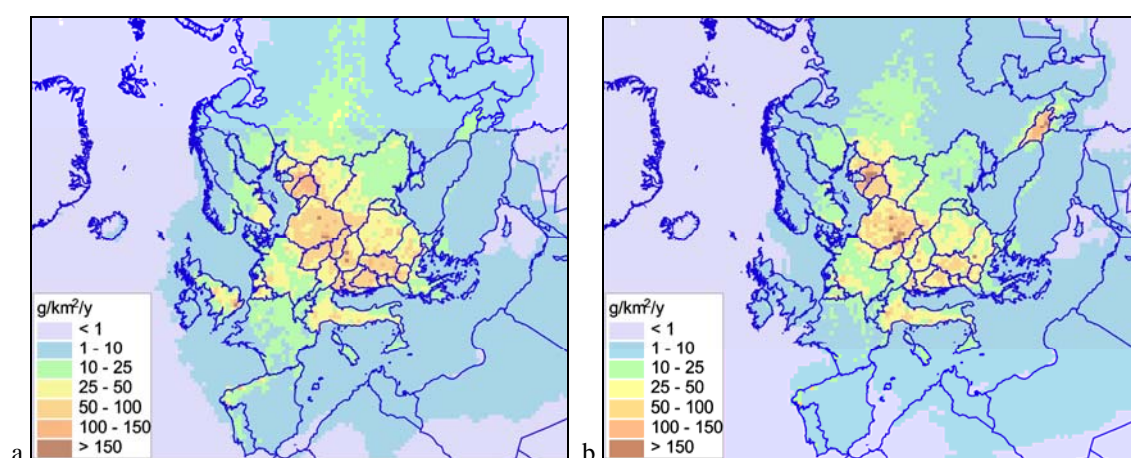


Fig. 35. Spatial distribution of I_P total annual depositions for 1990 (a) and 2003 (b), $\text{g}/\text{km}^2/\text{y}$

4.1.5. Polychlorinated dibenzo(p)dioxins and dibenzofurans

Calculations of pollution levels by polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs) in the EMEP region were performed with the $50 \times 50 \text{ km}^2$ model for the period from 1990 to 2003. Modeling was carried out for the indicator congener 2,3,4,7,8-PeCDF. The congener 2,3,4,7,8-PeCDF is chosen for calculations since it makes maximum contribution to the overall toxicity of emissions (about 40%). As it was shown in the previous EMEP status report on POPs [Dutchak et al., 2004] the calculations with the properties of the indicator congener 2,3,4,7,8-PeCDF can be used for the evaluation of long-range transport of the entire PCDD/F mixture. This assumption can lead to the differences in the spatial distribution of PCDD/F air concentrations of about 30% in comparison to the results obtained in modeling of distribution of 17 toxic congeners of PCDD/Fs. For the calculation of transboundary transport the discrepancies can be about 5%. The emission data used in the calculations are based on official data and expert estimates in case when official information is not available [Pacyna et al., 1999] (Section 2.2).

The assessment of long-term trends of air pollution by PCDD/Fs, its spatial distribution and transboundary transport in 2003 is given below. All estimates of pollution levels are given in NATO toxic equivalency units (TEQ).

Pollution trends in period 1990 – 2003. Total PCDD/F emission of European countries has reduced from 1990 to 2003 by 63%. According to model results obtained on the basis of these data annual total depositions of PCDD/Fs over Europe have decreased by 47% (Fig. 36). The decrease of annual mean concentrations of PCDD/Fs is accounted for 41% during this period. Annual mean deposition fluxes of PCDD/Fs undergo significant interannual variations. It can be seen that level of depositions obtained for 2003 is higher than in previous year in spite of decreasing of emission. Similar variations were also obtained for some other pollutants, for instance, for B[a]P and some heavy metals. The reason for this increase can be connected with anomalies in meteorological conditions of 2003. In particular, the summer of 2003 was one of the warmest in Western and Central parts of European region. Two distinct periods of heat wave in June and in the end of July beginning of August were recorded [Levinson and Waple, 2005].

Variations of PCDD/F emissions and depositions in individual European countries in period 1990-2003 are given in Fig. 37. In comparison to B[a]P described above changes in PCDD/Fs pollution levels are more uniform. This can be explained by higher fraction of gaseous phase and subsequently more essential re-emission. In course of emission reduction the process re-emission can play significant role and make substantial contribution to air concentrations and deposition fluxes. Significant reduction of national emissions (more than 50%) can be noted for about 20 countries, namely, the Netherlands, Czech Republic, France, Belgium, Switzerland, and etc (Fig. 37a). According to modelling results annual total depositions in these countries has been decreased by about 60%. Countries with lowest decrease of emissions are given in Fig. 37b. Corresponding changes in depositions over their territories are in a range 35-60%.

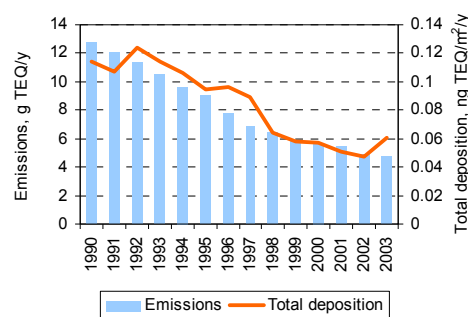


Fig 36. Decrease of PCDD/F total annual depositions over Europe in comparison with variation of emissions in period 1990-2003

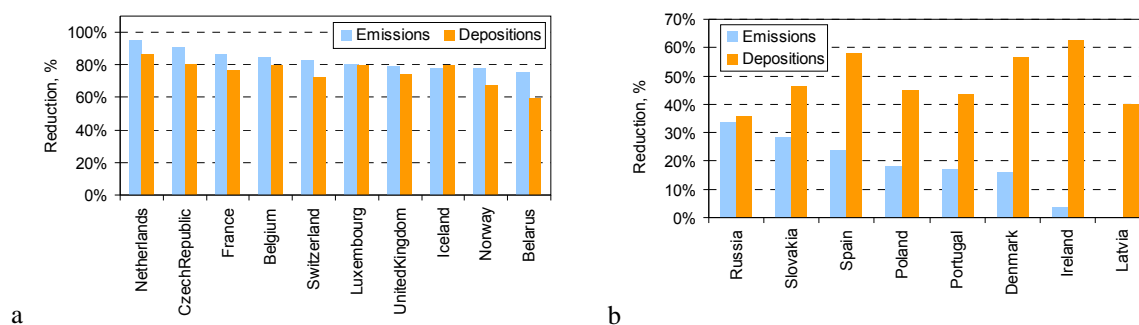


Fig. 37. Countries with highest (a) and lowest (b) reduction of PCDD/Fs emission in 1990-2003 and corresponding reduction of total depositions

Pollution levels in 2003. The distribution of PCDD/F environmental content between different environmental media (the atmosphere, soil, vegetation and seawater) for 2003 is shown in Fig. 38. Due to significant persistence of PCDD/Fs in soil (in accordance with model parameterization PCDD/F half-life in soil amounts to about 60 years) the pollutant is mainly accumulated in this media. According to model estimates, over 95% of overall environmental content of PCDD/Fs is contained in soil (including forest litter as the upper soil layer).

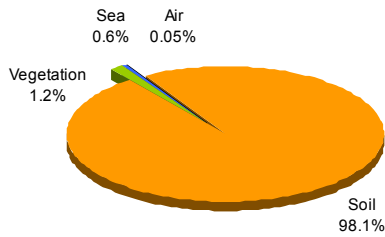


Fig. 38. Distribution of PCDD/Fs between main environmental compartments

Spatial distribution of PCDD/F air concentrations within the EMEP region for 2003 is presented in Fig. 39a. Mean value of PCDD/F air concentrations over European countries in 2003 accounts for 2.6 fg TEQ/m³. Averages of air concentrations over European countries vary from 0.02 fg TEQ/m³ to 15 fg TEQ/m³. Regions with high levels of air concentrations are located in central, eastern and southern parts of Europe.

The mean value of PCDD/F deposition flux (Fig.39b) over European countries in 2003 amounts to 0.55 ng TEQ/m²/y. High values of deposition fluxes (exceeding 1 ng TEQ/m²/y) were obtained for the southern and eastern parts of the region as well as in Northern Europe and a part of the United Kingdom. High values of deposition fluxes over these regions are conditioned by relatively higher deposition velocities characteristic for forested areas.

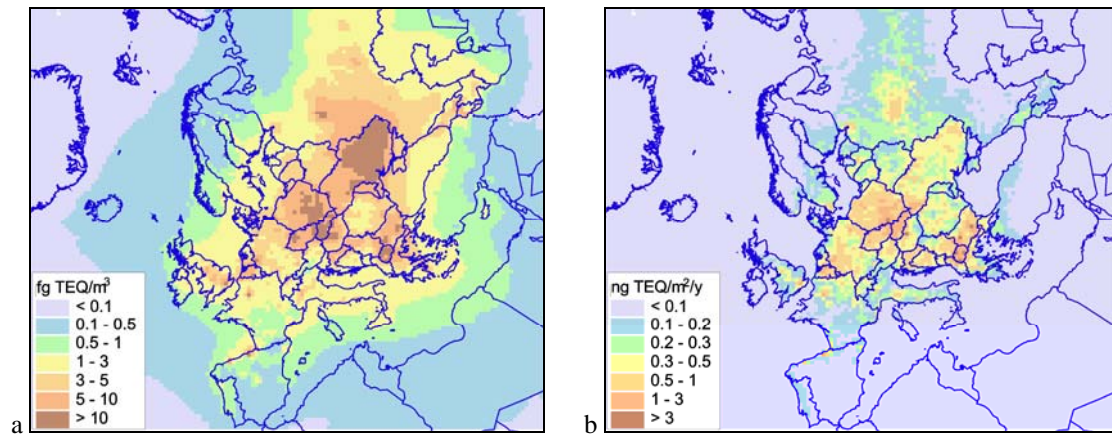


Fig.39. Spatial distribution of PCDD/F air concentrations (a) and depositions (b) for 2003

Spatial distribution of PCDD/F concentrations accumulated in upper 5 cm layer of soil within the EMEP region in period 1990-2003 is shown in Fig. 40. Average value of soil concentrations in the upper soil layer amounts to about 55 pg TEQ/g. However, over vast territories of Europe soil contamination levels exceed 100 pg TEQ/g. Regions with high soil concentrations correspond to regions with high levels of deposition density (Fig. 39b).

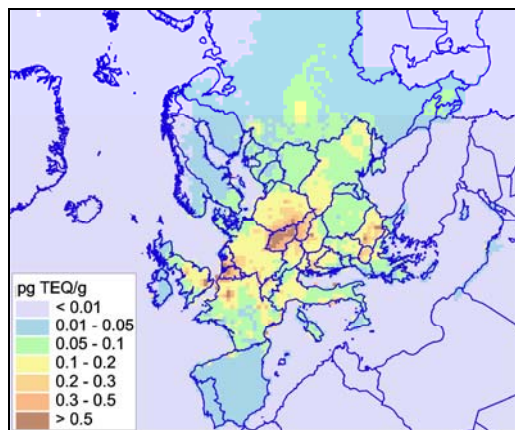


Fig.40. Spatial distribution of PCDD/F concentrations in upper soil layer (5cm) for 2003

Transboundary transport. Along with pollution levels in European countries in 2003 the contribution of each particular country to PCDD/F air pollution in different parts of European region and the contribution of the transboundary transport to the pollution in each particular European country were evaluated. As it was mentioned above, re-emission process can essentially contribute to air pollution. Therefore two types of transboundary transport were distinguished in modeling of PCDD/Fs: primary transport of a pollutant emitted into the atmosphere from the emission source, secondary transport of a pollutant re-emitted from the underlying surface (so-called grasshopper effect).

For the evaluation of primary transboundary transport calculations of transport from the sources of all European countries in 2003 were performed. The contribution of external sources to PCDD/F air pollution for the European countries typically varies from 30 to 70% (Fig. 41).

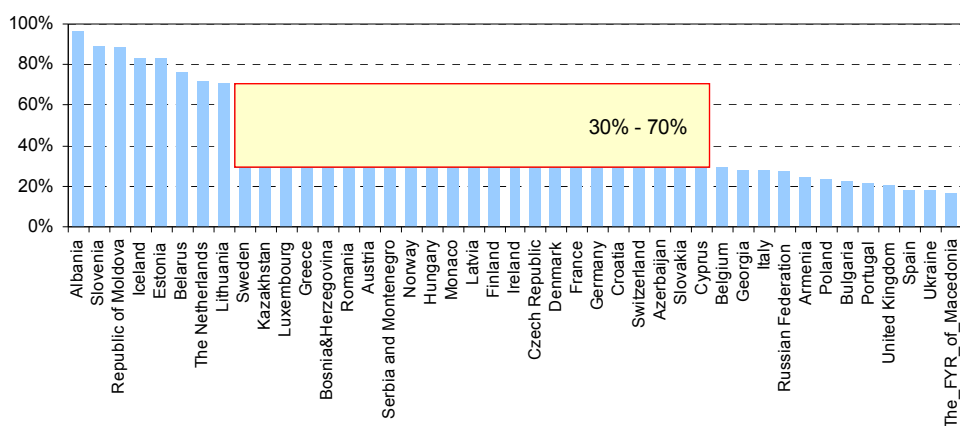


Fig. 41. Relative contributions of external emission sources to the PCDD/F annual mean air concentrations in EMEP countries

The contribution of external sources to PCDD/F pollution depends significantly on the levels of national emissions in the country under consideration as well as its geographic position. Particularly, in Albania, Slovenia, Republic of Moldova, Iceland and Estonia the contribution of external sources to air concentrations exceed 80%. At the same time, the contribution of transboundary transport to the total concentrations in Spain, Ukraine and the FYR of Macedonia does not exceed 20%.

Along with relative values the absolute contribution of external sources to PCDD/F mean annual air concentrations over European countries can be considered. The countries, where air concentrations exceed the average annual European value (2.6 fg TEQ/m³), are shown in Fig. 42 together with contributions of transboundary transport. The most significant influence of transboundary transport is obtained for the Republic of Moldova, Serbia and Montenegro, Belarus, the Netherlands, and Luxembourg.

Model results permit to estimate the contributions of countries to the transboundary transport of PCDD/Fs, i.e. the fraction of national emission deposited outside the territory of particular country. In Fig. 43 the fractions of national emissions deposited to the territory of other countries in 2003 are given. Significant fractions national emissions exported to depositions over Europe belong to Luxembourg, Monaco, Republic of Moldova, Lithuania, Slovakia, and etc.

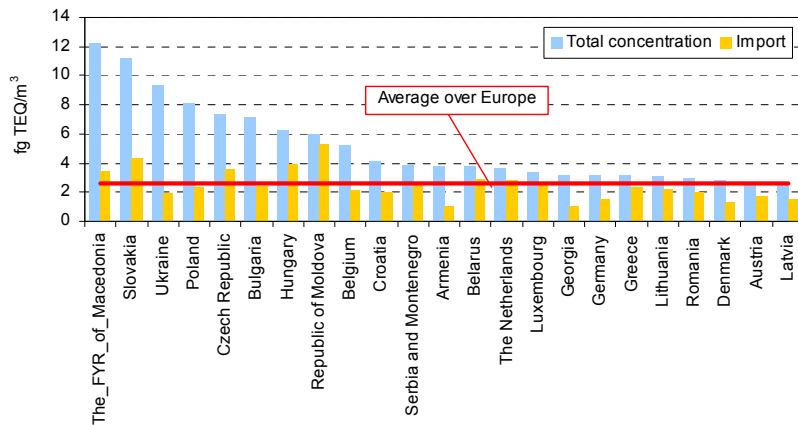


Fig. 42. Contributions of external emission sources to the PCDD/F mean annual air concentrations in some European countries. Annual mean value of air concentrations of PCDD/Fs over Europe in 2003 (2.6 fg TEQ/m³) is as a red line on the diagram

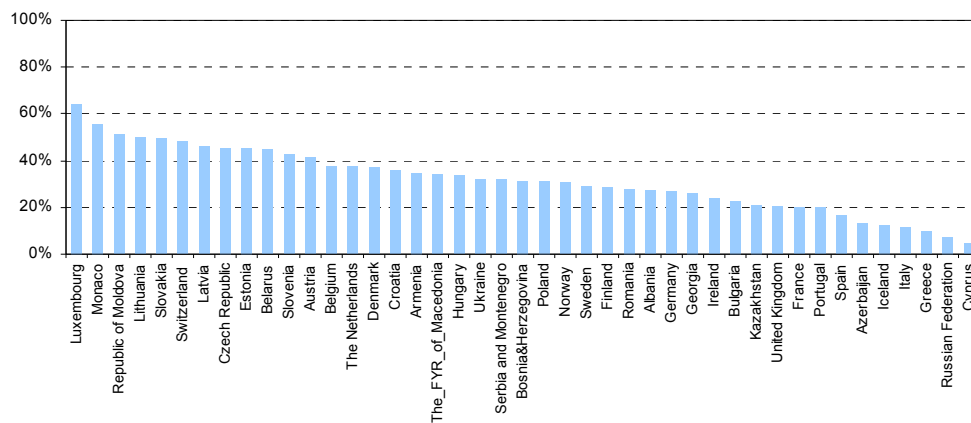


Fig. 43. Fractions of PCDD/F national emissions deposited to the territory of other countries

Additionally the contributions of external sources to depositions over particular countries can be analysed (Fig. 44). The most significant contribution originated from external sources to total depositions over European countries is a characteristic of Albania, Slovenia, Republic of Moldova, Iceland, Estonia, and Belarus (over 80%). In almost a half of the countries this contribution exceeds 50%.

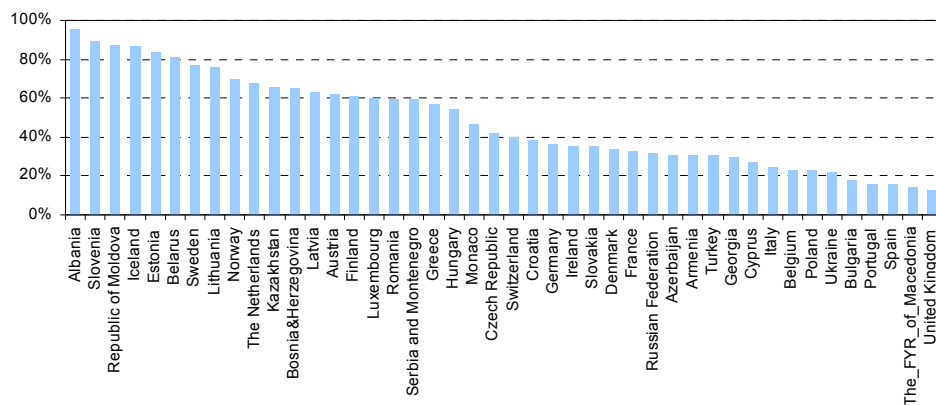


Fig. 44. Contributions of external emission sources to PCDD/F depositions to the territories of European countries

Country-specific information. Model calculations are also used for the analysis of primary transboundary transport for each European country. As an example, contributions of emission sources of various European countries to air concentrations over Slovenia, Austria and Portugal can be considered (Fig. 45).

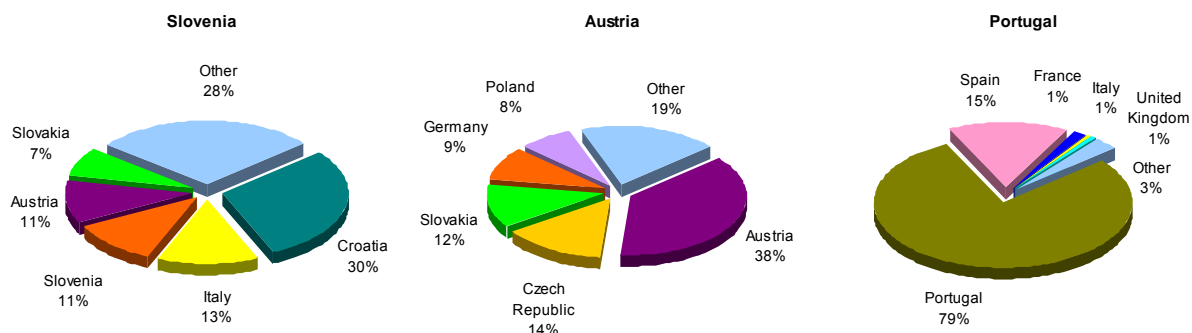


Fig. 45. Import diagrams for air concentrations in three European countries – Slovenia, Austria, and Portugal

In case of Slovenia the contribution of national emission sources to air concentrations is rather small (11%) due to comparatively small emissions (1.36 g TEQ/y). Thus air concentrations over Slovenia are formed mainly by the emission sources of neighboring countries, in particular, Croatia (30%), Italy (13%), Austria (11%), Slovakia (7%), and other (28%). For Austria the contribution of national emission sources to air pollution is 38%. Neighboring countries contribute 62%: Czech Republic (14%), Slovakia (12%), Germany (9%), Poland (8%), and other (19%). In case of Portugal air pollution is mainly determined by national emission sources (79%). Other countries contribute only 21% among which the most significant contribution belongs to Spain (15%).

Finally, model calculation allows evaluating contributions of secondary transport to air concentrations and depositions to European countries. As it was mentioned above this is a contribution of re-emission of previously deposited pollutant from underlying surface. The contributions of secondary transport to air concentrations over European countries are presented in Fig. 46.

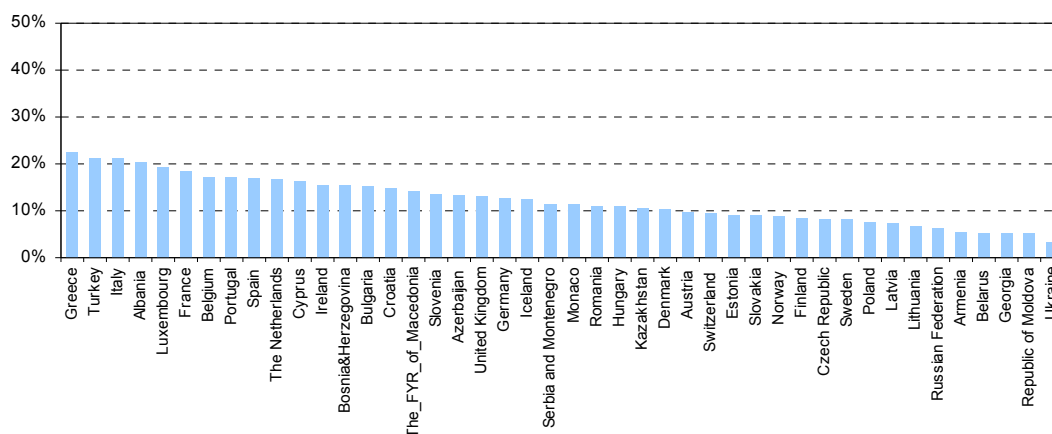


Fig. 46. Relative contributions of secondary PCDD/F transport (re-emissions) to air concentrations in European countries

For more than a half of European countries this contribution exceeds 10%. The contributions of secondary transport to depositions over European countries are higher (Fig. 47).

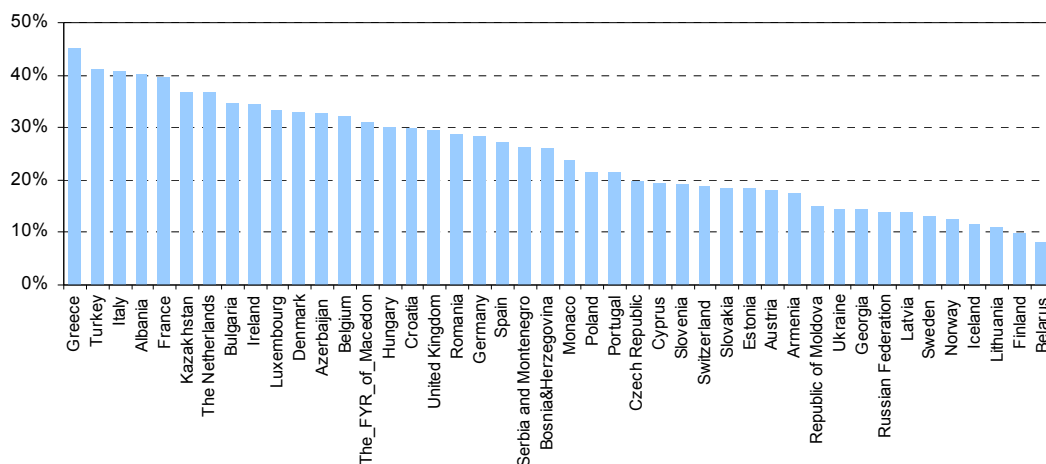


Fig. 47. Contributions of secondary PCDD/F transport (re-emissions) to depositions to European countries

For more than a half of European countries the contribution of secondary transport to depositions exceeds 20%. This shows that evaluation of air pollution in European countries require examination of secondary transport (re-emissions).

Model results on transboundary transport of PCDD/Fs for each European country are available in the Annex C of this report.

Comparison against measurements. Modeling results on PCDD/Fs concentrations in air and in precipitation and deposition fluxes were compared with available measurements for the period 1990-2003. At present PCDD/Fs are not included into the EMEP measurement programmes and for model validation the results of various measurement campaigns were used. On average the model underestimates available measurement data on air concentrations. For deposition fluxes model results are in a good agreement with measured ones. About 40% of calculated values of PCDD/F air concentrations agree within a factor of two with observed concentrations and over 60-70% within a factor of three. More detailed analysis of the comparison results will be given in the MSC-E Technical Report [Shatalov *et al.*, 2005b].

4.2. Pollution levels in the Northern Hemisphere

PCBs and HCB are the pollutants of significant long-range transport potential. Taking this into account the evaluation of pollution of European region by these POPs requires consideration of the influence of remote emission sources as well as re-emission of previously accumulated pollutants. Modeling of PCB and HCB transport and depositions within the northern hemisphere was carried out using the hemispheric version of MSCE-POP model. In current year the development of hemispheric MSCE-POP model was continued with the refinement of its ocean block. More detailed information on this subject is given in the Annex B.

4.2.1. Pollution levels of Hexachlorobenzene

The study of intercontinental transport of HCB within the northern hemisphere was continued during this year with calculations for the period 1990-2003. Modeling was performed on the basis of emission scenario described in Section 2.3 and in previous EMEP status report 3/2004 [Dutchak et al., 2004]. Several groups of emission sources were distinguished within the northern hemisphere: Europe, Russia, Central Asia, South-eastern Asia, and America. For the evaluation of HCB intercontinental transport several model run was 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 Europe and the Arctic region were estimated.

Spatial distribution of the pollution by HCB within the northern hemisphere for 2003 is present in Fig. 48. Along with maps of annual mean air concentrations and net annual deposition fluxes the distribution of annual emissions is also given in the figure.

Elevated levels of HCB air concentrations for 2003 ($30\text{-}80\text{ pg/m}^3$) are obtained for the countries of central and eastern parts of Europe and over European part of Russia. Relatively lower levels of pollution ($10\text{-}30\text{ pg/m}^3$) can be noted for the countries of Northern and Southern Europe. Significant level of HCB air concentrations (above 80 pg/m^3) is a characteristic of India. The pattern of HCB annual net deposition fluxes over land surfaces is similar to the spatial distribution of air concentrations. Elevated levels of fluxes can be noted over European part of Russia, countries of Eastern Europe, east coast of America, and in South-eastern Asia ($3\text{-}12\text{ g/km}^2/\text{y}$). Over the seawater re-volatilization fluxes of HCB are obtained for 2003.

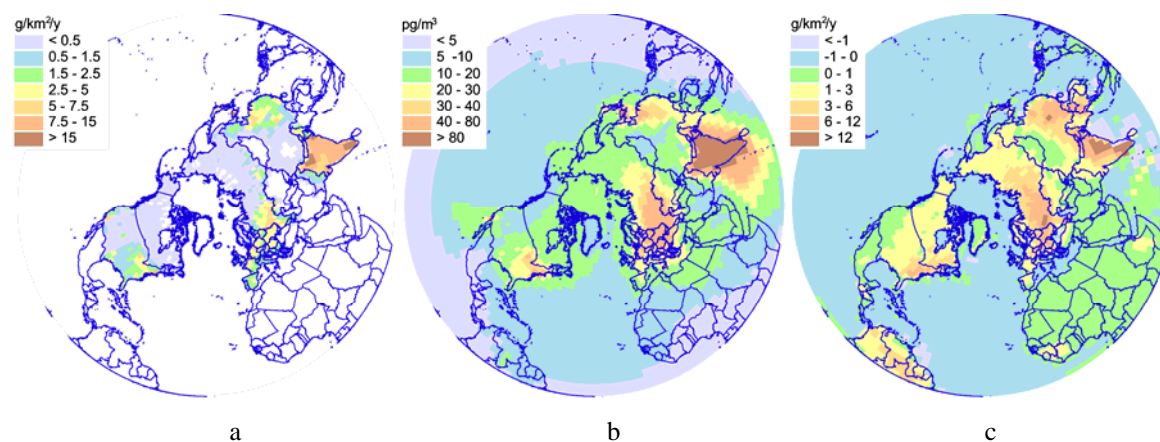


Fig. 48. Spatial distribution of HCB emissions, $\text{g/km}^2/\text{y}$ (a), annual mean air concentrations, pg/m^3 (b), and net depositions, $\text{g/km}^2/\text{y}$ (c) for 2003

Atmospheric pathways of distribution of HCB annual mean concentrations originated from emission sources in Europe, Southeastern Asia, and Northern America for 2003 are presented in Fig. 49.

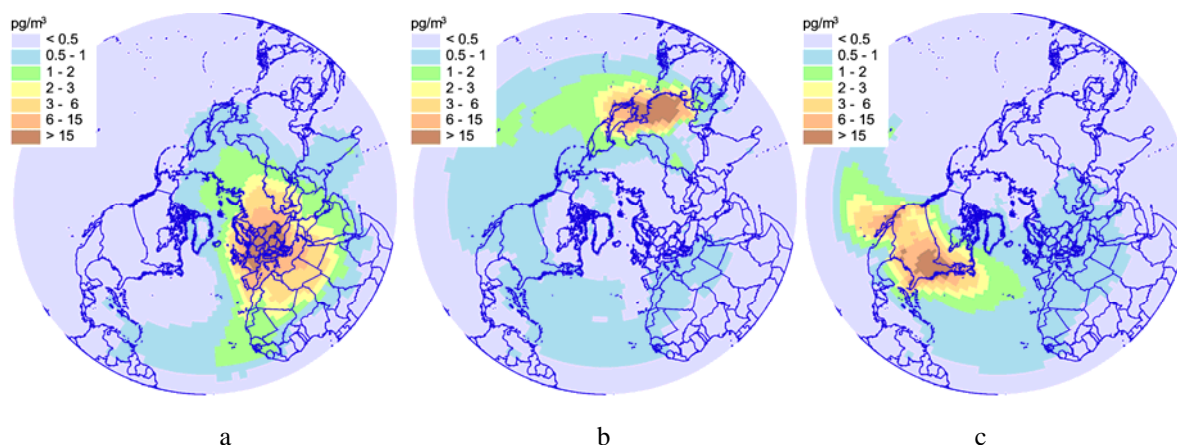


Fig. 49. Spatial distribution of annual mean air concentrations of HCB (pg/m^3) for 2003 originated from emission sources in different regions: Europe (a), South-eastern Asia (b), and North America (c)

It can be seen that European emission sources of HCB can essentially contribute to the contamination of the Arctic region, European part of Russia, and northern Africa. HCB from emission sources of North America reaches Northern Atlantic region and countries of Western Europe due to prevailing westerly winds. Emission sources of South-eastern Asia under favorable conditions can contribute to the pollution of North America. Peculiarities of atmospheric transport of HCB from emission sources of Europe and North America in different months of 2003 are shown in Fig. 50 and Fig. 51. It can be seen that the Arctic region is mainly affected by European emission sources in winter and early spring that is exemplified by the map of monthly mean air concentrations for March 2003. For summer time atmospheric transport in southern direction is more pronounced.

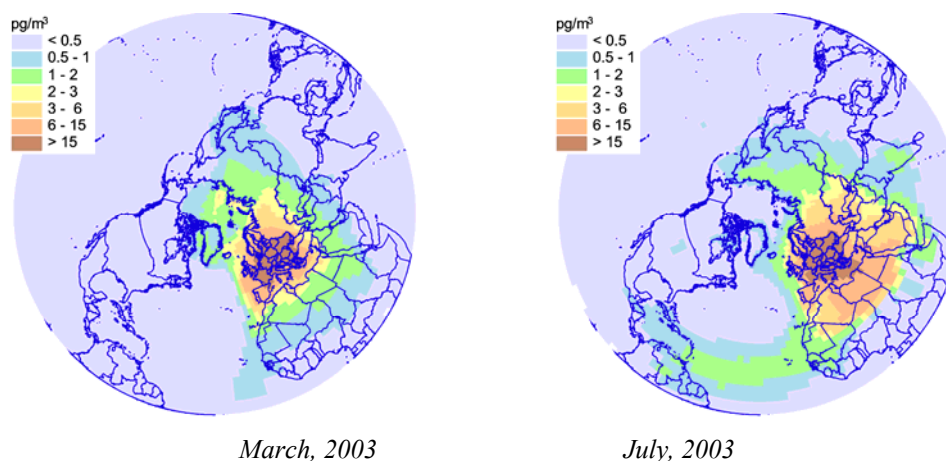


Fig. 50. Spatial distribution of monthly mean air concentrations of HCB (pg/m^3) from European emission sources in March and July of 2003

Distribution of pollution from North American emission sources is mainly occurred in north-eastern direction across the Northern Atlantic in March and in eastern direction in July of 2003 (Fig. 51).

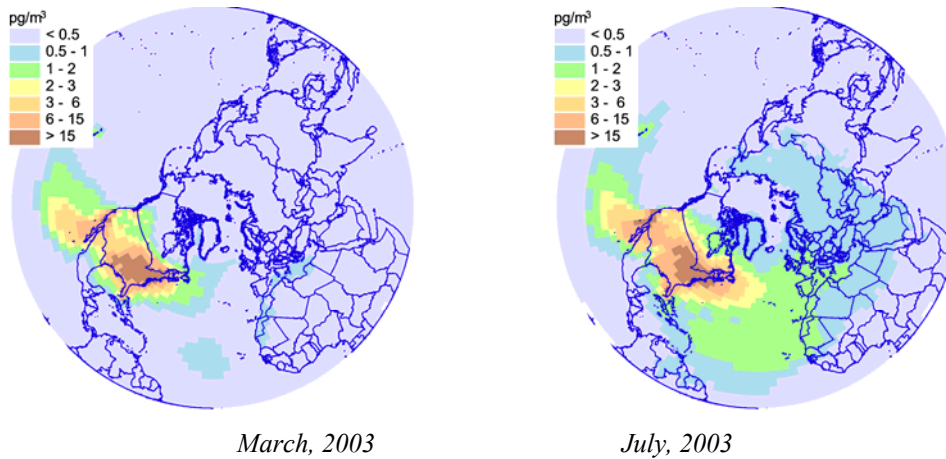


Fig. 51. Spatial distribution of monthly mean air concentrations of HCB (pg/m^3) from emission sources of North America in March and July of 2003

On the basis of calculations performed for the selected groups of emission sources the intercontinental transport of HCB was estimated. The distribution of HCB depositions originated from emission sources of Europe, South-eastern Asia and America for 2003 is presented in Fig. 52. Particularly, 48% of total HCB depositions over land from European emission sources is deposited over Europe, 35% over Russia, 8% over Central Asia, and about 9% in other parts of the northern hemisphere. In case of South-eastern Asia and North America major part of total HCB depositions over land (about 80%) belongs to their own territories.

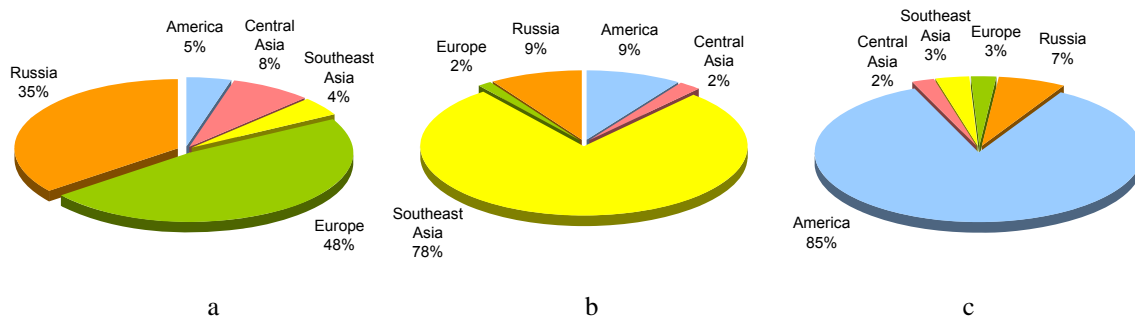


Fig. 52. Distribution of HCB depositions originated from emission sources of Europe (a), Southeast Asia (b), and North America (c) in 2003

Contributions of selected groups of emission sources to annual HCB depositions over Europe and over the Arctic region for 2003 are presented in Fig. 53. Major part of total HCB depositions over Europe (79%) is originated from European emission sources, 14% from Russia, and 7% from other emission sources. Relatively high contribution of European emission sources and small contribution of other sources to depositions over Europe can be explained by peculiarities of atmospheric conditions in 2003.

In case of the Arctic region the highest contribution to the depositions is made by Russian emission sources (44%). Significant contributions belong also to emission sources of Europe (22%), America (14%), and South-eastern Asia (12%).

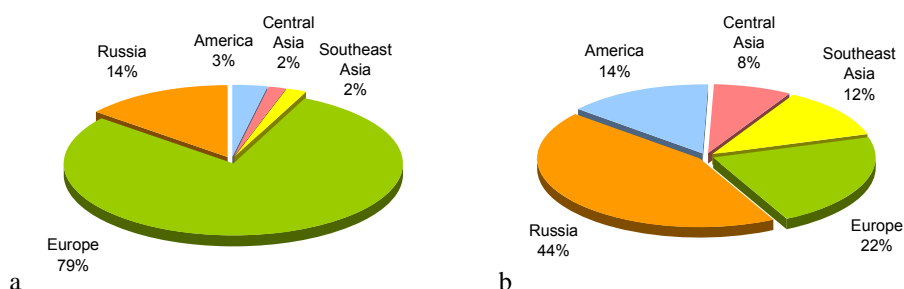


Fig. 53. Contributions of selected groups of emission sources of HCB to depositions over Europe (a) and the Arctic region (b) in 2003

4.2.2. Pollution levels of Polychlorinated biphenyls

Evaluation of intercontinental transport of PCBs and pollution of European region by emission sources outside Europe was carried out on the example of individual congener PCB-153. Computations were performed for the period 1990-2003 using the global emission inventory of 22 PCB congeners [Breivik *et al.*, 2002]. It should be mentioned that this inventory was prepared for the period 1930-2000. Therefore in computations it was assumed that the level of PCB emission in 2001-2003 was the same as in 2000. Similar to HCB several groups of emission sources were considered, namely: Europe, Russia, Africa and Central Asia, South-eastern Asia, and America (Section 2.4). The most significant annual emissions of PCB-153 among these group of sources belong to European region (5.9 tonnes) and North America (2.2 tonnes).

To evaluate PCB intercontinental transport several model run was made for each of the selected groups of emission sources. On the basis of obtained modeling results pathways of PCB-153 atmospheric transport were described and contributions of selected emission sources to depositions over Europe and the Arctic region were estimated.

In Fig. 54 the spatial distribution of PCB-153 annual emission fluxes, computed air concentrations, and net annual depositions for 2003 are illustrated. Elevated levels of emissions can be noted for European region in comparison to other parts of the northern hemisphere.

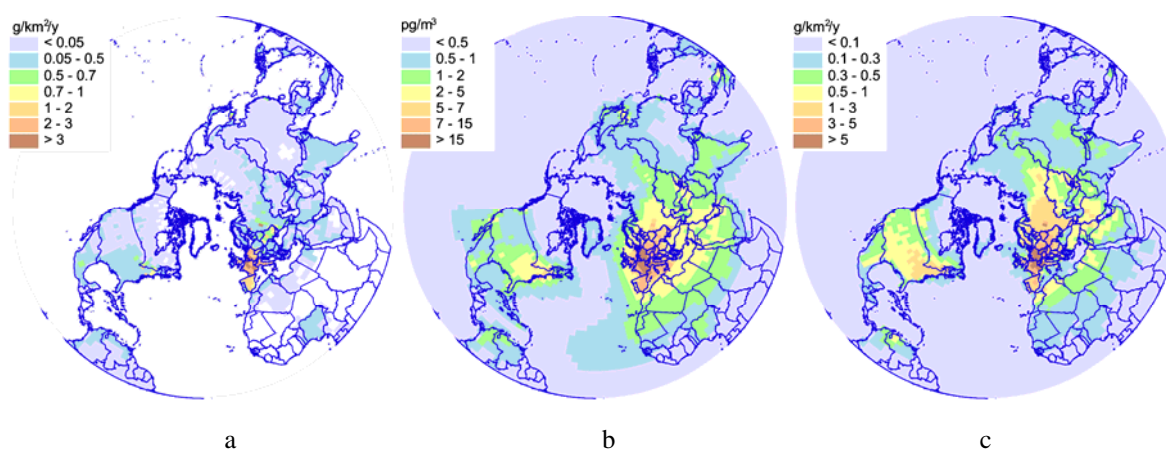


Fig. 54. Spatial distribution of PCB-153 emissions, g/km²/y (a), annual mean air concentrations, pg/m³ (b), and net depositions, g/km²/y (c) for 2003

Spatial distribution of annual mean air concentrations and net annual deposition fluxes of PCB-153 for 2003 resulted from all considered emission sources is shown in Fig 54b and c. Significant levels of air concentrations (about 15 pg/m^3 and higher) and net annual depositions (about 5 $\text{g}/\text{km}^2/\text{y}$) in Europe can be noted for France, Belgium, the Netherlands, and Germany. Relatively lower levels of pollution are obtained for the countries in eastern and northern parts of Europe. In most of the countries PCB-153 air concentrations are within a range of 1-15 pg/m^3 with lowest levels in northern parts of Norway, Sweden, Finland, and Russia. Similar spatial variation is the characteristic of net annual deposition flux of PCB-153.

PCB-153 is one of the heavy PCB congeners which can be characterized by relatively low volatility, significant persistence and lipophilicity. In comparison to HCB it has lower long-range transport potential. However due to long-term application of PCBs all over the world it is widely distributed and accumulated in soils and seawater compartments especially in regions of significant emissions.

On average the spatial dispersion of air concentrations of this congener is not rather essential. At the same time, due to variability of atmospheric transport pathways, during some episodes PCB-153 can be transported far from the actual emission sources as exemplified by several maps of monthly mean air concentrations for March and July of 2003 in Fig. 55 and 56.

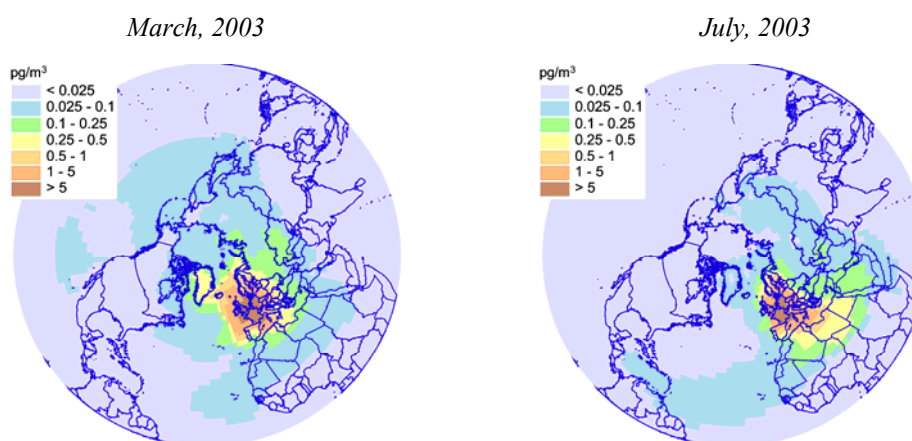


Fig. 55. Spatial distribution of monthly mean air concentrations of PCB-153 (pg/m^3) from European emission sources in March and July of 2003

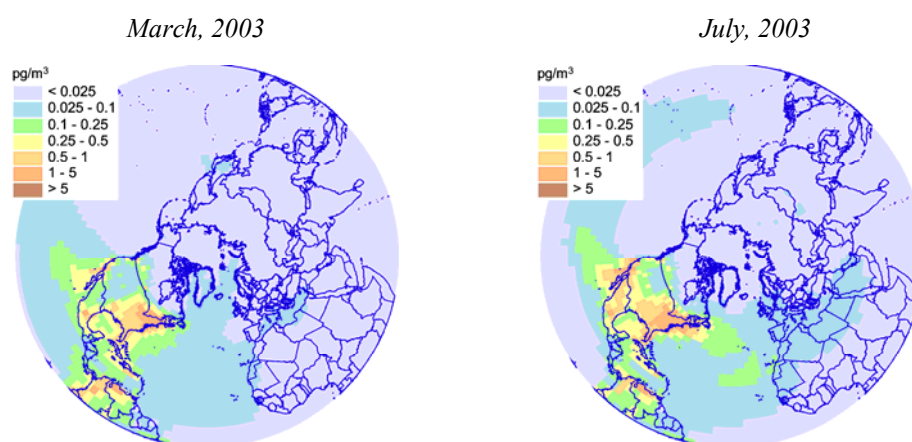


Fig. 56. Spatial distribution of monthly mean air concentrations of PCB-153 (pg/m^3) from emission sources of North America in March and July of 2003

The calculations performed allow to assess the distribution of pollution from various groups of sources over the northern hemisphere. To estimate intercontinental transport of PCB-153 several regions were selected, namely, Europe, America, Russia, Southeast Asia, and Central Asia and Africa. Fig. 57 illustrates the distribution of PCB-153 depositions originated from emission sources of Europe and America for 2003. Particularly, 77% of total PCB-153 depositions over land from European emission sources is deposited over Europe, 12% over Russia, and about 11% in other parts of the northern hemisphere. In case of America major part (92%) of total PCB-153 depositions over land is deposited over its region and 8 % reaches Europe, Russia, and Asia in total.

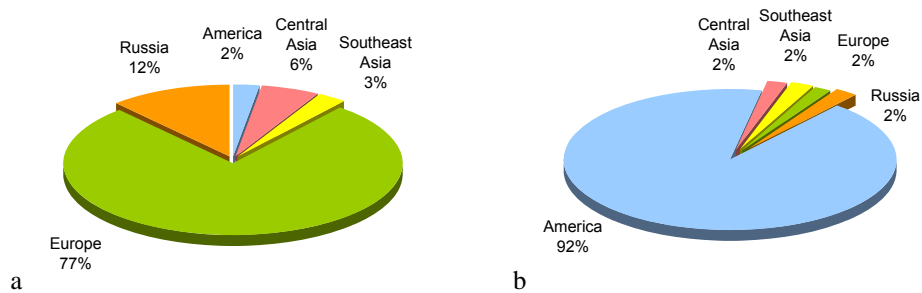


Fig. 57. Distribution of PCB-153 depositions originated from emission sources of Europe (a) and North America (b) in 2003

In Fig. 58 the contributions of selected groups of emission sources to annual PCB-153 depositions over the Arctic region for 2003 are presented. According to modeling results about 5% of the total emission of PCB-153 in 2003 is deposited over the Arctic region. Major contribution to the depositions is made by European emission sources (62%). Significant contributions belong also to Russian (20%) and American (11%) PCB emission sources. It should be mentioned that this distribution is conditioned by the contributions of selected groups of emission sources to the total emission of PCB-153 within the northern hemisphere. In particular, contribution of European emission sources is accounted for about half of the total emission for 2003.

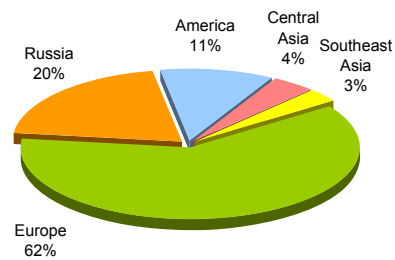


Fig. 58. Contributions of selected groups of emission sources of PCB-153 to depositions over the Arctic region in 2003

5. CO-OPERATION

For the monitoring and model assessment of POP pollution in the EMEP region, the EMEP Centres 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, this year according to the Work-plan on the Implementation of the Convention in 2005 (ECE/EB.AIR/83/add.2, item 2.7) MSC-E continued to support preparatory work of the Task Force on POPs for the review of the Protocol on POPs, and to carry out the POP model intercomparison study. MSC-E participated in international projects such as German Canadian Project "Quality of measuring data on atmospheric inputs of POPs" and 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. Also MSC-E kept on obtaining information possibly helpful for the development of the risk-assessment approach.

5.1. Task Force on POPs

In 2005 MSC-E collaborated with the Task Force on POPs contributing to the preparatory work for the review of the Protocol. MSC-E attended two meetings of the Task Force (informal meeting in Rome, 2005 and third meeting in Vienna, 2005). The main aims of the meetings were to assist the Working Group on Strategies and Review in preparation of the sufficiency and effectiveness review of the Protocol on POPs and to review the dossiers on Penta-BDE and PFOS.

At the request of the Task Force on POPs [EB.AIR/WG.5/2004/1] MSC-E took part in the work on gathering best available scientific information on the effects of POP deposition. Within this work Information Note 5/2004 "EMEP Contribution to the Preparatory Work for the Review of the CLRTAP Protocol on Persistent Organic Pollutants" [Mantseva et al., 2004] was prepared by MSC-E. The model results on atmospheric transport and deposition of POPs presented in this Note are included in the chapter "Best available scientific information on effects of deposition of POPs" of the sufficiency and effectiveness review of the Protocol on POPs.

Besides, the synthesis document on the best available emission data was submitted to the Task Force on POPs. It is devoted to the description of data on emissions of POPs included into Annexes I-III of the Protocol on POPs within a 13-year-long period (1990–2002). The brief review of country submitted emission data is fully included in the Integrated Summary of the components of technical input for reviewing the Protocol on POPs.

To support the work on peer review of dossiers of new substances that may be proposed by Parties for inclusion into annexes to the Protocol, model evaluation of long-range atmospheric transport potential and persistence of PentaBDE was done by MSC-E [Vulykh et al., 2004] and delivered to the Task Force on POPs. This year EMEP/MSCE Information Note 10/2005 "New Substances. Model Assessment of Potential for Long-range Transboundary Atmospheric Transport and Persistence of Endosulfan" [Vulykh et al., 2005] was prepared. In the note the model results are presented for two endosulfan isomers (α - and β -).

To characterize long-range transport potential, model estimates of half-life of the considered endosulfan isomers in the atmosphere (Half-life in air_{calc.}, $T_{1/2}^{air}$) calculated with allowance of all processes removing them from the atmosphere are used. LRTP is also illustrated by Transport Distance (TD) 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 ($T_{1/2}^{env}$) 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 are presented in Table 4.

Table 4. Long-range transport potential and overall persistence of two isomers of endosulfan

Congeners	Long-range transport potential		Overall persistence
	$T_{1/2}^{air}$ (days)	TD (km)	$T_{1/2}^{env}$ (months)
α -endosulfan	1	1621	0.01
β -endosulfan	5	2041	0.16

Due to peculiarities in the physical-chemical properties of two isomers of endosulfan, their ability to the atmospheric transport is somewhat different. According to the calculated values, β -endosulfan possesses higher LRTP and is more persistent isomer than α -endosulfan.

Spatial distributions of air concentrations provide complementary information on the main directions of atmospheric transport and the regions most affected by endosulfan pollution from the conventional emission source. The spatial distribution of air pollution in the EMEP domain by α -endosulfan and β -endosulfan from a point source with conventional intensity located in France (given in relative units) is shown in Fig. 59 a, b, respectively.

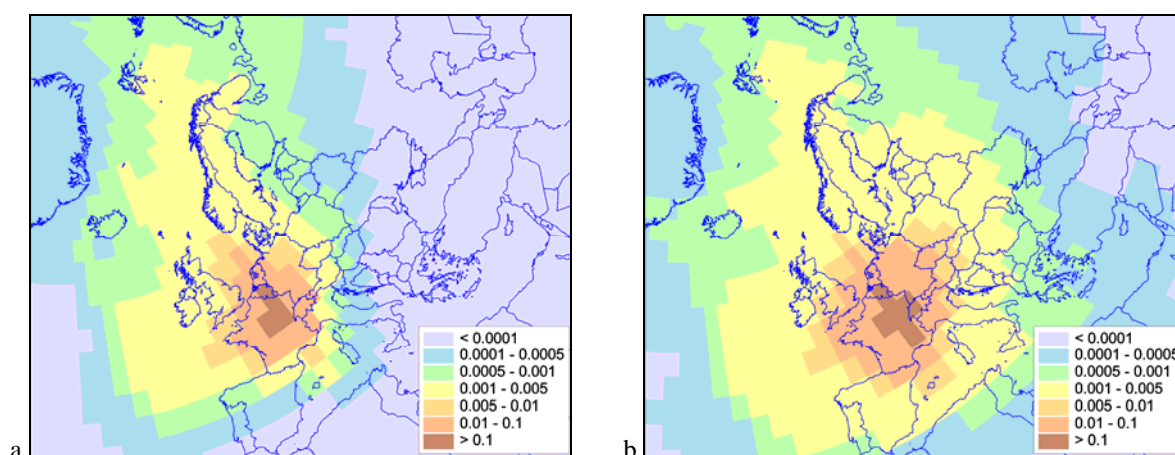


Fig. 59. Spatial distribution of α -endosulfan (a) and β -endosulfan (b) concentrations in the above-ground air in the EMEP domain, relative units

With a source of emissions in Europe (France), the area of atmospheric air contamination by α -endosulfan having concentration 1000 times lower than the initial concentration, covers the entire Central Europe, Scandinavia and approaches Greenland. In case of β -endosulfan the source affects even larger area with the emissions being equal.

Similar information on dicofol is planned to be prepared in 2006.

5.2. Co-operation with national experts

In 2005 MSC-E in co-operation with national experts continued the work on the intercomparison of POP multi-compartment models. The third EMEP expert meeting on intercomparison of POP models (Moscow, February 2005) was attended by 15 experts from Canada, Germany, Japan, the Netherlands, Norway, Switzerland, the United Kingdom, the USA and MSC-E. At the meeting results on computational experiments performed by the participants in the framework of Stage II were discussed in depth and an extended work-programme and time-schedule of Stage III were elaborated. Stage III of the POP model intercomparison study is aimed at the comparison relative order in ranking a number of chemicals with respect to LRTP and P_{over} calculated by different models.

Results of Stage II are presented in the MSC-E Technical Report 4/2005 [Shatalov *et al.*, 2005a]. It is also planned that the final report on the results of the POP model intercomparison study will be prepared in 2006.

MSC-E also keeps on cooperation with national experts in the field of POP measurements from the Czech Republic, Germany, Canada and Norway.

During 2004-2005 MSC-E has participated in the study in the framework of German Canadian Project "Quality of measuring data on atmospheric inputs of POPs". The contribution of MSC-E was focused on evaluation of benzo[a]pyrene (B[a]P) and lindane (γ -HCH) atmospheric transport and deposition from European and North American emission sources and pollution levels within the European and OSPAR regions. Special attention was paid to joint interpretation of the comparison of modeling results with available measurements obtained at EMEP, OSPAR and German sites as well as at Canadian IADN¹ (Great Lakes Region) and NAPS² sites. Evaluation of contamination of OSPAR and European region was carried out using hemispheric multicompartiment MSCE-POP model was used. To obtain more detailed pattern of contamination in the OSPAR region and in Europe, hemispheric calculations were refined by application of regional version of MSCE-POP model with spatial resolution $50 \times 50 \text{ km}^2$. Description of modeling results obtained and their comparison with available measurements are summarized in the report [Gusev *et al.*, 2005b].

Model results provide information on pollution levels of B[a]P and γ -HCH in different regions of northern hemisphere and on contributions of European and North American emission sources to the concentrations and depositions. Spatial distribution of computed annual mean air concentrations of B[a]P and γ -HCH over the northern hemisphere is illustrated in Fig. 60.

Thus major contributions both for B[a]P and γ -HCH air concentrations and depositions over OSPAR Convention waters, in particular, over the Greater North Sea, the Celtic Sea, and the Bay of Biscay regions belong to European emission sources (about 80-90%). At the same time contributions of European and North American emissions sources for the Arctic Waters and the Wider Atlantic sub-regions are comparable. North American sources contribute about 60% to B[a]P depositions in the Wider Atlantic region. For γ -HCH contribution of North American sources in these two sub-regions is about 25%. Other sources of γ -HCH (including Chinese emission sources and re-emission) contribute about 30-45%.

¹ IADN: Integrated Atmospheric Deposition Network.

² NAPS: National Air Pollution Surveillance.

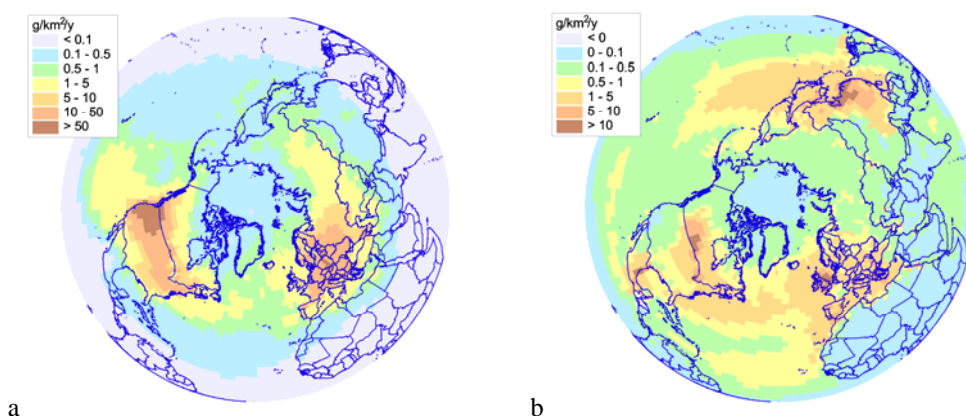


Fig. 60. Spatial distributions of annual deposition of B[a]P (a) and γ -HCH (b) in 2000, $\text{g}/\text{km}^2/\text{y}$

Comparison of model results with measurements has shown reasonable agreement between observed and calculated B[a]P concentrations. For the most of monitoring sites used for the comparison calculated mean annual concentrations agree with measurements within a factor of two. Model predictions of monthly mean concentrations represent main peculiarities of seasonal variations of B[a]P in air and in precipitation.

Comparison of model results with measurement data has shown good agreement for air concentrations of γ -HCH. For the most of measurements used calculated annual mean values agree with measured within a factor of two. Comparison of calculated mean annual concentrations in precipitation with measurements has shown underestimation of observed γ -HCH concentrations on average by a factor of 3. The underestimation of mean annual concentrations is a result of underestimated peak values measured at Knokke (BE4), De Zilk (NL91), Westerland (DE1), Zingst (DE9), and Lista (NO99) occurred in spring time. Possible reason of this underestimation can be connected with uncertainties of lindane emissions, in particular, with uncertain or missing data on application of lindane in some European countries.

5.3. Task Force on Hemispheric Transport of Air Pollution

MSC-E took part in the first meeting of the Task Force on Hemispheric Transport of Air Pollution held in Brussels in June 2005. It informed TFMM participants of the progress of EMEP activity in the field of hemispheric transport of selected persistent organic pollutants. Special attention was paid to the relevant studies of other international organizations such as AMAP, OSPAR, and UNEP.

TFHTAP welcomed the offer of MSC-E to present at the second Task Force meeting information on the results of mercury and POP model intercomparison study, contribution to the protocol reviews, and assessment of intercontinental transport of mercury and selected volatile POPs.

5.4. Working Group on Effects

In the current year MSC-E participated in the 6-th Subregional meeting on effect-oriented activities held by the Working Group on Effects (Moscow, September, 2004). At this meeting MSC-E presented the main results of model assessment of environmental pollution by POPs obtained with the help of multicompartiment transport model MSCE-POP underlining the information on deposition and concentrations of POPs in Eastern European countries. The main emphasis was given to MSCE-POP model estimates that may be of interest for the development of the effect-based/risk assessment approach aiming at the evaluation of POP impact on different ecosystems and human health. The evaluation of POP depositions to various types of underlying surface, accumulation dynamics in different environmental compartments and spatial distribution of POP concentrations in these media was described.

As an example of information potentially useful for risk assessment B[a]P depositions to various types of underlying surface in 2003 are demonstrated in Fig. 61. Calculations show that deposition densities over areas covered by forests (Fig. 61a) are much larger than depositions averaged over grid cells taking into account areas covered by different types of underlying surface. On the opposite, deposition densities for bare soil are essentially lower than average values. This means that, according to calculation results, B[a]P is mainly deposited to forests and this ecosystem accumulates B[a]P in maximum extent.

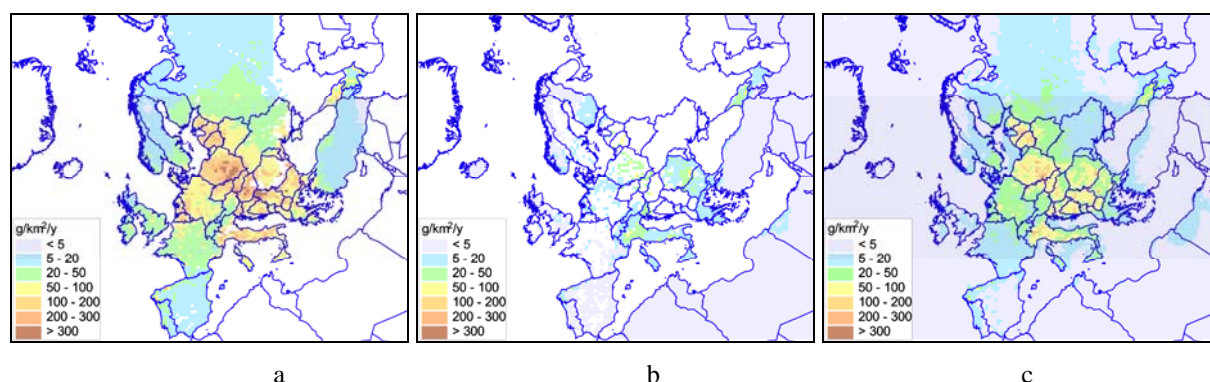


Fig. 61. Densities of B[a]P depositions to different types of underlying surface in 2003, g/km²/y: depositions to coniferous forest (a); depositions to bare soil (b); depositions averaged over grid cells (c)

CONCLUSIONS

During 2004 and 2005 the EMEP Centres continued their work aimed at evaluation of environmental pollution of European region by persistent organic pollutants (PAHs, PCDD/Fs, PCBs, HCB, and HCHs) on the basis of monitoring and modeling. The investigations carried out include collecting and analysis of information on observed levels of concentrations of POPs in air and in precipitation, as well as information on emissions of POPs; modeling of temporal and spatial trends of pollution by POPs on regional and hemispheric scales; evaluation of transboundary transport of POPs between the European countries and intercontinental transport within the northern hemisphere. Particular attention was paid to the development of the MSCE-POP regional model and preparation of the background documents for the EMEP/TFMM Workshop on the review of the MSC-E models to be held in Moscow in October 2005. The main conclusions of the studies are summarized below.

Monitoring of POPs

- Measurements of POP concentrations in air and in precipitation, and of deposition fluxes for 2003 were performed at 12 monitoring sites of EMEP and submitted to CCC. 4 monitoring sites submitted data on parallel measurements of POP concentrations in air and in precipitation. The spatial distribution of monitoring sites across the Europe is non-uniform and requires further improvement.
- Measurements of PCBs for 2003 indicate lowest levels of concentrations in Northern Europe (Sweden, Norway, Finland) and relatively higher levels in Central Europe (the Czech Republic). The tendency of prevailing of lighter congeners in air samples made at northern sites comparing to samples from Central Europe can be noted.

Emission data

- The official data on POP emissions (PAHs, PCDD/Fs, PCBs, HCB) for the period from 1990 to 2003 (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 emissions of POPs and their spatial distribution over 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 2003. 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 18 - 33% depending on particular PAH, and emissions of PCDD/Fs by 63%.

Model review and evaluation

- In course of preparation to the review of MSCE-POP regional model, as a first step, MSC-E has prepared a detailed description of the MSCE-POP regional model and an information on model sensitivity to variations of pollutant-specific and environmental parameters. Uncertainties of model results caused by uncertainties in pollutant-specific data and environmental parameters were estimated. These uncertainties taking into account both pollutant-specific can reach 50% for air concentrations and 70% for depositions. The results of model sensitivity together with the

description of the model are summarized in the EMEP/MSCE Technical Report 5/2005 [Gusev *et al.*, 2005a] and were presented at the sixth TFMM meeting in Zagreb, Croatia in April 2005.

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, and HCB. Trends in pollution of European region by PAHs and PCDD/Fs were estimated for the period 1990-2003. Transboundary transport of B[a]P and PCDD/Fs in 2003 was evaluated. For PCBs and HCB evaluation of pollution levels was performed on the hemispheric scale.
- Annual mean B[a]P deposition flux over the Europe has decreased by 20% on average during the period 1990-2003. Average value of B[a]P annual air concentrations over Europe amounts to 0.2 ng/m³ in 2003 and annual mean deposition flux to 16 g/km²/y. At the same time in some regions of Central and Eastern Europe levels of air concentrations exceed 1 ng/m³.
- Total annual depositions of B[b]F, B[k]F, and I_P in European countries in period 1990-2003 have decreased by 21%, 28%, and 15%, correspondingly. The most pronounced changes in deposition levels can be noted for countries of western and southeastern parts of Europe.
- Following the reduction of emission PCDD/F annual total depositions over Europe have decreased by 47% and annual mean air concentrations by 41% in period 1990-2003. Mean value of PCDD/F annual air concentration over Europe in 2003 is accounted for 2.6 fg TEQ/m³ varying from 0.02 to 15 fg TEQ/m³ for most of European countries. Mean level of annual PCDD/F deposition fluxes is about 0.6 ng TEQ/m²/y.
- The transboundary transport of B[a]P and PCDD/Fs between European countries was evaluated. The contribution of the external sources to air concentrations and depositions of B[a]P and PCDD/Fs is essential and varies typically from 30 to 70%. In case of PCDD/Fs noticeable contribution to air concentrations and depositions over Europe belongs also to the re-emission from underlying surface.
- The POP intercontinental transport of HCB and PCB-153 for 2003 was carried using the hemispheric version of MSCE-POP model. Contributions of selected groups of HCB and PCB emission sources to the contamination of the European region and the Arctic in 2003 were estimated.
- In the framework of co-operation with national experts MSCE-E participated in German Canadian Project "Quality of measuring data on atmospheric inputs of POPs" where it contributed with the study of pollution levels of B[a]P and γ -HCH within the OSPAR region, over Europe, and on the hemispheric scale.
- The development of hemispheric MSCE-POP model was continued during this year with the refinement of its ocean block, in particular, the descriptions of the processes of diffusion, partitioning to particulate organic matter, and sedimentation within the seawater for POPs were refined.

REFERENCES

- Aas W. and K. Breivik [2005] Heavy metal and POP measurements 2003. EMEP/CCC report 9/2005.
- Baart A., Berdowski J., van Jaarsveld J. and K.Wulffraat [1995] Calculation of atmospheric deposition of contaminants on the North Sea. TNO-MEP-R 95/138, Delft, The Netherlands.
- Berdowski J.J.M., Baas J., Bloos J.P.J., Visschedijk A.J.H. and P.Y.J.Zandveld [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, UBA-FB report 104 02 672/03, Apeldoorn, 239 p.
- Breivik K., A. Sweetman, J.M. Pacyna and K.C. Jones [2002] Towards a global historical emission inventory for selected PCB congeners – a mass balance approach. 1. Global production and consumption. *Science of the Total Environment*, v. 290, pp. 181-198.
- Dutchak S., V.Shatalov, E.Mantseva, O.Rozovskaya, N.Vulykh, M.Fedyunin, W.Aas, K.Breivik, S.Mano [2004] Persistent Organic Pollutants in the Environment" EMEP Status Report 3/2004.
- Gusev A., E. Mantseva, V. Shatalov, B.Strukov [2005a] Regional multicompartment model MSCE-POP EMEP/MSCE- Technical Report 5/2005, June 2005.
- Gusev A., Y.-F.Li, E. Mantseva, V.Shatalov, O.Rozovskaya, N.Vulykh [2005b] Evaluation of B[a]P and γ -HCH transport from European and North American emission sources and assessment of deposition to the OSPAR region Report was prepared in the framework of EMEP as a contribution to German Canadian Project "Quality of measuring data on atmospheric input of POPs " EMEP/MSCE-E Technical Report 12/2005. June 2005.
- Jaward F.M., N.J. Farrar, T. Harner, A.J. Sweetman and K.C. Jones [2004] Passive Air Sampling of PCBs, PBDEs, and Organochlorine Pesticides Across Europe. *Environmental Science and Technology*, v.38, pp. 34-41.
- Levinson D.H. (ed) and A.M. Waple (ed) [2005] State of the Climate in 2003. Bulletin of the American Meteorological Society. V.85, No. 6, pp.881-881.
- Mantseva E., S. Dutchak, O. Rozovskaya, V. Shatalov [2004] EMEP Contribution to the Preparatory Work for the Review of the CLRTAP Protocol on Persistent Organic Pollutants. MSC-E Information Note 5/2004.
- Pacyna J.M. et al. [1999] Final report for Project POPCYCLING-Baltic. EU DGXII, Environment and Climate Program ENV4-CT96-0214. Available on CD-rom including technical report, the emission and environmental databases as well as the POPCYCLING-Baltic model. NILU, P.O. Box 100, N-2027 Kjeller, Norway.
- Shatalov V., E. Mantseva, A. Baart, P. Bartlett, K. Breivik, J. Christensen, S. Dutchak, S. Gong, A. Gusev, K. M. Hansen, A. Hollander, P. Huang, K. Hungerbühler, K. Jones, G. Petersen, M. Roemer, M. Scheringer, J. Stocker, N. Suzuki, A. Sweetman, D. van de Meent, F. Wegmann [2005a] POP model intercomparison study. Stage II. Comparison of mass balance estimates and sensitivity studies. EMEP/MSCE-E Technical Report 4/2005. June 2005.
- Shatalov V., A. Gusev, S. Dutchak, I.Holoubek, E. Mantseva, O. Rozovskaya, A.Sweetman, B. Strukov, N. Vulykh [2005b] Modelling of POP contamination in European region: Evaluation of the model performance. EMEP/MSCE-E Technical Report 7/2005, June 2005.
- Shatalov V., M.Fedyunin, E.Mantseva, B.Strukov, N.Vulykh [2003] Persistent Organic Pollutants in the Environment. MSC-E Technical Report 4/2003. June 2003.
- Taniysau S., K. Kannan, I. Holoubek, A. Ansorgova, Y. Horii, N. Hanari Nyamashita and K.M. Aldous [2003] Isomer-specific analysis of chlorinated biphenyls, naphthalenes and dibenzofurans in Delor: polychlorinated biphenyl preparations from the former Czechoslovakia. *Environmental Pollution*, v.126, pp. 169-178.
- Tsibulsky V., V. Sokolovsky, S. Dutchak and V. Shatalov [2001] MSC-E contribution to the HM and POP emission inventories. Technical Note 7/2001. June 2001.
- Vulykh N., E. Mantseva, V. Shatalov [2005] EMEP contribution to the Preparatory Work for the Review of the CLRTAP Protocol on POPs. New Substances: Model Assessment of Potential for Long-range Transboundary Atmospheric Transport and Persistence of Endosulfan. MSC-E Information Note 10/2005. May 2005.
- Vulykh N., S.Dutchak, E. Mantseva, V. Shatalov [2004] EMEP contribution to the Preparatory Work for the Review of the CLRTAP Protocol on POPs. New Substances. Model Assessment of Potential for Long-range Transboundary Atmospheric Transport and Persistence of PentaBDE. MSC-E Information Note 10/2004. December 2005.
- Wania F and Y. Su [2004] Quantifying the Global Fractionation of Polychlorinated Biphenyls. *Ambio*, v.33, pp. 161-168.
- Wania F. and C.B. Dugani [2003] Assessing the Long-Range Transport Potential of Polybrominated Diphenyl Ethers: A Comparison of Four Multimedia Models. *Environmental Toxicology and Chemistry*, v. 22, pp. 1252-1261.

EMEP WORK-PLAN on POPs for 2005

2.2. Atmospheric measurements and modelling

Description/objectives: Assess the results of implementing the protocols to the Convention, provide the measurement and modelling tools necessary for further international air pollution abatement policies, including the review of protocols, and ensure support for the compilation and evaluation of information on transboundary air pollution. The Task Force on Measurements and Modelling, led by the United Kingdom and co-chaired by WMO, with the assistance of the centres, supports the Steering Body and its Bureau by: (i) reviewing and assessing the scientific and operational activities of EMEP related to monitoring and modelling; (ii) evaluating their contribution to the effective implementation and further development of the protocols; and (iii) drawing up specific proposals. It provides for closer collaboration among the Parties to the Convention, the centres, other bodies under the Convention, other international bodies and the scientific community in strengthening scientific communication and cooperation in air pollution monitoring and modelling.

Main activities and time schedule:

(c) ... CCC, in cooperation with MSC-E and MSC-W, will continue to examine approaches to combine modelling data with observations. ...

(d) The Task Force will, in close collaboration with experts from Parties and the EMEP centres, prepare for the evaluation of the MSC-E modelling of heavy metals and POPs. MSC-E will host a workshop on the review in autumn 2005;

2.7. Persistent organic pollutants (POPs)

Description/objectives: Improve the monitoring and modeling data on concentrations, depositions and transboundary fluxes of selected POPs. Study further the physical-chemical processes of POPs in different environmental compartments, taking into account their transport within the EMEP region and on the hemispheric/global scale. Develop reliable emission data for the POPs listed in the Protocol, as well as a preliminary data set for other substances. Support preparatory work for the review of the Protocol on POPs, and in particular the Task Force on POPs.

Main activities and time schedule:

(a) MSC-E will prepare information for 2002 on: evaluation of PAHs (benzo[a]pyrene (BaP), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF) and indeno[1,2,3-cd]pyrene) and PCDD/Fs concentration and deposition fields; evaluation of transboundary transport of BaP in 2002 and pilot assessment of source-receptor relationships for all toxic congeners of PCDD/Fs (country-to-country matrices); trend analysis of environmental contamination by four indicator PAHs and PCDD/Fs in the EMEP domain (1990-2002); and an assessment of the hemispheric pollution by PCBs and HCB. It will analyse contributions from emission sources of the Northern hemisphere to the contamination of the European region in 2000 and the contribution from European sources to the contamination of different regions;

(b) For the model review, MSC-E will evaluate uncertainties in modelling, measurement and emission data, in cooperation with CCC and the Task Force on Emission Inventories and Projections;

(c) MSC-E will further develop its models with respect to: the redistribution between different phases and sedimentation in the marine environment; the gas/aerosol partitioning process in the atmosphere; and the distribution in the atmosphere taking into account spatial and temporal variations of OH radical concentrations. It will complete the second stage of the model intercomparisons and prepare the third stage;

(d) In cooperation with MSC-E, CCC will complement EMEP monitoring data with data from other international and national programmes. It will evaluate data quality and data representativity, and, in cooperation with MSC-E, will compare the observations with model estimates. Both centres will cooperate with UNEP to harmonize the global POPs monitoring strategy with the EMEP strategy;

(e) CCC and MSC-E, in consultation with the Task Force on Emission Inventories and Projections and with Parties, will improve the POPs emission data quality with specific emphasis on PAHs, PCDD/Fs, PCBs and HCB. They will adjust European emission inventories for POPs to the modelling requirements. CCC will develop profiles of chemical species of the selected POPs and collate information on the height of major point sources. It will perform screening studies for new substances.

MODEL MODIFICATION: DESCRIPTION OF POP BEHAVIOR IN THE MARINE ENVIRONMENT

The global ocean is one of the most accumulative compartments for many of POPs distributed in the environment on a planetary scale. Due to various physical processes POPs are transported horizontally and vertically within the seawater, accumulated in bottom sediments, move from the atmosphere to the ocean and back. The following most significant processes (Fig. B.1) affect concentration of POPs in the global ocean:

- ocean horizontal and vertical currents,
- three-dimensional large-scale turbulent diffusion,
- redistribution of POPs between the dissolved phase and phases adsorbed on various suspended particles and dissolved organic compounds (Fig. B.2),
- sedimentation with particles,
- degradation of POP molecules.

In Polar regions the POP ocean-atmosphere exchange is to a large extent affected by sea ice cover.

Because of the importance of the processes occurring near the ocean-atmosphere boundary, the upper mixed ocean layer with the depth varying in time and space should be taken into account.

To describe the concentration field of lipophilic POPs (e.g. PCB-153) values of specific organic carbon concentrations in the marine environment are required. Three-dimension sets of the particulate organic matter (POM) concentrations and sedimentation rates have been computed from the model of sedimentation of particles subjected to coagulation and degradation. The annual cycle of concentrations and particle rates has been derived from the data on primary productions in the photic layer. Characteristics obtained for POM are in a qualitative agreement with measurements. Non-uniform distribution of the POM flow results in the convergence of POP fluxes and growth of POP depositions at the bottom in the points of increased sedimentation. For the most part such zones are located in coastal fishery areas.

The proposed physical processes are taken into consideration in the ocean block of the hemispheric MSCE-POP multicompartment transport model.

Several experiments with the refined ocean block of the MSCE-POP model were carried out. The role of sedimentation processes in the PCB-153 distribution dynamics has been studied in the following two experiments. In the first experiment the flux of PCB-153 from the atmosphere was assumed to be constant and

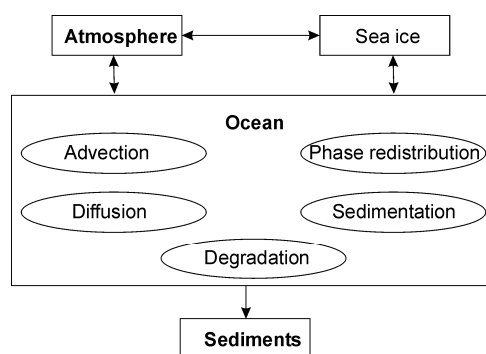


Fig. B.1. Block-scheme of the POP ocean transport model

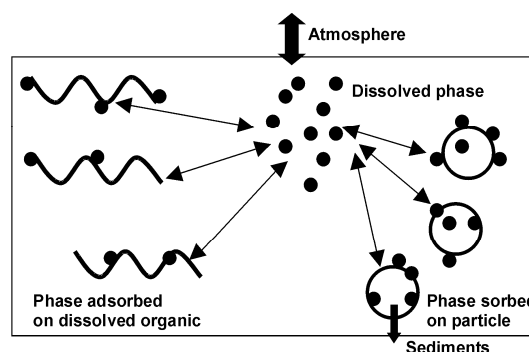


Fig. B.2. Phases of POPs in the marine environment

uniform over the whole sea area of the Northern Hemisphere. As a result of model run for 10 year period the distribution of the pollutant flux to the bottom appeared to be significantly non-uniform (Fig. B.3.).

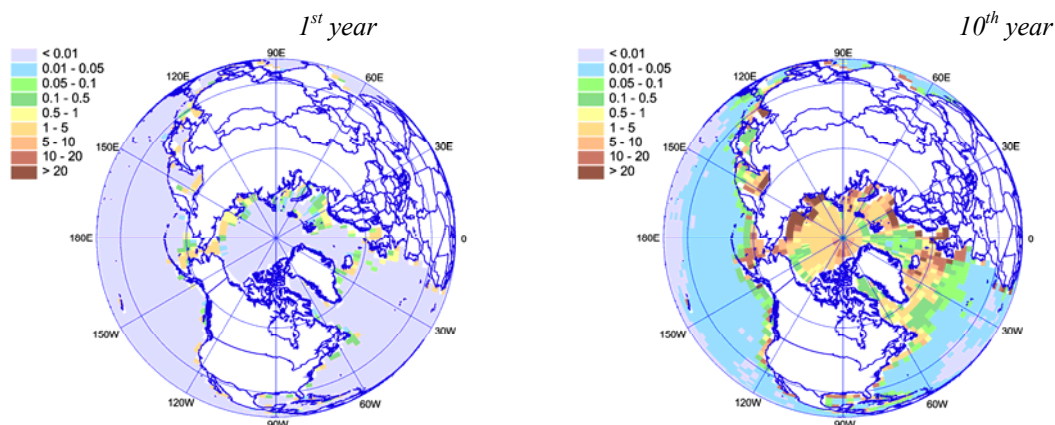


Fig. B.3. Values of the PCB-153 fluxes to sediments/PCB-153 fluxes to the ocean through the upper surface ratio (f_n)

From these figures it is seen that PCB-153 deposited to the bottom sediments mainly in the coastal zones and in relatively shallow water areas. The map of PCB -153 deposition correlates with the distribution of the organic matter flow to the bottom of the ocean (B.4 b).

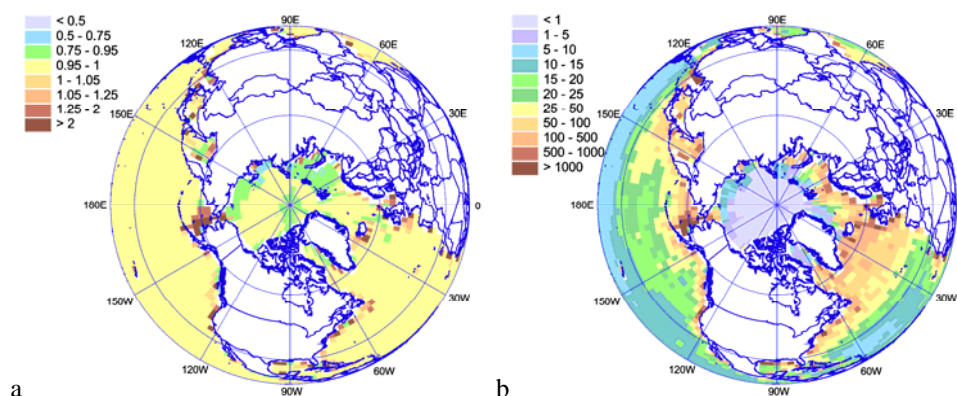


Fig. B.4. Distribution of the accumulated PCB-153 sedimentation fluxes with dynamics/ accumulated PCB-153 sedimentation fluxes without any dynamics ratio in the 10th year of the experiment (a) and field of mean annual organic carbon flow to the bottom of the ocean computed by the model, mmol OC/m²/y

In the second experiment the dynamics of the PCB-153 distribution in the ocean with the non-zero initial concentration and zero flux to the ocean from other environmental compartments has been analyzed. When organic particles in the upper layers of the ocean are formed some of the PCB-153 in dissolved phase is sorbed on the particles and sediments to the bottom. The organic matter productivity, concentration and flow to the ocean are essentially non-uniform (Fig. B.4b). Areas with the increased productivity are characterized by the PCB-153 convergence as it is shown in Fig B.4a. It is shown that zones of PCB-153 deposition to the bottom of the ocean are the zones of PCB-153 collection (horizontal convergence) from the adjacent ocean areas. These zones represent sinks of PCB-153 concentrations. Such pattern of the PCB-153 redistribution in the ocean is

worth noting since coastal and shallow water zones with the intensive flow of organic matter to the bottom of the ocean are normally areas of fishery.

The operating capacity of the model was also assessed in the experiment with the data of global emission inventory of PCBs [Breivik *et al.*, 2002]. Following the dynamics of emissions the concentration in the upper 100 m layer of the ocean was increasing from 1970 to 1985 (Fig. B.5). After 1985 it was decreasing (Fig. B.5). As for the horizontal distribution of the PCB-153 concentrations maximum values were noted in the region of the North Sea. Comparatively high concentrations were also obtained in the Arctic Ocean area.

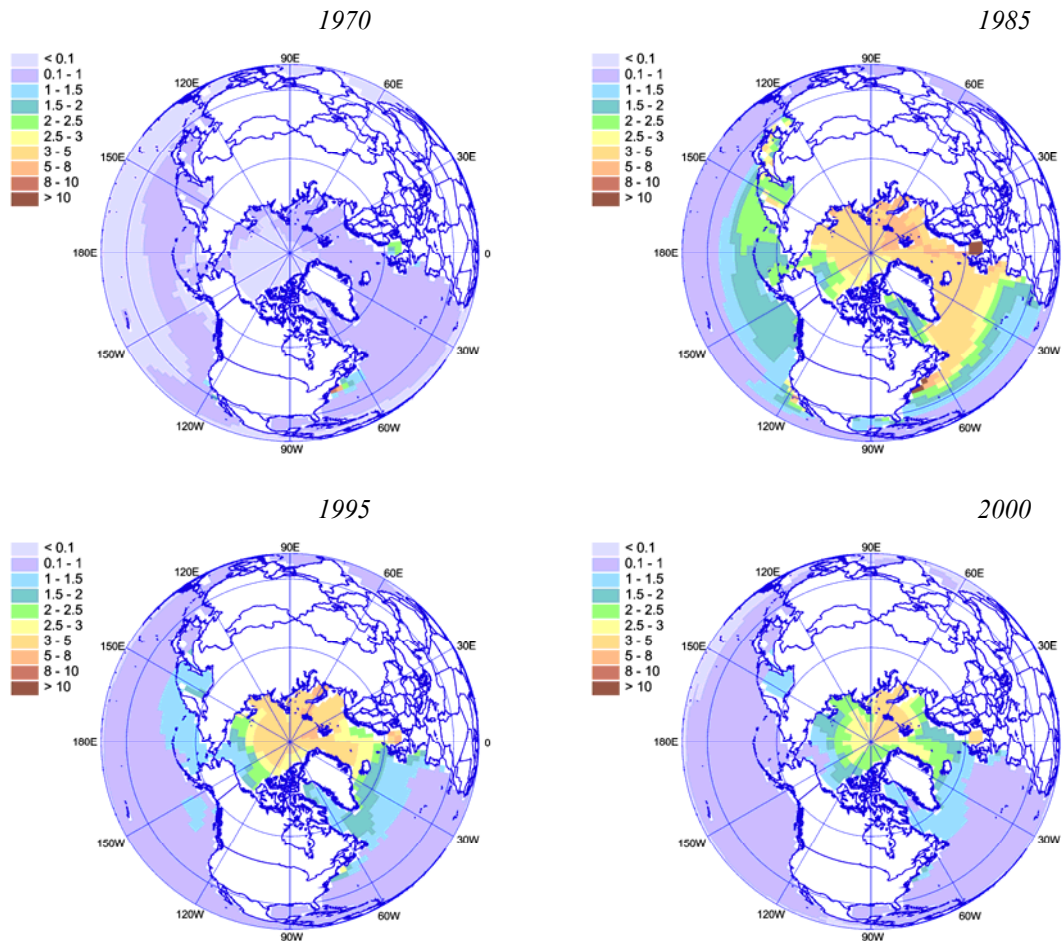


Fig. B.5. Values of C_o in the upper 100 m layer, ng/m^3

The computed data for the PCB-153 concentration in the marine environment were compared with available measurements. On average, the agreement between the calculation data and observed values concentrations in seawater is within the order of magnitude. The deviations of calculated data can be explained by the sufficiently high degree of averaging in the model calculations whereas measurements normally are performed locally under specific hydrometeorological conditions. Additional uncertainties may also be introduced by the shortcomings of the modeling approach.

COUNTRY-TO-COUNTRY DEPOSITION MATRICES FOR 2003

Table C.1. Codes of countries

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 C.2. Matrix of B[a]P country-to-country depositions in 2003, kg/y

Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	
AL	47	0.1	3.2	0.3	16	1.3	43	3.0	1.2	231	0.004	5.9	6.9	0.9	0.7	AL
AM	0.02	33	0.1	29	0.3	0.1	0.9	0.8	0.1	0.8	0.01	0.3	0.6	0.2	0.2	AM
AT	0.2	0.03	703	0.2	10	24	3.3	8.6	61	20	0.001	204	259	9.0	3.0	AT
AZ	0.1	15	0.5	429	1.1	0.5	3.5	4.4	0.3	3.3	0.024	1.3	2.5	0.8	1.0	AZ
BA	1.9	0.1	28	0.3	1445	5.5	23	8.2	5.2	319	0.003	42	36	4.3	2.3	BA
BE	0.01	0.003	1.7	0.02	0.4	630	0.2	0.6	3.1	0.6	0.000	4.0	86	3.7	0.6	BE
BG	2.3	0.3	13	1.9	42	5.5	2100	28	3.8	390	0.015	30	32	5.2	5.2	BG
BY	0.4	0.2	15	1.1	12	14	22	2811	6.1	38	0.003	60	87	32	67	BY
CH	0.05	0.004	11	0.0	1.6	7.3	0.6	0.7	316	2.1	0.000	6.1	44	1.9	0.6	CH
CS	7.8	0.1	28	0.7	330	7.0	194	14	6.0	3633	0.006	53	45	5.4	3.5	CS
CY	0.01	0.03	0.1	0.1	0.1	0.05	0.4	0.2	0.0	0.4	0.732	0.1	0.2	0.04	0.03	CY
CZ	0.1	0.02	113	0.2	10	21	4.5	11	23	29	0.001	1668	302	12	3.4	CZ
DE	0.1	0.04	139	0.3	5.5	388	3.6	17.4	231	12	0.002	491	5312	98	10	DE
DK	0.01	0.004	1.6	0.0	0.4	12	0.2	2.2	1.3	0.8	0.000	9.2	58	243	1.0	DK
EE	0.04	0.02	2.9	0.1	1.7	6.0	2.3	66	1.4	4.7	0.001	12	30	16	823	EE
ES	0.2	0.02	5.1	0.1	5.2	6.1	2.8	2.1	5.3	7.8	0.001	7.4	16	1.9	0.8	ES
FI	0.2	0.1	11	0.6	6.4	28	8.7	94	6.8	18	0.002	41	117	65	191	FI
FR	0.6	0.03	29	0.2	19	173	8.6	7	132	26	0.003	48	339	20	5.1	FR
GB	0.1	0.01	2.6	0.1	1.3	57	0.9	2.2	3.8	2.3	0.001	8	66	9	1.1	GB
GE	0.2	14	0.8	46	1.9	0.7	6.3	5.8	0.4	5.9	0.035	2.2	3.6	1.1	1.1	GE
GR	12	0.3	6.7	1.6	21	3.2	289	12	2.4	133	0.035	13	17	2.6	2.4	GR
HR	1.0	0.1	66	0.2	232	5.7	12	9.3	6.7	177	0.002	61	43	4.4	2.5	HR
HU	0.5	0.05	115	0.3	53	8.2	21	17	8.3	247	0.001	134	69	6.1	3.4	HU
IE	0.01	0.0	0.5	0.02	0.3	5.9	0.2	0.4	0.8	0.5	0.000	1.0	7.7	1.2	0.2	IE
IS	0.01	0.0	0.4	0.01	0.3	2.6	0.2	0.3	0.6	0.5	0.000	1.0	5.2	1.1	0.1	IS
IT	5.0	0.1	101	0.6	119	22	42	14	85	153	0.012	62	102	10	4.7	IT
KZ	0.2	0.9	2.2	12	3.6	2.9	10	31	1.1	11	0.014	6.7	13	4.8	8.2	KZ
LT	0.1	0.03	6.4	0.2	3.6	10	4.6	223	2.9	11	0.001	29	56	27	31	LT
LU	0.0	0.00	0.3	0.003	0.04	12	0.03	0	0.6	0.1	0.000	0.7	12.2	0.3	0.1	LU
LV	0.1	0.03	5.4	0.2	3.0	10	4.0	185	2.6	8.9	0.001	24	55	27	131	LV
MC	0.0	0.00	0.003	0.0	0.002	0	0.001	0	0.0	0.002	0.000	0.003	0.005	0.001	0.000	MC
MD	0.1	0.05	1.1	0.3	2.2	0.9	10	18	0.4	8.4	0.001	3.8	5.4	1.6	1.9	MD
MK	8.6	0.1	3.9	0.3	16	1.4	126	3.4	1.1	278	0.004	7.9	8.1	1.1	0.8	MK
NL	0.01	0.002	1.2	0.02	0.3	136	0.2	0.6	1.6	0.5	0.000	4.2	145	5.4	0.6	NL
NO	0.05	0.02	4.8	0.1	2.0	33	1.7	12	4.6	4.2	0.001	19	102	67	6.8	NO
PL	0.6	0.1	92	0.8	35	66	26	250	31	103	0.003	768	697	122	35	PL
PT	0.01	0.003	0.7	0.02	0.6	1.1	0.3	0.4	0.8	0.7	0.000	1.1	2.7	0.2	0.1	PT
RO	2.8	0.4	38	2.3	113	15	346	85	11	616	0.008	86	91	15	14	RO
RU	2.7	6.7	81	61	83	120	149	2126	42	236	0.078	274	575	243	1124	RU
SE	0.1	0.04	14	0.3	5.6	59	6.5	49	9.4	15	0.003	61	248	285	34	SE
SI	0.1	0.01	79	0.1	10	2.6	2.1	3.6	4.5	15	0.001	24	21	1.7	1.0	SI
SK	0.4	0.04	54	0.3	23	7.8	13	20	7.1	71	0.001	174	64	7.6	4.2	SK
TR	1.7	23	9	33	21	7.8	176	57	4.0	79	0.528	25	38	10	10	TR
UA	1.4	0.9	36	5	52	22	96	673	12	171	0.015	111	141	39	48	UA
	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	

Table C.2. Matrix of B[a]P country-to-country depositions in 2003, kg/y (continued)

Receptors ↓ Emitters →

	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	
AL	5.3	0.5	5.6	1.4	4.3	20	4.7	14	1.1	0.01	80	0.8	4.8	0.0	1.5	AL
AM	0.3	0.1	0.3	0.2	138	0.5	0.1	0.4	0.2	0.001	2.5	1.1	1.0	0.0	0.3	AM
AT	17	2.6	65	14	1.7	0.9	12	72	7.7	0.08	257	0.8	21	0.0	6.9	AT
AZ	1.5	0.6	1.4	1.0	370	1.8	0.5	1.5	0.8	0.01	9.2	14	5.8	0.0	2.1	AZ
BA	11	1.7	17	4.8	5.1	6.0	138	149	3.4	0.03	206	1.5	16	0.0	5.0	BA
BE	15	0.6	119	20	0	0.1	0.3	0.6	7.8	0.04	9.5	0.1	2.3	0.0	1.0	BE
BG	10	3.1	15	6.3	26	46	14	65	4.4	0.05	99	7.0	36	0.0	12	BG
BY	15	25	25	17	14	2.9	7.0	46	11.0	0.13	58	7.5	1537	0.0	317	BY
CH	16	0.5	96	7.1	0.3	0.2	1.8	2.9	4.3	0.03	261	0.1	2.5	0.0	1.0	CH
CS	13	2.4	21	7	10	18	79	211	4.5	0.05	199	2.6	24	0.0	7.7	CS
CY	0.1	0.0	0.2	0.1	1.0	0.4	0.1	0.2	0.1	0.00	1.4	0.1	0.2	0.0	0.1	CY
CZ	13	3.1	48	14	1.5	0.7	9.0	86	7.2	0.06	67	0.8	26	0.0	8.1	CZ
DE	96	11	537	125	2.8	1.0	4.1	25	55	0.41	162	1.3	56	0.0	20	DE
DK	7.1	1.3	13	15	0.3	0.1	0.2	1.7	7.1	0.06	4.3	0.1	8.0	0.0	2.3	DK
EE	6	56	9.1	10	1.5	0.4	0.9	5.8	5.6	0.07	9.0	0.8	275	0.0	277	EE
ES	1909	0.7	89	8.8	0.8	0.8	3.8	7.4	12	0.12	101	0.4	4.1	0.0	1.5	ES
FI	29	1471	46	48	4.9	1.4	3.4	19	31	0.42	44	2.4	322	0.0	164	FI
FR	552	5.0	3426	80	2.0	3.2	14	24	61	0.33	639	0.8	22	0.0	8.6	FR
GB	66	1.0	89	550	0.7	0.3	0.8	2.0	160	0.37	24	0.3	6.6	0.0	2.2	GB
GE	2.1	0.7	2.2	1.3	2059	3.1	0.8	2.7	1.1	0.01	15	4.2	6.9	0.0	2.4	GE
GR	10	1.4	11	3.7	23	414	7	25	2.8	0.03	116	4.5	17	0.0	5.4	GR
HR	9	1.9	18	4.6	3.2	3.2	481	283	3.2	0.03	234	1.1	19	0.0	5.7	HR
HU	9	2.3	21	6.4	3.1	2.9	79	2180	3.9	0.04	133	1.1	29	0.0	8.3	HU
IE	18	0.2	15	22	0.1	0.1	0.2	0.4	267	0.10	5.0	0.1	1.1	0.0	0.4	IE
IS	6	0.1	6.0	5.1	0.1	0.1	0.2	0.5	6.0	3.80	4.5	0.03	1.0	0.0	0.2	IS
IT	73	3.7	139	19	10	26	98	123	14	0.11	8760	2.8	30	0.0	10	IT
KZ	5.3	5.3	6.1	4.8	53	3.2	1.6	6.6	3.7	0.04	22	1290	38	0.0	15	KZ
LT	10	13	16	12	2.6	0.8	2.1	14	7.2	0.09	19	1.2	4069	0.0	296	LT
LU	1.3	0.1	18	1.1	0.02	0.01	0.03	0	0.5	0.00	1.2	0.01	0.3	0.0	0	LU
LV	10	25	16	14	2.5	0.7	1.7	12	8	0.09	15	1.3	1632	0.0	1628	LV
MC	0.004	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.00	0.2	0.0	0.001	0.0	0.0	MC
MD	1.2	1.0	1.9	1.3	3.4	0.8	0.8	4.6	0.9	0.01	8.2	1.8	16	0.0	4.7	MD
MK	3.7	0.5	4.7	1.5	4.2	25	4.8	19	1.1	0.01	49	0.9	5.5	0.0	1.8	MK
NL	11	0.6	45	22	0.1	0.1	0.2	0.5	7.5	0.05	5.6	0.1	2.4	0.0	1.0	NL
NO	36	12	49	82	1.3	0.4	1.2	5.9	55	0.68	21	0.5	46	0.0	15	NO
PL	44	25	112	61	9.2	3.8	24	198	34	0.32	181	4.8	475	0.0	95	PL
PT	147	0.1	7.1	1.3	0.1	0.1	0.5	1.0	1.6	0.02	10	0.1	0.7	0.0	0	PT
RO	22	8.2	38	16	29	22	41	266	11	0.10	237	11	107	0.0	32	RO
RU	156	808	224	197	726	28	41	221	134	1.77	433	857	3170	0.0	1572	RU
SE	46	76	78	98	2.9	1.1	3.6	19	57	0.63	49	1.3	207	0.0	69	SE
SI	4.3	0.8	9.2	2.0	1.0	0.6	58	52	1.3	0.01	171	0.4	7.7	0.0	2	SI
SK	7.4	2.8	18	6.4	2.7	1.8	18	268	3.9	0.04	84	1.0	36	0.0	10	SK
TR	18	6.0	20	12	444	55	8.4	35	8.9	0.09	139	22	65	0.0	21	TR
UA	26	24	44	28	68	14	27	214	19	0.18	183	48	454	0.0	124	UA
	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	

Table C.2. Matrix of B[a]P country-to-country depositions in 2003, kg/y (continued)

Receptors ↓ Emitters →

	MC	MD	MK	NL	NO	PL	PT	RO	RU	SK	SI	SE	UA	Total, kg/y	
AL	0.0	0.9	82	0.9	0.8	31	0.6	55	3.0	9.0	2.9	1.2	15	707	AL
AM	0.0	0.1	0.2	0.1	0.2	3.4	0.04	2.6	3.7	0.4	0.1	0.2	4.3	228	AM
AT	0.0	0.4	1.1	13	6.5	242	1.9	24	3.4	121	78	9.1	13	2297	AT
AZ	0.0	0.4	0.7	0.5	0.9	14	0.2	10	34	1.4	0.5	1.3	20	958	AZ
BA	0.0	1.2	7.1	3.6	3.4	174	1.2	119	4.6	69	33	5.0	28	2934	BA
BE	0.0	0.0	0.1	37	3.1	12	1.8	1.3	0.3	1.1	0.4	2.7	0.8	968	BE
BG	0.0	13	58	4.2	4.7	186	1.1	852	28	48	10	7.7	160	4376	BG
BY	0.0	9.3	3.5	12	26	1275	2.0	152	121	64	10	55	467	7444	BY
CH	0.0	0.1	0.2	2.8	1.9	16	1.6	3.0	0.4	3.1	5.2	1.8	1.3	823	CH
CS	0.0	3.4	125	4.9	4.3	227	1.5	419	8.9	90	23	6.9	57	5898	CS
CY	0.0	0.0	0.1	0.03	0.04	1.0	0.0	0.9	0.4	0.2	0.1	0.1	0.9	10	CY
CZ	0.0	0.4	1.1	14.8	6.6	957	1.5	26	3.9	233	18	12	15	3770	CZ
DE	0.0	0.6	0.9	263	41	763	12	29	7.3	52	8.4	59	22	9064	DE
DK	0.0	0.0	0.1	14	15	54	1.2	1.7	0.7	3.1	0.4	23	2.0	505	DK
EE	0.0	0.6	0.4	6.0	18	163	1.0	14	23	8.8	1.2	57	21	1935	EE
ES	0.0	0.2	1.0	3.1	4.1	32	128	10	1.2	7.2	7.0	2.0	4.8	2401	ES
FI	0.0	1.7	1.6	26	134	424	4.7	47	75	27	5.1	354	57	3932	FI
FR	0.0	0.5	3.7	42	23	142	42	32	3.7	25	28	19	13	6017	FR
GB	0.0	0.1	0.3	38	13	31	11	4.9	1.1	2.9	1.3	6.2	3.4	1168	GB
GE	0.0	0.8	1.3	0.6	1.2	22	0.3	18	32	2.5	0.8	1.6	30	2303	GE
GR	0.0	4.3	139	2.2	2.4	77	1.2	161	18	18	5.7	3.5	77	1668	GR
HR	0.0	0.7	4.1	3.7	3.4	227	1.0	76	3.9	109	238	5.3	24	2386	HR
HU	0.0	0.9	5.2	5.8	4.3	442	1.0	276	5.3	561	78	6.8	60	4608	HU
IE	0.0	0.0	0.1	3.4	2.0	4.7	3.7	0.9	0.2	0.5	0.3	0.9	0.7	365	IE
IS	0.0	0.0	0.1	1.7	2.7	5.5	1.0	0.7	0.1	0.6	0.3	0.8	0.5	60	IS
IT	0.0	1.8	21	11	8.6	268	8.5	120	9.4	87	173	11	43	10793	IT
KZ	0.0	1.7	1.7	2.6	6.0	75	0.9	40	231	6.8	1.7	9.2	109	2049	KZ
LT	0.0	1.4	0.9	9.2	19	581	1.4	30	28	22	3.1	46	49	5659	LT
LU	0.0	0.0	0.0	0.8	0.2	1.6	0.1	0.2	0.04	0.2	0.1	0.3	0.1	52	LU
LV	0.0	1.1	0.7	10	23	394	1.6	25	29	18	2.5	64	42	4434	LV
MC	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.003	0.0	0.003	0.006	0.001	0.001	0.3	MC
MD	0.0	66	1.0	0.8	1.3	64	0.2	182	7.8	5.2	0.8	2.2	128	561	MD
MK	0.0	1.1	547	1.0	0.9	38	0.4	65	3.2	11.7	2.9	1.4	17	1268	MK
NL	0.0	0.02	0.1	301	3.8	13	1.5	1.1	0.3	1.0	0.3	3.3	0.8	717	NL
NO	0.0	0.3	0.4	33	690	127	6.6	9.4	4.7	8.7	2.1	100	10	1575	NO
PL	0.0	4.1	5.8	58	53	17224	6.0	170	39	511	40	109	248	21963	PL
PT	0.0	0.0	0.1	0.5	0.6	4.9	347	1.3	0.2	1.0	0.8	0.3	0.8	536	PT
RO	0.0	52	31	11	13	680	2.7	6655	43	174	32	20	428	10420	RO
RU	0.0	41	24	112	341	3535	24	836	9393	267	52	624	2235	31177	RU
SE	0.0	1.2	1.2	62	370	556	7.8	33	15	30	6	1683	36	4297	SE
SI	0.0	0.2	0.7	1.6	1.4	85	0.5	17	1.5	38	512	2.2	7.3	1143	SI
SK	0.0	0.8	3.3	5.8	5.2	895	0.9	94	5.5	1478	29	8.6	51	3485	SK
TR	0.0	13	16	6.4	10	235	2.2	319	126	33	8.0	13	344	2477	TR
UA	0.0	72	14	19	33	1941	3.4	900	352	221	29	56	4482	10803	UA
	MC	MD	MK	NL	NO	PL	PT	RO	RU	SK	SI	SE	UA	Total, kg/y	

Table C.3. Matrix of PCDD/Fs country-to-country depositions in 2003, g TEQ/y
 Receptors ↓ Emitters →

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	
AL	1149	15	69	13	123	40	2219	15	35	1849	1.6	163	157	25	1.2	AL
AM	0.4	9966	2.7	1926	2.3	4.5	56	5.0	2.0	6.5	4.8	11	15	7.5	0.4	AM
AT	2.0	5.3	18386	6.5	62	591	113	33	1496	111	0.3	6110	5714	227	4.8	AT
AZ	1.4	3651	12	30387	10	22	238	29	8.8	28	12	46	72	39	2.5	AZ
BA	23	14	431	13	12947	135	743	29	113	2028	0.9	947	670	102	3.4	BA
BE	0.1	0.4	33	0.6	2.0	37103	7.2	2.4	83	2.9	0.0	94	2088	97	1.0	BE
BG	35	65	223	72	284	146	174154	116	89	2859	3.5	696	611	145	8.1	BG
BY	3.9	21	195	31	62	254	789	14671	97	187	0.9	1151	1255	634	95	BY
CH	0.4	0.6	185	0.7	8.6	173	22	2.3	11655	10	0.1	140	1100	47	0.9	CH
CS	128	22	414	23	2409	153	8134	48	115	34058	1.5	1141	764	124	5.0	CS
CY	0.4	13	2.4	8.4	2.0	3.5	52	2.3	2	6.2	1436	8.0	11	3.4	0.2	CY
CZ	1.5	5.2	2476	7	64	556	145	47	548	164	0.3	59407	7387	299	5.6	CZ
DE	1.4	7.6	3053	11	32	11568	135	70	6898	70	0.8	13667	156256	2852	17	DE
DK	0.1	1.2	34	1.9	3.0	430	10	11	35	5.9	0.1	268	1464	13084	2.1	DK
EE	0.4	2.2	35	3.4	7.3	113	73	210	22	19	0.1	203	417	339	1674	EE
ES	2.0	2.9	99	4.4	36	240	138	10	169	52	0.5	205	442	65	1.6	ES
FI	1.9	9.0	149	17	31	635	343	320	123	85	0.8	803	1787	1431	303	FI
FR	6.4	5.4	505	7.8	116	6462	311	26	3957	137	0.8	1119	7998	560	8.5	FR
GB	0.5	2.8	59	5.5	6.9	2593	36	13	137	13	0.2	226	1705	297	2.4	GB
GE	1.0	3043	8.4	2197	7.0	14	189	20	6.0	21	7.0	33	47	25	1.6	GE
GR	284	103	201	103	224	130	21364	76	96	1300	13	496	506	105	5.2	GR
HR	15	10	1076	10	2217	162	459	34	166	1278	1	1511	905	115	4	HR
HU	6.6	10	1998	12	354	214	680	72	169	1721	0.4	3909	1463	171	5.9	HU
IE	0.2	0.8	14	1.6	2.2	373	12	3.2	48	4.0	0.1	41	269	56	0.6	IE
IS	0.1	0.2	5.2	0.4	1.2	59	5.0	0.9	9.0	1.9	0.03	18	73	23	0.1	IS
IT	103	41	2107	38	1078	785	2645	67	3029	1336	5.3	1899	2744	301	9.1	IT
KZ	2.2	156	42	507	25	91	491	185	29	68	3.9	173	293	177	17	KZ
LT	0.8	3.4	69	4.3	15	133	136	917	36	42	0.2	485	638	437	38	LT
LU	0.02	0.1	6.8	0.1	0.3	275	1.0	0.3	18	0.4	0.01	16	340	8.3	0.1	LU
LV	0.7	3.6	64	5.0	13	167	127	613	35	37	0.2	408	700	539	222	LV
MC	0.00	0.00	0.04	0.00	0.01	0.03	0.02	0.00	0.08	0.01	0.00	0.05	0.09	0.01	0.00	MC
MD	1.0	6.6	17	8.1	12	21	354	97	8.1	43	0.2	82	96	43	3.7	MD
MK	161	12	75	12	114	40	6287	15	31	2464	1.2	199	170	28	1.3	MK
NL	0.1	0.4	28	0.6	1.2	8712	5.9	2.7	46	2.3	0.04	113	4124	154	1.0	NL
NO	0.5	3.2	72	5.1	11	758	71	44	87	23	0.4	389	1607	1604	10	NO
PL	7.1	23	1596	32	199	1527	844	1182	630	534	0.9	21476	14438	3286	53	PL
PT	0.1	0.3	10	0.6	2.4	37	11	1.2	18	3.3	0.0	24	58	6.5	0.2	PT
RO	34	81	562	91	684	349	13581	354	220	4043	2.1	1864	1580	395	23	RO
RU	33	1219	1228	2845	491	2899	6604	9650	822	1308	25	5738	9728	5893	1928	RU
SE	1.5	6.4	197	11	29	1332	253	178	170	73	1.0	1257	3996	8204	50	SE
SI	1.2	2.8	1485	2.9	61	77	67	13	104	78	0.2	624	497	46	1.6	SI
SK	3.7	8.9	933	11	123	187	407	78	138	363	0.3	5162	1216	192	6.6	SK
TR	23	6172	186	1691	142	248	10150	299	107	511	219	655	845	342	18	TR
UA	17	166	570	208	299	504	4078	3730	243	903	4.1	2456	2522	1047	88	UA
	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CS	CY	CZ	DE	DK	EE	

Table C.3. Matrix of PCDD/Fs country-to-country depositions in 2003, g TEQ/y (continued)

Receptors ↓ Emitters →

	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	
AL	76	4.6	120	94	41	1359	252	181	10	0.4	812	5.7	6.4	3.9	7.4	AL
AM	5.5	1.5	10	20	1503	30	6.7	7.0	2.5	0.1	26	11	1.7	0.3	2.3	AM
AT	175	27	1243	823	13	37	536	763	63	2.5	2406	4.4	23	78	29	AT
AZ	25	9.2	43	95	3724	107	27	27	12	0.4	104	132	10	1.4	14	AZ
BA	121	17	312	266	35	206	7897	1418	26	1.1	1650	7.6	17	14	20	BA
BE	154	6.6	4550	1990	0.9	2.4	10	8.2	81	1.4	78	0.4	2.7	687	4.3	BE
BG	125	30	291	383	200	2670	598	805	36	1.6	885	38	41	13	50	BG
BY	90	194	361	726	70	95	233	502	71	3.5	348	42	1862	18	1327	BY
CH	204	4.6	2093	376	1.6	8.9	67	29	29	0.7	2627	0.5	2.4	23	3.8	CH
CS	139	22	340	323	61	749	4173	2109	31	1.3	1474	13	25	15	30	CS
CY	3.7	0.6	7.7	12	22	45	5.8	6.2	1.3	0.04	23	1.3	0.8	0.3	1.0	CY
CZ	130	33	968	842	13	28	391	1399	60	2.1	592	4.5	32	69	37	CZ
DE	1015	115	14257	9308	21	41	166	353	544	16	1545	8.0	66	2587	91	DE
DK	69	16	356	1687	2.9	3	12	25	117	4.1	40	0.9	11	15	12	DK
EE	34	572	127	448	7.4	11	26	58	41	1.9	54	3.6	279	6	1242	EE
ES	72171	9.1	2700	850	6.6	38	206	97	171	5.8	1178	2.9	5.8	26	8.1	ES
FI	207	17842	778	2414	29	54	120	217	254	13	320	12	305	35	622	FI
FR	15890	55	132472	8489	14	117	671	280	761	15	6634	5	25	1583	38	FR
GB	995	14	3053	109665	6.4	12	33	30	3558	24	218	3	11	85	14	GB
GE	16	5.5	29	60	22280	76	20	22	7.4	0.2	71	25	6.6	1.0	9.1	GE
GR	207	18	335	343	302	48037	520	451	35	1.3	1601	40	27	12	33	GR
HR	123	20	389	296	24	130	37687	2609	29	1.1	2481	6.2	20	17	24	HR
HU	90	27	384	374	24	124	4540	45727	31	1.4	1073	6.6	35	23	38	HU
IE	371	3.0	658	2848	1.7	3.7	11	8.4	9880	8.2	73	0.9	2.7	17	3.2	IE
IS	50	1.0	102	272	0.5	2.1	5.1	3.9	50	135	28	0.1	0.7	3.0	0.7	IS
IT	1531	46	4494	1615	114	1564	6098	1578	160	5.0	158004	21	40	90	49	IT
KZ	62	70	136	362	381	111	72	89	43	1.8	171	15198	55	5.4	83	KZ
LT	40	91	169	409	11	20	56	134	39	1.7	93	5.0	5357	8.7	1292	LT
LU	15	0.7	1339	84	0.1	0.4	1.4	1.2	4.8	0.1	13	0.04	0.3	1460	0.5	LU
LV	50	218	197	586	12	19	49	114	55	2.3	89	6	1948	10	8550	LV
MC	0.08	0.00	0.73	0.07	0.00	0.01	0.06	0.03	0.01	0.00	2.32	0.00	0.00	0.00	0.00	MC
MD	10	11	31	67	20	25	30	55	7	0.3	54	10	24	1.5	25	MD
MK	54	5.2	100	97	34	1453	225	212	10	0.4	469	5.1	6.6	3.9	7.9	MK
NL	121	7.1	1712	2622	1.0	1.9	6.2	7.6	101	2.3	44	0.4	2.9	66	5	NL
NO	226	138	809	3786	8.9	16	51	69	392	19	179	3.0	47	35	57	NO
PL	362	246	1958	3221	59	140	977	2870	264	11	1391	25	580	131	401	PL
PT	2284	1.0	174	117	0.6	3.0	16.2	8.8	20.9	0.7	81	0.3	0.6	3.5	0.9	PT
RO	217	82	640	872	219	821	1610	2823	82	3.4	1721	60	125	31	138	RO
RU	1331	8674	4032	10725	6715	1088	1581	2599	1142	65	3216	7104	3453	180	6980	RU
SE	301	856	1347	4971	20	42	130	216	469	20	365	8.0	209	63	271	SE
SI	42	8.3	181	130	7.0	19	3832	494	12	0.4	1679	2.4	8.2	8.6	9.6	SI
SK	66	29	317	363	21	69	712	5167	31	1.3	574	5.9	39	18	43	SK
TR	249	66	466	839	4790	3167	384	491	90	3.6	1203	147	88	18	108	TR
UA	242	266	778	1538	540	554	1032	2530	155	7.1	1340	321	616	39	616	UA
	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KZ	LT	LU	LV	

Table C.3. Matrix of PCDD/Fs country-to-country depositions in 2003, g TEQ/y (continued)

Receptors ↓ Emitters →

	MC	MD	MK	NL	NO	PL	PT	RO	RU	SK	SI	SE	UA	Total, kg TEQ/y	
AL	4.3	16	12455	16	5.1	492	9.2	326	169	327	6.8	6.7	807	23	AL
AM	0.2	2.5	23	2.4	1.7	65	1.0	20	248	16	0.2	2.0	295	14	AM
AT	18	5.9	97	202	37	3367	25	130	166	4384	199	46	683	48	AT
AZ	0.9	11	99	12	10	296	4.8	82	2723	64	1.0	12	1486	44	AZ
BA	11	16	624	55	18	2235	15	561	203	1916	59	25	1249	37	BA
BE	1.5	0.4	4.9	849	15	161	26	6.5	18	32	0.7	15	41	48	BE
BG	7.2	186	7639	67	29	2487	19	5128	1289	1457	18	42	7276	211	BG
BY	3.8	153	310	133	124	16884	20	737	6194	1641	12	228	25907	78	BY
CH	21	0.7	19	45	10	185	24	13	16	76	7.1	8.4	56	19	CH
CS	10	44	15813	66	22	2877	18	2150	370	2542	39	33	2363	83	CS
CY	0.2	1.6	25	1.5	0.7	38	0.5	13	58	12	0.2	0.8	133	2	CY
CZ	7.7	6.4	104	247	38	16384	22	142	205	8436	31	61	800	102	CZ
DE	28	10	75	5835	259	11436	178	157	382	1467	13	322	1124	246	DE
DK	0.7	0.9	5.7	427	181	880	28	12	45	104	0.7	249	124	20	DK
EE	0.7	6.9	31	71	116	1767	11	53	1093	197	1.4	286	815	10	EE
ES	25	3.4	103	72	30	480	5064	65	78	228	14	13	288	85	ES
FI	4.5	24	142	348	1016	4958	57	219	4022	683	6.9	2580	2563	46	FI
FR	941	7.9	299	893	137	1869	1187	159	189	706	48	102	630	195	FR
GB	3.8	1.7	25	1160	115	586	433	30	78	104	2.2	41	240	126	GB
GE	0.6	9.2	72	7.4	6.3	214	2.9	68	1950	51	0.7	7.4	1160	32	GE
GR	11	92	24576	57	21	1523	25	1230	1303	865	18	27	5207	112	GR
HR	17	11	418	64	20	3096	15	390	187	3235	586	28	1186	61	HR
HU	10	15	526	96	27	7181	14	1832	268	22839	157	39	3498	100	HU
IE	1.2	0.5	8.6	127	23	122	185	8.0	20	25	0.7	7.9	64	15	IE
IS	0.4	0.2	4.8	25	16	53	14	2.6	5.6	12	0.3	3.4	16	1	IS
IT	499	38	3331	249	62	4405	160	832	628	3094	388	69	2820	208	IT
KZ	1.8	35	157	51	58	1214	17	250	15349	222	3.1	68	7456	44	KZ
LT	1.1	17	61	76	82	6849	10	116	1333	483	3.2	171	1979	22	LT
LU	0.2	0.1	0.7	16	1.3	22	2.2	1.0	2.3	4.8	0.1	1.5	5.7	3.6	LU
LV	1.1	14	56	102	135	4209	15	100	1302	396	2.8	313	1676	23	LV
MC	4.51	0.00	0.02	0.01	0.002	0.13	0.01	0.01	0.01	0.07	0.01	0.003	0.05	0.01	MC
MD	0.5	1963	85	11	8.6	905	2.1	1433	435	158	1.2	13	9204	15	MD
MK	3.1	17	88811	18	5.7	533	7.0	354	160	377	6.4	7.6	804	103	MK
NL	0.8	0.4	3.4	8762	25	187	28	5.9	19	32	0.4	19	45	27	NL
NO	2.9	4.9	35	474	6256	1586	74	49	251	235	3.3	513	487	20	NO
PL	18	61	523	795	330	319883	76	810	2015	18298	64	606	13821	416	PL
PT	1.3	0.3	6.0	10	3.1	53	16428	5.3	8.4	23	1.1	1.2	30	19	PT
RO	16	1066	3168	163	79	9126	35	52844	2056	5089	49	112	23843	131	RO
RU	37	719	2487	1596	2464	44607	345	4560	666157	7210	77	3682	135810	979	RU
SE	5.5	17	107	825	3483	6818	94	158	738	768	8.0	11994	1706	52	SE
SI	8.2	3.0	54	29	8.5	1162	5.7	87	72	1160	1484	12	362	14	SI
SK	5.4	13	294	87	29	13620	11	480	280	63076	46	44	3054	97	SK
TR	10	239	1811	126	76	3514	40	1905	7529	1125	15	90	19362	70	TR
UA	13	1865	1381	269	222	28952	48	5647	23087	7284	44	330	352587	449	UA
	MC	MD	MK	NL	NO	PL	PT	RO	RU	SK	SI	SE	UA	Total, kg TEQ/y	

