CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

EMEP

Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

Assessment of POP Transport and Accumulation in the Environment

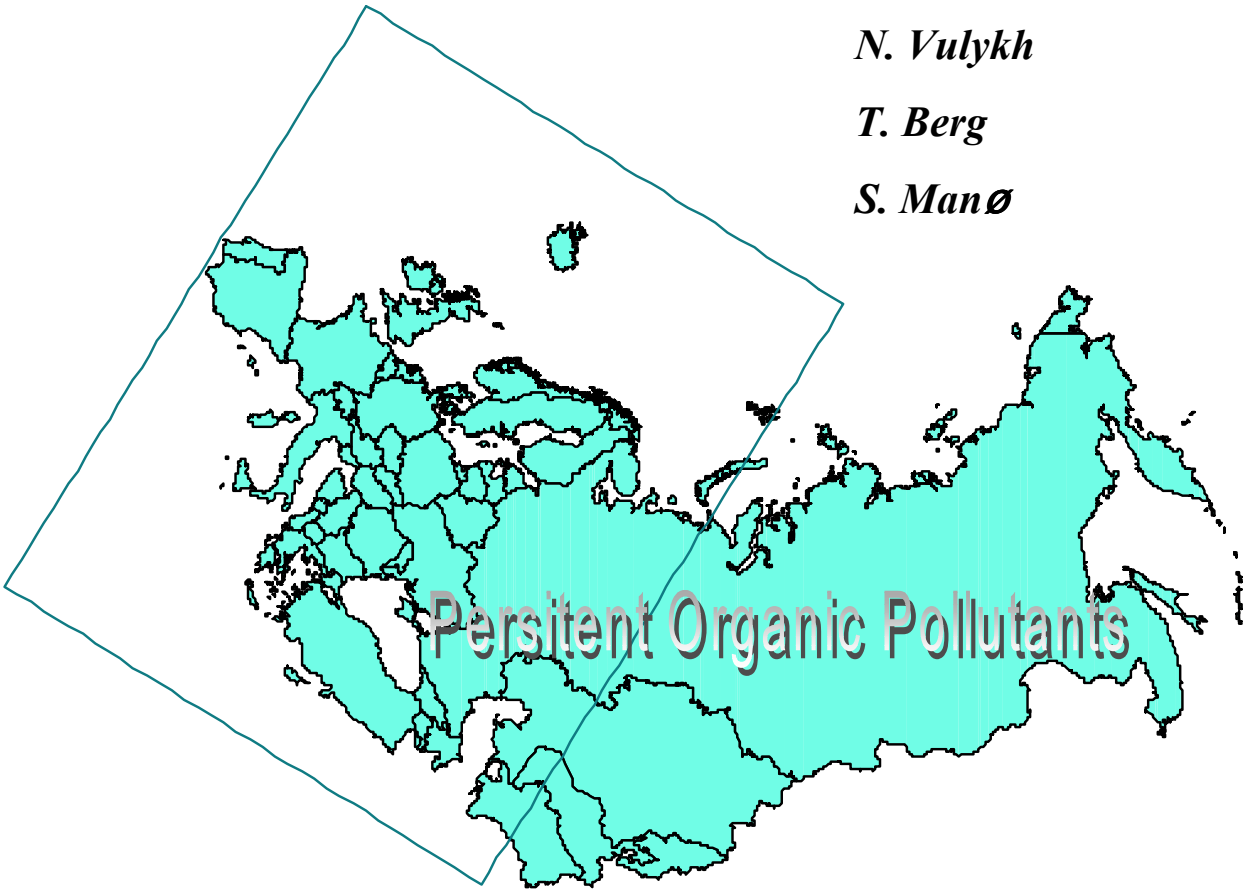
V. Shatalov

A. Malanichev

N. Vulykh

T. Berg

S. Manø



MSC-E/CCC Technical Report 7/2002

June 2002

Meteorological Synthesizing Centre - EAST

MSC-E/CCC Technical Report 7/2002

June 2002

ASSESSMENT OF POP TRANSPORT AND ACCUMULATION IN THE ENVIRONMENT

METEOROLOGICAL SYNTHESIZING CENTRE - EAST

Viktor Shatalov, Alexander Malanichev, Nadezhda Vulykh

CHEMICAL COORDINATING CENTRE

Torunn Berg, Stein Manø

with contributions from:

Ivan Holoubek	Czech Republic
Audrone-Regina Milukaite	Lithuania
Olga Afinogenova	MSC-East
Elena Mantseva	MSC-East

METEOROLOGICAL SYNTHESIZING CENTRE-EAST
Ul. Arhitektor Vlasov, 51, 117393 Moscow, Russia
tel.: 007 095 128 90 98
fax: 007 095 125 24 09
e-mail: msce@msceast.org
home page: http://www.msceast.org/

NORWEGIAN INSTITUTE FOR AIR RESEARCH
Postboks 100, N-2007 Kjeller, Norway
tel.: +47 63 89 80 00
fax: +47 63 89 80 50
e-mail: torunn.berg@nilu.no
home page: http://www.nilu.no/projects/ccc/index.html

EXECUTIVE SUMMARY

This Technical Report describes the progress in evaluation of benzo[a]pyrene (B[a]P) and dioxins/furans (PCDD/F) transport and pollution in the EMEP region. The work is fulfilled in accordance with the EMEP work-plan 2002 [ECE/EB.AIR/75, Annex VI].

Tentative evaluation of B[a]P transport between European countries was presented in [*Shatalov et al.,* 2000, 2001]. At the present time the model is modified, i.e. description of wet deposition process is refined and the pollutant inflow from outside the EMEP region is introduced.

On the base of the modified model calculations of B[a]P long-range transport, "country-to-country" matrices for atmospheric concentrations and depositions as well as the evaluation of contamination levels in European countries for 1999 with spatial resolution 50x50 km are performed. For this purpose available official B[a]P emission data together with expert estimates are used. Calculations show that:

- Model modification improves the agreement between calculation and measurement data. Calculated concentration values exceed measured ones 1.3 times on the average, that is better than in calculations of the previous year (factor 2.4). The ratio of measured to calculated deposition fluxes is within a factor of 4 (0.9 on the average). Model modification results in some changes in the evaluation of transboundary fluxes between European countries.

- Model evaluation of contamination of European region shows that for the most part of Europe annual air concentration values are within the range of 0.1 – 1 ng/m^3. Over the vast areas of Central Europe a high level of mean annual air concentrations (exceeding the limit value of 1 ng/m^3 accepted in some European countries) is achieved. For these areas considerable deposition fluxes (100 – 180 g/km^2/y) are specified. Total deposition to the EMEP region in 1999 amounted to 184 tonnes, which is about a half of annual emissions.

- For most of European countries the total contribution from external sources to concentrations and depositions to a country exceeds the contribution from national sources. The highest contribution from external emission sources to atmospheric concentrations is calculated for Slovakia, the Czech Republic and Hungary (0.3 - 0.5 ng/m^3). Considerable values of B[a]P export are characteristic of Germany, Poland and France.

At the previous stage of investigations, pilot calculation of PCDD/F long-range transport for the period 1970 - 98 was made.

Currently, congener composition of PCDD/F mixture in various media is investigated. Eight congeners reasonably describing PCDD/F toxicity in the environment are selected. It is shown that physical-chemical properties of one of them ("indicator congener") can be used for evaluation of PCDD/F transport and accumulation at the initial stages of investigations with reasonable accuracy. Development of PCDD/F transport model is continued. Main attention is paid to the refinement of the model description of exchange in the atmosphere/vegetation/soil system. Besides, degradation rate constants for the atmosphere and soil are refined.

On the basis of the modified model the evaluation of PCDD/F accumulation dynamics in environmental compartments for the period from 1970 to 1999 is performed and estimates of contamination levels in environmental media for 1999 are given. Calculations of the PCDD/F mixture were carried out on the properties of the "indicator congener". Available PCDD/F official emission data

for the considered period together with expert estimates are used in calculations. Finally, the evaluation of media response time to an emission scenario reduction is made.

As a result of PCDD/F modelling the following results has been achieved:

- According to calculations, soil compartment accumulates a huge fraction of the total PCDD/F toxicity in the environment.

- The analysis of long-term trends of PCDD/F pollution for 1970 – 1999 shows that the dynamics of soil concentrations severely lags behind the emission dynamics. By the end of the calculation period (1999) the value of re-emission flux from soil amounts to about 20% of European average anthropogenic emission flux. So, soil contamination is able to support pollution levels in other environmental compartments for sufficiency long time (decades).

- At the end of calculation period (1999) PCDD/F soil concentrations range from 0.01 to 20 pg TEQ/g. PCDD/F contamination levels in air exceed 1 fg TEQ/m^3 for most of Europe.

- The agreement between calculated and measured concentrations in the atmosphere and soil is improved. Namely, calculated data on atmospheric concentrations and depositions agree with measurements within a factor of 6. More than half the measured soil concentrations agree with calculations within a factor of 4.

- On the basis of additional PCDD/F transport simulations under the assumption of full emission cessation PCDD/F environmental half-life is evaluated as 30 years.

To refine the evaluation of PCDD/F long-range transport and accumulation in the forthcoming year it is planned to perform simulations of the mixture of eight selected congeners.

The analytical laboratory intercomparison on POP measurements is continued. Preliminary results of Round 1 (analysis of standards) were promising, showing that most laboratories are able to analyse standards within ±30%.

More detailed information on contamination levels and transboundary fluxes of B[a]P and PCDD/Fs in European countries can be found in Internet www.emep.int, www.msceast.org.

CONTENTS

INTRODUCTION

The progress in evaluation of benzo[a]pyrene (B[a]P) and dioxins/furans (PCDD/F) transport and pollution in the EMEP region is presented in this report. The work is fulfilled in accordance with the EMEP work-plan for 2002. The report consists of three chapters. First two describe the results on B[a]P and PCDD/F modelling, respectively. The third written by CCC is devoted to description of measurement activities under EMEP.

In the first chapter evaluation of B[a]P contamination in the European region and its transboundary transport is presented. At the previous stages of the work B[a]P concentrations in air, precipitation, vegetation, soil and seawater were calculated for the period 1970 to 1997. Long-term trends of the content variations in these media were investigated [*Shatalov et al.,* 2001]. The comparison of calculation results with measurements demonstrated that for better consistency of model results with observations it is necessary to refine the parameterization of B[a]P wet deposition scheme since calculated concentrations in precipitation were underestimated. Besides it was necessary to make allowance for B[a]P import from remote sources outside the EMEP region. First of all it was essential for the refinement of pollution levels over territories adjacent to the borders of the calculation domain (the North Atlantic and the Arctic). Such modifications were incorporated into the model.

The goal of the present stage of investigations is to evaluate the B[a]P contamination levels and transboundary fluxes for European countries using improved version of MSCE-POP model.

On the basis of modified model the evaluation of concentrations and depositions for European region for 1999 is done. The results were compared with measurement data on B[a]P concentrations in the atmospheric air, precipitation and depositions.

For transboundary transport assessment “country-to-country” deposition and concentration matrices with spatial resolution 50x50 km and meteorological information for 1999 are calculated. The analysis of the role of transboundary transport in formation of air pollution and deposition fluxes is presented. The transboundary pollution of European countries is analysed on the example of depositions and air concentrations from national and external B[a]P sources in the United Kingdom.

The second chapter is aimed at evaluation of PCDD/F long-range transport and accumulation. At the previous stages of the work [*Shatalov et al*., 2001] a primary evaluation of contamination levels in environmental media for 1998 was done. In particular, it was shown that main accumulation media for PCDD/F is soil. The comparison with measurements carried out at these stages showed the necessity of improvement of soil/atmosphere exchange model description.

The goals of the present stage are: further investigations of PCDD/F long-range transport and accumulation with emphasis on description of PCDD/F behaviour in soil; investigation of long-term trends in the environmental pollution by dioxins/furans for the period from 1970 to 1999; evaluation of pollution spatial distribution in environmental media by the end of the calculation period (1999); estimation of media response to emission reduction on the base of the refined description of exchange processes between the atmosphere and soil.

For investigation of PCDD/F long-range transport contributions of particular congeners to the overall toxicity of PCDD/F mixture (toxicity profiles) in environmental media is of importance. A set of eight congeners responsible for more than 75% of the mixture toxicity is selected. This makes it possible to

evaluate contributions of individual congeners to the total toxicity and to investigate behavioural peculiarities of individual congeners in the environment.

To refine the agreement between calculated and measured data the model description of atmosphere/soil exchange module is improved. Namely, soil layer thickness is changed for more accurate representation of the vertical profile of PCDD/F concentration distribution in soil. Besides, degradation rates in the atmosphere, soil and seawater are refined for the above eight congeners.

By the modified model, a number of calculations were performed. Long-term trends of PCDD/F accumulation in the environmental media, contamination levels in Europe, and media response to emission reduction were evaluated. Emission data for calculations are prepared on the basis of official data and expert estimates. Calculation results were compared with available measurements.

The third part presents the description of measurement activities undertaken under EMEP. It contains the overview of measurement sites included into the EMEP monitoring network, summaries of measurement data on POPs and the description of the quality of POP monitoring data (including the preliminary results on analytical laboratory intercomparison on POP measurements).

Detailed information on the above described investigations is presented in this report and in MSC-E Technical Note 1/2002 [*Vassilyeva and Shatalov*, 2002].

Chapter 1

EVALUATION OF B[A]P TRANSBOUNDARY TRANSPORT AND AIR CONTAMINATION FOR 1999

In this chapter evaluation of B[a]P contamination and transboundary transport in the European region for 1999 is described.

1.1. Model development

1.1.1. *Model modification*

The comparison of last year calculation results with measurements demonstrated that for better consistency of model results with observations it is necessary to refine the parametrization of B[a]P wet deposition scheme since calculated concentrations in precipitation were 2 times underestimated [*Shatalov et al.*, 2001]. Besides, it was necessary to make allowance for B[a]P import from remote sources outside the calculation domain. First of all it is essential for the refinement of pollution levels over territories adjacent to the borders of the calculation domain (the North Atlantic and the Arctic). In this section we describe modifications of wet deposition and B[a]P inflow description and the effect of them on B[a]P long-range transport.

One of the reasons for underestimation of concentration in precipitation can be a low value of washout ratio W_p for B[a]P atmospheric particle phase used in the model parameterization. To investigate this problem the experimental calculations were carried out [*Shatalov et al*., 2001] and the information on W_p values estimated on the base of measurements was collected from literature sources. Monitoring information is summarized in Table 1.1.

From Table 1.1 one can see that the range of washout ratio values varies from $1.7 \cdot 10^3$ to $9.8 \cdot 10^6$. According to experimental calculation results [*Shatalov et al.,* 2001] the value of $W_p = 10^5$ was accepted. It should be noted, that this value is within the range, estimated on the base of measurements. New W_p value results in better agreement between calculations and measurements both for concentrations in precipitation and deposition fluxes and made a small impact on calculated air concentrations. This year new washout ratio value has been included into the model parameterization and induced changes of calculation results are examined. The second model modification is accounting the B[a]P inflow across the EMEP boundary from sources located outside the domain. As it was shown in [*Shatalov et al.,* 2000], the export of the pollutant outside the calculation grid is about 35%. Thereby, the inflow of B[a]P to the EMEP region may be also tangible.

The consideration of B[a]P inflow from outside the EMEP region made out by prescribing concentration values on the boundary of the calculation domain. The values of these concentrations were chosen on the basis of measurements in remote regions (Table 1.2) available in [*Berg and Hjellbrekke,* 1998,1999].

Table 1.1. Values of washout ratio obtained by different authors

W_p	Analysis method	Note	Source
$1.7 \cdot 10^3$	Experiment (calculated mean value)	Rain, Portland, Oregon, 1984.	*Ligocki et al*., 1985
$2.6 \cdot 10^5$	Calculations based on the ratio of mean annual concentrations in air and precipitation. It is assumed that measured concentrations in precipitation are measured for particle phase only.	Eagle Harbor Station, lake Superior 1990 – 93	Calculations based on *Hoff et al*., 1996
$1.8 \cdot 10^5$		Sleeping Bear Dunes Station, lake Michigan 1990 – 93	
$1.2 \cdot 10^5$		Stugeron Point Station, lake Erie 1990 - 93	
$6.7 \cdot 10^5$		Point Petre Station, lake Ontario 1990 - 93	
$1.2 \cdot 10^5$	Recommended mean value	For particles with d < 0.95 μm	*Baart et al*., 1995
$2.5 \cdot 10^4$	Experiment	winter rain, Minnesota December 12, 1991	*Franz and Eisenreich.,* 1998
$2.0 \cdot 10^6$	Experiment (calculation of mean value)	snow, Minnesota, December 13 – 14 and 19 – 20, 1991 and March 8 – 9, 1992	*Franz and Eisenreich.,* 1998
$9.8 \cdot 10^6$	Calculation using experimental data from [*Franz and Eisenreich.,* 1998]	snow, Minnesota, December 13, 1991	*Wania et al*., 1999

Table 1.2. B[a]P air concentration values in remote regions.

Sampling location	Coordinates	B[a]P air concentrations in remote regions, ng/m^3				
		1992	1993	1994	1995	1996
Alert, Canada	82° 30′ N 62° 20′ W	0.143 0.001 - 3.42	1.324 0.001 - 9.908	2.563 0.001 - 6.69		
Tagish, Canada	60° 20′ N 134° 15′ W	0.004 0.003 - 0.005	0.004 0.001 - 0.012	0.021 0 - 0.061		
Zeppelinfjell, Spitsbergen, Norway (NO42)	78° 54′ N 11° 53′ E			**0.013** **0 - 0.202**	**0.009** **0 - 0.09**	11.63 0 – 100
Pallas, Finland (FI96)	67° 58′ N 24° 07′ E					**0.015** **0.004 - 0.053**
Rorvik, Sweden (SE3)	57° 25′ N 11° 56′ E			0.07 0.01 - 0.52	0.12 0 - 0.89	0.09 0 - 0.28
Dunai Island, Russia	73° 59′ N 124° 30′ E		0.605 0 - 4			

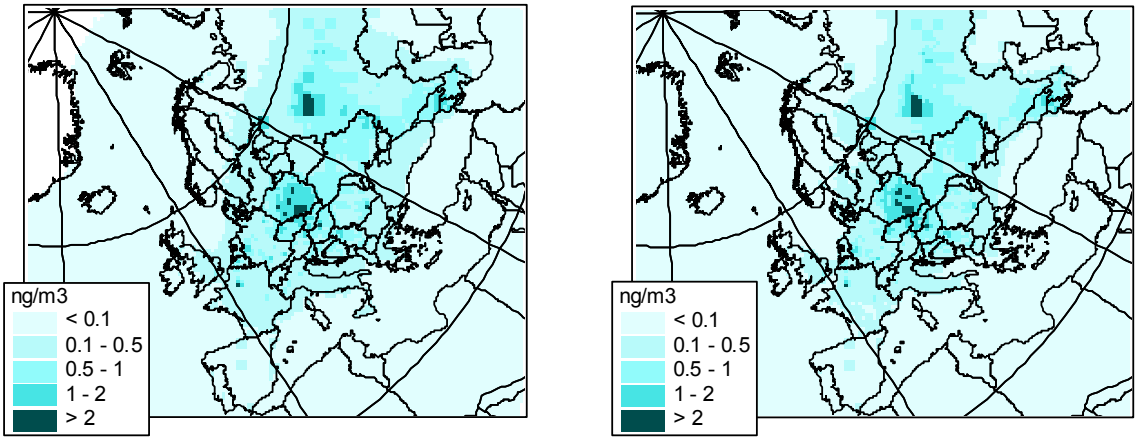
As seen from the data of Table 1.2, B[a]P concentrations vary in a wide range (from 0.001 to 12 ng/m^3). Minimum average annual concentrations close to 0.01 ng/m^3 were obtained at stations Tagish, Spitsbergen and Pallas. Two last stations (marked bold) are located at the boundary of the EMEP region. At this stage, as a first approximation, the value 0.01 was chosen as air concentration at the boundary of the EMEP calculation domain. In future the refinement of this value on the basis of the hemispheric modelling results is planned.

Below two calculation runs of B[a]P transport using previous and new parametrization are compared. These runs use the same emission scenario including expert estimates [*Pacyna et al.*, 1999] and meteorological data for 1998 and 1999 respectively.

Figure 1.1 demonstrates maps of spatial distribution of concentrations over the surface atmospheric layer obtained with the previous (a) and new (b) paramentrization.

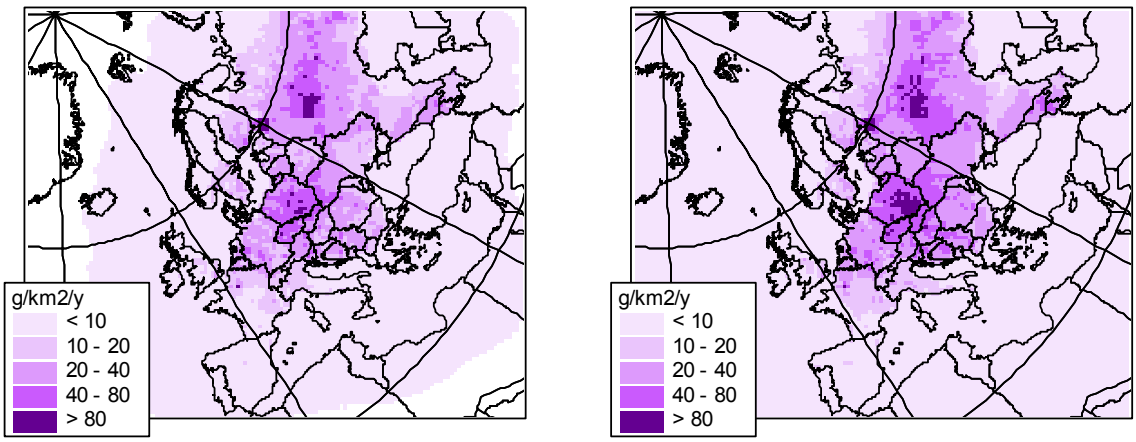
As follows from Figure 1.1 new parametrization has brought about a moderate change in B[a]P air concentrations. One of the most distinctive differences is the increase of concentrations over Greenland and near the North Pole. Besides one can see some local decrease of concentrations over France, Sicily, the Black and the Caspian Seas.

Figure 1.2 illustrates total deposition fields calculated with the previous and new parametrization.



a - previous parametrization **b** - new parametrization

Figure 1.1. *B[a]P concentration in the surface air layer, ng/m^3*



a - previous parametrization **b** - new parametrization

Figure 1.2. *B[a]P total depositions, g/km^2/y*

The comparison of the maps shows that the new parametrization resulted in deposition increase actually over the whole EMEP region. The consideration of export from outside the calculation domain increased depositions in periphery regions (Greenland, the Atlantic, the Arctic and Africa). Zones with depositions more than 80 g/km^2/y noticeably extended. Maximum values of fluxes raised from 213 to 252 g/km^2/y. On the whole during the year 292 tonnes of B[a]P have fallen out to the EMEP region. It is by 60 tonnes (25%) more compared with the results obtained with the previous parametrization. Depositions from external sources to the EMEP domain are 2.6 tonnes.

The model results appeared to be rather stable with regard to new parameterization. However, some changes in transboundary fluxes were revealed. Figure 1.3 gives pie charts showing depositions from European countries where most valuable changes were found. These charts are obtained by calculations on the previous and new parameterization. The comparison of these charts indicates the changes in the main countries – receptors.

As to depositions from the Czech Republic (Fig 1.3.a) among four main receptors Austria appeared instead of the Ukraine, for Georgia (Fig.1.3.b) - Kazakhstan replaced Azerbaijan, for Norway (Fig. 1.3.c) - the Arctic region appeared instead of Russia.

Figure 1.4 presents differences in depositions to individual European countries and regions calculated by previous and modified model versions.

Thus in depositions to Finland (Fig. 1.4.a) among four main contributors, Germany appeared instead of the Ukraine, for Lithuania (Fig. 1.4.b) - Belarus replaced Germany, for Yugoslavia (Fig. 1.4.c) - Macedonia appeared instead of Bulgaria.

Thus, the introduction of new parametrization (refined washout ratio and the consideration of external sources) has brought about the following changes:

- Concentrations in the surface air underwent some changes: they increased in remote regions due to the export from external sources through the EMEP boundary. The consideration of external sources inflicted a insignificant impact on the pollution of the central part of the EMEP region.

- Total deposition fluxes increased by 15-20% on the average.

- Contribution of transboundary transport to depositions to some countries in the EMEP domain were changed.

In the next section we discuss the reliability of the obtained results by comparing them with monitoring data.

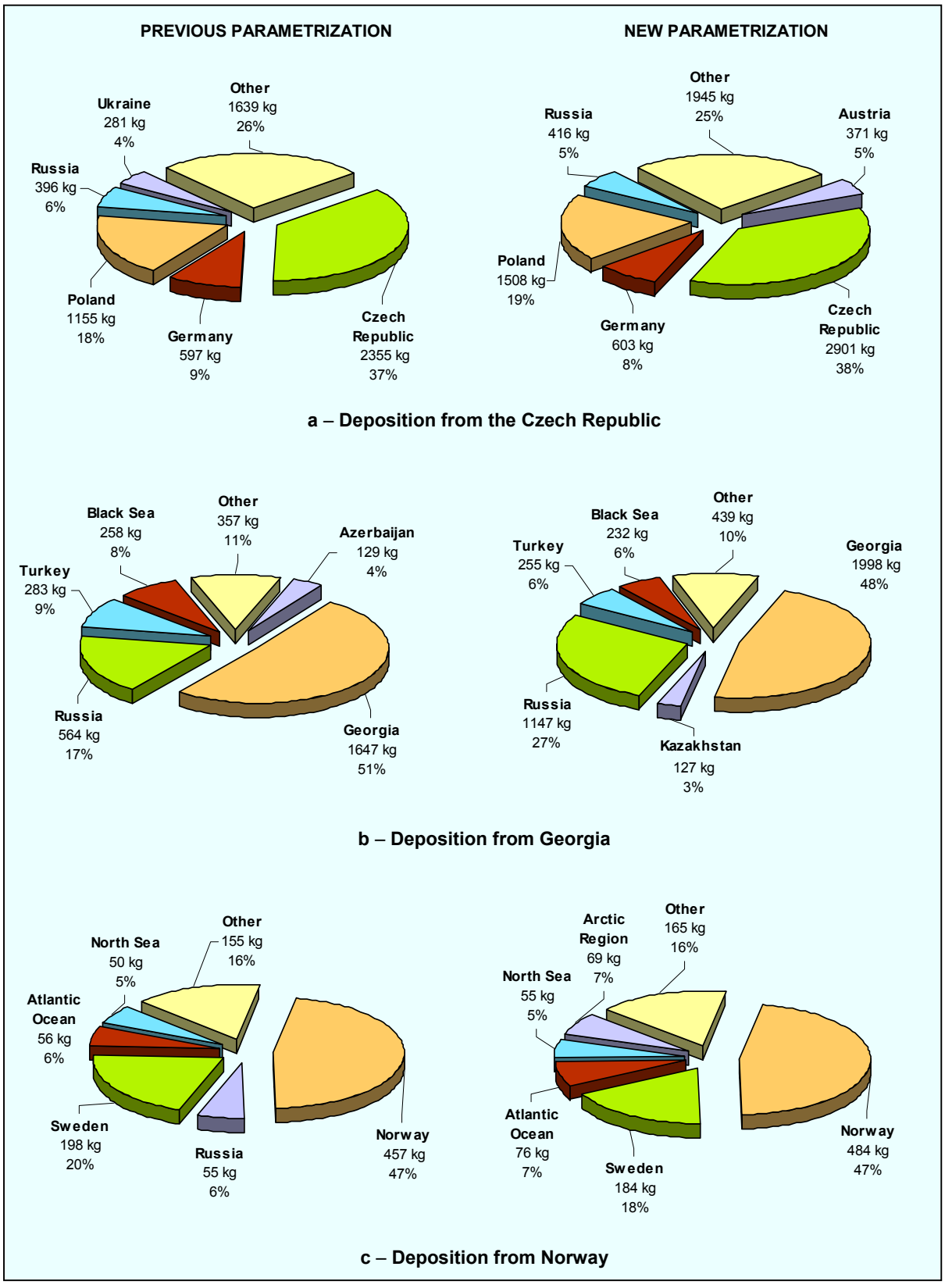


Figure 1.3. *B[a]P depositions from some European countries*

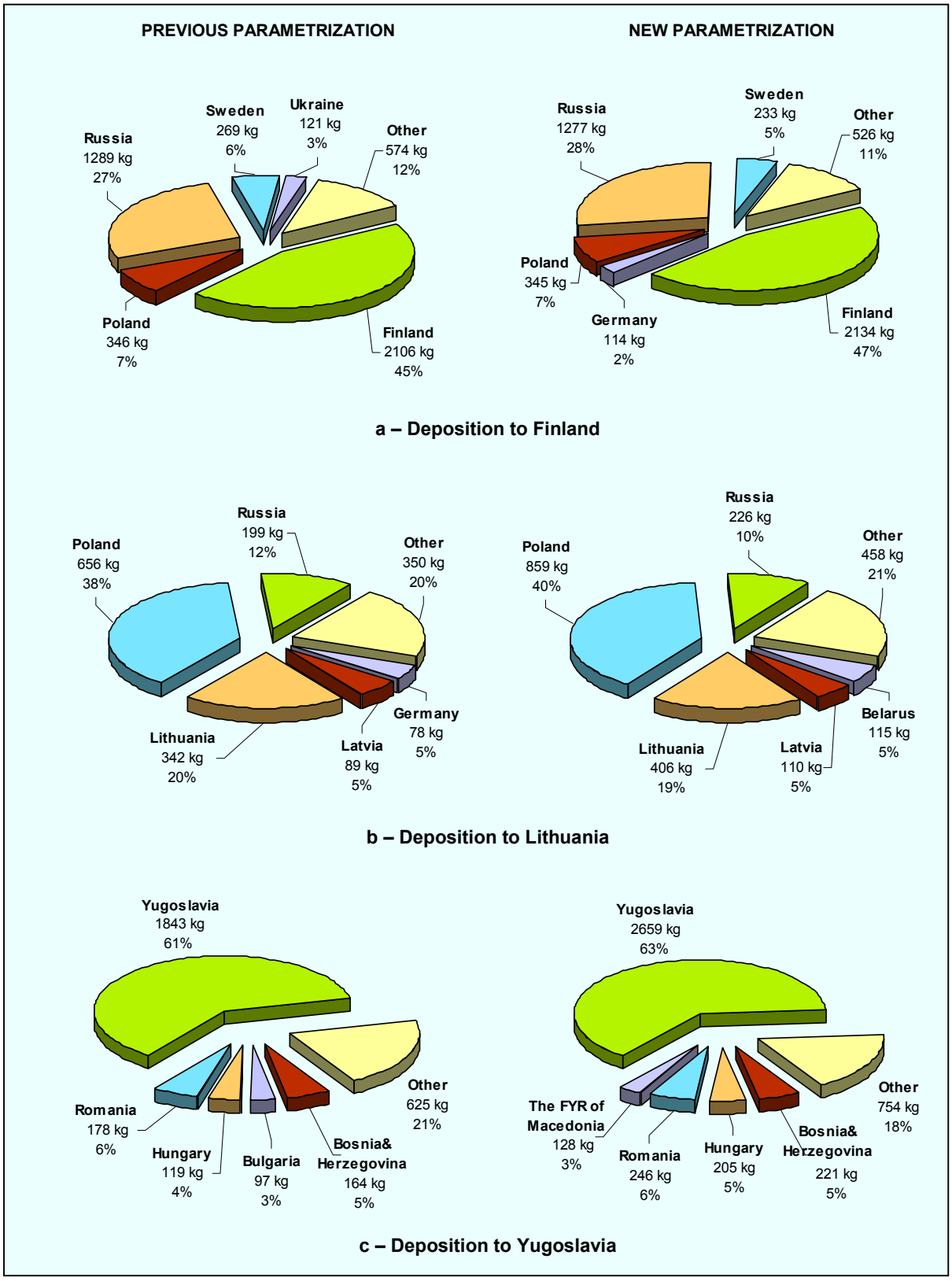


Figure 1.4. *B[a]P depositions to some European countries*

1.1.2. Model verification

Model estimates of B[a]P tansboundary transport were verified by means of the comparison of the model results with monitoring data. Annual mean values are used. In order to make the comparison comprehensive and more complete four years 1996-99 were calculated with spatial resolution 50x50 km and with the use of emission expert estimates [*Pacyna et al.*, 1999]. In the comparison available B[a]P measurement data obtained at EMEP stations for the indicated years and in the course of national measurement campaigns were used. The location of monitoring stations are shown in Figure 1.5. The comparison results of concentrations in air and precipitation and deposition fluxes are discussed below.

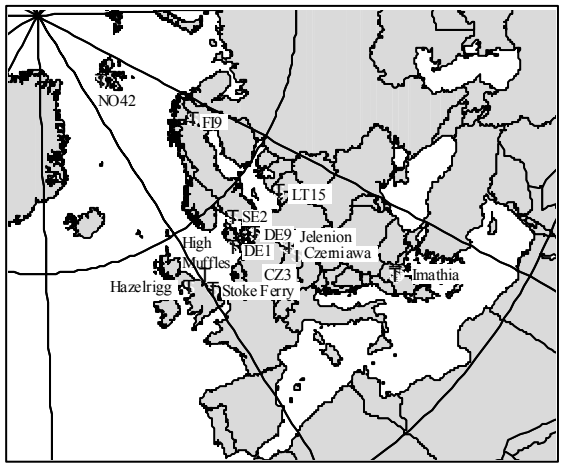


Figure 1.5. *The location of monitoring stations, which data on B[a]P are used in the comparison*

Air concentrations

Figure 1.6 demonstrates the comparison of measured and calculated mean annual total (gas + particles) B[a]P air concentrations for 1996 - 99. To represent on one diagram rather wide range of values – from 0.007 to 1.7 ng/m^3 we used the logarithmic scale. Measured and calculated values used in the comparison, their ratios and appropriate references are presented in Table 1.3.

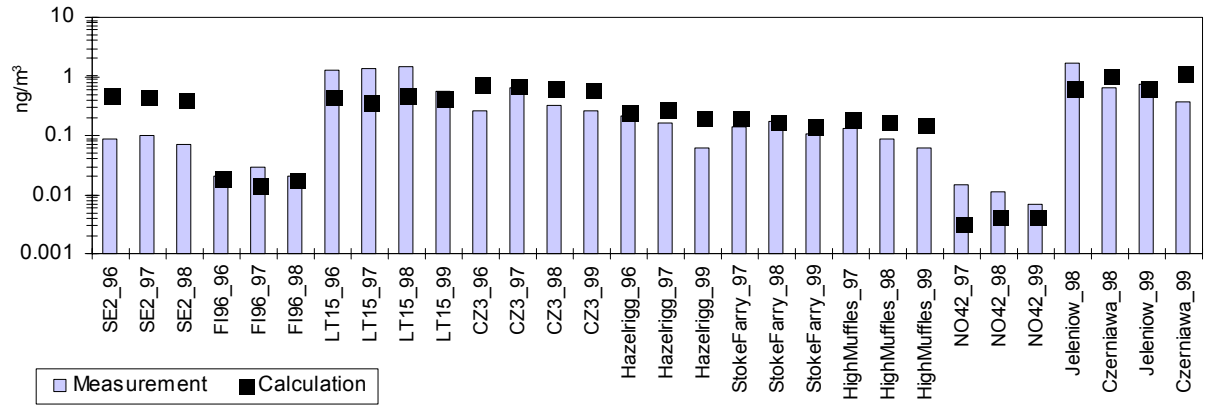


Figure 1.6. *The comparison of measured and calculated mean annual B[a]P concentrations in the surface air for 1996 - 99, ng/m^3*

Table 1.3. The comparison of measured and calculated mean annual B[a]P concentrations in the surface air for 1996-99, ng/m^3

Station	Country	Year	Measurement	Calculation	Ratio	References
SE2	Sweden	1996	0.09	0.48	0.19	*Berg et al*, 1998
		1997	0.10	0.46	0.22	*Brorstrom-Lunden,* 2000
		1998	0.07	0.40	0.17	
FI96	Finland	1996	0.02	0.02	1.04	*Berg et al*, 1998
		1997	0.03	0.01	2.02	*Brorstrom-Lunden,* 2000
		1998	0.02	0.02	1.14	
LT15	Lithuania	1996	1.26	0.46	2.75	*Milukaite,* 2001
		1997	1.33	0.37	3.56	
		1998	1.47	0.48	3.07	
		1999	0.58	0.43	1.33	
CZ3	Czech Republic	1996	0.26	0.75	0.35	*Holoubek et al,* 2000
		1997	0.64	0.69	0.93	
		1998	0.32	0.63	0.51	
		1999	0.27	0.61	0.45	
Hazelrigg	UK	1996	0.21	0.24	0.88	*Coleman et al.,* 1998, 2001
		1997	0.16	0.27	0.59	
		1998	0.14	0.19	0.7	
		1999	0.06	0.20	0.29	
Stoke Ferry		1997	0.14	0.20	0.71	
		1998	0.17	0.18	0.96	
		1999	0.11	0.14	0.81	
High Muffles		1997	0.13	0.18	0.72	
		1998	0.09	0.17	0.53	
		1999	0.06	0.15	0.40	
NO42	Norway	1997	0.015	0.003	4.63	*Berg et al*, 1999,2000,2001
		1998	0.01	0.004	2.62	
		1999	0.007	0.004	1.65	
Jeleniow	Poland	1998	1.68*	0.621	2.70	*Abraham et al.,* 2000
Czerniawa		1998	0.64*	1.05	0.61	
Jeleniow		1999	0.75*	0.625	1.20	
Czerniawa		1999	0.37*	1.14	0.32	
Averaged			**0.37**	**0.37**	**1.24**	
Min			**0.007**	**0.003**	**0.17**	
Max			**1.7**	**1.1**	**4.63**	

*- particle phase only

The ratio of measured to calculated data varies from 0.17 to 4.6 whereas measured values can differ from each other more than 200 times. For example, mean values of measured concentrations at station NO42 (Norway) is 0.007 ng/m^3, at Jeleniow station (Poland) – 1.7 ng/m^3. Consequently the model reasonably describes air concentrations in a sufficiently wide range of values.

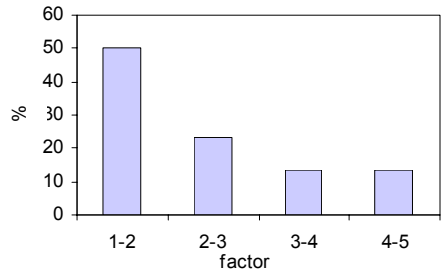


Figure 1.7. *Distribution of ratios of measured and calculated values according to the factor*

The mean value of measurement/calculation ratios is 1.2. More than a half of the ratios does not exceed a factor of 2, two thirds are within a factor of 3 (Figure 1.7). The diagram of Figure 1.7 shows percent of ratios, which is within the range of a certain factor. The factor implies the ratio of maximum of C_{meas}, C_{calc} to their minimum.

The highest discrepancy between measured and calculated data is observed for stations SE2 and NO42. For station SE2 the calculated values are 5 times higher than measured ones. This difference was analysed in the previous report [*Shatalov et al.,* 2001]. It was determined that the overestimation of the calculated concentration may be explained by the overestimated emission in the cell where station SE2 is located. The overestimation of emissions obviously is caused by close location of Göteborg city.

It should be mentioned that the consideration of B[a]P import from outside the region boundaries improved the consistency of calculated and measured values for remote regions. For example, the relation between of measured and calculated results for station NO42 reduced almost 2 times. It is 1.6 – 4.6 times instead of 4.7 – 8.9 [*Shatalov et al.,* 2001].

Concentrations in precipitation

Figure 1.8 illustrates measured and calculated mean annual concentrations in precipitation for 1996 – 99. Numerical values used in the comparison, their relations and appropriate references are given in Table 1.4. These ratios vary from 0.31 to 1.2 (factor 4). The calculated concentration values exceed measured ones on the average 1.3 times. It is better than in the previous year (factor 2.4). The correlation is 0.54. Obviously better results are obtained due to the refined coefficient of washout.

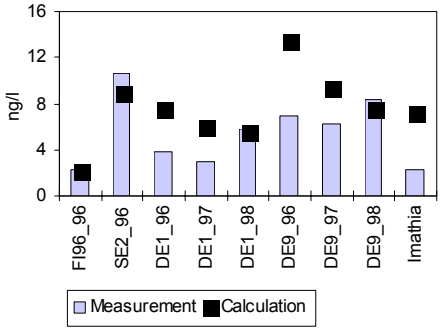


Figure 1.8. *The comparison of measured and calculated mean annual concentrations of B[a]P in precipitation in 1996-99, ng/l*

Table 1.4. The comparison of measured and calculated mean annual concentrations of B[a]P in precipitation in 1996 - 99, ng/l

Station	Country	Year	Measurement	Calculation	Ratio	Reference
FI96	Finland	1996	2.2	2.14	1.03	*Berg et al,* 1998
SE2	Sweden	1996	10.58	8.89	1.19	
DE1	Germany	1996	4.14	7.51	0.52	
		1997	2.93	5.98	0.49	
		1998	5.80	5.50	1.06	
DE9		1996	6.91	13.50	0.51	
		1997	6.26	9.31	0.67	
		1998	8.40	7.47	1.12	
Imathia	Greece	1996-97	2.23	7.25	0.31	*Manoli et al,* 2000
Mean			**5.5**	**7.5**	**0.77**	
Min			**2.2**	**2.1**	**0.31**	
Max			**10.6**	**13.5**	**1.19**	

More detailed analysis of the calculated concentrations can be made with the availability of simultaneous measurements of B[a]P content in precipitation and air. At present such measurements are reported for stations FI96 and SE2 for 1996. For station FI96 the calculated concentrations in air and precipitation actually coincide with measurements. For station SE2 the discrepancy between measured and calculated concentrations in precipitation is lower than for air (the ratio between measured and calculated values is 1.2 and 0.2 respectively). Obviously this fact may be explained by the peculiarity of sampler collecting bulk deposition (dry and wet) whereas in calculations of concentrations in precipitation only wet depositions are considered. On the whole the agreement between measured and calculated values has been refined compared with results of the previous year: mean value of measurement/calculation ratio is 0.77 (being in the range of 0.31 − 1.19) against 2.4 (ranged from 1.5 to 3.7) obtained by previous calculations.

Deposition fluxes

Figure 1.9 demonstrates measured and calculated values of total deposition fluxes for 1996-99. Numerical values of compared measured and calculated data, their ratios and appropriate references are presented in Table 1.5. Note that the calculated flux value includes the components of dry and wet deposition. The measurement/calculation ratio varies from 0.23 to 2.2 with mean value 0.9 whereas previous year calculations give this ratio in the range from 0.3 to 2.8 with the average 1.1. The correlation is 0.54.

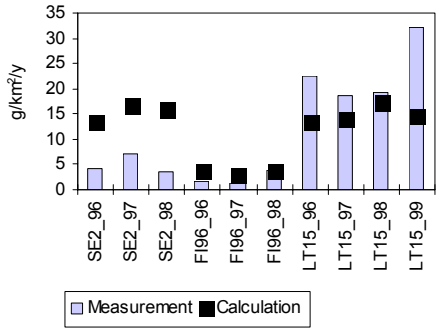


Figure 1.9. *The comparison of measured and calculated total fluxes of B[a]P for 1996-99, g/km^2/y*

Table 1.5. The comparison of measured and calculated deposition fluxes of B[a]P in 1996-99, g/km^2/y

Station	Country	Year	Measurement	Calculation	Ratio	Reference
SE2	Sweden	1996	4.02	13.54	0.30	*Brorstrom-Lunden,* 2000
		1997	6.92	16.64	0.42	
		1998	3.64	16.04	0.23	
FI96	Finland	1996	1.72	4.01	0.43	
		1997	1.20	2.95	0.41	
		1998	4.00	3.89	1.03	
LT15	Lithuania	1996	22.40	13.45	1.67	*Milukaite*, 2001
		1997	18.60	14.17	1.31	
		1998	19.26	17.35	1.11	
		1999	32.15	14.92	2.16	
Mean			**11.4**	**11.7**	**0.90**	
Min			**1.2**	**2.9**	**0.23**	
Max			**32.2**	**17.3**	**2.16**	

The best agreement between calculated and measured data was obtained for Finnish station FI96 and Lithuanian LT15. However, the measured flux for station LT15 for 1999 is 2 times higher than the calculated value. It may be explained by the local increase of emissions in the considered period.

For station SE2 the calculated values for 1996 - 99 are 3 - 4.5 times higher than measured ones. It is similar to the overestimation of calculated air concentrations (about 5 times). Hence it may be concluded that although for this station the substantial discrepancy of absolute values is obtained obviously resulted from uncertain emission data, the model parametrization of B[a]P scavenging from the atmosphere is satisfactory.

Basic results:

- Introduction of the refined parametrization of B[a]P wet deposition and the consideration of B[a]P import from outside the calculation domain on the whole improved the agreement between measured and calculated values of concentrations in air and precipitation. In particular, agreement between calculations and measurements was improved in remote regions, e.g. at NO42 site (Spitsbergen).

- More than 75% of the comparisons of measured and calculated air concentrations are within a factor of 3 the rest is within a factor of 6. A large share of the comparisons of concentrations in precipitation is within a factor of 2 except for one station (factor 4). 80% of comparisons of deposition fluxes are within a factor of 3 the rest of values is within a factor of 4. The mean calculation/measurement ratio are 1.2, 0.8 and 0.9. and correlations – 0.46, 0.54 and 0.54.

- The consideration of simultaneous measurements of concentrations in the atmosphere and precipitation pointed out that the difference between observed concentrations in precipitation mainly corresponds to differences in air concentrations.

- For the evaluation of the quality of parametrization of scavenging processes more complete set of simultaneous measurements in air and precipitation is required.

1.2. Model evaluation of B[a]P transboundary transport and air contamination

1.2.1. *B[a]P emissions*

In accordance with the POP Protocol of the Convention on Long-range Transboundary Pollution of 1979 polycyclic aromatic hydrocarbons (PAH) are referred to chemicals, which annual emissions the Parties are obliged to reduce by applying effective measures. For emission inventory the Protocol identifies 4 PAH as indicator compounds – they are benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene and indeno(1,2,3-cd)pyrene. In this report we consider the most carcinogenic compound among these polyaromatic hydrocarbons – benzo[a]pyrene.

National data and expert estimates of emissions in the EMEP domain

National data on PAH emissions for 1990-99 submitted by countries to the ECE Secretariat are demonstrated in Table 1.6. Denmark, Lithuania, Norway and Poland submitted totals of 4 indicator PAH. Hungary and Germany provided totals of 6 Borneff PAH (in addition to 4 indicator compounds this group includes fluoranthene and benzo[ghi]perylene). Russia has estimated the emission of benzo[a]pyrene only. Other countries do not identify PAH included to the inventory. It complicates the use of national data in model calculations.

Table 1.6. Official PAHs emission data, t/y

Country	UN/ECE reported official emission data									
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Austria	547	505	483	496	477	521	515	481	468	460
Belgium	354 [a]			294 [a]	235	274	184	187	187	190
Bulgaria	677					443	410	364	384	286
Croatia	11						9.3	9.2	8.6	7.9
Czech Republic	752	747	1131	1115	951	1357	971	657	657	557
Denmark [b]					10	10	11	11	10	11
Finland	16	13	13	13	17	17	17	16	16	17
France	2054	2467	2308	2282	1943	1971	2109	1886	1927	1896
Germany [c]	420				396					
Hungary [c]	132	122	87	81	72	68	63	60	54	55
Lithuania [b]								71	53	44
Luxembourg					1.1	0.6	0.7	0.4	0.3	0
Netherlands	172		142		139	128	109	107	73	73
Norway [b]	15	15	14	15	15	15	15	15	15	14
Poland [b]	163	178	176	170	236	242	230	201	181	181
Republic of Moldova	6.2	4.9	4	3.3	3.1	4.3	3.6	5.1	4.8	4.4
Russian Federation [d]	18	17	16	15	15	15	15	15	15	15
Slovakia	42					19		19	16	17
Slovenia	24				18	17	17	19	18	18
Spain	301	307	284	288	281	233	252			
Sweden	182		153			153				
Ukraine								2.9	0.77	
United Kingdom	243	230	209	159	149	122	67	55	49	44

[a] Referring to Flanders only
[b] Sum of 4 indicator PAHs
[c] Sum of 6 Borneff PAHs
[d] Benzo[a]pyrene only

Emission data for four individual compounds are available only for Denmark, Lithuania and Poland. Table 1.7 presents data on benzo[a]pyrene contribution to total emissions of 4 indicator PAH and 6 Borneff PAH calculated on the base of information submitted by these countries. For example, in 1998 the share of benzo[a]pyrene in total emissions of 4 PAH in Denmark, Lithuania and Poland was 27, 27 and 30 percent respectively. Benzo[a]pyrene fraction in total emissions of 6 Borneff PAH in Denmark and Lithuania was 12 and 17 percent. Using these relationships it is possible to estimate benzo[a]pyrene emissions from other countries, which provided total emissions of 4 or 6 PAH. Certainly these assessments will be very rough since the fraction of this or that polyaromatic hydrocarbon (in this case it is benzo[a]pyrene) in the total emission varies from country to country.

Table 1.7. B[a]P fraction in PAH emissions, %

Country	1994	1995	1996	1997	1998	1999
Denmark						
Fraction B[a]P of 4 indicator PAHs, %	27	27	27	27	27	27
Fraction B[a]P of 6 Borneff PAHs, %	9	9	12	12	12	13
Lithuania						
Fraction B[a]P of 4 indicator PAHs, %					27	
Fraction B[a]P of 6 Borneff PAHs, %					17	
Poland						
Fraction B[a]P of 4 indicator PAHs, %					30	30

Emission data of Table 1.8 were used in calculations of B[a]P transboundary transport in 1999. These data are a mixture of official and expert estimates for different years. Benzo[a]pyrene official emissions are available only for Denmark, Poland and Russia. B[a]P emissions from Germany, Hungary, Lithuania and Norway were estimated as an appropriate fraction of provided national total (on the base of the data presented in table 1.7 it was assumed that B[a]P fraction of 4 indicator PAH is 30% and of - Borneff PAHs – 15%). For the rest of countries expert estimates were used [*Pacyna et al.,* 1999; *Tsybulsky,* 2001; *Berdowski et al*., 1997]. In [*Pacyna et al.,* 1999] B[a]P emissions and their spatial distribution are estimated. In [*Tsybulsky,* 2001] emissions of each Borneff PAHs for the CIS and Baltic countries are estimated. In [*Berdowski et al.,* 1997] total emission of 6 Borneff PAHs and their spatial distribution in European countries are estimated. Table 1.8 contains also comments on the data used for each European country.

Emission spatial distribution

Benzo[a]pyrene emission spatial distribution obtained on the base of the data in Table 1.8 is illustrated in Figure 1.12.a. Bulgaria (for 1990 and 1995), Finland (for 1990 - 99) and Spain (for 1990 - 96) submitted data on PAH spatial distribution. These spatial distributions were used for preparation of B[a]P emission fields for these countries. For Iceland and Cyprus we used the emission distribution estimated by [*Berdowski et al.,* 1997]. For the rest of countries emission spatial distribution estimated by [*Pacyna et al.,* 1999] was used.

Table 1.8. Emissions of B[a]P in European countries in 1999 used in calculations, t/y

Country	B[a]P emissions	Comments
Albania	0.22	Expert estimates for 1995 [*Pacyna et al*., 1999]
Armenia	0.27	Expert estimates for 1997 [*Tsibulsky*, 2001]
Austria	6.1	Expert estimates for 1995 [*Pacyna et al*., 1999]
Azerbaijan	2.1	Expert estimates for 1997 [*Tsibulsky*, 2001]
Belarus	3.6	Expert estimates for 1997 [*Tsibulsky*, 2001]
Belgium	3.4	Expert estimates for 1995 [*Pacyna et al*., 1999]
Bosnia&Herzegovina	4.5	Expert estimates for 1995 [*Pacyna et al*., 1999]
Bulgaria	6.7	Expert estimates for 1995 [*Pacyna et al*., 1999]
Croatia	4.7	Expert estimates for 1995 [*Pacyna et al*., 1999]
Cyprus	0.03	Evaluated as 15 % of 6 Borneff emissions in 1990 [*Berdowski et al*., 1997]
Czech Republic	14	Expert estimates for 1995 [*Pacyna et al*., 1999]
Denmark	3.0	Official data for 1999
Estonia	3.7	Expert estimates for 1997 [*Tsibulsky*, 2001]
Finland	6.9	Expert estimates for 1995 [*Pacyna et al*., 1999]
France	26	Expert estimates for 1995 [*Pacyna et al*., 1999]
Georgia	11	Expert estimates for 1997 [*Tsibulsky*, 2001]
Germany	59	Evaluated as 15 % of 6 Borneff emissions (official data for 1994)
Greece	2.9	Expert estimates for 1995 [*Pacyna et al*., 1999]
Hungary	8.2	Evaluated as 15 % of 6 Borneff emissions (official data for 1999)
Iceland	0.95	Evaluated as 15 % of 6 Borneff emissions in 1990 [*Berdowski et al*., 1997]
Ireland	1.2	Expert estimates for 1995 [*Pacyna et al*., 1999]
Italy	14	Expert estimates for 1995 [*Pacyna et al*., 1999]
Kazakhstan	4.3	Expert estimates for 1995 [*Pacyna et al*., 1999]
Latvia	6.9	Expert estimates for 1997 [*Tsibulsky et al*., 2001]
Lithuania	12	Estimated as 30 % from 4 indicator PAH emissions (official data for 1999)
Luxembourg	0.00	Official data on PAH emissions for 1999
Netherlands	2.3	Expert estimates for 1995 [*Pacyna et al*., 1999]
Norway	4.1	Evaluated as 30 % from 4 indicator PAH emissions (official data for 1999)
Poland	53	Official data for 1999
Portugal	1.6	Expert estimates for 1995 [*Pacyna et al*., 1999]
Republic of Moldova	0.35	Expert estimates for 1997 [*Tsibulsky*, 2001]
Romania	19	Expert estimates for 1995 [*Pacyna et al*., 1999]
Russian Federation	15	Official data for 1999
Slovakia	6.7	Expert estimates for 1995 [*Pacyna et al*., 1999]
Slovenia	2.4	Expert estimates for 1995 [*Pacyna et al*., 1999]
Spain	9.6	Expert estimates for 1995 [*Pacyna et al*., 1999]
Sweden	6.8	Expert estimates for 1995 [*Pacyna et al*., 1999]
Switzerland	1.7	Expert estimates for 1995 [*Pacyna et al*., 1999]
The FYR of Macedonia	1.9	Expert estimates for 1995 [*Pacyna et al*., 1999]
Ukraine	16	Expert estimates for 1997 [*Tsibulsky* , 2001]
United Kingdom	12	Expert estimates for 1995 [*Pacyna et al*., 1999]
Yugoslavia	11	Expert estimates for 1995 [*Pacyna et al*., 1999]
Total	371	

Countries, which official data were used in calculations are given in bold

1.2.2. *Evaluation of deposition and concentrations*

In this section we describe B[a]P concentration and deposition spatial distribution for 1999 obtained with the use of the emission scenario including expert estimates and official emission data (Table 1.8). Figure 1.10 shows fields of mean annual concentrations (1.10.a) and maximum diurnal concentrations (1.10.b) in the surface air in 1999.

In the major part of European countries mean air concentrations are within the range of 0.1-1 ng/m^3. Values lower than 0.1 ng/m^3 are characteristic of Spain, Scandinavia, the northern part of the United Kingdom and Russia.

Over vast areas of Poland and in some regions of Germany, the Czech Republic, Slovakia, Hungary, Yugoslavia and Georgia mean annual concentrations are high – more than 1 ng/m^3. It exceeds B[a]P limit value set up in a number of European countries [*Ежегодник,* 1994; *Policy on* …, 1994, *Zurek et al*., 2000]. Maximum values of mean annual concentrations do not exceed 5 ng/m^3.

In the report of the previous year we substantiated the significance of the consideration of maximum diurnal concentrations, which on the average are 5 times higher than mean annual ones (Figure 1.10). It is conditioned both by the influence of meteorological parameters and seasonal irregularity of emissions. Essential diurnal concentrations (more than 1 ng/m^3) are obtained for the majority of countries of Central Europe. In 1999 particularly high concentrations (more than 2 - 20 ng/m^3) were observed in Poland, Germany, the Czech Republic (the Black Triangle), Lithuania and Latvia. These concentrations were obtained for a cold season when B[a]P emissions are highest. It is clearly seen in Figure 1.11.a demonstrating B[a]P air concentration variations over European land in 1999. To a great extent this variation is defined by seasonal variations of emissions degradation and by dry deposition velocity [*Shatalov et al.,* 2000]. Figure 1.11.b gives a map of a number of days with mean diurnal concentrations exceeding 1 ng/m^3. It is seen that for the most part of European territory the exceedance may last more than 100 days.

The value of deposition fluxes over the continental part of the EMEP region (Fig. 1.13) varies from 1 to 180 g/km^2/y. Essential fluxes (100 - 180 g/km^2/y) are observed in regions with intensive emissions: Poland, the Czech Republic, Slovakia, Germany, Lithuania and Latvia. On the whole in 1999 184 tonnes of B[a]P deposited to the EMEP domain that amounts to half of the annual emissions.

It should be mentioned that emission values for some countries used in calculations for 1999 may be somewhat different from those used in calculations for 1998 [*Shatalov et al.,* 2001] that affects the calculation results. The pollution estimate for 1998 was made with the use of two emission scenarios including both expert estimates and available official emission data. The greatest difference in emission data, which resulted in a noticeable change of pollution levels for 1999, is in Germany (Table 1.9). According to official data emissions increased more than twice in this country in 1999. It resulted in almost two-fold increase of mean annual concentrations and deposition fluxes to this country. Besides the contamination of the neighboring countries and regions was enhanced.

The information about pollution levels in individual countries can be found in Internet: http://www.msceast.org/countries/idex.html. For example, Figure 1.14 gives maps of emissions, total depositions and mean annual concentrations for Finland.

Table 1.9. The comparison of emissions and calculated values of mean annual air concentration and deposition fluxes in Germany for 1998 and 1999

	1998	1999
Emissions, tonnes	26.4 (expert estimates)	59 (official data)
Atmospheric concentrations, ng/m^3	0.46	0.8
Depositions, g/km^2/y	26.8	56

As seen from the figure 1.14 powerful emission sources are located in the south of the country. Here emission fluxes reach 250 g/km^2/y. They are 6 times lower than maximum values (1560 g/km^2/y) for Europe. This region is characterized by relatively high depositions (to 47 g/km^2/y) and mean annual air concentrations (0.9 ng/m^3).

Basic results:

- Essential deposition fluxes (100-180 g/km^2/y) are observed in regions with appreciable B[a]P emissions: Poland, the Czech Republic, Slovakia, Germany, Lithuania and Latvia.

- For the majority of European countries mean annual concentrations in the surface air are within the range of 0.1-1 ng/m^3. Over vast areas of Poland and in some regions of Germany, the Czech Republic, Slovakia, Hungary, Yugoslavia and Georgia mean annual concentrations are high: from 1 to 5 ng/m^3.

- Maximum mean diurnal air concentrations on the average are 5 times higher than mean annual concentrations. High values of diurnal concentrations 2 – 20 ng/m^3 are characteristic of Poland, Germany, the Czech Republic (the Black Triangle), Lithuania and Latvia. In the same countries mean diurnal concentrations exceeding 1 ng/m^3 can occur for more than 100 days a year. Detailed information about spatial distribution of emission, concentration and deposition fluxes in the surface air in European countries can be found in Internet (http://www.msceast.org/countries/idex.html).

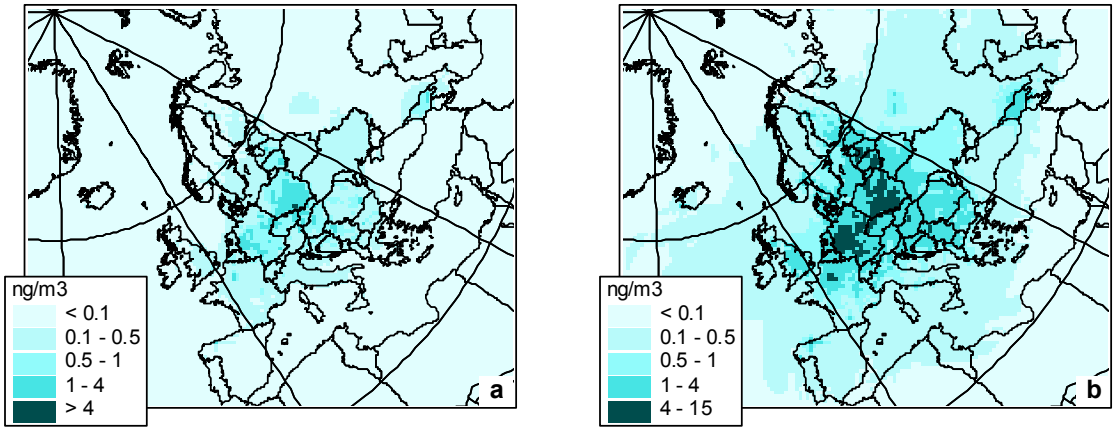


Figure 1.10. **a** - *B[a]P annual mean concentrations and* **b** - *maximum mean diurnal concentrations in the surface air in 1999, ng/m^3*

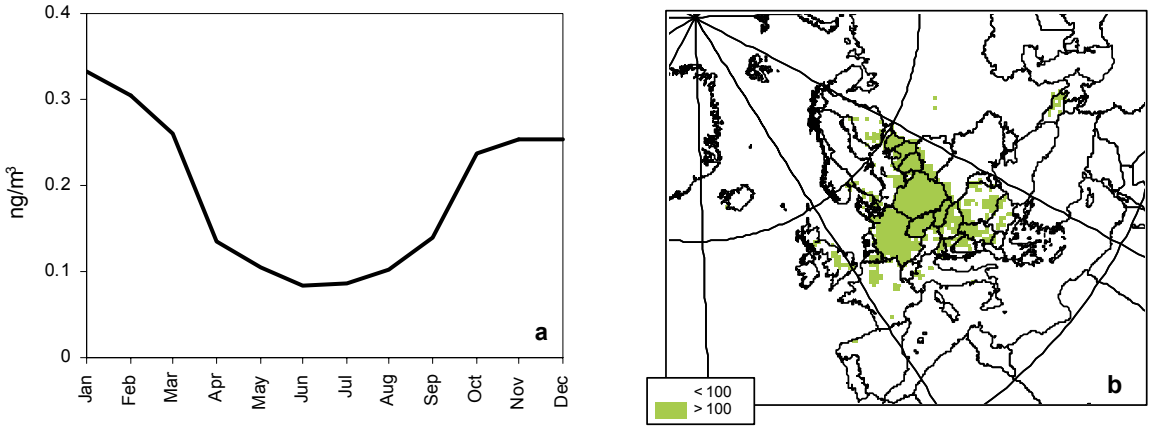


Figure 1.11. **a** - *Seasonal variations concentration of B[a]P air concentrations over land in 1999, ng/m^3;* **b** - *number of days with mean diurnal concentrations exceeding 1 ng/m^3 in 1999*

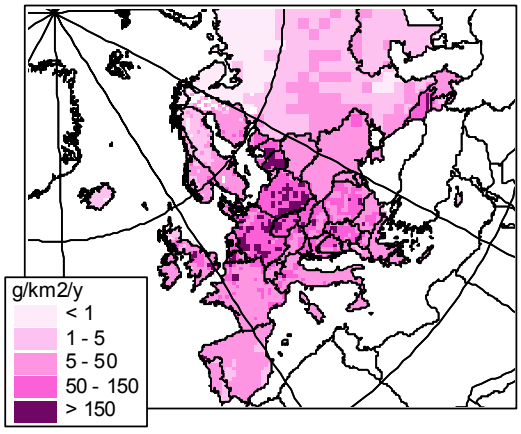


Figure 1.12. *B[a]P emissions, g/km^2/y*

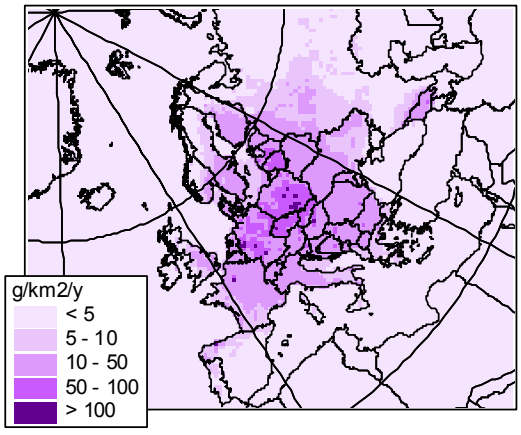
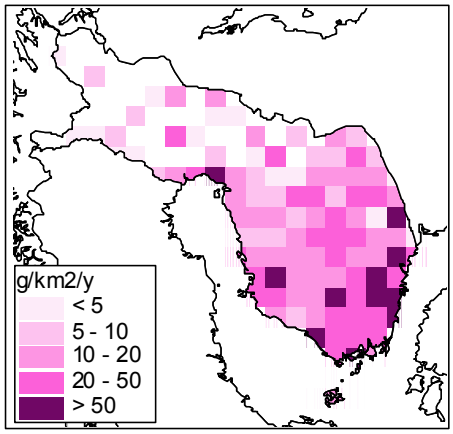
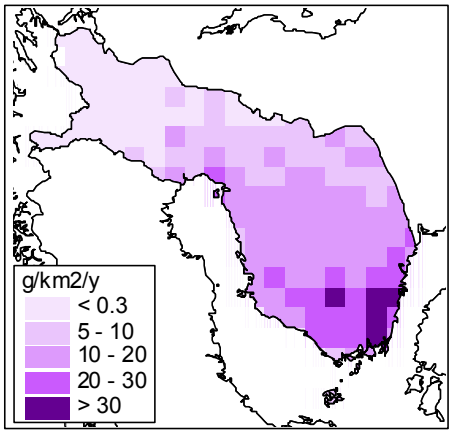


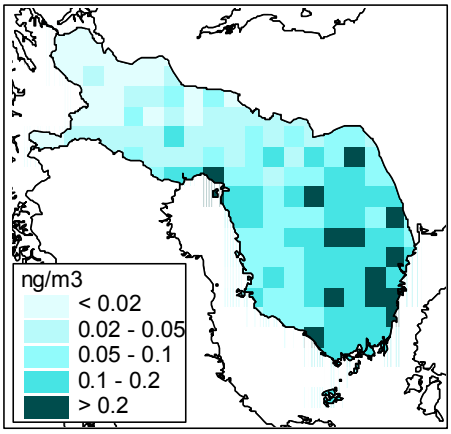
Figure 1.13. *B[a]P depositions in 1999, g/km^2/y*



Emission, g/km^2/y



Total depositions, g/km^2/y



Mean annual concentrations in the surface air, ng/m^3

Figure 1.14. *B[a]P pollution levels in Finland in 1999*

1.2.3. *Assessment of transboundary transport*

For the evaluation of transboundary transport in 1999 "country-to-country" matrices of depositions and concentrations were calculated with spatial resolution 50x50 km. In calculation expert estimates and available official emission data (section 1.2.1) for 1999 were used.

Air concentrations

One of the main pathways of B[a]P penetration to human organism is the respiratory system. Therefore investigations of the impact of transboundary fluxes on the formation of air concentrations are important. In this context the matrix of "country-to-country" mean annual concentrations was calculated (see Annex B). Each element of the matrix gives the contribution to a country-emitter corresponding to the column to air concentration of country/region of the corresponding row. Total air concentrations of countries are given in the last column.

The greatest contribution to air concentration from external sources (import) was obtained for Slovakia (482 pg/m^3), the Czech Republic (338 pg/m^3) and Hungary (323 pg/m^3). The predominance of internal emission sources over external ones is observed in the Czech Republic, Poland, Lithuania, Yugoslavia and some other countries (Fig.1.15).

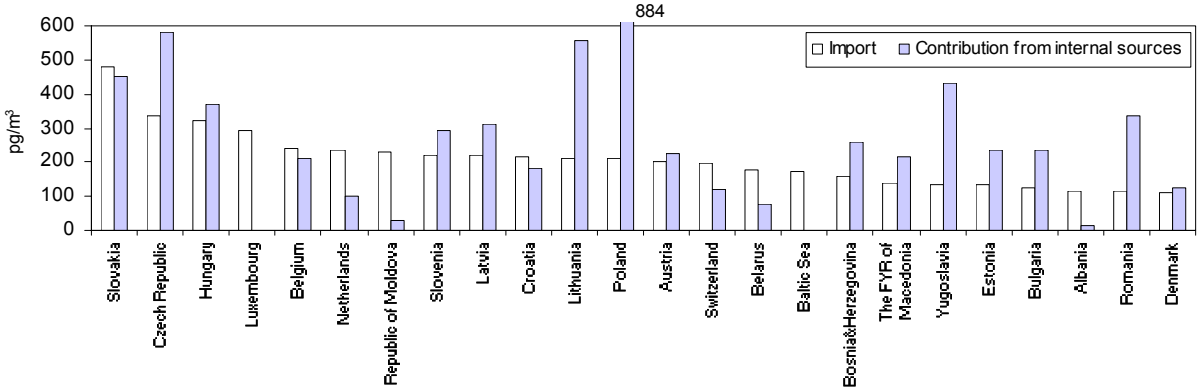


Figure 1.15 *B[a]P air concentrations in European countries caused by external (import) and internal sources in 1999, pg/m^3*

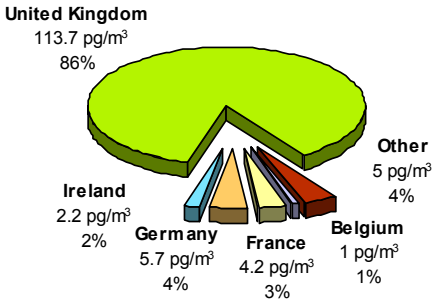


Figure 1.16. *Contributions of external (import) and internal B[a]P emission sources to air concentrations of the UK in 1999*

On the example of the UK more detailed consideration of the effect of external and internal B[a]P emission sources on the formation of mean annual concentrations is given. Concentration values from the indicated sources are shown in the pie chart of Figure 1.16.

As seen from the diagram, the greatest contribution to concentrations is made by national sources (86%). Then come external sources located in Germany (4%), France (3%), Ireland (2%) and Belgium (1%).

Depositions

This section is dedicated to the analysis of "country-to-country" deposition matrices (Table B.1 Annex B). In particular deposition fluxes from individual countries to European countries/regions and depositions to a given country from European countries were assessed on the base of these matrices. Each element of the matrix represents B[a]P quantity deposited in 1999 from a country corresponding to a given column to a country/region corresponding to a given row. The last column gives total depositions to corresponding countries.

Depositions from countries. B[a]P emitted by a country enters the atmospheric air, degrades there or falls out to its own territory and to territories of other countries and it is transported outside the calculation grid. Pollutant deposition from a given country to other countries and regions within the EMEP domain is called export. The scheme clarifying these processes is shown in Figure 1.17.

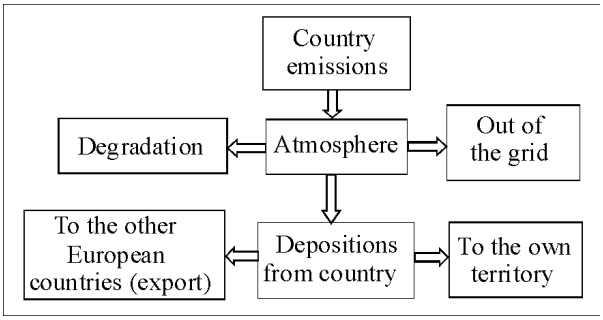


Figure 1.17 *B[a]P emission distribution from each European country*

According to the emission scenario used in calculations in 1999 appreciable quantities of B[a]P export (not less than 4 tonnes) were calculated for Germany (13.6 t/y), Poland (13 t/y), France (5.6 t/y), the Czech Republic (4.9 t/y), Romania (4.9 t/y). The export values from these and a number of other countries are given in Figure 1.18.

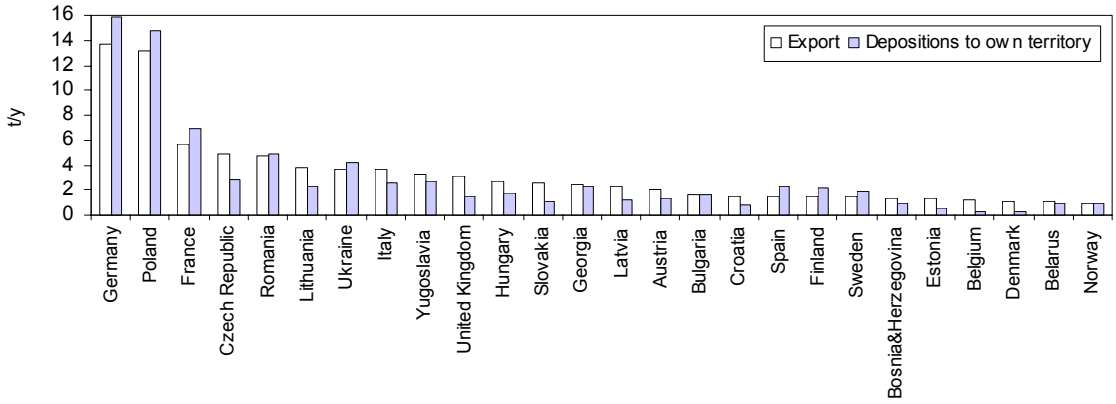


Figure 1.18. *B[a]P depositions from some European countries to other countries/regions (export) and to own territory, t/y*

The figure 1.18 evidences that for such countries as Germany, Poland, France, Romania, the Ukraine, Spain, Finland and Sweden B[a]P depositions to own territory exceed the export. For other countries given in the figure a reverse pattern is observed. Three countries: Germany, Poland and France make the greatest total contributions to European region (export + depositions to own territory) – 30, 28 and 13 t/y, respectively.

The calculations made allow us to analyse in detail fields of depositions from each country. For instance, Figure 1.19 shows B[a]P deposition field from the United Kingdom and the pie chart demonstrating the distribution of depositions from the United Kingdom with European countries/regions. As seen from the chart the highest amount of depositions falls out to the United Kingdom itself (1470 kg, 31%). Then come the North Sea (639 kg, 14%), the Atlantic Ocean (538 kg, 12%), France (298 kg, 6%) and Germany (260 kg, 6%). Deposition to the other regions amounts to 1430 kg (31%).

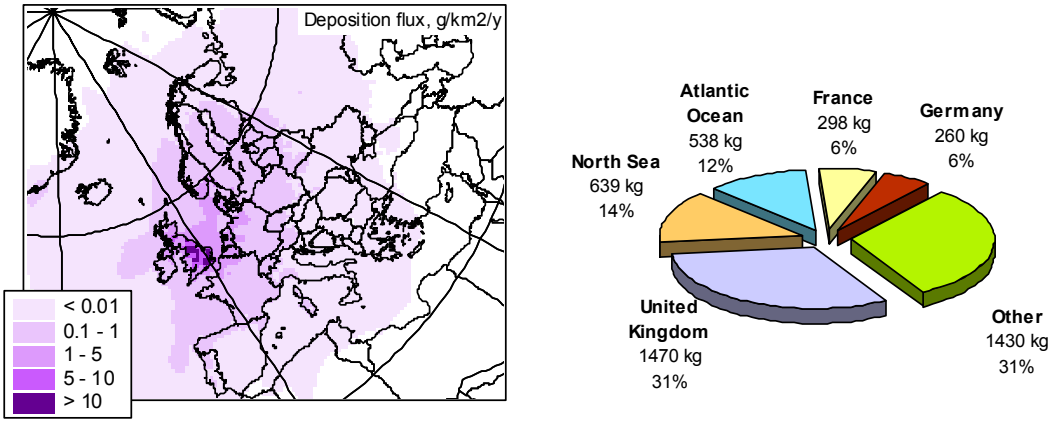


Figure 1.19. *B[a]P depositions from the UK emission sources to the European countries and regions in 1999*

Depositions from the United Kingdom may be considered from the viewpoint of their significance for the pollution of other countries and regions. Figure 1.20 shows fractions of deposition from UK sources in total depositions to various European countries.

As it follows from the figure depositions to the United Kingdom make up 75% of all depositions to this country. UK depositions to the North Sea, Ireland, Norway, Denmark and the Netherlands make up 31, 18, 9, 8 and 6% correspondently.

Depositions to countries. The analysis of B[a]P depositions to European countries and regions will be also exemplified by the United Kingdom. Figure 1.21 shows B[a]P emission fluxes and total deposition flux.

The comparison of these two maps shows that the most intensive depositions are observed in the southern part of the UK reaching 29 g/km^2/y. In the same region essential emission flux of B[a]P, which value reaches 380 g/km^2/y, takes place. The lowest depositions are in the north of the country (0.5 - 5 g/km^2/y). The average value of deposition is 8.4 g/km^2/y.

The role of B[a]P transport in depositions to the UK may be estimated by calculations and represented in the form of a map with percent of deposition from external sources to the UK (1.22.a) and a pie chart of the contribution of European countries to B[a]P depositions to the UK (Fig. 1.22.b).

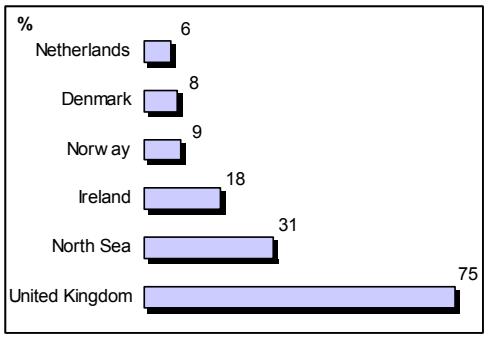


Figure 1.20. *B[a]P deposition fraction from the UK sources in total deposition to some European countries and regions in 1999*, %

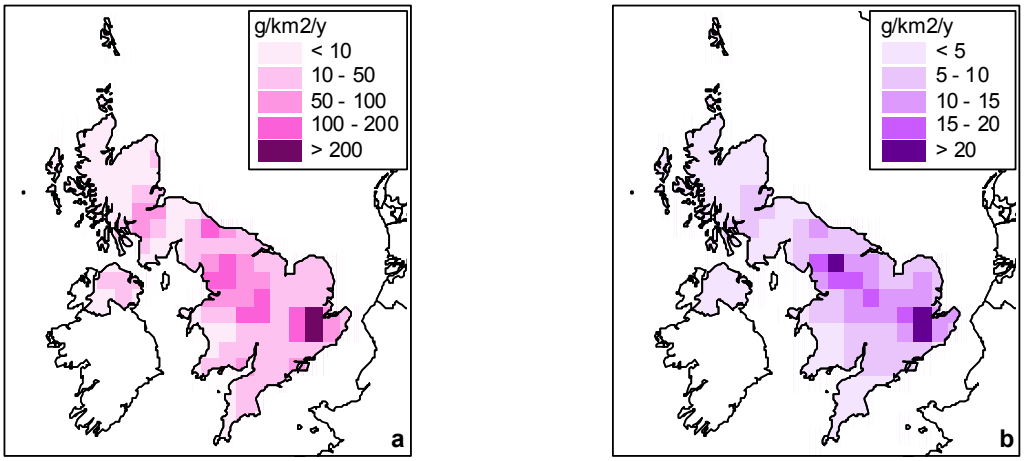


Figure 1.21. *B[a]P emission flux - **a** and total deposition flux - **b** to the UK in 1999, g/km^2/y*

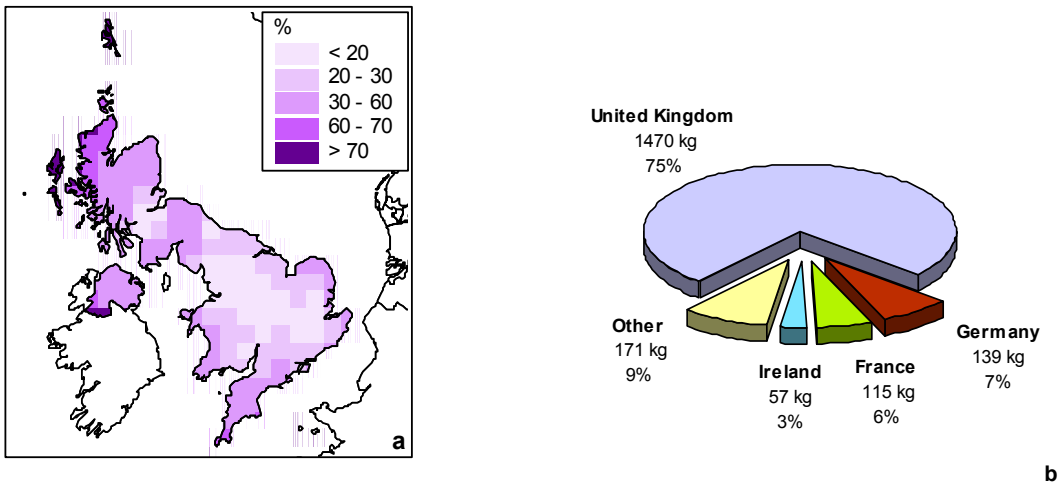


Figure 1.22. ***a*** *– percent of depositions from external sources to the UK in 1999*
b *– contributions of European countries to B[a]P depositions to the UK in 1999*

As seen from the map the fraction of depositions from external sources is highest in the north of the UK (50 - 70%) where own emission sources are insignificant. It should be mentioned that in this region the absolute value of deposition is not high (0.5 - 5 g/km^2/y).

As clear from the diagram (Fig.1.22.b) 75% (1470 kg) of deposition is accounted for sources of the UK itself. 7% (140 kg) gives Germany, 6% (115 kg) – France, 3% - Ireland (57 kg), 1% (29 kg) – boundary sources. The fraction of other sources is 9% (171 kg). Remote sources imply sources located outside the EMEP grid. On the whole this diagram is comparable qualitatively with the diagram of Figure 1.16 above showing air concentrations in the UK. For instance, the deposition from national sources is 75% and concentration is 86%. The main external sources of depositions and concentrations are located in the same countries except for one source, which input to the UK air pollution is estimated as 1% (Belgium).

For other European countries the aggregated information about main sources of pollution is given in Table 1.9. It provides deposition values from three main countries-sources contaminating the considered country/region. In addition it gives values of emissions and total deposition to countries/regions and the contribution of external sources to depositions to them.

Table 1.9. B[a]P main sources contaminating European countries

Country/region-receptor	Depositions from main countries-sources						Emis	Total dep	% external sources
	kg	%	kg	%	kg	%	kg	kg	%
Albania	Yugoslavia, 135	39	Macedonia, 53.8	16	Italy, 29.	8	221	347	93
Armenia	Georgia, 66.8	44	Azerbaijan, 43.9	29	Armenia, 35.2	23	272	151	77
Austria	Austria, 1417	40	Germany, 678	19	Czech Rep, 371	10	6109	3541	60
Azerbaijan	Azerbaijan, 288	64	Georgia, 123.2	27	Armenia, 11.5	3	2061	449	36
Belarus	Poland, 1498	33	Belarus, 890	20	Lithuania, 544	12	3626	4487	80
Belgium	Belgium, 329.8	34	France, 291.4	30	Germany, 221	23	3350	961	66
Bos&Herzegov.	Bosn&Herz, 981	49	Yugoslavia, 261	13	Croatia, 236	12	4517	2004	51
Bulgaria	Bulgaria, 1564	56	Romania, 657	23	Yugoslavia, 197	7	6703	2811	44
Croatia	Croatia, 754	36	Bosn&Herz, 234	11	Slovenia, 225	11	4663	2108	64
Cyprus	Cyprus, 1.7	14	Greece, 1.5	12	Romania, 1.4	12	27	12	86
Czech Republic	Czech Rep, 2898	52	Germany, 1201	21	Poland, 703	13	14250	5595	48
Denmark	Denmark, 203.7	35	Germany, 177.3	30	Un King, 46.3	8	3045	590	65
Estonia	Estonia, 557	44	Latvia, 203	16	Poland, 119	9	3688	1263	56
Finland	Finland, 2127	54	Poland, 286	7	Germany, 256	7	6877	3910	46
France	France, 6908	71	Germany, 1188	12	Spain, 368	4	26383	9669	29
Georgia	Georgia, 2324	94	Azerbaijan, 55	2	Russia, 27	1	11374	2460	6
Germany	Germany, 15923	79	France, 1091	5	Poland, 639	3	59367	20052	21
Greece	Greece, 343	30	Bulgaria, 234	21	Yugoslavia, 111	10	2888	1125	70
Hungary	Hungary, 1717	41	Slovakia, 446	11	Poland, 378	9	8184	4191	59
Iceland	Iceland, 60.5	58	Boundary, 11.5	11	Germany, 5.9	6	948	104	42
Ireland	Ireland, 137.8	54	United King, 47	18	Germany, 20.3	8	1237	257	46
Italy	Italy, 2616	60	France, 384	9	Germany, 338	8	13896	4381	40
Kazakhstan	Kazakhstan, 706	49	Russia, 149	10	Georgia, 142	10	4259	1450	51
Latvia	Latvia, 1190	44	Lithuania, 620	23	Poland, 325	12	6849	2714	56
Lithuania	Lithuania, 2271	59	Poland, 717	18	Latvia, 270	7	11993	3877	41
Luxembourg	Germany, 32.7	47	France, 22.2	32	Belgium, 7.	10	0	70	100
Malta	Italy, 0.2	28	France, 0.1	14	Germany, 0.06	9	0	1	100
Netherlands	Germany, 281	36	Netherlands, 169	22	France, 124.1	16	2289	783	78
Norway	Norway, 895	52	Germany, 177	10	Un. King, 151	9	4108	1735	48
Poland	Poland, 14847	70	Germany, 2353	11	Cz. Rep., 1498	7	53463	21295	30
Portugal	Portugal, 299.8	58	Spain, 145.6	28	Boundary, 24	5	1638	518	42
Moldova	Romania, 224.8	44	Ukraine, 91.3	18	Moldova, 39.8	8	350	514	92
Romania	Romania, 4910	64	Yugoslavia, 613	8	Hungary, 355	5	18972	7660	36
Russia	Russia , 6527	31	Poland, 2615	12	Ukraine, 1628	8	15312	20934	69
Slovakia	Slovakia, 1109	37	Poland, 688	23	Hungary, 378	12	6701	3027	63
Slovenia	Slovenia, 383.8	45	Austria, 109.7	13	Croatia, 93.9	11	2409	846	55
Spain	Spain, 2280	77	France, 306	10	Portugal, 147	5	9602	2970	23
Sweden	Sweden, 1854	38	Germany, 700	14	Poland, 440	9	6773	4855	62
Switzerland	Switzerland, 269	29	Germany, 228	24	France, 178.8	19	1653	931	71
Macedonia	Macedonia, 309	48	Yugoslavia, 156	24	Bulgaria, 62.5	10	1902	645	52
Turkey	Georgia, 274	20	Romania, 221	16	Bulgaria, 168	12	0	1362	100
Ukraine	Ukraine, 4138	41	Poland, 1794	18	Romania, 1080	11	16091	9978	59
United Kingdom	Un. King, 1470	75	Germany, 139	7	France, 115	6	11986	1951	25
Yugoslavia	Yugoslavia, 2665	65	Romania, 248	6	Bos&Herz, 221	5	11236	4080	35
Africa	Italy, 184	17	France, 141	13	Boundary, 99	9		1104	100
Asia	Boundary, 113.8	26	Azerbaijan, 87.9	20	Georgia, 72.4	17		430	100
Atlantic Ocean	Boundary, 1709	36	Un. King., 538	11	Germany, 513	11		4743	100
Baltic Sea	Germany, 807	23	Poland, 754	21	Sweden, 306	9		3575	100
English channel	France, 113.5	39	Un. King., 88.5	31	Germany, 35.5	12		289	100
North Sea	Un. King., 639	31	Germany, 454	22	France, 245	12		2050	100
Mediterran. Sea	Italy, 1498	25	France, 885	15	Greece, 399	7		6030	100
Black Sea	Romania, 418	24	Ukraine, 381	21	Georgia, 266	15		1778	100
Caspian Sea	Georgia, 135.1	36	Azerbaijan, 71.	19	Kazakhstan, 61	16		373	100
Arctic Region	Boundary, 399	27	Germany, 153	10	Norway, 128	9		1476	100

To analyse the effect of transboundary transport on the pollution of European countries Figure 1.23 demonstrates B[a]P deposition fluxes from external (import) and internal sources for European countries with mean import flux exceeding mean value of 11 g/km^2/y. The countries are given in the order of import decrease.

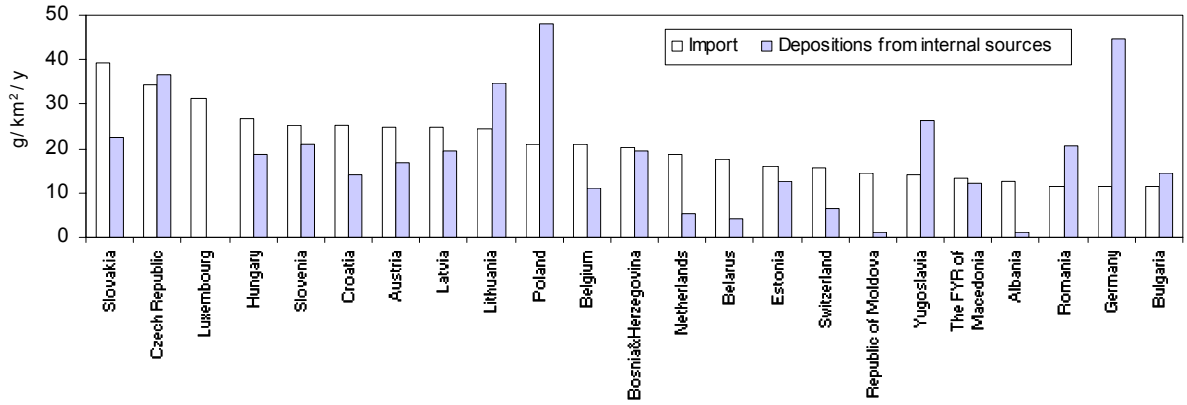


Figure 1.23. *B[a]p deposition flux to European countries from external sources (import) and internal sources, g/km^2/y*

As it follows from the diagram the highest deposition fluxes from external sources are obtained for Slovakia, the Czech Republic and Luxembourg (40, 34 and 31 g/km^2/y slightly respectively). It should be mentioned that for the majority of considered countries the import slightly exceed depositions from national sources. Exceptions are the Czech Republic, Lithuania, Poland, Yugoslavia, Romania and Germany, i.e. countries with own substantial emissions.

More detailed information about the transboundary transport and its role in the formation of depositions and concentrations in European countries can be found in Internet: http://www.msceast.org/countries/index.html.

According to the official and estimated emission the modelling results show that.:

- B[a]P transport from the majority of European countries to territories of other countries and the EMEP region (export) are comparable with depositions to their own territories thereby indicating an essential role of transboundary transport for the pollution of European countries and regions.

- Great contribution to depositions to European countries and regions is made by Germany (30 t/y), Poland (28 t/y) and France (13 t/y).

- High deposition fluxes from external sources receive Slovakia, the Czech Republic and Luxembourg (40, 34 and 31 g/km^2/y).

- High air concentrations caused by external sources were obtained for Slovakia, the Czech Republic and Hungary (482, 338 and 323 pg/m^3).

1.3. Conclusive remarks

Due to model modification, its verification and calculations of B[a]P pollution levels and transboundary fluxes in European region in 1999 the following results were obtained:

The refined parametrization of wet deposition and the consideration of B[a]P input from remote sources located outside the EMEP domain increased total deposition in Europe by 15 - 20% and provided more accurate description of concentration in precipitation and deposition fluxes. In addition better estimates of B[a]P air concentrations are obtained for remote regions. For example, the discrepancy between measured and calculated data for NO2 station (Spitzbergen) decreased more than 2 times.

The comparison of calculation results against measurements show that more than 75% of calculated air concentrations are within a factor of 3 with respect to measurements and the rest is within a factor of 6. Concentrations in precipitations and depositions are described more precisely than concentrations (factor 4).

Deposition fluxes (100 - 180 g/km^2/y) are observed in regions with considerable B[a]P emissions: Poland, the Czech Republic, Slovakia, Germany, Lithuania and Latvia.

For the major part of the territory of European countries mean annual concentration values are within the range of 0.1 - 1 ng/m^3. Over vast territories of Poland, in some regions of Germany, the Czech Republic, Slovakia, Hungary, Yugoslavia and Georgia of mean annual concentrations is rather high – from 1 to 5 ng/m^3. At the same time for mean diurnal concentrations in winter, when B[a]P emission are maximum high concentration level (more than 1 ng/m^3) is reached in the majority of countries in Central Europe. Particularly high values in 1999 (2 – 20 ng/m^3) were calculated for Poland, Germany, the Czech Republic (the Black Triangle), Lithuania and Latvia. In these countries mean diurnal concentrations exceeding 1 ng/m^3 may be observed during more than 100 days per year.

The analysis of transboundary transport was carried out on the base of examination of “country-to-country” matrixes for depositions and concentrations. It showed that contribution of external emission sources of B[a]P into contamination of some European countries are rather considerable. For instance, it was demonstrated, that long-range transport contribution to the air concentration may reach a value of 0.5 ng/m^3.

Chapter 2

EVALUATION OF PCDD/F LONG-RANGE TRANSPORT AND ACCUMULATION (1970 – 1999)

This chapter is devoted to investigation of PCDD/F behaviour in the environment from the viewpoint of toxicity and tentative evaluation of their long-range transport and accumulation.

2.1. Investigation of PCDD/F environmental behaviour and model modification

2.1.1. *Congener composition of PCDD/F mixture*

PCDD/F mixture contains 210 congeners combined in 14 homologue groups with different chlorination level. Among these congeners toxicity coefficients are assigned to 17 congeners belonging to 10 homologue groups. Physical-chemical properties of these congeners are different leading to differences in their environmental behaviour. For this reason to improve the assessment of the environmental pollution by dioxins/furans it is necessary to take into account not only the total toxicity but also congener composition of the mixture (in emissions and in environmental media). In the previous report [*Shatalov et al.,* 2001] 8 congeners of PCDD/F, which make a decisive contribution to the mixture toxicity were distinguished. Calculations of the long-range transport and accumulation were carried out for four of them (Table 2.1).

Table 2.1. Congeners selected for modelling (congeners considered earlier [*Shatalov et al*., 2001] are given in bold)

Homologue group	Congener
TCDD	2,3,7,8-TCDD
PeCDD	**1,2,3,7,8-PeCDD**
HxCDD	1,2,3,6,7,8-HxCDD
	1,2,3,7,8,9-HxCDD
TCDF	2,3,7,8-TCDF
PeCDF	**2,3,4,7,8-PeCDF**
HxCDF	**1,2,3,6,7,8 HxCDF**
	1,2,3,4,7,8- HxCDF

The aims of this section are:

- Investigation of the homologue toxicity profiles in different media (i.e. the contribution of PCDD/F different homologue groups to the total toxicity of the mixture).
- Investigation of behavioural peculiarities of PCDD/F different congeners in the environment.

For this purpose 8 model runs of PCDD/F long-range transport and accumulation were made. In each run physical-chemical properties of one of eight congeners were subsequently assigned to the whole mixture. Then calculations of PCDD/F mixture transport with emissions distributed between the eight congeners in accordance with the results obtained in [*Vulykh and Shatalov*, 2001] were carried out.

Homologue toxicity profiles. In this section we describe homologue toxicity profiles defined as a result of calculations of PCDD/F mixture transport and accumulation during 1970 – 99 with the use of emission expert estimates and the comparison with profiles calculated on the base of measurement data. The comparison of toxicity profiles for the atmosphere, soil and vegetation is shown in Figure 2.1 a – c. Toxicity profile in seawater is not considered since measurements in seawater are not available at this stage.

The greatest contribution to total toxicity of dioxins/furans mixture is made by groups of penta- and hexachlorinated dibenzofurans (PeCDF and HxCDF). Then come (in the order of contribution significance to the total toxicity) groups of penta- and hexaclorinated dibenzo-p-dioxins and tetrachlorinated dibenzo-p-dioxins and dibenzofurans. The contribution of the rest of homologue groups to the mixture toxicity in air is only 25%.

Like in the case of the atmosphere congeners from the group of penta- and hexachlorodibenzofurans make an essential contribution to soil and vegetation toxicity. However, the homologue profile of toxicity in vegetation is characterized by appreciable contributions of light congeners from the group of tetrachlorodibenzo-p-dioxins and tetrachlorodibenzofurans. The input of hepta- and octachlorodibenzo-p-dioxins/furans is less than 25% both for vegetation and soil.

The results indicate that ***modelling results are in a reasonable agreement with measured PCDD/F toxicity profiles in the considered media.***

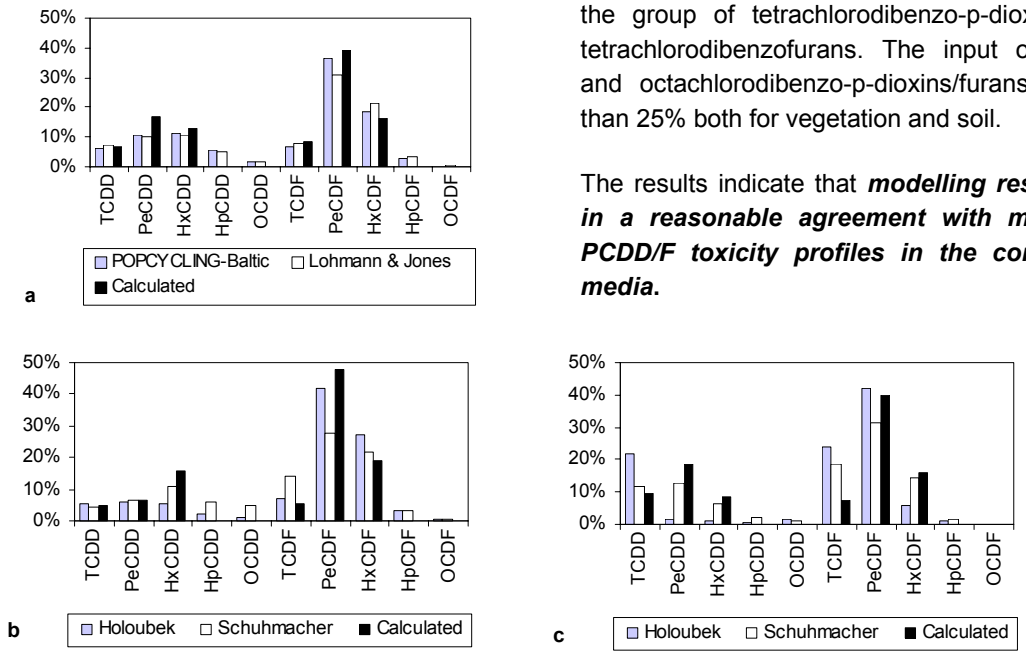


Figure 2.1. *Homologue toxicity profiles of PCDD/F mixture in the* ***a****- atmosphere,* ***b*** *- soil and* ***c*** *- vegetation obtained by modelling and measurements* [*Pacyna et al.,* 1999*; Lohmann and Jones,* 1998, *Holoubek et al.,* 2000*; Schumacher et al.,* 2000]

Behavioural peculiarities of different congeners in the environment. The aim of this section is to investigate behavioural peculiarities of different congeners of PCDD/F in the environment from the viewpoint of total toxicity. There are 17 PCDD/F congeners contributing to the total toxicity of PCDD/F

mixture. Earlier [*Shatalov et al.,* 2001] eight congeners covering about 75% of total toxicity were selected. At this stage we examine differences in the environmental behaviour of the selected congeners and perform the selection of an "indicator congener" whose physical-chemical properties can be used for the evaluation of PCDD/F mixture toxicity in the environment at the first stages of investigation.

To fulfill this task we have first simulated long-range transport and accumulation of the whole PCDD/F mixture for 1970 – 1999 under the assumption that overall emission toxicity is distributed between the selected 8 congeners according to their fractions in emissions determined in [*Vulykh and Shatalov,* 2001] (multi-congener run). In this simulation physical-chemical properties of each particular congener are used for the evaluation of the corresponding fraction transport.

Then eight model runs for the same period were performed under the assumption that the total mixture toxicity is determined by a single congener (mono-congener runs). Thus, eight model runs were done with physical-chemical properties of each of 8 selected congeners. The comparison of multi-congener run results with those of each mono-congener runs allows one to select an "indicator congener" as the congener describing the mixture behaviour most closely.

Below the deviations of calculation results of eight mono-congener runs from those of multi-congener run are examined. Figure 2.2 displays overall PCDD/F toxicity in soil calculated by mono-congener and multi-congener runs. The diagram shows that PCDD/F toxicity in soil calculated with the use of single congener properties can be essentially higher or lower than that for the mixture transport.

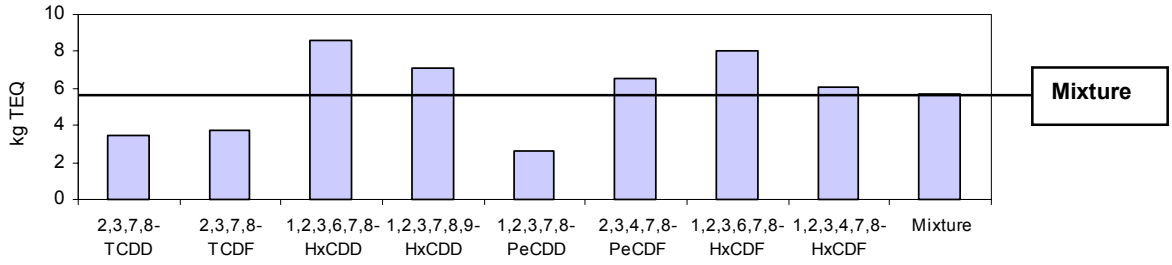


Figure 2.2. *PCDD/F toxicity accumulated in soil, kg TEQ. Calculations by mono-congener (2,3,7,8-TCDD, …) and multi-congener (Mixture) model runs for 1999.*

A diagram of deviations (%) of budget values of eight mono-congener runs from those of the multi-congener run in main media averaged over the last simulated year (1999) is displayed in Fig. 2.3. The diagram indicates a significant difference between the behaviour of selected congeners and PCDD/F mixture. The difference in soil for the majority of congeners and mixture reaches 40 – 50% and the difference for seawater may reach 90%.

Differences in spatial distribution of concentrations for selected PCDD/F congeners in soil and vegetation can reach 200% and more (Fig. 2.4).

As seen from Figures 2.3 and 2.4 minimum deviations are characteristic of the congener 2,3,4,7,8-PeCDF. So, ***at the initial stage of investigations it is possible to use properties of "indicator congener" 2,3,4,7,8-PeCDF with inaccuracy of 60% for vegetation and 30-40% in other compartments*** since the uncertainty of emission expert estimates is even higher (within an order of

magnitude according to [*Pacyna et al.,* 1999]). Below the results of simulations using physical-chemical properties of the indicator congener are described. However, the behaviour of different PCDD/F congeners in the environment is different and to improve ***the assessment of dioxins/furans long-range transport and accumulation it is necessary to simulate the mixture of eight selected congeners*.**

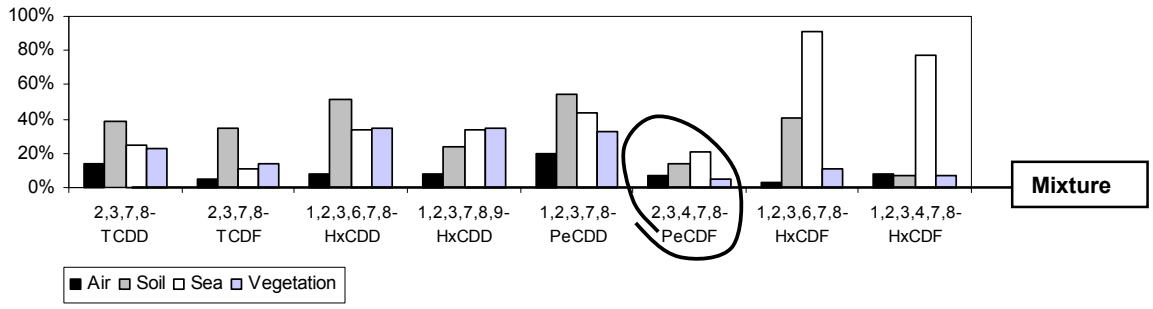


Figure 2.3. *The difference in mean annual media content obtained in mono-congener runs from the results of multi-congener run for 1999*

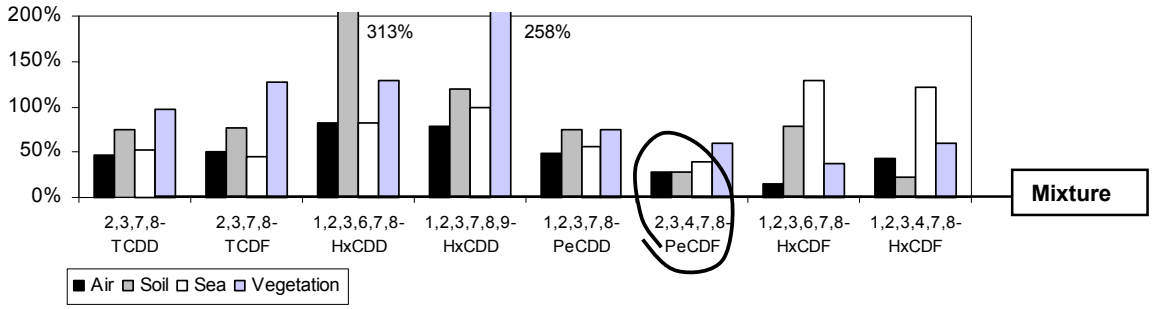


Figure 2.4. *The difference in spatial pollution distribution in media obtained in mono-congener runs from the results of multi-congener run for 1999*

2.1.2. *Model modification*

According to calculation results and measurement data* [*Shatalov et al.,* 2001] *about 80% of PCDD/F toxicity is accumulated in soil. To improve the consistency between calculated and observed data (especially PCDD/F concentrations in soil) the following modifications are made:

- Refinement of atmosphere/soil exchange scheme by selection of soil layer thickness for more accurate description of the calculated vertical pollution profile in soil.
- Refinement of pollutant redistribution between leaves/needles and soil in the forest ecosystem.
- Refinement of PCDD/F degradation rate constants for main environmental compartments.

Here we describe model modifications and changes in modelling results due to these modifications.

Atmosphere/soil exchange. A correct description of atmosphere/soil exchange to a great extent depends on proper parametrization of processes occurred in soil. The previous version of the soil scheme considered a number of processes: vertical diffusion, redistribution between soil compartments, POP transport with soil water. The vertical profile in the scheme was described by mean pollutant concentration in 5 subsequent layers with depths 0.005, 0.005, 0.01, 0.02 and 0.11 m. According to calculation results 99% of the whole mixture toxicity was concentrated in the upper layer (0.005 m). Since, the vertical toxicity profile influences significantly the process of atmosphere/soil exchange, the flux of atmosphere/soil gaseous exchange was described with considerable uncertainties.

In this context a special investigation of POP vertical distribution was carried out [MSC-E Technical Note 1/2002]. In this study vertical diffusion, transport with soil solution flux, dynamic redistribution between the dissolved and solid (organic carbon) phase and sorption on dissolved organic were explored. Alongside a number of model numerical experiments for the optimization of the soil scheme with regard to the refinement of soil layer depths were performed. As a result layer thickness were determined as 0.001, 0.0005, 0.002 and 0.03 m. The further work improvement of soil module is ongoing.

Vertical concentration profiles obtained by the modified model are given in Figure 2.5 for two years from the calculation period: 1970 and 1999.

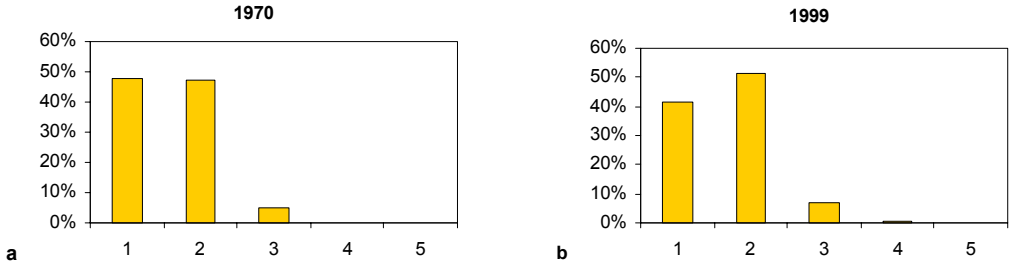


Figure 2.5. *Vertical profiles of soil pollution by dioxins/furans during the period of* ***a*** *- accumulation and* ***b*** *- clearance, percent of pollution content in soil layers 1-5*

The diagrams in Figure 2.5 show the percent of PCDD/F toxicity in soil layers 1-5 (renumerated from top to bottom). It is evident that the pollutant is distributed mainly between three upper layers. The diagrams demonstrate different character of PCDD/F toxicity distribution in the periods of accumulation and clearance. In the first case the pollution decreases with depth and in the second case the pollution content declines in the upper layer mainly due to re-emission. ***This representation of calculated toxicity vertical profile with modified soil layers leads to better understanding of the gaseous exchange flux between the atmosphere and soil*** defined mainly by the pollution concentration in the upper soil layer. More detailed discussion of modelling results is presented below.

Redistribution of pollutant scavenged to forest ecosystems between soil and vegetation. At earlier stages it was supposed that all the particle phase scavenged to forest ecosystems is accumulated in forest trees and the removal from leaves/needles to soil has not been taken into account. This has led to essential overestimation of concentrations in vegetation and underestimation of concentrations in forest soils. The latter according to measurement data (see e.g. [*Shatalov et al.,* 2001]) can be essentially larger compared with concentrations in soils not covered by forests.

Measurements of deposition fluxes over and under the forest trees [*Brorstrom-Lunden and Lofgren,* 1998] show that the fraction of a pollutant accumulated by forest trees is about 30% for PCBs and 20% for B[a]P. Similar data for PCDD/Fs were not found in the literature. However, as a first approximation it is assumed that about 1/3 of the pollutant in the particle phase scavenged to forest is accumulated by forest trees (in leaves/needles) and the rest is transported to soil being washed away from leaves/needles.

The description of changes caused by the above model modifications follows:

Toxicity distribution between environmental components. PCDD/F toxicity distribution between main natural media (the atmosphere, soil, seawater, vegetation and forest litter) calculated by the previous and modified model versions is demonstrated in Figure 2.6.

As seen from the diagrams the modification resulted in the increase of soil toxicity fraction (from 66% to 77%).

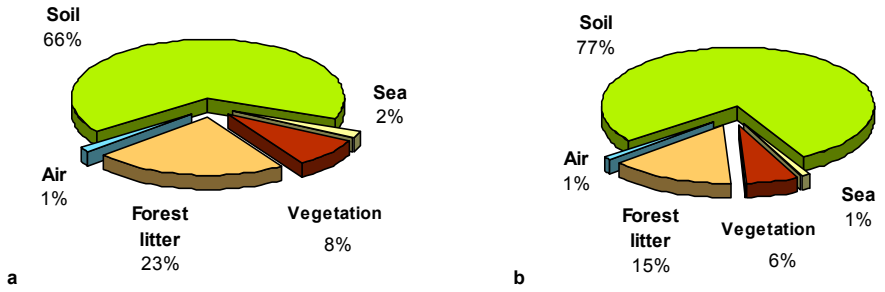


Figure 2.6. *PCDD/F toxicity distribution between main natural media calculated by the previous -* ***a*** *and modified -* ***b*** *model versions*

Thus ***due to the above described modifications the calculated accumulation in soil essentially increased.*** Let us analyze the corresponding changes in spatial distributions.

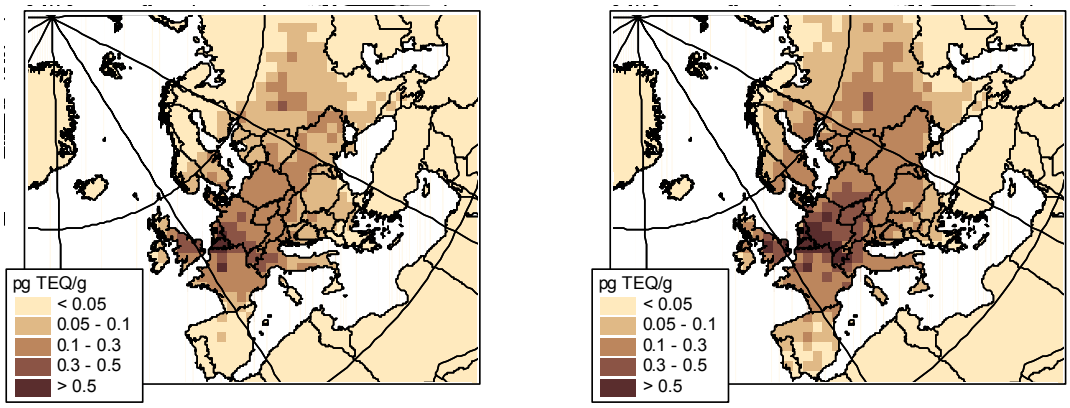
Spatial distribution of contamination in the EMEP region. Mean soil concentrations in European countries calculated by the modified model version have also changed. The comparison of soil concentrations calculated by previous and modified model versions is shown in Figure 2.7.

On the average over Europe concentrations increased as much as 1.5 times. In some regions the more than 3-fold increase of soil concentrations compared with previous calculations takes place. For the most part of Europe the increase of soil concentrations is 1.5 – 2.5 times.

Similar picture takes place for surface air concentrations (Fig. 2.8).

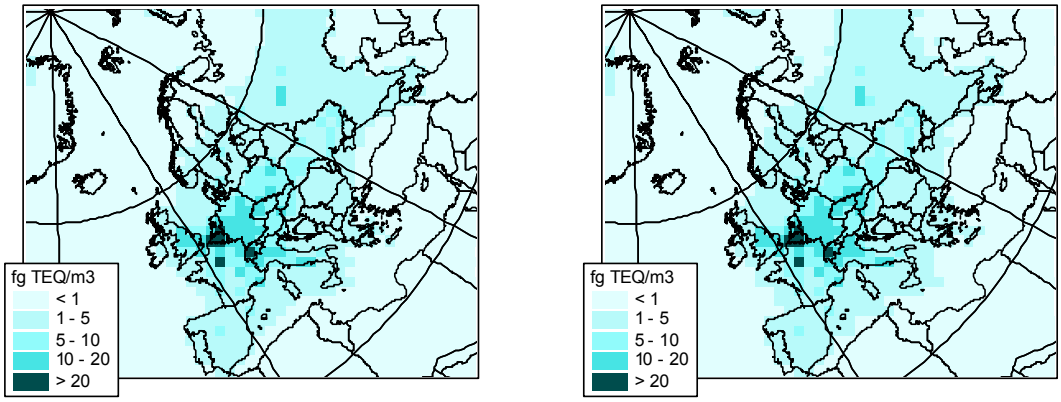
For atmospheric concentrations somewhat less increase compared with soil concentrations takes place. On the average in Europe atmospheric concentrations have increased 1.25 times with about two-fold maximal increase.

The analysis indicates that ***the modification of the atmosphere/soil exchange processes improves the consistency between calculated and measured values, but the model still underestimates soil concentrations*** (the previous model version underestimated soil concentrations within two orders of magnitude).



Previous version Modified version

Figure 2.7. *Soil concentrations calculated by previous and modified model versions, pg TEQ/g*



Previous version Modified version

Figure 2.8. *Concentrations in the surface air calculated by previous and modified model versions, fg TEQ/m^3*

Refinement of PCDD/F degradation parameters in environmental media. Further model modification concerned the refinement of PCDD/F degradation rate constants in main environmental media. Main attention is paid to the atmosphere (as to main transport media) and to soil (as to main accumulating media).

The values of degradation rate constants according to various literature sources can differ significantly from one another. For example, degradation rate constants in the atmosphere calculated on the basis of data from two literature sources differ 6 times (Table 2.2 where the corresponding half-lives are presented as well).

For modelling refined degradation rate constants in the atmosphere due to [Atkinson, 1996] are used.

The values of degradation rate constants in soil and seawater taken from different sources can differ even more (20 – 30 times, Table 2.3).

Table 2.2. PCDD/F degradation rate constants in the atmosphere calculated on the basis of literature data

	[*Atkinson,* 1991]		[*Atkinson,* 1996]	
	Degradation rate constants, s^{-1}	Half-lives, days	Degradation rate constants, s^{-1}	Half- lives, days
Winter	$1.6 \cdot 10^{-7}$	50	$2.7 \cdot 10^{-8}$	297
Spring/fall	$1.4 \cdot 10^{-6}$	6	$2.4 \cdot 10^{-7}$	33
Summer	$3.6 \cdot 10^{-6}$	2	$6.0 \cdot 10^{-7}$	13
Average	$1.6 \cdot 10^{-6}$	5	$2.7 \cdot 10^{-7}$	29

Table 2.3. PCDD/F degradation rate constants in soil and seawater according to different literature sources

	[*Mackay et al.,* 1992]		[*Sinkkonen and Paasivirta,* 2000]	
	Degradation rate constants, s^{-1}	Half- lives, days	Degradation rate constants, s^{-1}	Half- lives, days
Soil	$1.1 \cdot 10^{-8}$	708	$3.5 \cdot 10^{-10}$	22900
Seawater	$3.5 \cdot 10^{-7}$	23	$1.5 \cdot 10^{-8}$	550

As it was mentioned above, the previous model version using more rapid degradation rate in soil strongly underestimated soil concentrations. Thus, usage of degradation rates proposed by* [*Sinkkonen and Paasivirta*, 2000] *seems to be more reasonable*. As shown below,*** usage of these degradation rate constant leads to more accurate agreement between calculated and measured data. ***These values are used also in other models describing PCDD/F fate in the environment* [*Beyer and Matthies,* 2001].**

Below the comparison between spatial distributions of PCDD/F concentrations in various media (the atmosphere, soil, seawater and vegetation) obtained by calculations with the help of the model with modified atmosphere/vegetation/soil exchange scheme and with:

- **variant 1** – previous degradation rate constants
- **variant 2** – modified degradation rate constants

is presented.

Atmospheric concentrations. Figure 2.9 shows maps of PCDD/F concentration distribution in the atmosphere obtained in calculations made by variants 1 and 2.

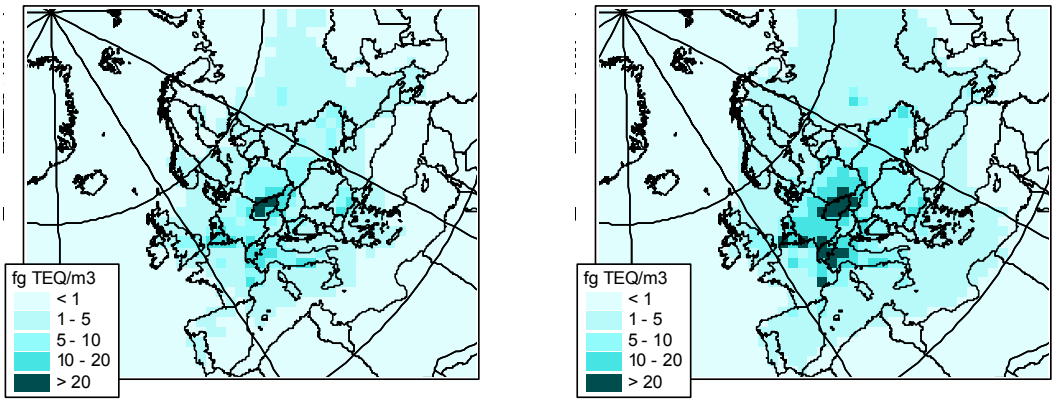
The comparison of the maps reveals that the modification of degradation rate constants resulted in the atmospheric concentration increase. More detailed analysis shows that in Europe the concentration increase is 70% on the average and can reach more than two times.

Soil concentrations. Figure 2.10 presents spatial distribution of concentrations over the upper soil layer (5 mm) calculated by the first and second variants.

The comparison of the maps indicates that soil concentrations defined with the use of modified degradation rate constants essentially increase. On the average concentrations in Europe increased as much as 18 times and in some regions the growth reached more than 30 times.

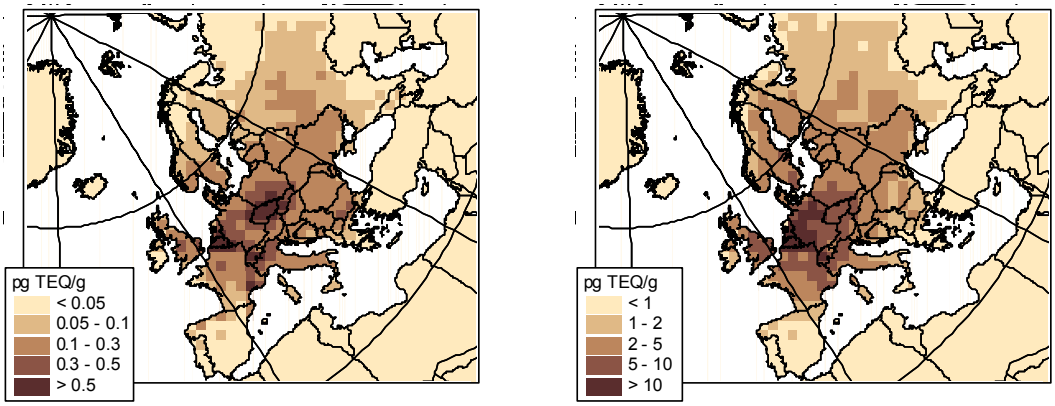
Spatial distribution of soil concentrations is also modified. The latter is caused by the fact that at half-life in soil accepted in the model soil concentrations are affected not only by emissions of recent years

but also by emissions at earlier stages of accumulation (see below comparison of emission spatial distributions for 1990 and 1999, Figure 2.19 at page 57).



Variant 1 Variant 2

Figure 2.9. *PCDD/F atmospheric concentrations in 1999 calculated by variants 1 and 2 for 1999, fg TEQ/m^3*



Different scales: concentration levels differ more than 30 times!

Variant 1 Variant 2

Figure 2.10. *PCDD/F concentrations in the upper soil layer calculated by variants 1 and 2 for 1999, pg TEQ/g*

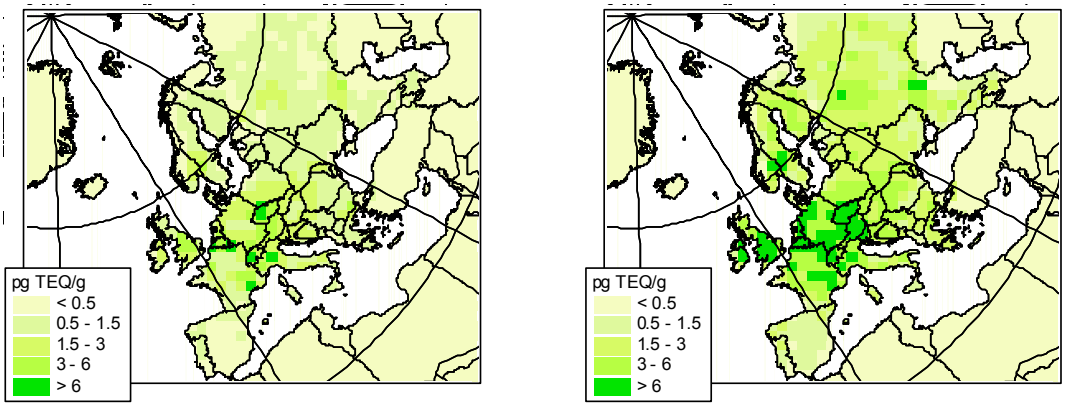
Concentrations in vegetation. Figure 2.11 presents maps of spatial concentration distribution in vegetation calculated by variants 1 and 2.

The maps in the figure show that like in the case of atmospheric concentrations the modification of degradation rates in soil and seawater gives rise to concentrations in vegetation. Concentrations in vegetation in Europe increase 2.4 times on the average and in some regions – more than 3 times.

Concentrations in seawater. Similar to the case of soil, modification of degradation rates resulted in essential increase of seawater concentrations (approximately by an order of magnitude, Fig. 2.12).

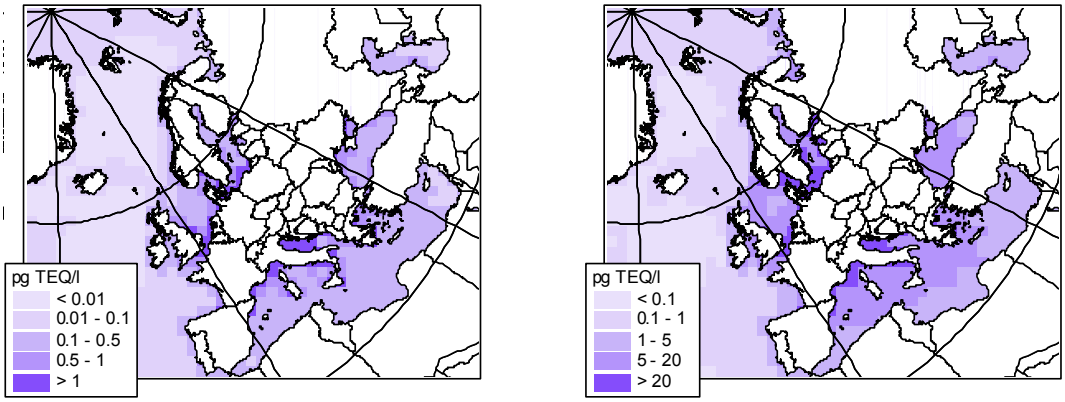
Thus the comparison of calculation results obtained with two variants of degradation rates points out that ***the modification of these rates leads to the increase of concentrations in the main natural media*** (the atmosphere, soil, seawater and vegetation). This induces an increase of PCDD/F toxicity accumulated by all the accumulating media: soil, vegetation and seawater. However, since the largest increase takes place in soil concentrations, the fraction of toxicity accumulated in soil increases as well.

The comparison of values defined by the modified model with measurements is made below.



Variant 1 Variant 2

Figure 2.11. *PCDD/F concentrations in vegetation calculated by variants 1 and 2 for 1999, pg TEQ/g*



Different scales: concentration levels differ by about an order of magnitude!

Variant 1 Variant 2

Figure 2.12. *PCDD/F concentrations in seawater (pg TEQ/l) calculated by variants 1 and 2 for 1999*

2.1.3. *Comparison of calculations against measurements*

Atmospheric concentrations. Table 2.4 gives the comparison of calculation results obtained by the modified model for the period from 1970 to 1999 with available measurement data. The first part of the table contains data on clean regions and the second – on contaminated regions.

Table 2.4. PCDD/F concentrations calculated and measured in the surface air, fg TEQ/m^3

Location	Year	Measured		Calculated		Factor	Source of measurement data
		Range	Aver	Range	Aver		
Sweden, Stockholm, suburban	1986 – 87		13	2.07 - 18.4	6.05	2.15	*Broman et al.,* 1991a
Sweden, Stockholm, countryside	1986 – 87		4.4			1.38	
Sweden, Stockholm, open coastal	1986 – 87		2.6			2.33	
Sweden, SE2	1989	28 - 55	46	4.78 - 12.6	8.39	5.36	*Tysklind et al*., 1993
Sweden, SE2	1990	3.7 - 31.5	14	3.6 – 12.6	7.75	2.1	
Netherlands	1992	9 - 63	31	11.5 - 97.1	45.7	1.47	*Buckley-Golder et al.,* 1999
Germany, Bavaria	1992 – 93	3 -179	22.5	7.85 - 63.9	22.1	1.02	
Germany, rural area	1992	25 - 70		8.55 - 63.9	23.9		*Fiedler*, 1996
Germany, Baden-Wuerttemberg, rural	1993 –94	8 - 54	21	7.38 - 52.4	20	1.05	*Wallenhorst et al*., 1997
Germany, Baden-Wuerttemberg, rural, with elevated regions	1993 –94	5 - 49	18			1.11	
Germany, Bayereuth	1995	4.3 - 49	17	5.78 - 64.3	20.5	1.21	*Pacyna et al.,* 1999
Germany, DE9	1995	11.0 - 45.3	29	4.25 - 12.4	7.5	3.87	
Germany, Bayereuth	1996	19.6 - 63.4	43	5.59 - 56.3	17.6	2.44	
Germany, DE9	1996	17.5 - 50.8	36	4.26 - 8.76	6.7	5.37	
Austria	1993 - 94	11 - 110	36.4	10.9 - 28.8	15.6	2.33	*Buckley-Golder et al.,* 1999
Czech Republic, CZ3	1994 – 95	2 - 156	38.4	37.4 - 124.8	63.4	1.65	*Holoubek et al*., 2000
UK, Hazelrigg	1993	58 - 190	101		17.85	5.66	Coleman et al., 1998
	1994	9 - 49	24		20.8	1.15	
	1995	5 - 83	35		22	1.59	
UK,Lancaster,north-west coast	1996	7 - 16.6			10.96		*Lee et al.,* 1999
UK, High Muffles	1996 – 97	2.2 - 16	6		10.1	1.68	*Buckley-Golder et* al., 1999
UK, Hazelrigg	1996 – 97	5 - 29	17		14.18	1.20	*Coleman et al.,* 1998
UK, Stoke Ferry	1997	2.1 - 21	19		10.95	1.74	
UK, Hazelrigg	1997	8-18	11		9.25	1.19	*Lohmann and Jones,* 1998
UK, East coast	1997	2 - 6	4	3.62 - 13.3	7.36	1.84	
UK, Lancaster	V, 1997	7.1 - 17.6	10.1		9.25	1.09	*Lohmann et al.,* 1999a
UK, Lancaster	X-XII, 1997	5.5 - 220	38		9.25	4.0	*Lohmann et al.,* 1999b
UK, North York Moors	V, 1997	2.1 - 6.1	3.7		9.25	2.5	*Lohmann et al*., 1999a
Ireland, Mace Head	V, 1997	2.9 - 4.2	3.6		1	3.60	
Czech Republic, Zlin	1990	<0.2 - 299		16.6 - 58.6	42.7		*Holoubek et al.,* 2000
Czech Republic, Praha	1994	16 - 161			51.4		
	1995	13.3 - 637			75.4		
	1996	36.8 - 10700			63.3		
Italy, Rome	1990-91	50-280	85	3.55 - 27.7	14.8	5.74	*Lohmann and Jones,* 1998
Germany, urban area	1992	70 - 350		8.55 - 63.9	23.9		*Fiedler*, 1996
Austria, Leoben/Donawitz	1999 - 2000	81.6 - 491	194	10.5 - 24.4	14.1	13.77	*Moche and Thanner, cited by Holoubek,* 2000

Thus *the calculated values agree with measured ones within a factor of 6. Among 27 available measurements 22 are within a factor of 3 with respect to calculations*. In polluted regions the calculated values appeared to be considerably lower than the observed values.

Deposition fluxes. Table 2.5 presents the comparison of calculated deposition fluxes with available measurement data.

Table 2.5. Measured and calculated deposition fluxes, pg TEQ/m^3/day

Location	Year	Measured		Calculated	Factor	Source
		Range	Aver			
Germany, rural area	1992	5 – 20	12.5	2.47	5.06	*Fiedler*, 1996
Belgium, Flanders	1993	11 – 18	15	3.46	4.33	*Lohmann and Jones*, 1998
Germany, Baden-Wuerttemberg, rural	1993 –94	9 – 16	13	2.11	6.17	*Wallenhorst et al.*, 1997
Belgium	1994		15	3.21	4.67	*Lohmann and Jones*, 1998
Belgium, Eksel	1997		3.1	1.91	1.62	*Buckley-Golder et al.*, 1999
Belgium, Mol	1997		0.7	1.91	0.37	

Like in the case of atmospheric concentrations ***the calculated deposition fluxes in remote regions are consistent with measurements within a factor of 6.***

Soil concentrations. Table 2.6 demonstrates the comparison of available measured concentrations of PCDD/F in soil with calculated values.

Table 2.6. PCDD/F measured and calculated concentrations in soil, pg TEQ/g

Location	Year	Measured		Calculated		Factor	Source
		Range	Mean	Range	Mean		
Austria	1993	1.6 – 31	6.9	13.2 – 29	19.6	2.84	*Weiss*, 1998
Czech Republic	1994	12 – 54	30.2	15.4 – 32.5	24.4	1.24	*Holoubek et al.*, 2000
Czech Republic	1994 - 95	12 – 54	28.7	15.4 – 32.6	24.6	1.17	
Belgium	1992	2.1 – 2.3		32 – 65	41		*Buckley-Golder et al.*, 1999
Germany, rural	up to 1998	0.002 – 112	6.1	1.46 – 59	30.9	5.07	
Germany, rural and conurbation	up to 1998	0.3 – 8.9	3.2	1.46 – 59	30.9	9.66	
Ireland	1997	2 – 13.3	7.5	0.17 – 7.91	3.62	2.07	
Italy	1993	1.9 – 3.1	2.4	0.33 – 25.2	11.79	4.91	
Luxembourg	1993	1.4 – 3.6			41.3		
Netherlands	1991	2.2 – 16.4	4	1.87 – 65.6	37.48	9.37	
Spain	1996	0.08 -- 8.4	0.54	0.12 – 7.2	3.14	5.81	
UK	up to 1995	0.78 -- 17	5.2	0.37 – 30.2	15.3	2.94	
Spain	1996 – 97	7 – 14	12	0.12 – 7.2	3.11	3.86	*Schumacher et al.*, 2000

The analysis of data presented in the table shows that the modified model describes soil concentration with an accuracy of an order of magnitude. ***More than a half of measurements are within a factor of 4 with respect of calculations and only in two cases the agreement is beyond a factor of 6.***

Concentration in vegetation. Calculated and measured PCDD/F concentrations in vegetation are given in Table 2.7.

The comparison of measurements with calculated values shows that the ***model considerably overestimates concentrations in vegetation***. This can be related with the fact that the model does not take into account the degradation in vegetation. In future the degradation will be considered.

Table 2.7. Calculated and measured PCDD/F concentrations in vegetation, pg TEQ/g

Location	Year	Measured		Calculated		Source
		Range	Mean	Range	Mean	
Austria	1993	0.3 − 1.9	0.7	5.9 − 16.1	12.3	*Weiss*, 1998
Czech Republic	1994	0.07 -- 0.6	0.2	5.8 − 25.2	15.3	*Holoubek et al*., 2000
Germany	1993	0.27 − 3.45	1.12	4.4 − 36.4	11.9	*Buckley-Golder et al.,* 1999
Luxembourg	1993 − 94	0.6 − 0.8			35.3	
Spain	1996 − 97	0.52 -- 1.0	0.7	0.001 − 4.14	1.38	*Schumacher et al.,* 2000
Spain	1997	0.12 − 1.14	0.23	0.001 − 3.81	1.35	*Schuhmacher et al*, 1998
Finland	1996	1 − 7	3	0.08 − 6.58	3.53	*Sinkkonen et al*., 1997
UK, archive herbage	1979 − 88		1.17	0.007 − 36.8	13.6	*Kjeller et al.,* 1991

Concentration in seawater. Table 2.8 demonstrates calculated and measured PCDD/F concentrations in the Baltic Sea in 1988 [*Broman et al*., 1991.b].

Table 2.8. Calculated and measured PCDD/F concentrations in the Baltic Sea in 1988, pg TEQ/l

Location	Latitude	Longitude	Measured	Calculated	Factor
Southern Baltic, offshore	56° 07′40″	16° 29′50″	4.78	57.67	12
Middle Baltic, offshore	58° 15′38″	17° 32′68″	3.07	68.08	22
Middle Baltic, coastal	58° 16′01″	16° 56′82″	4.35	68.08	16
Northern Baltic, offshore	58° 43′38″	18° 14′37″	3.01	68.08	23
Northern Baltic, coastal	58° 48′23″	17° 37′57″	2.18	68.08	31
Aland Sea, offshore	59° 52′33″	19° 20′12″	4.38	35.53	8
Bothnian Sea, offshore	62° 37′68″	18° 35′06″	9.07	33.88	4
Bothnian Sea, coastal	62° 51′22″	18° 16′10″	4	33.88	8
Bothnian Bay, offshore	64° 33′60″	21° 53′69″	5.14	36.73	7

The comparison shows that *the* ***model appreciably overestimates concentrations in seawater***. Most likely it is connected with the overestimation of PCDD/F half-life in sea. In future it is supposed to modify the model.

Basic results:

- Calculated atmospheric concentrations of PCDD/F agree with observations within a factor of 6. Among 27 measured values for 22 of them the consistency with calculations is within a factor of 3. In clean regions calculated deposition fluxes also agree with measurements within a factor of 6.
- The modified model describes soil concentrations with an accuracy of an order of magnitude. For more than a half of measurements the consistency with calculated data is within a factor of 4 and only in two cases the agreement is beyond a factor of 6.
- The model considerably overestimates concentrations in vegetation and seawater. In future it is supposed to modify the model for the improvement of the agreement between measured and calculated data especially from the viewpoint of model description of degradation processes in these media.

2.2. Model evaluation of PCDD/F transport and accumulation

This section provides tentative evaluation of pollution levels of natural media in the EMEP region made by calculations of PCDD/F long-range transport and their accumulation in environmental compartments for the period from 1970 to 1999 with the use of the modified model and properties of the "indicator congener" 2,3,4,7,8-PeCDF. For 1970 – 89 we used expert estimates of emissions [*Pacyna et al*., 1999] and for the last decade (1990 – 99) – available official data complemented by expert estimates. The initial pollution level was imitated by the model spin-up for 15 years with emissions and meteorology of 1970.

The section content is as follows. First we describe emission data (section 2.2.1). Then investigation of long-term trends of media pollution from 1970 to 1999 is carried out (section 2.2.2). Further evaluation of spatial distribution of pollution in various environmental compartments in the end of simulation period (1999, section 2.2.3) and comparison between calculations and measurements (section 2.2.4) are presented.

2.2.1. *Emissions of dioxins/furans*

In accordance with the Protocol on Persistent Organic Pollutants to the Convention on Long-Range Transboundary Air Pollution of 1998 polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) are referred to substances in respect to which the Parties are obliged to reduce annual emissions by taking appropriate effective measures.

National data and expert estimates of emissions in the EMEP domain. Model calculations presented in this report were performed with emission data involving national data submitted by countries to the UN ECE Secretariat. 23 countries have reported emission data for 1990 - 99 (Table 2.9).

Austria, the Czech Republic, Finland, France, Hungary, Poland, Russia and the United Kingdom submitted emission data for all the years (1990 - 99). Information for individual years is reported by Belarus, Belgium, Bulgaria, Croatia, Cyprus, Denmark, Germany, Lithuania, Luxembourg, the Netherlands, Norway, Slovakia, Slovenia, Spain and Sweden.

In model calculations of transboundary transport for 1990-99 we used available official data (Table 2.9) and expert estimates of emissions [*Pacyna et al*., 1999].

Table 2.9. Dioxins/furans official emission data, g TEQ/y

Country	UN/ECE reported official emission data									
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Austria	92.06	85.35	70.86	64.28	58.38	61	60.21	56.67	53.83	50.62
Belarus								16.4	15.68	15.19
Belgium	448[1]				147.6 [2]	437.5	108.1	122.6	116.3	135.9
Bulgaria	554.2					456	340.9	309.6	288.4	245.3
Croatia	178.6						97.35	95.04	110.8	97.96
Cyprus	0.772									
Czech Republic	1252	1220	1220	1140	1135	1135	921.5	830.2	766.7	643.2
Denmark										95
Finland [3]	35.4	34.8	33.1	34.7	41.5	40.7	39.8	39.1	39.5	41.1
France	2206	2268	2312	2392	2363	2107	1865	1253	836	558
Germany	1196					309				
Hungary	157	151	126	122	104	116	108	103	93.6	37.6
Lithuania								5.62	5.97	5.03
Luxembourg	40				23	24	16	16	8	
Netherlands	618		505		143	74.2	60.7	55.3	43.8	34.8
Norway						125	105	105		
Poland	368.3	349.2	338.1	396.6	360.9	387.7	366.2	347.7	290.4	287.4
Russian Federation	991	947	901	878	82 5	769	637	614	606	625
Slovakia	224.5					372.7		194.2	187.6	161
Slovenia	8.60				5.67	4.94	4.91	3.82	3.53	3.51
Spain	181	190	200	196	185	157	155			
Sweden	93			33						
UK	1142	1123	1098	1049	953.2	819.5	588.6	384.2	361.0	345.7

[1] - Refers to Flanders only

[2] - Refers to Brussels and Wallonia only

[3] - Emissions prior to 1994 are underestimated and will be updated

Table 2.10 presents emission data set for 1990 - 99 involving official, expert and calculated (in the case when no data were available for some years) values. Below there is some clarification how emissions were calculated for countries, for which national data were absent for some years.

- Belarus and Lithuania – official data are submitted for 1997 - 99. For 1990 expert estimates [*Pacyna et al*., 1999] were used. For 1991 - 96 emissions were assessed by linear interpolation between the expert value for 1990 and official data for 1997.

- Belgium provided emission data for 1990 and 1994 - 99. Data for 1990 and 1994 are incomplete since they include emissions of some provinces (Table 2.9). Therefore for 1990 expert estimates [*Pacyna et al*., 1999] were used and emissions for 1991 - 94 were defined by linear interpolation between expert estimate for 1990 and official data for 1995.

- Bulgaria, Croatia, the Netherlands, Slovakia, and Slovenia – lacking data were estimated by the interpolation between available official data.

- Cyprus – official data for 1990 were used for the whole period of 1990 - 99.

- Denmark – official data are reported for 1999. For 1990 - 98 emissions were presumed to be equal to those of 1999 since expert estimates for 1990 and 1995 [*Pacyna et al*., 1999] are lower than official emission data for 1999.

- Germany submitted data for 1990 and 1995. Emissions for 1991-94 were estimated by linear interpolation and for 1996-99 we used the emission for 1995.

- Luxembourg – official data are reported for 1990 and 1994 - 98. Emissions for 1991 - 93 were assessed by linear interpolation and for 1999 we used emissions for 1998.

- Norway provided data for 1995 - 97. For 1990-94 emissions were assumed to be equal to those for 1995 since expert estimates for 1990 [*Pacyna et al*., 1999] are lower than official emission data for 1995. Emissions for 1998 and 1999 were taken equal to those for 1997.

- Spain – official data are submitted for 1990 - 96. Emissions for 1997 - 99 were taken the same as in 1996.

- In calculations for Sweden emissions for 1991 and 1992 were defined by linear interpolation between data for 1990 and 1993 (Table 2.9). Emissions for 1994 - 99 were taken the same as in 1993.

For countries, which did not report their national emission data expert estimates for 1990 and 1995 [*Pacyna et al*., 1999] were used and for 1996 - 99 - estimates for 1995.

Table 2.10. Emissions of dioxins/furans to the atmosphere in the EMEP domain used for modelling (Official data are given in bold), g TEQ/y

Country	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Albania	3	3	3	3	3	3	3	3	3	3
Armenia	43	40	37	34	31	28	28	28	28	28
Austria	**92**	**85**	**71**	**64**	**58**	**61**	**60**	**57**	**54**	**51**
Azerbaijan	77	71	66	61	55	50	50	50	50	50
Belarus	107	94	81	68	55	42	29	**16**	**16**	**15**
Belgium	511	496	482	467	452	**438**	**108**	**123**	**116**	**136**
Bosnia&Herzegovina	19	19	20	20	21	22	22	22	22	22
Bulgaria	**554**	535	515	495	476	**456**	**341**	**310**	**288**	**245**
Croatia	**179**	165	152	138	124	111	**97**	**95**	**111**	**98**
Cyprus	**1**	1	1	1	1	1	1	1	1	1
Czech Republic	**1252**	**1220**	**1220**	**1140**	**1135**	**1135**	**922**	**830**	**767**	**643**
Denmark	95	95	95	95	95	95	95	95	95	**95**
Estonia	15	14	14	13	13	12	12	12	12	12
Finland	**35**	**35**	**33**	**35**	**41**	**41**	**40**	**39**	**40**	**41**
France	**2206**	**2268**	**2312**	**2392**	**2363**	**2107**	**1865**	**1253**	**836**	**558**
Georgia	72	67	62	56	51	46	46	46	46	46
Germany	**1196**	1019	841	664	486	**309**	309	309	309	309
Greece	155	148	142	135	129	122	122	122	122	122
Hungary	**157**	**151**	**126**	**122**	**104**	**116**	**108**	**103**	**94**	**38**
Iceland	0	0	0	0	0	1	1	1	1	1
Ireland	17	17	16	16	15	14	14	14	14	14
Italy	870	856	842	827	813	799	799	799	799	799
Kazakhstan	38	35	32	30	27	24	24	24	24	24
Latvia	13	13	13	13	13	12	12	12	12	12
Lithuania	24	21	19	16	13	11	8	**6**	**6**	**5**
Luxembourg	**40**	36	32	27	**23**	**24**	**16**	**16**	**8**	8
Netherlands	**618**	562	**505**	324	**143**	**74**	**61**	**55**	**44**	**35**
Norway	125	125	125	125	125	**125**	**105**	**105**	105	105
Poland	**368**	**349**	**338**	**397**	**361**	**388**	**366**	**348**	**290**	**287**
Portugal	41	40	38	37	36	34	34	34	34	34
Republic of Moldova	18	16	15	13	11	10	10	10	10	10
Romania	129	119	110	100	91	81	81	81	81	81
Russian Federation	**991**	**947**	**901**	**878**	**825**	**769**	**637**	**614**	**606**	**625**
Slovakia	**225**	254	284	313	343	**373**	283	**194**	**188**	**161**
Slovenia	**9**	8	7	6	**6**	**5**	**5**	**4**	**4**	**4**
Spain	**181**	**190**	**200**	**196**	**185**	**157**	**155**	155	155	155
Sweden	**93**	73	53	**33**	33	33	33	33	33	33
Switzerland	242	230	218	206	194	181	181	181	181	181
The FYR of Macedonia	8	8	8	9	9	9	9	9	9	9
Ukraine	925	855	784	713	642	571	571	571	571	571
United Kingdom	**1142**	**1123**	**1098**	**1049**	**953**	**820**	**589**	**384**	**361**	**346**
Yugoslavia	46	48	49	51	52	54	54	54	54	54
Total, kg TEQ/y	13	12	12	11	11	9.8	8.3	7.2	6.6	6.1

Figure 2.13 shows the percent variations of emission contributions, for which official data are available to the total emission in the EMEP region during 1990 - 99. These emissions amount to more than a half of the overall emissions in the region. Their fraction varies within the range from 51% (1991) to 72% (1990).

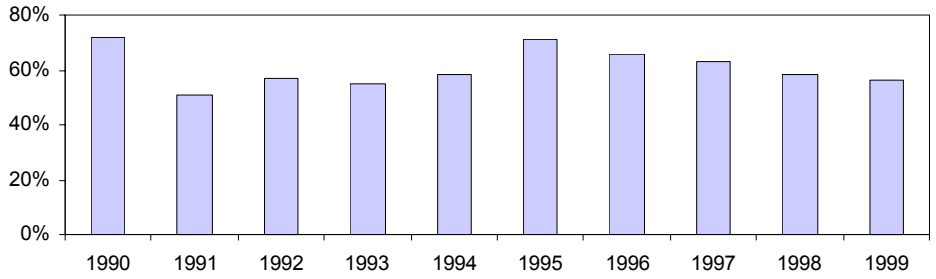


Figure 2.13. *Percent of official emission data reported, to the total emissions in the EMEP region*

Trend of dioxins/furans emissions in the EMEP domain. According to the data of Table 2.10 the total emission in the EMEP domain tends to decline. During 1990-99 emissions of dioxins/furans decreased 2 times. However, one should keep in mind that for many countries in model calculations expert estimates of emissions [*Pacyna et al*., 1999] were used. The uncertainty of these data is about an order of magnitude. Official data reported by countries are not always complete and reliable. Besides the majority of countries submitted national data not for all years. For this reason emission total values in the region should be considered as approximate ones.

The variation of dioxins/furans emissions in the EMEP domain in 1970 - 99 is illustrated by Figure 2.14 Emission values for 1970, 1975, 1980, 1985, 1990 and 1995 denoted as “expert estimates” correspond to estimates calculated under “POPCYCLING-Baltic” project [*Pacyna et al.,* 1999]. Emission values for 1990-99 denoted as “official and expert estimates” correspond to the data of Table 2.10. As seen from the figure, for the region on the whole in 1990 and 1995 “official and expert estimates” slightly exceed expert estimates (approximately by 15%).

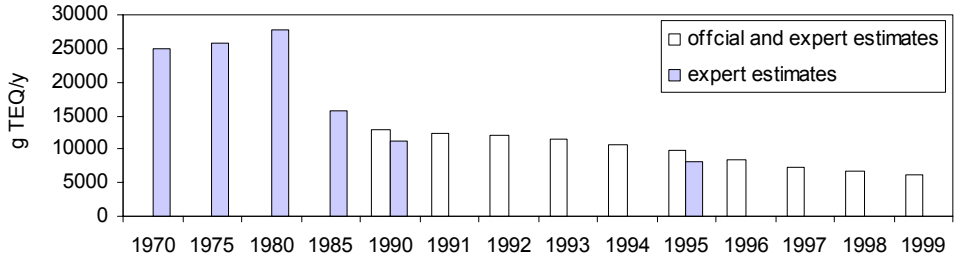


Figure 2.14. *Emissions of dioxins/furans in the EMEP region, g TEQ/y*

Trends of dioxins/furans emissions in individual countries. Relative and absolute variations of emissions of dioxins/furans in individual countries, which submitted official data for 1990 and 1999 are

shown in Table 2.11. For the sake of comparison data for the whole EMEP region are presented. Actually in all the countries but Finland emissions decrease. The emission value for 1990 - 94 calculated for Finland is underestimated and will be recalculated. According to official data maximum relative decline of emissions took place in the Netherlands (18 times). The maximum contribution to emission reduction in the EMEP region is made by France. This contribution is almost a quarter of the emission reduction value during the considered period. The reduction of emissions in 1999 in comparison with emissions in 1990 for the EMEP region and in individual countries (%) is illustrated in Figure 2.15.

Table 2.11. Reduction of dioxins/furans emissions in individual countries

Country	E_{1990}/E_{1999}	$100*(E_{1990} - E_{1999})/E_{1990}$, %	$E_{1990}-E_{1999}$, g TEQ
EMEP region	2	52	6863
Netherlands	18	94	583
Hungary	4	76	119
France	4	75	1648
UK	3	70	796
Slovenia	2	59	5
Bulgaria	2	56	309
Czech Republic	2	49	608
Croatia	2	45	81
Austria	2	45	41
Russian Federation	2	37	366
Slovakia	1	28	64
Poland	1	22	81
Finland	1	-16	-6

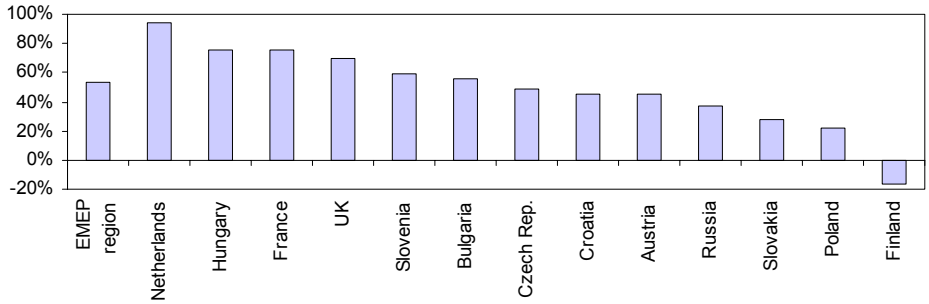


Figure 2.15. *Reduction of dioxins/furans emissions in individual countries, %*

Spatial distribution of emissions in individual countries. Four countries submitted data on spatial distribution with resolution 50x50 km: Bulgaria (for 1990 and 1995), Finland (for 1990-99), France (for 1995) and Spain (1990-96). These data were included in model calculations. For other countries spatial distribution estimates by [*Pacyna et al.*, 1999] were used. Spatial distribution of dioxins/furans emissions recalculated for the grid with resolution 150x150 km is demonstrated in Figure 2.19 (see in page 57). As it follows from the figure during the considered period an essential emission reduction is observed in Central Europe.

2.2.2. *Investigations of long-term trends*

Pollution dynamics in the period of 1970 - 99. In this section we present calculated trends of accumulation and clearance of natural media during 1970 – 99.

Figure 2.16 shows the dynamics of dioxins/furans emission in Europe and their content in the atmosphere and soil during the considered period (due to preliminary character of calculated concentrations in vegetation and seawater trend analysis for these media is not performed).

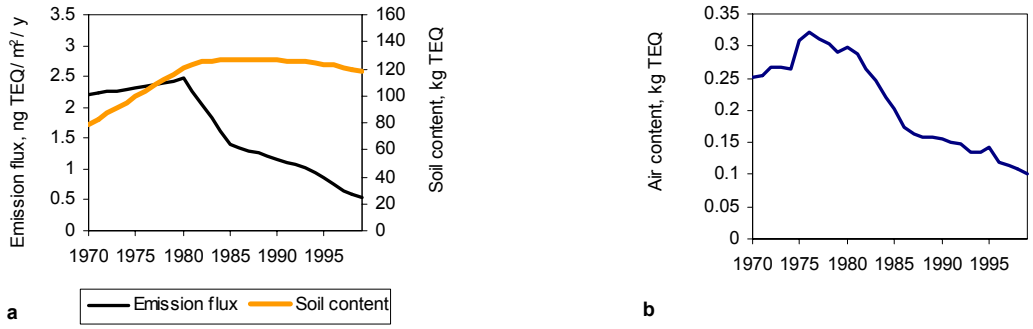


Figure 2.16. *The comparison of dynamics of European emissions and the content in the atmosphere and soil:* ***a*** *– emission flux, ng TEQ/m²/y and soil content, kg TEQ,* ***b*** *–air content, kg TEQ*

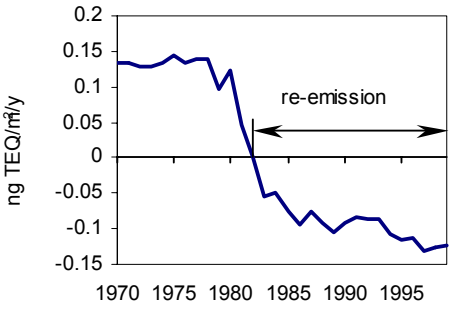


Figure 2.17. *Dynamics of total deposition flux (gas+particles, wet+dry) from the atmosphere to land calculated*

Diagram 2.16.a shows that the dynamics of PCDD/F soil content drastically lags behind from that of emissions. The total emission reduction in Europe as much as 4.6 times during the calculated period leads to rather moderate (only by 2%) decrease of accumulation in soil. It happens due to an essential half-life of PCDD/F in soil accepted in the model ($T_{1/2}$ is about 60 years).

The analysis of PCDD/F air content dynamics shows that the decrease is 2.9 times during the considered period while the emission decrease is as much as 4.6 times. This discrepancy obviously is due to re-emission flux from soil to the atmosphere. Figure 2.17 demonstrates the dynamics of total deposition flux (gas+particles, wet+dry) from the atmosphere during 1970 – 99. Negative flux values imply that the re-emission flux predominates over other deposition fluxes.

It follows from the figure that in accordance with calculations beginning with 1992 the re-emission flux dominates over the deposition flux. By the end of the considered period the re-emission flux from soil reaches 20% of the mean emission flux for Europe.

Above we discussed the calculated dynamics of accumulation and clearance of environmental media in Europe. Model calculations make it possible to detect pollution trends in different European countries. As an example we consider trends of the pollution of air and soil in Germany in comparison with its emissions (Fig. 2.18).

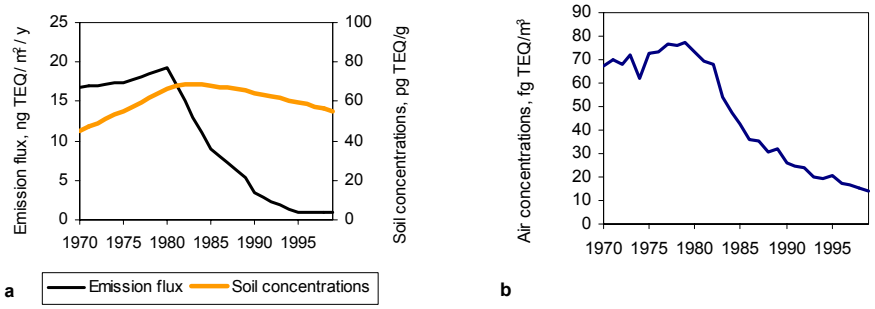


Figure 2.18. *Pollution trends in the atmosphere and soil in Germany in comparison with its emissions (calculations for 1970-99);* ***a*** *– emission flux, ng TEQ/m²/y and soil concentrations, pg TEQ/g,* ***b*** *– air concentrations, fg TEQ/m³*

The comparison of the soil pollution dynamics with the emission dynamics indicates that like in the case of Europe as a whole the trend of soil pollution drastically lags behind from emission reduction: namely in Germany 22-fold emission decrease is followed by 25% soil pollution decline.

Air pollution cuts down as much as 5.6 times during the considered period that is close to an average reduction over Europe while the emission reduction in Germany is faster than in Europe as a whole (22 times versus 4.6 times). It points out to the fact that the transboundary transport makes a substantial contribution to air pollution in Europe.

The information about trends in pollution of environmental compartments by dioxins/furans can be found in Internet www.emep.int, www.msceast.org.

2.2.3. *Evaluation of pollution levels of environmental compartments in 1999*

This section provides estimates of the pollution level of natural media in the EMEP region in 1999 made by calculations of PCDD/F long-range transport and their accumulation in environmental compartments during 1970 - 99.

The atmosphere. Figure 2.20 gives the map of atmospheric concentration spatial distribution of PCDD/F in 1999.

As evident from the maps spatial distribution of PCDD/F air concentrations is in a good agreement with spatial distribution of emissions (Fig.2.19.b). According to calculation results actually in entire Europe dioxins/furans pollution levels exceed 1 fg TEQ/m^3. Sufficiently high levels of PCDD/F air concentrations (exceeding 5 fg TEQ/m^3) are observed over a considerable part of Europe. In particular for the Czech Republic and Switzerland the calculated levels exceeded 10 fg TEQ/m^3, which corresponds to emission expert estimates for these countries [*Pacyna et al.,* 1999].

Soil. Figure 2.21 shows the map of concentration distribution in the upper soil layer (5 mm) defined for 1999 from calculations for 1970 - 99.

PCDD/F soil concentrations range between 0.01 to 20 pg TEQ/g. Since PCDD/F concentration in soil is a result of long-term accumulation for the whole period (from 1970 to 1999) the correlation between

concentration distribution in soil and the emission density in 1999 is worse than between emission density and air concentrations. Relatively high soil concentrations in the region of the Scandinavian Peninsula and in the north of Russia are explained by the role of PCDD/F scavenging to forests with subsequent washout to soil.

The highest soil concentrations (more than 5 pg TEQ/g) are characteristic of Germany, Belgium, the Netherlands, Luxembourg and partially of the Czech Republic, France, Switzerland and the United Kingdom.

Vegetation. PCDD/F spatial distribution of concentrations in vegetation is presented in Figure 2.22. We remark that values of concentrations in vegetation may be changed essentially in the course of further model modification and hence the calculated spatial distribution cannot be viewed as an estimate of real pollution levels in vegetation and is presented just as an illustration of model output.

Concentrations in vegetation are on the average about 1 pg TEQ/g reaching 10 – 20 pg TEQ/g in some regions. High calculated concentrations in vegetation (more than 3 pg TEQ/g) were defined for Central Europe and the United Kingdom.

Seawater. PCDD/F concentration distribution in seawater is mapped in Figure 2.23. Similar to concentrations in vegetation these concentrations can be essentially changed during further model modifications. The calculated spatial distribution of seawater concentrations cannot be viewed as an estimate of real pollution levels in seawater.

Concentrations exceeding 10 fg TEQ/l were obtained in regions closely located to major emission sources (the Mediterranean, the North and the Black Seas). Relatively high concentrations in seawater in the region of the northern boundary of the Scandinavian Peninsula are explained by the pollution transport with sea currents.

Deposition fluxes. Figure 2.24 shows the map of spatial distribution of total atmospheric depositions (except for the flux of gaseous exchange) in the EMEP region. The spatial distribution of the gaseous flux will be analysed below.

Like in the case of air concentration spatial distribution of total depositions correlates well with spatial distribution of emissions and air concentrations.

To evaluate the importance of re-emission for the environmental pollution by dioxins/furans it is of interest to consider spatial distribution of density of gas exchange flux between the atmosphere and the underlying surface. It is demonstrated on the map presented in Figure 2.25. The negative value of the gas flux implies the existence of re-emission, i.e. mean annual flux from the underlying surface to the atmosphere.

The calculated values of the gas flux over sea are positive and over the land they are negative. It means that there is re-emission over land and the re-emission flux is comparable with the flux value defined by the rest of scavenging processes (dry deposition of the particle phase and total wet deposition, see Fig. 2.24).

On the base of calculated spatial distribution over the EMEP domain PCDD/F mean pollution levels and depositions for all European countries were calculated (Annex C).

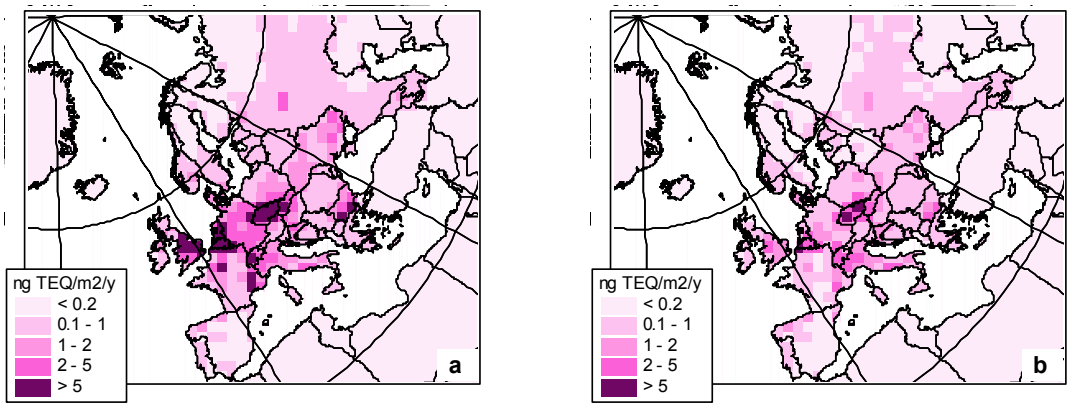


Figure 2.19. *Spatial distribution of dioxins/furans emission flux in 1990 - **a** and 1999 - **b** with spatial resolution 150x150 km used in modelling, ng TEQ/m^2/y*

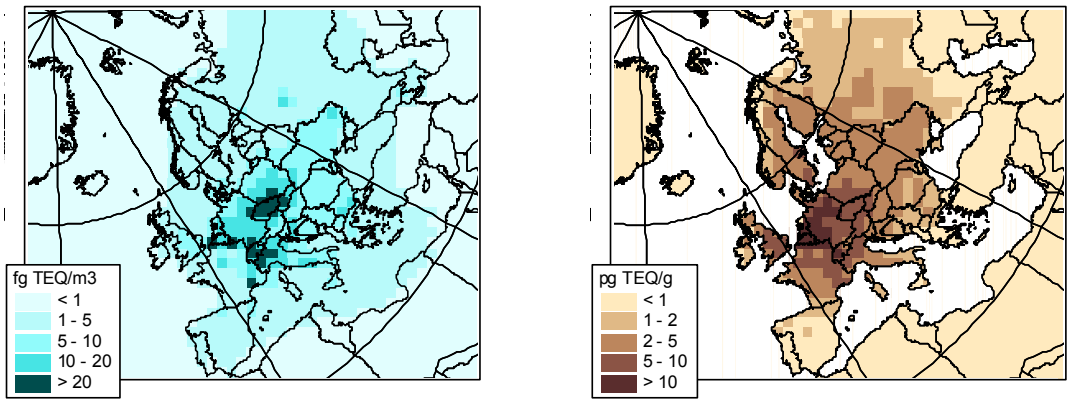


Figure 2.20. *Spatial distribution of concentrations in the surface air in 1999, fg TEQ/m^3*

Figure 2.21. *PCDD/F calculated concentrations in the upper soil layer in 1999, pg TEQ/g*

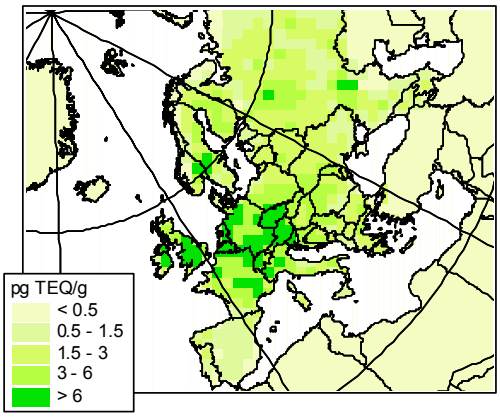


Figure 2.22. *Calculated PCDD/F concentrations in vegetation in 1999, pg TEQ/g*

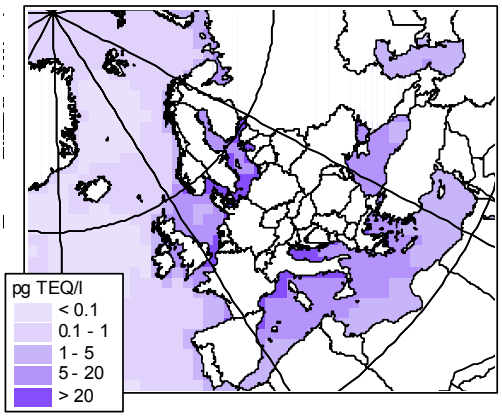


Figure 2.23. *Calculated PCDD/F concentration distribution in seawater in 1999, fg TEQ/l*

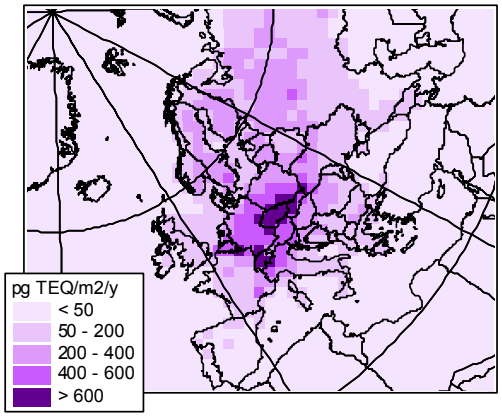


Figure 2.24. *The calculated flux of PCDD/F total (dry aerosol + wet) depositions in 1999, pg TEQ/m^2/y*

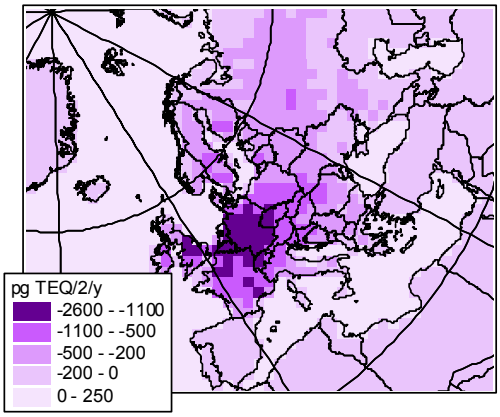


Figure 2.25. *The calculated flux of gas exchange with the underlying surface. The negative value of the gas flux implies the availability of re-emission, pg TEQ/m^2/y*

2.3. Evaluation of media response to emission reduction

The aim of this section is the evaluation of the response of environmental media to the reduction of PCDD/F emissions to air. For this purpose the simulation of PCDD/F mixture long-range transport for the period from 2000 to 2010 was carried out with the use of properties of 2,3,4,7,8-PeCDF on the assumption of full emission cessation during this time interval. Concentrations in media calculated for the end of 1999 in the previous model run were used as initial data. The specified clearance rates may be considered as the upper estimate of concentration reduction rate in natural compartments resulted from emission decline.

Figure 2.26 gives maps of the dynamics of PCDD/F content in the atmosphere and soil during the calculation period (2000 – 2010).

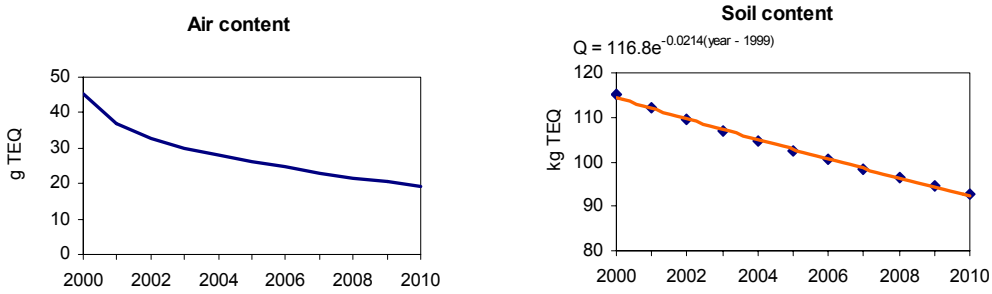


Figure 2.26. *Dynamics of PCDD/F content in the atmosphere and soil (calculations for 2000 – 2010)*

The plot of soil content is supplied by the corresponding exponential trend. Such trends approximate well clearance dynamics both in soil and in the whole environment (see below). At the same time the clearance dynamics of the atmosphere cannot be approximated by exponential dependence with sufficient accuracy. This medium is characterized by a rapid pollution decline at the initial stage of clearance and relatively slow decrease at subsequent ones. Therefore the clearance of the atmosphere cannot be described by the characteristic half-life (i.e. by the period, during which a pollutant quantity is cut down by half). To describe this process it is necessary to simulate the clearance dynamics for the whole period considered.

Coefficients of the exponential dependence defined for soil concentrations make it possible to calculate corresponding environmental half-lives. Figure 2.27 shows the dynamics of calculated PCDD/F content in the whole environment together with its exponential extrapolation up to 2035. The obtained extrapolation allows us to estimate the PCDD/F environmental half-life as about 30 years.

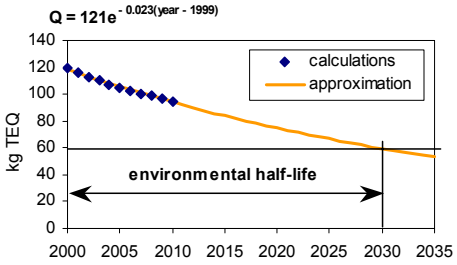


Figure 2.27. *Dynamics of PCDD/F toxicity Q in the whole environment (calculations and extrapolation up to 2035), kg TEQ*

On model assumptions the half-life in soil is approximately 60 years (550000 hours) in accordance with [*Sinkkonnen and Paasivirta*, 2000] and half-lives in other compartments are considerably shorter. Thus due to redistribution between media and degradation in them the characteristic clearance time in the environment appeared to be appreciably shorter than the half-life in soil.

2.4. Conclusive remarks

In the current year the environmental behaviour of dioxins/furans was further investigated. It has been shown that main PCDD/F medium-accumulator is soil. The accumulation in seawater and vegetation is essentially less significant. Toxicity profiles of PCDD/F mixture in various environmental media were evaluated. As a result of the comparison it was found that calculation estimates for toxicity profiles are

in a reasonable agreement with measurements. Differences in the behaviour of various congeners have been investigated and the estimates of inaccuracy due to usage of “indicator congener” properties for calculations of long-range transport and accumulation of PCDD/F mixture has been refined.

The work on model modification was continued. The modification of model description of atmosphere/soil exchange process including the refinement of degradation rate constants in soil was carried out. This modification resulted in better agreement between calculated and measured data on PCDD/F soil concentrations. The comparison of calculated against measured data shows that for concentrations in atmosphere and soil the agreement between measured and calculated values is mainly within a factor of 6. To refine the agreement between calculated and measured concentrations in seawater and vegetation further investigations are required.

The analysis of calculated trends for the period from 1970 to 1999 of PCDD/F concentrations in the environmental media shows that soil contamination decreases much slower than emissions. As a result re-emission from soil takes place beginning from the middle of 80^{th}. By the end of calculation period the re-emission flux has become comparable with that of anthropogenic emissions.

Additional calculations carried out for the period from 2000 to 2010. To evaluate media response to emission reduction calculations with full emission cessation were carried out. It is shown that the characteristic time of the overall PCDD/F toxicity reduction (that is, the time by which total toxicity in whole environment is twice reduced) is estimated as about 30 years.

Chapter 3

ANALYSIS OF AVAILABLE POP MEASUREMENT DATA FOR 2000

3.1. Introduction

Persistent organic pollutants (POPs) were included in EMEP's monitoring program in 1999. However, already in 1995, co-operation concerning POPs between EMEP and other international programs was extended. This co-operation included the establishment of a database and collection of already available data on POPs among the participants. A number of countries have been reporting POPs within the EMEP area in connection with different national and international programmes such as HELCOM, AMAP, OSPAR, MEDPOP. The following POPs are included in this report: PCBs, γ-HCH and benzo[a]pyrene.

3.2. The measurement sites

The location of the measurement sites for which there are data reported for POPs for 2000 are given in Table 3.1.

Table 3.1. List of monitoring stations included in the POPs data base

Country	Station codes	Station name	Location		Height above sea, m
			Lat.	Long.	
Belgium	BE4	Knokke	51° 21' N	3° 20' E	0
Czech Republic	CZ3	Kosetice	49° 35' N	15° 05' E	633
Germany	DE1	Westerland	54° 55' N	8° 18' E	12
	DE9	Zingst	54° 26' N	12° 44' E	1
Finland	FI96	Pallas	67° 58' N	24° 7' E	566
Iceland	IS91	Stórhöfdi	63° 24' N	20° 17' W	118
Ireland	IE2	Turlough Hill	53° 02' N	6° 24' W	420
Lithuania	LT15	Preila	55° 21' N	21° 04' E	5
Norway	NO42	Spitsbergen, Zeppelinfjell	78° 54' N	11° 53' E	474
	NO99	Lista	58° 06' N	6° 34' E	13
Sweden	SE2	Rørvik	57° 25' N	11° 56' E	10
	SE12	Aspvreten	58° 48' N	17° 23' E	20

The site codes used are the new EMEP codes introduced during 1992. Stations without standard EMEP codes have been coded with the country ISO code and numbers from 90 and higher.

3.3. Summaries of the data

Annual summaries of POPs in precipitation and air are given in Annex D.1 and Annex D.2, respectively. The definitions are as follows:

W. Mean: the precipitation weighted arithmetic mean value

Min: the minimum value reported for a specific component

Max: the maximum value reported for a specific component

Num bel: the number of data below the detection limit

Num samples: the number of samples for a specified component

Samp flag: a flag which gives information on the resolution of the reported data. The code used in this report is:

- D: daily
- D1: one day each week
- D2: two days each week
- W: weekly
- WC: weekly with change the first day each month
- W1: one week each month
- W2: two-weekly
- W4: four-weekly
- M: monthly
- Y: yearly

QA: a flag which gives information on the quality of the data

Arit mean: the arithmetic mean value used for air components only

Arit sd: the arithmetic standard deviation from the arithmetic mean value. It is computed for air components only

Geom mean: the geometric mean value used for air components only

50%: the 50 percentile

A more detailed description of the flags is given in [*Berg and Hjellbrekke*, 1998].

Monthly averages of POPs are given in Annex D.3 and D.4. The monthly mean values of precipitation data are precipitation weighted arithmetic averages. Average air concentrations are arithmetic averages of the reported values.

The units used for the results in this report are given in Table 3.2.

Table 3.2. *Units used for the measured components*

Components	Units
Amount of precipitation	mm
POPs in precipitation	ng/l
Benzo[a]pyrene in air	ng/m^3
PCBs in air	pg/m^3

3.4. Quality of the monitoring data

To provide sufficiently accurate data for EMEP's needs, data with expected lower accuracy have been flagged (QA) in the tables with annual summaries and monthly means. The definitions of the quality flags are as follows:

1. High detection limit
2. Site location not regionally representative
3. Sampling problems
4. Analytical problems
5. Sampling site at high altitude
6. Concentration level low compared to stations in the neighbourhood
7. Extremely long sampling time
8. Sum of wet deposition + deposited particles on the funnel. Unit: ug/m^2 month
9. Estimated values
10. Extremely high single sample concentrations

The data have been checked for outliers. Extremely high values, outside four times standard deviation in a lognormal distribution, have been flagged in the EMEP database and are excluded from this report.

It is generally difficult to give full credit to the information content in the POP data. Different sampling and analysis techniques make it difficult to compare data. For example, the Icelandic station has generally lower concentrations than the high Arctic NO42, which is reasonable, considering the geographical location in relation to known source areas, but the differences are also due to different data handling and analysis techniques. Iceland subtracts blanks, whereas Norway does not. A few data with extremely high detection limits are not included in the report (precipitation data from Ireland)

IS02 and NO42 are dominated by the low-chlorinated PCBs. CZ03 shows a more balanced composition of individual PCB congeners. There is a marked seasonal trend, with higher concentrations in the summer months than in autumn and winter.

Benzo[a]pyrene (also other PAHs) is rapidly destroyed by UV. In the absence of local sources, therefore, a pronounced seasonal trend in air is to be expected, which is seen especially for CZ03 (Figure 3.1). Different methods are used for the different stations, and the results from LT15 are e.g. given as deposition rates, $\mu g/m^2$ month.

We will have more knowledge on the quality of the data when the analytical intercomparison on POPs, carried out in the framework of EMEP, is finished late 2002. Preliminary results from Round 1 (analysis of standards) were promising, showing that most laboratories are able to analyse standards within ± 30%. Figures 3.2 and 3.3 show the results for benzo[a]pyrene and PCB28. In the next step of the intercomparison, real samples have been analysed, but these data are still not processed.

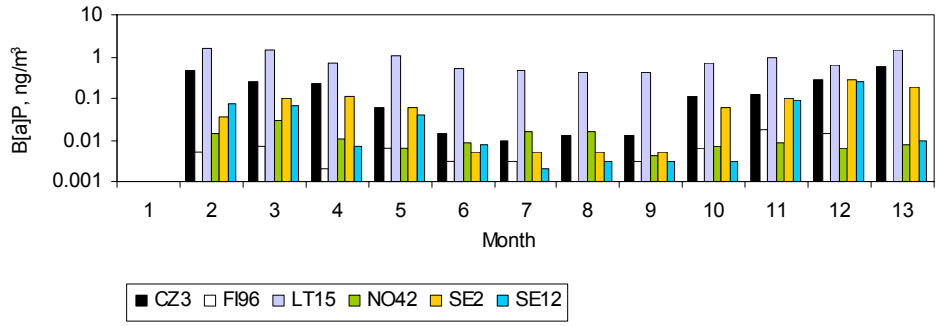


Figure 3.1. *Concentrations of benzo[a]pyrene in air (gas+aerosol) at EMEP stations in 1999*

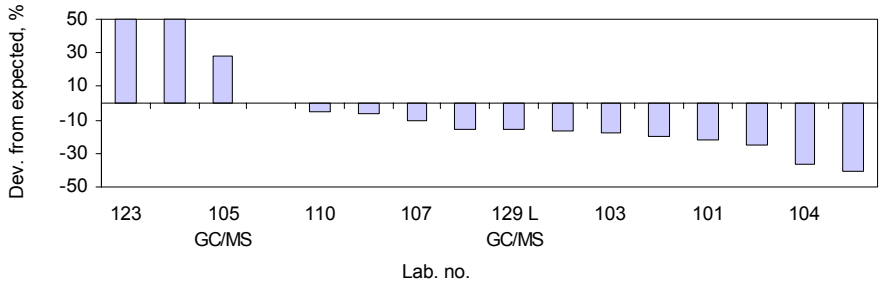


Figure 3.2. *EMEP, analytical intercomparison on POPs measurements: Preliminary results for benzo[a]pyrene*

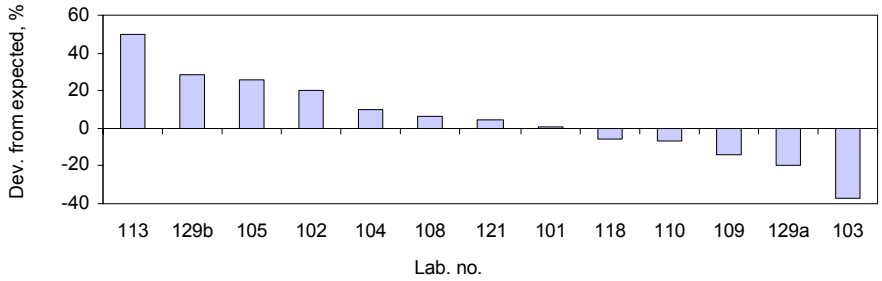


Figure 3.3. *EMEP, analytical intercomparison on POP measurements: Preliminary results for PCB-28*

CONCLUSIONS

B[a]P

The refined parametrization of wet deposition and the account of B[a]P inflow from external sources across the EMEP domain boundaries resulted in the increase of total depositions (by 15 - 20% on the average) and provided more accurate description of concentrations in precipitation and deposition fluxes. More than 75% of calculation/measurement comparisons are within a factor of 3 the rest of them are within a factor of 6. Concentrations in precipitation and depositions are better described by the model than concentrations in the atmosphere (factor 4 and 5).

Substantial deposition fluxes (100 - 180 g/km^2/y) are observed in Poland, the Czech Republic, Slovakia, Germany, Lithuania and Latvia. In the major part of Europe B[a]P surface air concentrations are within 0.1 - 1 ng/m^3. Over some areas of Poland and Germany, the Czech Republic, Slovakia, Hungary, Yugoslavia and Georgia mean annual concentrations are in the range 1 - 5 ng/m^3. Over an essential part of Europe maximum mean diurnal B[a]P concentrations exceed 1 ng/m^3 during more than 100 days a year.

The analysis of transboundary fluxes revealed that high B[a]P total depositions to the EMEP region are produced by Germany (30 t/y), Poland (28 t/y) and France (13 t/y). Greatest B[a]P depositions from external sources to European countries and regions receive Slovakia, the Czech Republic and Luxembourg (40; 34 and 31 g/km^2/y).

PCDD/F

Model results provide a reasonable description of concentration levels in the atmosphere and soil. Most part of PCDD/F environmental toxicity is accumulated in soil (about 80%). The modification of atmosphere/soil exchange module improved the agreement between calculated and measured data on PCDD/F concentrations in soil. More than a half of measurements are within a factor of 4 with respect to calculated values.

The analysis of calculated trend of PCDD/F contamination for the period from 1970 to 1999 shows that soil contamination decreases much slower than emissions. As a result re-emission from soil takes place beginning from the middle of 80th. By the end of calculation period the re-emission flux has become comparable with that of anthropogenic emissions.

Over most part of European territory levels of air concentration pollution by dioxins/furans are within 1 – 5 fg TEQ/m^3. In the Czech Republic and Switzerland calculated pollution levels are more than 10 fg TEQ/m^3. High soil concentrations (more than 5 pg TEQ/g) are characteristic of Germany, Belgium, the Netherlands, Luxembourg and partly of the Czech Republic, France, Switzerland and the United Kingdom. Calculated air concentrations and deposition fluxes agree with measurements within a factor of 6.

The model considerably overestimates concentrations in vegetation and seawater. To improve the situation it is necessary to refine degradation parameters for these media.

PCDD/F half-life in the environment is about 30 years. Therefore PCDD/F may keep the environmental pollution during decades even under full emission cessation.

To improve the evaluation of PCDD/F toxicity in the environment it is necessary to simulate long-range transport of eight selected congeners.

Acknowledgements

The authors are grateful to Prof. Kevin Jones and Dr. Peter Coleman for fruitful discussions and measurement data put at our disposal for model validation. The authors are very thankful to Mr. Sergey Dutchak, Dr. Elena Mantseva, Mr. Alexey Gusev, Mr. Michael Fedyunin and Ms. Marina Varygina for the valuable remarks. The authors are very much indebted to Mrs. Larissa Zayavlina for thorough editing the text as well as to Ms. Irina Strizhkina and Mrs. Svetlana Yevdokimova for its preparing and publishing.

REFERENCES

Abraham J., R.Ciechanovicz-Kusztal, M. Drueke, G.J.Opyd, J. Keder, W.Kulaszka and J. Novak [2000] Common report on air quality in the Black Triangle Region 1999./ CHMU, WIOS, LfUG, UBA, pp.117

Atkinson R., [1991] Atmospheric lifetimes of dibenzo-p- dioxins and dibenzofurans, *Science of the Total Environment*, v. 104, pp. 17-33.

Atkinson R. [1996] In Issues in Environmental Science and Technology; Hester R.E., Harrison R.M., Eds.; The Royal Society of Chemistry; Cambridge, UK., v. 6, pp. 53-72 cited by.

Brubaker W.W. and R.A. Hites [1997] Polychlorinated Dibenzo-p-dioxins and Dibenzofurans: Gas-Phase Hydroxyl Radical Reactions and related atmospheric removal, *Environ. Sci. Technol*., v. 31, N. 6, pp. 1805-1810.

Baart A., J.Berdowski, J.van Jaarsveld 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. et al. [1997] The European Emission Inventory of heavy metals and persistent organic pollutants for 1990. 104 02 672/03. TNO Institute of Environmental Science, Energy Research and Process Innovation.

Berg T. and A.-G.Hjellbrekke [1998] Heavy metals and POPs within the ECE region. Supplementary data for 1989-1996. Kjeller, Norwegian Institute for Air Research (NILU EMEP/CCC-Report 7/98).

Berg T. and A.-G.Hjellbrekke [1999] Heavy metals and POPs within the ECE region 1997. Kjeller, Norwegian Institute for Air Research (NILU EMEP/CCC-Report 7/99).

Berg T., A.-G.Hjellbrekke and R. Larsen [2001] Heavy metals and POPs within the EMEP region 1999/EMEP/CCC –Report 9/2001.

Berg T. and A.G.Hjellbrekke [2001] Heavy metals and POPs within the ECE region. Supplementary data for 1989-1996. Kjeller, Norwegian Institute for Air Research (EMEP/CCC-Report 7/98).

Beyer A and M.Matthies [2001] Criteria for Atmospheric Long-range Transport Potential and Persistence of Pesticides and Industrial Chemicals. Umweltforschungsplan des Bundesministerium fur Umwelt, Naturschutz und Reaktorsicherheit. Stoffbewertung, Gentechnik, Forderkennzeichen (UFOPLAN) 299 65 402.

Broman D., C. Näf and Y. Zebühr [1991a] Long –Term High- and Low-Volume Air Sampling of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans and Polycyclic Aromatic Hydrocarbons along a Transect from Urban to Remote Areas on the Swedish Baltic Coast, *Environ. Sci.Technol.*, v. 25, N.11, pp. 1841 – 1850.

Broman D., C.Näf, C. Rolff and Y. Zebühr [1991b] Occurence and Dynamics of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans and Polycyclic Aromatic Hydrocarbons in the Mixed Surface Layer of Remote Coastal and Offshore Waters of the Baltic, *Environ. Sci. Technol.*, v.25, N. 11, pp. 1850 – 1864.

Brorström-Lunden E. and C. Löfgren [1998] Atmospheric fluxes of persistent semivolatile organic pollutants to a forest ecological system at the Swedish west coast and accumulation in spruce needls, *Environmental Pollution*, v. 102, pp. 139-149.

Brorström-Lunden E., E.Junedal, H.Wingfors and S.Juntto [2000] Measurements of the Atmospheric Concentrations and the Deposition Fluxes of Persistent Organic Pollutants (POPs) at the Swedish West Coast and in the northern Fennoscandia. IVL Swedish Environmental Research Institute report, L00/14, Gotterborg 2000-03-17.

Buckley-Golder D., P.Coleman, M.Davies, K.King, A.Petersen, J.Watterson, M.Woodfield, H.Fiedler and A.Hanberget [1999] Compilation of EU Dioxin Exposure and Health Data, European Commission DG Environment and UK Department of the Environmental Transport and the Regions.

Bulgakov A., Ioannisian D., Moisseev B., Zelenko A., Resnyansky Yu. and L.Erdman [1998] Review of physical – chemical properties of polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) in respect to their long-range transport in the atmosphere // Long-range transport of selected POPs. Part II. Physical-chemical properties of dioxins and furans and factors influencing the transport and accumulation of Persistent Organic Pollutants. EMEP/MSC-E Report 2/98, Part II.

Coleman P.J., B.J.Donovan, G.W.Campbell, J.D.Watterson, K.C.Jones, R.G.M.Lee and A.J. Peters [1998] Results from the Toxic Organic Micropollutants (TOMPS) network: 1991 to 1997, National Environmental technology Centre, Report AEAT – 2167, Issue 2, May 1998.

Dutchak S., A. Malanichev, V.Shatalov, O.Travnikov and J.Munthe [2000] Long-Range Transport of Persistent Organic Pollutants and Heavy Metals: Progress and Scientific Needs, EUROTRAC Newsletter, N. 22, pp. 21 - 26.

Erdman L., A.Gusev and N.Pavlova [2001] Atmospheric Input of Persistent Organic Pollutants to the Mediterranean Sea. WMO, Map Technical Reports Series No.130

Fiedler H. [1996] Sources of PCDD/PCDF and Impact on the Environment. *Chemosphere*, v.32, N.1, pp.55-64

Franz T.P. and S.J. Eisenreich [1998] Snow scavenging of polychlorinated biphenyls and polycyclic aromatic hydrocarbones in Minnesota. *Environ. Sci Technol*., v.32, pp.1771-1778.

Govers H.A.J. and H.B.Krop [1998] Partition constants of chlorinated dibenzofurans and dibenzo-p-dioxins. *Chemosphere*, v.37, N.9-12, pp.2139-2152.

Harrad S. J and D. J. T. Smith [1997] Evaluation of a terrestrial food chain model for estimating foodstuff concentrations of PCDD/Fs, *Chemosphere*, vol. 34, No.8, pp.1723-1737.

Hoff R.M., W.M.J. Strachan, C.W. Sweet, C.H.Chan, M.Shackleton, T.F.Bidleman, K.A.Brice, D.A.Butniston, S.Cussion, D.F.Gatz, K.Harlin and W.H. Schroeder [1996], Atmospheric deposition of toxic chemicals to the Great Lakes: a review of data through 1994, *Atmos. Environ.*, v. 30, N.20, pp. 3505-3527.

Holoubek I., A. Ansorgová, V. Shatalov, S. Dutchak and J. Kohoutek [2001] Regional background monitoring of PBT compounds. The comparison of the results from measurements and monitoring, *Environ. Sci. Pollut. Res.,* 8, pp.201-211.

Holoubek I., A.Kocan, I.Holoubkova, K.Hilscherova, J.Kohoutek, J.Falandysz and O. Roots [2000] Persistent, Bioaccumulative and Toxic Chemicals in Central and Eastern European Countries – State – of – the – Art Report, /TOCOEN REPORT No. 150a. RECETOX-TOCOEN s.r.o., Brno, Czech Republic, May 2000, pp. 253.

Horstmann M. and M.S.McLachlan [1998] Atmospheric deposition of semivolatile organic compounds to two forest canopies, *Atmos. Environ.,* v.32, N.10, pp.1799-1809.

Howard P. H. and W.P.Meylan, Ed. by. [1997] Handbook of physical properties oforganic chemicals, CRC Press, Inc.

Kakareka S and T. Kukharchyk [2002] Expert estimates of PVDD/F and PCB emissions for some European countries. MSC-E Technical Note 2/2002.

Karikhoff S. W. [1981] Semiempirical estimation of sorbtion of hydrophobic pollutants on natural sediments and soil, *Chemosphere*, v.10, pp.833-846.

Kjeller L.-O., K.C.Jones, A.E.Johnston and C. Rappe [1991] Increases in the Polychlorinated Dibenzo-p-dioxin and – furan Content of Soils and vegetations since the 1840s, *Environ. Sci. Technol*., v. 25, No. 9, pp. 1619 – 1627.

Lee R., N.Green, R.Lohmann and K. Jones [1999] Seasonal, anthropogenic, air mass, and meteorological influences on the atmospheric Concentrations of polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs): Evidence foe the importance of diffuse combustion sources, *Environ. Sci. Technol*., v. 33. N. 17, pp. 2864 –2871.

Lee R.G.M. and K.C.Jones [1999] The Influence of Meteorology and Air Masses on Daily Atmospheric PCB and PAH Concentrations at a UK Location. *Environ. Sci. Technol*, v. 33, N. 5, pp. 705-712

Ligocki M.P., C.Leuenberger and J.F.Pankow [1985] Trace organic compounds in rain –III. Particle scavenging of neutral organic compounds, *Atmos. Environ.*, v. 19, pp. 1619-1626.

Lohmann R., N.J. Green and K.C. Jones [1999a] Atmospheric Transport of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs) in Air Masses Across the United Kingdom and Ireland: Evidence of Emissions and Depletion, *Environ. Sci. Technol.*, v. 33, N. 17, pp. 2872 – 2878.

Lohmann R., N.J.Green and K.C. Jones [1999b] Detailed Studies of the Factors Controlling Atmospheric PCDD/F Concentrations, *Environ. Sci. Technol.*, v. 33, N. 24, pp. 4440 – 4447.

Lohmann R. and K.Jones [1998] Dioxins and furans in air and deposition: Dioxins and furans in air and deposition: A review of levels, behaviour and processes, *Sci. Total Environment,* v. 219, pp. 53-81.

Mackay D., W.Y. Shiu and K.C. Ma [1992] Illustrated handbook of physical-chemical properties and environmental fate for organic chemicals, vol. II, Polynuclear aromatic hydrocarbons, polychlorinated dioxins and dibenzofurans, Lewis Publishers, Boca Raton, Ann Arbor, London, Tokyo, 597 p.

Mackay D., E. Webster, I. Cousins, T. Cahill, K. Foster and T. Gouin [2001]. An introduction to multimedia models. Final report prepared as a background paper for OECD Workshop, Ottawa, October 2001

Malanichev A., V.Shatalov, N.Vulykh and B.Strukov [2002] Modelling POP hemispheric transport. MSC-E Technical Report 8/2002.

Manoli E., C.Samara, I.Konstantinou and T.Albanis [2000] Polycyclic aromatic hydrocarbons in the bulk precipitation and surface waters of Northern Greece, *Chemosphere*, v.41, pp. 1845-1855.

Mantseva E., A.Malanichev and N.Vulykh [2002] Polyaromatic hydrocarbons in the environment. MSC-E Technical Note 9/2002.

Milukaite A. [2001] Investigation of benzo[a]pyrene in atmospheric air and deposition on the eastern coast of the Baltic Sea. /EUROTRAC-2. MEPOP Atmospheric cycling of mercury and persistent organic pollutants. Annual report 1998/1999. Munich, Germany, pp.37-41.

Paasivirta J., S.Sinkkonen, P.Mikkel and F.Wania [1999] Estimation of vapor pressures, solubilities and Henry's law constants of selected persistent organic pollutants as functions of temperature. *Chemosphere*, v.39, N.5, pp.811- 832.

Pacyna J.M. et al., [1999] Technical Report. Appendix 1 to Executive Final Summary Report. Environmental Cycling of Selected Persistent Organic Pollutants (POPs) in the Baltic Region (Popcycling-Baltic project). Contract No. ENV4-CT96-0214. CD-Rom

Pekar M., N.Pavlova, A.Gusev, V.Shatalov, N.Vulikh, D.Ioannisian, S.Dutchak, T.Berg and A.-G. Hjellbrekke [1999] Long-range transport of selected persistent organic pollutants. Development of transport models for polychlorinated biphenyls, benzo[a]pyrene, dioxins/furans and lindane Joint report of EMEP Centres: MSC-E and CCC, EMEP Report 4/99.

Policy on Polycyclic Aromatic Hydrocarbons in the Environment [1994] Ministry of Housing, Spatial Planning and Environment. The Netherlands.

Schwarzenbach R.P., P.M.Gschwend and Imboden D.M. [1993] Environmental organic chemistry, John Wiley and Sons Inc., New York.

Sijm D.T.H.M., H.Wever, P.J. de Vries and A.Opperhuizen [1989] Octan-1-ol/ water partition coefficients of polychlorinated dibenzo-p-dioxins and dibenzofurans: experimental values determined with a stirring method, *Chemosphere,* v.19, N.1-6, pp.263-266.

Shatalov V., A. Malanichev and S. Dutchak [2002] POP contamination on European and hemispherical scales EMEP Status Report 4/2002.

Shatalov V., A. Malanichev, N. Vulykh, T. Berg and S. Manø [2001] Assessment of POP Transport and Accumulation in the Environment. EMEP Report 4/2001.

Shatalov V., A.Malanitchev, T.Berg and R.Larsen [2000] Investigation and assessment of POP transboundary transport and accumulation in different media, EMEP Report 4/2000, Part 1,2.

Schumacher M., S.Granero, J.Rivera, L.Mueller, J.Llobet and J.Domingo [2000] Atmospheric deposition of PCDD/Fs near an old municipal solid waste incinerator: levels in soil and vegetation, *Chemosphere*, v. 40, N.6, pp. 593-600.

Schumacher M., J.L.Domingo, J.M.Llobet, W.Suenderhauf and L. Mueller [1998] Temporal variation of PCDD/F concentrations in vegetation samples collected in the vicinity of a municipal waste incinerator (1996 – 1997), *Science Total Environ.*, v. 218, pp. 175 – 183.

Sinkkonen S., N.Kämäräinen, J.Paasivirta and R. Lammi [1997] PCDDs, PCDFs, PCDTs, PCBs and some other organochlorine compounds in pine needls exposed to pulp and paper mill emissins and effects of waste combustion on the concentrations, *Chemosphere*, v. 35, N. 10, pp. 2193 – 2202.

Sinkkonen S. and J.Paasivirta [2000] Degradation half-life times of PCDDs, PCDFs and PCBs for environmental fate modeling. *Chemosphere*, v. 40, pp.943-949.

Tysklind M., I.Fängmark, S.Marklund, A.Lindskog, L.Thaning and C. Rappe [1993] Atmospheric Transport and Transformation of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans, *Environ. Sci. Technol.*, v. 27, N. 10, pp. 2190-2197.

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. and V.Shatalov [2001] Investigation of dioxin/furan composition in emissions and in environmental media. Selection of congeners for modelling. MSC-E Technical Note 6/2001.

Vassilyeva G. and V. Shatalov [2002] Behaviour of persistent organic pollutants in soil. MSC-E Technical Note 1/2002.

Wallenhorst Th., P.Krauss and H. Hagenmaier [1997] PCDD/F in ambient air and deposition in Baden-Württemberg, Germany, *Chemosphere*, v. 34, No. 5-7, pp. 1369 – 1378.

Wania F., D.Mackay anf J.T. Hoff [1999] The importance of snow scavenging of polychlorinated biphenyl and polycyclic aromatic hydrocarbon vapors, *Environ. Sci. Technol.*, v. 33, No.1, pp. 195-197.

Weiss P. [1998] Persistente organische Schadstoffe in Hintergrund-Waldgebieten Oesterreichs, Monographien Band 97, Wien, p.242.

Zurek J., K. Rymwid-Mickiewicz and W. Mill [2000]. Status report Poland 2000. Institute of environmental protection.

Ежегодник состояние загрязнения атмосферы в городах на территории России 1993 г.[1994] Санкт-Петербург, 328 с.

Annex A

Model parametrization

Here we describe the model parametrization for eight congeners involved in calculations at this stage (2,3,7,8-TCDD, 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,6,7,8-HxCDF and 1,2,3,4,7,8-HxCDF). To harmonize the parametrization of all eight congeners some parameters of four congeners considered at the previous stage (1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF, 1,2,3,6,7,8-HxCDF and 1,2,3,4,7,8-HxCDF) were refined.

Table A.1 contains the parameterization for the above listed eight congeners accepted at present for model estimation of PCDD/F mixture toxicity congener composition in various environmental compartments.

In the parametrization of new congeners the following literature sources were used:

1. Temperature dependence of the Henry's law coefficient and saturated vapour pressure over subcooled liquid were taken from [*Bulgakov and Ioannisian*, 1998]

2. Washout ratios for the particle phase are taken from [*Lohmann and Jones*, 1998] except for 2,3,7,8-TCDD, for which the mean value of the congeners was used.

3. Values of coefficients specifying the particle phase washout rate over land and sea were taken the same as those used for earlier employed congeners [*Shatalov et al*., 2000].

4. Degradation rates in the atmosphere due to the interaction with OH-radical were calculated on the base of reaction rate constants taken from [*Atkinson,* 1996 cited in *Brubaker and Hites*, 1997]. The degradation rates are calculated for the gas phase. It is presumed PCDD/F degradation in the particle phase may be neglected. As it was mentioned above in sea and soil two variants of degradation are considered. In the first variant values are taken of the handbook [*Mackay et al*., 1992]. Since data on HxCDD degradation rates in soil and sea were not found in the literature these degradation rates were taken the same as for HxCDF. In calculations by the second variant degradation coefficients were taken from [*Sinkkonen and Paasivirta*, 2000].

5. In the estimation of octanol/water partition coefficient we used works [*Mackay et al*.., 1992; *Govers and Krop*, 1998; *Howard and Meylan*, 1997; *Paasivirta et al*., 1999; *Sijm*, 1989; *Harrad and Smith,* 1997]. Corresponding coefficients K_{oc} were calculated by the fomular from [*Karikhoff*, 1981].

6. For the calculation of temperature dependences of octanol/air partition coefficient we used basic values of K_{oa} at 25^0C taken from [*Horstmann and McLachan*, 1998] together with temperature dependence coefficients for Henry's law constant.

7. Values of molecular diffusion coefficients were calculated on the base of the work [*Schwarzenbach et al*., 1993].

Besides parameterization for earlier used congeners were refined with the use of the following sources:

Washout ratios for the particle phase were taken from [*Lohmann and Jones*, 1998].

Table A.1. Model parametrization (values kept as in [*Shatalov et al*., 2000] are shaded)

		Dibenzo(p)dioxins				Dibenzofurans			
		TCDD	PeCDD	HxCDD		TCDF		PeCDF	HxCDF
		2378	12378	123678	123789	2378	23478	123678	123478
1.Temperature dependence of Henry's constant: $H = H_0 \exp(-a_H(1/T - 1/T_0)$									
H_0		0.33	0.505	$3.07\cdot10^{-2}$	$4.2\cdot10^{-2}$	0.27	$8.14\cdot10^{-2}$	$7.97\cdot10^{-2}$	0.192
a_H		10104	10182	11366	11720	8998.5	10288	11089	11126
2. Washout ratio for the particle phase W									
W		14000	9300	10000	10000	19000	12000	9800	9800
3. Temperature dependence of vapor pressure over subcooled liquid: $p_{0L} = p^0_{0L}\exp(-a_P(1/T - 1/T_0)$									
p^0_{0L}		$8.11\cdot10^{-5}$	$1.06\cdot10^{-5}$	$2.8\cdot10^{-6}$	$3.19\cdot10^{-6}$	$1.31\cdot10^{-4}$	$2.69\cdot10^{-5}$	$1.2\cdot10^{-5}$	$1.26\cdot10^{-5}$
a_P		10113	11002	11059	11414	10002	10608	10696	10718
4.1. Dry deposition velocity over land: $V_d^{land} = (A_{soil}\, u_*^2 + B_{soil}) \cdot z_0^{C_{soil}}$									
A_{soil}		0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04
B_{soil}		0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
C_{soil}		0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
4.2. Dry deposition velocity over sea: $V_d^{sea} = (A_{sea}\, u_*^2 + B_{sea})$									
A_{sea}		0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15
B_{sea}		0.023	0.023	0.023	0.023	0.023	0.023	0.023	0.023
5. Dry deposition velocity over forest: $E = \alpha u_*^{\beta}\,(1 + \gamma)$									
α		0.06	0.06	0.06	0.06	0.06	0.06	0.06	0.06
β		0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
γ		0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
6. Degradation rate constants k_d									
Air	Winter	$9.45\cdot10^{-8}$	$5.04\cdot10^{-8}$	$2.43\cdot10^{-8}$	$2.43\cdot10^{-8}$	$5.49\cdot10^{-8}$	$2.7\cdot10^{-8}$	$1.26\cdot10^{-8}$	$1.26\cdot10^{-8}$
	Spring/fall	$8.40\cdot10^{-7}$	$4.48\cdot10^{-7}$	$2.16\cdot10^{-7}$	$2.16\cdot10^{-7}$	$4.88\cdot10^{-7}$	$2.4\cdot10^{-7}$	$1.12\cdot10^{-7}$	$1.12\cdot10^{-7}$
	Summer	$2.10\cdot10^{-6}$	$1.12\cdot10^{-6}$	$5.4\cdot10^{-7}$	$5.4\cdot10^{-7}$	$1.22\cdot10^{-6}$	$6.0\cdot10^{-7}$	$2.8\cdot10^{-7}$	$2.8\cdot10^{-7}$
Soil	Var. 1*)	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$	$1.13\cdot10^{-8}$
	Var. 2**)	$2.14\cdot10^{-10}$	$1.93\cdot10^{-10}$	$3.5\cdot10^{-10}$	$3.5\cdot10^{-10}$	$3.5\cdot10^{-10}$	$3.5\cdot10^{-10}$	$3.21\cdot10^{-10}$	$3.21\cdot10^{-10}$
Sea	Var. 1*)	$3.5\cdot10^{-7}$	$3.5\cdot10^{-7}$	$1.13\cdot10^{-7}$	$1.13\cdot10^{-7}$	$3.5\cdot10^{-7}$	$3.5\cdot10^{-7}$	$1.13\cdot10^{-7}$	$1.13\cdot10^{-7}$
	Var. 2**)	$4.81\cdot10^{-8}$	$2.67\cdot10^{-8}$	$1.3\cdot10^{-8}$	$1.3\cdot10^{-8}$	$3.01\cdot10^{-8}$	$1.46\cdot10^{-8}$	$6.88\cdot10^{-9}$	$6.88\cdot10^{-9}$
7. Molar volume V_{mol}									
V_{mol}		206	218.9	231.8	231.8	199.4	212.3	225.2	225.2
8. Octanol/water partition coefficient K_{ow}									
K_{ow}		$6.31\cdot10^{6}$	$6.31\cdot10^{6}$	$9.55\cdot10^{7}$	$2.0\cdot10^{7}$	$3.39\cdot10^{6}$	$6.3\cdot10^{6}$	$3.72\cdot10^{7}$	$3.39\cdot10^{7}$
9. Organic carbon/water partition coefficient K_{oc}									
K_{oc}		$2.59\cdot10^{3}$	$2.59\cdot10^{3}$	$3.92\cdot10^{4}$	$8.2\cdot10^{3}$	$1.39\cdot10^{3}$	$2.58\cdot10^{3}$	$1.53\cdot10^{4}$	$1.39\cdot10^{4}$
10. Temperature dependence of octanol/air partition coefficient $K_{oa} = K^0_{oa} \exp(a_K(1/T - 1/T_0))$									
K^0_{oa}		$8.31\cdot10^{10}$	$3.20\cdot10^{11}$	$1.22\cdot10^{12}$	$2.48\cdot10^{12}$	$1.3\cdot10^{10}$	$7.65\cdot10^{10}$	$3.44\cdot10^{11}$	$3.15\cdot10^{11}$
a_K		10104	10182	11366	11720	8998.5	10288	11089	11126
11. Molecular diffusion coefficients D									
Air: D_a		$5.58\cdot10^{-6}$	$5.40\cdot10^{-6}$	$5.24\cdot10^{-6}$	$5.24\cdot10^{-6}$	$5.67\cdot10^{-6}$	$5.48\cdot10^{-6}$	$5.32\cdot10^{-6}$	$5.32\cdot10^{-6}$
Water: D_w		$6.53\cdot10^{-10}$	$6.3\cdot10^{-10}$	$6.09\cdot10^{-10}$	$6.09\cdot10^{-10}$	$6.66\cdot10^{-10}$	$6.42\cdot10^{-10}$	$6.2\cdot10^{-10}$	$6.2\cdot10^{-10}$

*) variant 1 – degradation rates in soil and seawater taken from [*Mackay et al*., 1992].

**) variant 2 - degradation rates in soil and seawater taken from [*Sinkkonen, Paasivirta*, 2000].

Annex B

Country-to-country matrices for B[a]P air concentrations and depositions calculated for 1999

Table B.1. B[a]P country-to-country deposition matrix, kg/y

Country		al	am	at	az	by	be	ba	bg	hr	cy	cz	dk	ee	fi	fr	ge	de	gr	hu	is	ie	it	kz	lv
Albania	**al**	26	0	2	0	0	0	17	13	8	0	3	0	0	0	6	0	5	13	7	0	0	29	0	0
Armenia	**am**	0	35	0	44	0	0	0	0	0	0	0	0	0	0	0	67	0	0	0	0	0	0	1	0
Austria	**at**	0	0	1417	0	2	8	20	1	63	0	371	3	1	1	72	0	678	0	98	0	1	207	0	3
Azerbaijan	**az**	0	11	0	288	0	0	0	0	0	0	0	0	0	0	0	123	0	0	0	0	0	0	9	0
Belarus	**by**	0	0	41	0	890	9	10	9	12	0	165	17	27	13	40	2	305	2	61	0	1	18	2	153
Belgium	**be**	0	0	1	0	0	330	0	0	0	0	4	2	0	0	291	0	221	0	0	0	2	3	0	1
Bosnia&Herzegovina	**ba**	1	0	34	0	1	1	981	9	236	0	33	1	0	0	20	0	34	2	100	0	0	86	0	1
Bulgaria	**bg**	3	0	7	0	2	1	18	1564	12	0	13	1	1	0	10	3	19	49	23	0	0	29	1	2
Croatia	**hr**	1	0	100	0	1	2	234	5	754	0	58	1	0	0	30	0	64	1	175	0	0	131	0	1
Cyprus	**cy**	0	0	0	0	0	0	0	1	0	2	0	0	0	0	0	0	0	2	0	0	0	1	0	0
Czech Republic	**cz**	0	0	247	0	3	14	9	2	16	0	2898	6	1	1	75	0	1201	0	67	0	1	49	0	6
Denmark	**dk**	0	0	1	0	1	14	0	0	0	0	4	204	1	1	36	0	177	0	1	0	2	1	0	3
Estonia	**ee**	0	0	3	0	16	3	1	1	1	0	15	8	557	38	12	0	77	0	4	0	1	3	0	203
Finland	**fi**	0	0	7	0	28	13	2	3	3	0	37	39	198	2127	45	1	256	0	9	2	3	6	1	152
France	**fr**	0	0	22	0	1	190	3	1	7	0	40	14	2	2	6908	0	1188	0	4	2	21	218	0	4
Georgia	**ge**	0	21	0	55	0	0	0	1	0	0	1	0	0	0	0	2324	2	0	1	0	0	1	4	0
Germany	**de**	0	0	208	0	6	308	6	1	13	0	625	65	5	3	1091	0	15923	0	21	1	13	175	0	18
Greece	**gr**	13	0	7	0	1	1	23	234	16	0	8	0	0	0	20	1	17	343	14	0	0	64	0	1
Hungary	**hu**	1	0	167	0	4	3	107	15	209	0	182	2	1	1	26	1	125	2	1717	0	0	72	0	3
Iceland	**is**	0	0	0	0	0	0	0	0	0	0	1	1	0	1	2	0	6	0	0	60	1	0	0	1
Ireland	**ie**	0	0	0	0	0	2	0	0	0	0	2	1	0	1	12	0	20	0	0	1	138	0	0	0
Italy	**it**	2	0	153	0	2	11	68	9	123	0	83	3	1	1	384	0	338	7	48	0	2	2616	0	2
Kazakhstan	**kz**	0	2	5	11	8	1	3	10	3	0	13	2	4	4	7	142	36	2	10	0	0	4	706	9
Latvia	**lv**	0	0	7	0	54	5	2	1	3	0	36	15	107	19	22	0	151	0	10	0	1	5	0	1190
Lithuania	**lt**	0	0	12	0	89	6	4	1	4	0	64	16	19	11	23	0	183	0	16	0	1	7	0	270
Luxembourg	**lu**	0	0	0	0	0	7	0	0	0	0	1	0	0	0	22	0	33	0	0	0	0	1	0	0
Malta	**mt**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Netherlands	**nl**	0	0	1	0	0	120	0	0	0	0	5	3	0	0	124	0	281	0	0	0	2	2	0	1
Norway	**no**	0	0	3	0	3	18	1	1	1	0	11	53	9	27	57	0	177	0	4	4	11	3	0	14
Poland	**pl**	0	0	183	0	73	42	31	9	43	0	1498	62	14	11	169	1	2353	1	276	1	5	66	1	59
Portugal	**pt**	0	0	0	0	0	2	0	0	0	0	0	0	0	0	20	0	8	0	0	0	1	2	0	0
Republic of Moldova	**md**	0	0	4	0	2	0	5	14	4	0	8	0	0	0	3	1	13	2	13	0	0	5	0	2
Romania	**ro**	3	0	48	0	10	4	115	231	82	0	99	3	2	2	31	4	121	18	355	0	1	76	2	7
Russian Federation	**ru**	2	10	115	48	498	49	51	119	55	0	411	98	602	703	218	1254	1268	21	198	5	10	83	470	857
Slovakia	**sk**	0	0	126	0	4	2	19	5	32	0	305	2	1	0	15	0	116	1	378	0	0	24	0	2
Slovenia	**si**	0	0	110	0	0	1	12	1	94	0	27	0	0	0	11	0	42	0	29	0	0	64	0	0
Spain	**es**	0	0	1	0	0	13	1	0	1	0	2	3	0	1	306	0	59	0	0	1	4	19	0	1
Sweden	**se**	0	0	11	0	20	40	4	2	4	0	53	280	70	237	127	0	700	0	15	4	11	11	1	121
Switzerland	**ch**	0	0	31	0	0	5	1	0	3	0	12	1	0	0	179	0	228	0	1	0	1	158	0	0
The FYR of Macedonia	**mk**	8	0	2	0	0	0	9	62	5	0	2	0	0	0	4	0	4	20	5	0	0	15	0	0
Turkey	**tr**	2	16	10	29	3	2	20	168	16	2	16	1	1	1	27	274	34	85	20	0	0	60	4	3
Ukraine	**ua**	2	0	100	1	182	13	62	113	67	0	263	15	17	10	68	30	414	19	377	0	2	69	13	57
United Kingdom	**gb**	0	0	2	0	0	23	0	0	0	0	9	9	1	2	115	0	139	0	1	3	57	3	0	2
Yugoslavia	**yu**	10	0	27	0	1	1	221	101	107	0	41	1	0	0	22	1	42	13	164	0	0	89	0	1
Africa	**af**	4	0	16	0	1	6	26	62	24	0	19	2	1	1	141	1	82	81	17	0	1	184	0	1
Asia	**as**	0	6	1	88	1	0	2	10	2	1	2	0	0	0	4	72	5	13	2	0	0	10	37	1
Atlantic Ocean	**atl**	0	0	12	0	4	57	2	1	3	0	44	50	11	25	445	0	513	0	7	201	120	19	0	20
Baltic Sea	**bas**	0	0	14	0	23	31	5	2	5	0	73	178	167	295	101	0	807	0	18	1	5	12	0	221
English channel	**ech**	0	0	1	0	0	8	0	0	0	0	2	1	0	0	114	0	36	0	0	0	5	1	0	0
North Sea	**nos**	0	0	5	0	2	89	1	0	2	0	21	97	4	5	245	0	454	0	4	7	32	7	0	8
Mediterranean Sea	**med**	19	0	116	0	4	29	206	258	265	4	118	8	2	3	885	4	394	399	126	1	7	1498	1	5
Black Sea	**bls**	1	2	11	4	8	2	14	142	11	0	29	2	2	1	14	266	57	25	33	0	0	24	5	6
Caspian Sea	**cas**	0	3	1	71	1	0	1	2	0	0	2	0	1	0	1	135	5	1	1	0	0	1	61	1
Arctic Region	**arc**	0	0	4	0	11	12	2	2	2	0	18	23	25	91	44	2	153	0	6	54	10	4	2	35
	SUM	99	107	3397	641	1965	1500	2317	3188	2312	9	7745	1293	1854	3643	12643	4712	29562	1126	4438	353	474	6232	1327	3448

Table B.1 (continuation). B[a]P country-to-country deposition matrix, kg/y

Country		lt	lu	mt	nl	no	pl	pt	md	ro	ru	sk	si	es	se	ch	mk	tr	ua	gb	yu	bnd *	SUM
Albania	**al**	0	0	0	0	0	6	0	0	13	0	3	2	2	0	0	54	0	2	1	135	0	347
Armenia	**am**	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	151
Austria	**at**	10	0	0	5	1	220	0	0	17	1	108	147	5	3	32	1	0	7	13	22	1	3541
Azerbaijan	**az**	0	0	0	0	0	1	0	0	1	9	0	0	0	0	0	0	0	2	0	0	2	449
Belarus	**by**	544	0	0	8	7	1498	0	3	86	68	87	7	3	26	5	3	0	310	22	31	3	4487
Belgium	**be**	1	0	0	36	1	8	0	0	0	0	0	0	5	1	2	0	0	0	47	0	2	961
Bosnia&Herzegovina	**ba**	2	0	0	1	0	62	0	0	43	1	36	31	5	1	2	8	0	8	3	261	1	2004
Bulgaria	**bg**	4	0	0	0	0	33	0	3	657	7	12	3	2	1	1	60	0	71	2	197	1	2811
Croatia	**hr**	3	0	0	1	0	83	0	0	32	1	56	225	6	1	3	3	0	8	4	122	1	2108
Cyprus	**cy**	0	0	0	0	0	1	0	0	1	0	0	0	0	0	0	0	0	1	0	1	0	12
Czech Republic	**cz**	19	0	0	11	2	703	0	0	20	1	150	14	3	4	21	1	0	9	20	17	1	5595
Denmark	**dk**	6	0	0	15	8	31	0	0	1	0	1	0	2	26	1	0	0	1	46	0	3	590
Estonia	**ee**	114	0	0	3	5	119	0	0	5	19	5	1	1	24	1	0	0	10	9	3	1	1263
Finland	**fi**	163	0	0	12	61	286	0	0	16	91	12	1	4	234	2	1	0	29	48	6	12	3910
France	**fr**	7	0	0	66	8	68	20	0	3	1	4	5	368	10	138	0	0	2	298	3	41	9669
Georgia	**ge**	1	0	0	0	0	3	0	0	5	27	1	0	0	0	0	0	0	8	0	1	1	2460
Germany	**de**	42	0	0	244	12	639	2	0	12	4	28	12	32	30	216	0	0	14	259	7	14	20052
Greece	**gr**	1	0	0	1	0	17	0	1	93	2	7	5	5	0	1	97	0	17	3	111	2	1125
Hungary	**hu**	9	0	0	2	1	378	0	1	260	2	446	81	4	2	4	6	0	36	6	313	1	4191
Iceland	**is**	1	0	0	0	4	4	0	0	0	0	0	0	0	2	0	0	0	0	5	0	11	104
Ireland	**ie**	1	0	0	2	2	4	1	0	0	0	0	0	4	1	0	0	0	0	47	0	17	257
Italy	**it**	6	0	0	6	1	125	3	0	27	1	33	87	48	3	68	9	0	9	30	66	7	4381
Kazakhstan	**kz**	17	0	0	1	2	66	0	1	47	149	8	1	1	4	1	1	0	109	4	10	45	1450
Latvia	**lv**	620	0	0	5	6	325	0	0	11	18	12	2	2	34	2	0	0	23	15	6	2	2714
Lithuania	**lt**	2271	0	0	5	5	717	0	1	17	17	22	2	2	27	2	0	0	36	16	9	2	3877
Luxembourg	**lu**	0	0	0	1	0	1	0	0	0	0	0	0	0	0	1	0	0	0	2	0	0	70
Malta	**mt**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1
Netherlands	**nl**	1	0	0	169	1	12	0	0	0	0	0	0	4	2	1	0	0	0	50	0	2	783
Norway	**no**	23	0	0	17	895	80	0	0	4	5	4	1	4	125	1	0	0	5	151	2	20	1735
Poland	**pl**	258	0	0	35	14	14847	1	2	145	19	600	27	10	52	25	4	0	194	79	82	6	21295
Portugal	**pt**	0	0	0	1	1	2	300	0	0	0	0	0	146	0	0	0	0	0	10	0	24	518
Republic of Moldova	**md**	4	0	0	0	0	39	0	40	225	3	9	2	0	1	0	2	0	91	1	18	0	514
Romania	**ro**	18	0	0	3	1	340	0	27	4910	15	143	25	5	3	3	37	0	293	9	613	2	7660
Russian Federation	**ru**	1177	0	0	40	90	2615	2	18	596	6527	201	29	20	276	19	18	0	1628	143	165	225	20934
Slovakia	**sk**	8	0	0	2	1	688	0	0	69	1	1109	22	1	2	3	2	0	27	4	55	0	3027
Slovenia	**si**	1	0	0	1	0	30	0	0	5	0	17	384	2	0	2	0	0	2	2	8	0	846
Spain	**es**	1	0	0	7	3	7	147	0	1	0	0	1	2280	3	2	0	0	0	47	1	58	2970
Sweden	**se**	179	0	0	39	336	440	1	0	21	21	17	3	10	1854	5	1	0	25	162	10	20	4855
Switzerland	**ch**	1	0	0	2	0	16	0	0	1	0	1	2	6	1	269	0	0	0	9	1	1	931
The FYR of Macedonia	**mk**	0	0	0	0	0	5	0	0	26	0	2	1	1	0	0	308	0	4	0	156	0	645
Turkey	**tr**	6	0	0	1	0	46	0	4	221	30	12	6	6	1	2	22	0	129	5	70	7	1362
Ukraine	**ua**	151	0	0	10	6	1794	1	52	1080	189	330	32	7	18	8	21	0	4138	28	216	4	9978
United Kingdom	**gb**	4	0	0	16	9	25	2	0	1	0	1	0	15	7	2	0	0	1	1470	0	29	1951
Yugoslavia	**yu**	3	0	0	1	0	87	1	1	248	2	50	14	5	1	2	129	0	24	4	2665	1	4080
Africa	**af**	3	0	0	3	1	43	7	0	53	2	11	10	73	2	7	24	0	13	18	64	99	1104
Asia	**as**	1	0	0	0	0	6	0	0	13	12	1	1	1	0	0	3	0	11	1	7	114	430
Atlantic Ocean	**atl**	37	0	0	42	139	177	70	0	8	6	9	2	387	62	7	0	0	8	538	5	1709	4743
Baltic Sea	**bas**	257	0	0	29	43	754	1	0	19	39	21	3	7	306	5	1	0	24	87	11	9	3575
English channel	**ech**	1	0	0	4	1	3	1	0	0	0	0	0	12	1	1	0	0	0	89	0	7	289
North Sea	**nos**	15	0	0	92	101	91	2	0	3	1	5	1	17	55	3	0	0	3	639	2	40	2050
Mediterranean Sea	**med**	12	0	0	15	5	229	19	2	251	8	74	99	283	10	42	97	0	65	89	337	41	6030
Black Sea	**bls**	14	0	0	2	1	109	0	11	418	71	24	5	2	3	1	12	0	381	6	58	2	1778
Caspian Sea	**cas**	3	0	0	0	0	11	0	0	7	33	1	0	0	1	0	0	0	19	1	2	6	373
Arctic Region	**arc**	52	0	0	11	128	122	1	0	12	53	7	1	5	71	1	0	0	17	92	4	399	1476
	SUM	6075	0	0	967	1902	28015	586	174	9706	7460	3684	1295	3813	3294	910	927	0	7815	4634	5873	2997	184513

* **bnd –** depositions from emission sources located outside the EMEP grid

Table B.2. B[a]P country-to-country mean annual concentration matrix, pg/m^3

Country		al	am	at	az	by	be	ba	bg	hr	cy	cz	dk	ee	fi	fr	ge	de	gr	hu	is	ie	it	kz	lv
Albania	**al**	16	0	1	0	0	0	4	4	2	0	1	0	0	0	1	0	1	5	2	0	0	6	0	0
Armenia	**am**	0	27	0	17	0	0	0	0	0	0	0	0	0	0	0	28	0	0	0	0	0	0	0	0
Austria	**at**	0	0	226	0	0	1	1	0	5	0	39	0	0	0	5	0	69	0	11	0	0	17	0	0
Azerbaijan	**az**	0	3	0	73	0	0	0	0	0	0	0	0	0	0	0	27	0	0	0	0	0	0	1	0
Belarus	**by**	0	0	2	0	79	0	0	0	0	0	6	0	1	1	1	0	9	0	3	0	0	0	0	9
Belgium	**be**	0	0	0	0	0	213	0	0	0	0	1	1	0	0	100	0	99	0	0	0	0	1	0	0
Bosnia&Herzegovina	**ba**	0	0	5	0	0	0	259	1	47	0	5	0	0	0	2	0	6	0	12	0	0	8	0	0
Bulgaria	**bg**	0	0	1	0	0	0	1	235	1	0	1	0	0	0	1	0	2	4	2	0	0	2	0	0
Croatia	**hr**	0	0	15	0	0	0	46	0	180	0	8	0	0	0	3	0	10	0	28	0	0	14	0	0
Cyprus	**cy**	0	0	0	0	0	0	0	1	0	6	0	0	0	0	0	0	0	1	0	0	0	1	0	0
Czech Republic	**cz**	0	0	32	0	0	1	1	0	1	0	582	0	0	0	6	0	151	0	7	0	0	4	0	0
Denmark	**dk**	0	0	0	0	0	2	0	0	0	0	1	125	0	0	5	0	56	0	0	0	0	0	0	1
Estonia	**ee**	0	0	0	0	3	0	0	0	0	0	2	1	234	13	1	0	8	0	0	0	0	0	0	55
Finland	**fi**	0	0	0	0	0	0	0	0	0	0	0	0	3	76	0	0	2	0	0	0	0	0	0	2
France	**fr**	0	0	0	0	0	4	0	0	0	0	0	0	0	0	154	0	19	0	0	0	0	4	0	0
Georgia	**ge**	0	5	0	10	0	0	0	0	0	0	0	0	0	0	0	606	0	0	0	0	0	0	0	0
Germany	**de**	0	0	8	0	0	8	0	0	0	0	23	3	0	0	21	0	692	0	0	0	0	4	0	0
Greece	**gr**	1	0	0	0	0	0	1	19	1	0	1	0	0	0	1	0	1	45	1	0	0	3	0	0
Hungary	**hu**	0	0	26	0	0	0	11	1	30	0	23	0	0	0	2	0	15	0	368	0	0	4	0	0
Iceland	**is**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	20	0	0	0	0
Ireland	**ie**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	2	0	0	0	22	0	0	0
Italy	**it**	0	0	5	0	0	0	2	0	4	0	2	0	0	0	9	0	10	0	1	0	0	135	0	0
Kazakhstan	**kz**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0	0	0	24	0
Latvia	**lv**	0	0	1	0	10	0	0	0	0	0	3	1	27	3	1	0	10	0	1	0	0	0	0	312
Lithuania	**lt**	0	0	1	0	17	0	0	0	0	0	6	1	3	2	1	0	13	0	2	0	0	1	0	59
Luxembourg	**lu**	0	0	1	0	0	35	0	0	0	0	2	1	0	0	75	0	161	0	0	0	0	2	0	0
Malta	**mt**	0	0	0	0	0	0	1	0	1	0	1	0	0	0	3	0	2	0	0	0	0	8	0	0
Netherlands	**nl**	0	0	0	0	0	48	0	0	0	0	2	1	0	0	30	0	130	0	0	0	0	0	0	0
Norway	**no**	0	0	0	0	0	0	0	0	0	0	0	1	0	1	1	0	3	0	0	0	0	0	0	0
Poland	**pl**	0	0	6	0	3	1	1	0	1	0	65	1	0	0	3	0	57	0	10	0	0	2	0	2
Portugal	**pt**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	1	0	0	0	0	0	0	0
Republic of Moldova	**md**	0	0	1	0	1	0	1	3	1	0	3	0	0	0	1	0	6	0	6	0	0	1	0	1
Romania	**ro**	0	0	2	0	0	0	4	12	3	0	4	0	0	0	1	0	5	0	17	0	0	2	0	0
Russian Federation	**ru**	0	0	0	0	1	0	0	0	0	0	1	0	1	1	0	6	1	0	0	0	0	0	1	1
Slovakia	**sk**	0	0	38	0	1	0	4	0	6	0	93	0	0	0	2	0	24	0	120	0	0	3	0	0
Slovenia	**si**	0	0	59	0	0	0	4	0	59	0	10	0	0	0	3	0	18	0	13	0	0	29	0	0
Spain	**es**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	0	1	0	0	0	0	0	0	0
Sweden	**se**	0	0	0	0	0	0	0	0	0	0	1	6	1	4	1	0	7	0	0	0	0	0	0	1
Switzerland	**ch**	0	0	12	0	0	1	0	0	1	0	3	0	0	0	45	0	77	0	0	0	0	49	0	0
The FYR of Macedonia	**mk**	5	0	1	0	0	0	3	26	2	0	1	0	0	0	1	0	2	9	2	0	0	4	0	0
Turkey	**tr**	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	4	0	1	0	0	0	0	0	0
Ukraine	**ua**	0	0	1	0	3	0	1	1	1	0	4	0	0	0	1	1	6	0	6	0	0	1	0	1
United Kingdom	**gb**	0	0	0	0	0	1	0	0	0	0	0	0	0	0	4	0	6	0	0	0	2	0	0	0
Yugoslavia	**yu**	1	0	3	0	0	0	24	8	13	0	4	0	0	0	1	0	4	1	18	0	0	4	0	0
Africa	**af**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	1	0	0
Asia	**as**	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0
Atlantic Ocean	**atl**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	1	0	0	1	0	0	0	0
Baltic Sea	**bas**	0	0	0	0	1	1	0	0	0	0	3	13	10	18	2	0	31	0	1	0	0	0	0	11
English channel	**ech**	0	0	0	0	0	1	0	0	0	0	0	0	0	0	19	0	6	0	0	0	1	0	0	0
North Sea	**nos**	0	0	0	0	0	3	0	0	0	0	1	4	0	0	6	0	16	0	0	0	1	0	0	0
Mediterranean Sea	**med**	0	0	0	0	0	0	1	1	1	0	1	0	0	0	4	0	2	3	0	0	0	9	0	0
Black Sea	**bls**	0	0	0	0	0	0	0	5	0	0	1	0	0	0	0	11	1	0	1	0	0	0	0	0
Caspian Sea	**cas**	0	0	0	5	0	0	0	0	0	0	0	0	0	0	0	10	0	0	0	0	0	0	4	0
Arctic Region	**arc**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Table B.2 (continuation). B[a]P country-to-country mean annual concentration matrix, pg/m^3

Country		lt	lu	mt	nl	no	pl	pt	md	ro	ru	sk	si	es	se	ch	mk	tr	ua	gb	yu	bnd *	SUM
Albania	**al**	0	0	0	0	0	1	0	0	4	0	1	0	0	0	0	26	0	1	0	55	0	132
Armenia	**am**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	74
Austria	**at**	1	0	0	0	0	14	0	0	1	0	14	16	0	0	4	0	0	0	1	2	0	428
Azerbaijan	**az**	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	106
Belarus	**by**	34	0	0	0	0	78	0	0	3	4	4	0	0	1	0	0	0	15	1	1	0	255
Belgium	**be**	0	0	0	21	0	2	0	0	0	0	0	0	1	0	0	0	0	0	13	0	0	456
Bosnia&Herzegovina	**ba**	0	0	0	0	0	8	0	0	5	0	4	5	0	0	0	1	0	1	0	47	0	418
Bulgaria	**bg**	0	0	0	0	0	3	0	0	66	0	1	0	0	0	0	7	0	4	0	24	0	358
Croatia	**hr**	1	0	0	0	0	10	0	0	3	0	6	46	0	0	0	0	0	1	1	23	0	397
Cyprus	**cy**	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	12
Czech Republic	**cz**	2	0	0	1	0	97	0	0	1	0	25	1	0	0	2	0	0	1	1	1	0	920
Denmark	**dk**	2	0	0	3	3	9	0	0	0	0	0	0	0	16	0	0	0	0	7	0	0	235
Estonia	**ee**	21	0	0	0	1	15	0	0	1	6	1	0	0	3	0	0	0	1	1	0	0	369
Finland	**fi**	2	0	0	0	1	3	0	0	0	2	0	0	0	3	0	0	0	0	0	0	0	98
France	**fr**	0	0	0	1	0	1	0	0	0	0	0	0	3	0	3	0	0	0	4	0	0	195
Georgia	**ge**	0	0	0	0	0	0	0	0	0	4	0	0	0	0	0	0	0	1	0	0	0	627
Germany	**de**	1	0	0	8	0	14	0	0	0	0	1	0	0	1	7	0	0	0	4	0	0	800
Greece	**gr**	0	0	0	0	0	1	0	0	6	0	0	0	0	0	0	9	0	1	0	7	0	99
Hungary	**hu**	1	0	0	0	0	39	0	0	34	0	72	11	0	0	0	1	0	3	1	46	0	691
Iceland	**is**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	24
Ireland	**ie**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	0	1	33
Italy	**it**	0	0	0	0	0	3	0	0	1	0	1	3	1	0	3	0	0	0	1	2	0	183
Kazakhstan	**kz**	0	0	0	0	0	1	0	0	1	3	0	0	0	0	0	0	0	1	0	0	1	36
Latvia	**lv**	117	0	0	0	0	31	0	0	1	3	1	0	0	3	0	0	0	2	1	0	0	531
Lithuania	**lt**	555	0	0	0	0	88	0	0	1	2	2	0	0	3	0	0	0	3	1	1	0	764
Luxembourg	**lu**	0	0	0	6	0	3	0	0	0	0	0	0	1	0	2	0	0	0	5	0	0	294
Malta	**mt**	0	0	0	0	0	1	0	0	0	0	0	0	1	0	0	0	0	0	0	1	0	21
Netherlands	**nl**	0	0	0	99	0	4	0	0	0	0	0	0	1	1	0	0	0	0	14	0	0	332
Norway	**no**	1	0	0	0	46	2	0	0	0	0	0	0	0	4	0	0	0	0	2	0	0	62
Poland	**pl**	9	0	0	1	0	884	0	0	4	1	27	1	0	1	1	0	0	7	1	3	0	1094
Portugal	**pt**	0	0	0	0	0	0	36	0	0	0	0	0	12	0	0	0	0	0	1	0	1	52
Republic of Moldova	**md**	1	0	0	0	0	15	0	30	137	1	4	1	0	0	0	0	0	40	0	7	0	261
Romania	**ro**	1	0	0	0	0	12	0	1	338	0	6	1	0	0	0	1	0	10	0	30	0	454
Russian Federation	**ru**	2	0	0	0	0	4	0	0	1	22	0	0	0	0	0	0	0	4	0	0	0	49
Slovakia	**sk**	1	0	0	0	0	152	0	0	11	0	449	4	0	0	0	0	0	4	1	12	0	931
Slovenia	**si**	1	0	0	0	0	10	0	0	1	0	5	291	0	0	1	0	0	1	1	3	0	510
Spain	**es**	0	0	0	0	0	0	2	0	0	0	0	0	54	0	0	0	0	0	1	0	1	65
Sweden	**se**	2	0	0	0	6	5	0	0	0	0	0	0	0	50	0	0	0	0	1	0	0	88
Switzerland	**ch**	0	0	0	1	0	3	0	0	0	0	0	1	1	0	122	0	0	0	2	0	0	319
The FYR of Macedonia	**mk**	0	0	0	0	0	1	0	0	7	0	1	0	0	0	0	216	0	1	0	73	0	354
Turkey	**tr**	0	0	0	0	0	1	0	0	2	0	0	0	0	0	0	0	0	1	0	1	0	14
Ukraine	**ua**	2	0	0	0	0	27	0	1	20	4	5	0	0	0	0	0	0	129	0	3	0	220
United Kingdom	**gb**	0	0	0	1	0	1	0	0	0	0	0	0	0	0	0	0	0	0	114	0	1	132
Yugoslavia	**yu**	0	0	0	0	0	7	0	0	20	0	5	1	0	0	0	17	0	1	0	430	0	566
Africa	**af**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	5
Asia	**as**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	4
Atlantic Ocean	**atl**	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	1	0	3	9
Baltic Sea	**bas**	14	0	0	1	1	42	0	0	1	2	1	0	0	18	0	0	0	1	2	0	0	173
English channel	**ech**	0	0	0	1	0	0	0	0	0	0	0	0	1	0	0	0	0	0	27	0	1	59
North Sea	**nos**	1	0	0	4	4	3	0	0	0	0	0	0	0	2	0	0	0	0	20	0	0	65
Mediterranean Sea	**med**	0	0	0	0	0	1	0	0	1	0	0	0	1	0	0	1	0	0	0	2	0	30
Black Sea	**bls**	0	0	0	0	0	3	0	0	17	2	1	0	0	0	0	0	0	15	0	2	0	63
Caspian Sea	**cas**	0	0	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	1	0	0	0	24
Arctic Region	**arc**	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	5

*** bnd –** concentrations formed from emission sources located outside the EMEP grid

Annex C

Contamination levels in European countries

Table C1. Average levels of PCDD/F contamination and depositions in European countries (TEQ)

Country	Air, fg/m^3	Soil, pg/g	Vegetation, pg/g	Seawater, pg/m^3	Deposition flux, pg/m^2/y	Emission flux, pg/m^2/y
Albania	4.65	1.16	1.15	18.4	-28.9*	91.6
Armenia	4.11	0.40	0.90		-11.3	899
Austria	14.1	6.89	8.20		-542	597
Azerbaijan	3.37	0.28	0.95	7.40	-2.12	574
Belarus	5.46	2.63	2.69		-190	75
Belgium	22	13.8	12.4	101	-1530	4370
Bosnia&Herzegovina	7.92	2.46	4.33	26.5	-262	431
Bulgaria	12.5	1.99	2.77	24.8	-114	2240
Croatia	10.4	2.89	5.10	33.8	-225	1610
Cyprus	1.05	0.11	0.00	3.56	25.6	85.7
the Czech Republic	35.1	9.17	11		-461	8170
Denmark	6.25	3.03	0.79	26.2	47.3	2020
Estonia	3.49	2.59	0.90	20	-51.2	272
Finland	2.01	2.77	2.65	16.8	-110	122
France	7.82	5.69	6.21	24	-725	1010
Georgia	4.43	0.51	1.30	11.9	-26.2	670
Germany	13.8	11.1	7.07	28.9	-1020	868
Greece	6.50	0.90	2.53	12	-15.9	910
Hungary	11.9	4.28	4.02		-281	409
Iceland	0.25	0.06	0.09	0.53	1.35	5.35
Ireland	1.08	1.28	4.07	3.63	-241	195
Italy	13.7	4.07	4.16	22.8	-129	2620
Kazakhstan	1.31	0.18	0.32	4.02	-10.1	39.9
Latvia	3.9	2.71	1.36	22.5	-118	193
Lithuania	4.76	3.09	2.36	22.9	-238	76.4
Luxembourg	23.3	14.5	18.9		-2090	3590
Malta	2.21	0.07	0.00	7.28	66.7	0
the Netherlands	12.8	12.1	7.21	49.6	-966	952
Norway	2.04	1.66	1.38	6.6	-14.1	320
Poland	13.4	5.35	3.76	38.8	-187	919
Portugal	1.96	0.84	1.14	3.11	-36.3	383
Moldova	7.27	2.73	4.02	27.8	-194	297
Romania	7.25	2.57	3.36	17.1	-158	341
Russian Federation	2.64	1.48	2.08	8.86	-82.6	167
Slovakia	21.2	5.70	4.87		-206	3300
Slovenia	11.3	4.90	7.16	59.4	-375	193
Spain	2.40	1.07	1.10	7.24	-78.8	310
Sweden	2.39	2.98	2.70	18.5	-132	73.5
Switzerland	25.2	9.87	9.28		-519	4340
Macedonia	8.24	1.66	2.35	20	-139	356
Turkey	1.60	0.11	0.18	8.74	2.02	0
the Ukraine	8.31	3.08	2.37	17.8	-74.2	950
The United Kingdom	4.66	5.24	5.80	11.6	-531	1350
Yugoslavia	7.53	2.72	3.45	23.3	-196	527

* Negative value of deposition flux means prevailing of re-emission process

Calculated levels of contamination for European countries (especially on concentrations in seawater and vegetation) are preliminary and will be refined in the course of further investigations.

Annex D1

Annual statistics on POPs in precipitation/deposition

BE0004R Knokke Belgium

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
Precip	-	28.6	98.1	0	14		M
gamma_HCH	18.56	3.00	64.00	0	13		M

DE0001R Westerland Germany

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
Precip	-	22.4	109.2	0	12		M
benzo_a_pyrene	3.213	1.200	11.900	3	12		M
gamma_HCH	6.24	1.80	29.00	1	12		M

FI0096R Pallas Finland

January 2000 - December 2000

Component (ng/m^2 day)	Mean mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	1.824	0.500	5.000	7	12	8	M
gamma_HCH	1.07	0.04	5.46	0	12	8	M
PCB_101	0.045	0.005	0.100	2	12	8	M
PCB_118	0.013	0.005	0.050	7	12	8	M
PCB_138	0.048	0.020	0.130	0	12	8	M
PCB_153	0.063	0.020	0.150	0	12	8	M
PCB_180	0.022	0.005	0.080	2	12	8	M
PCB_28	0.106	0.010	0.820	1	12	8	M
PCB_52	0.132	0.015	0.500	2	12	8	M

IE0002R Turlough Hill Ireland

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
Precip	-	42.3	390.0	0	12		M
gamma_HCH	2.27	0.50	8.50	10	10	1	M
PCB_101	2.462	0.500	14.500	10	10	1	M
PCB_118	2.494	0.500	14.500	10	10	1	M
PCB_138	2.274	0.500	16.500	10	10	1	M
PCB_153	2.274	0.500	16.500	10	10	1	M
PCB_180	1.895	0.500	8.500	10	10	1	M
PCB_52	1.645	0.500	8.500	10	10	1	M
Precip	-	42.3	390.0	0	12	1	M

IS0091R Storhofdi Iceland

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
Precip	-	2.5	65.0	0	24		W4
gamma_HCH	0.10	0.05	0.36	0	24		W4
PCB_101	0.010	0.002	0.128	21	24		W4
PCB_118	0.009	0.002	0.147	20	24		W4
PCB_138	0.015	0.003	0.140	19	24		W4
PCB_153	0.017	0.004	0.140	17	24		W4
PCB_180	0.012	0.002	0.143	20	24		W4
PCB_28	0.074	0.014	0.960	22	24		W4
PCB_52	0.029	0.006	0.360	21	24		W4

LT0015R Preila Lithuania

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	24.378	10.700	50.300	0	12		M

NL0091R

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
Precip	-	18.7	149.3	0	10		W4
gamma_HCH	10.38	5.00	70.00	8	11	1	W4

NO0099R Lista Norway

January 2000 - December 2000

Component	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
Precip	-	2.9	94.4	0	53		W
gamma_HCH	3.09	0.23	25.79	0	52		W

SE0002R Rorvik Sweden

January 2000 - December 2000

Component (ng/m2 day)	Mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	16.458	0.500	100.000	3	12	8	M
gamma_HCH	2.29	0.04	18.91	4	12	8	M
PCB_101	0.130	0.015	0.290	1	12	8	M
PCB_118	0.081	0.015	0.320	3	12	8	M
PCB_138	0.227	0.090	0.410	0	12	8	M
PCB_153	0.259	0.110	0.500	0	12	8	M
PCB_180	0.155	0.060	0.250	0	12	8	M
PCB_28	0.138	0.035	0.670	3	12	8	M
PCB_52	0.110	0.010	0.430	0	12	8	M

SE0012R Aspvreten Sweden

January 2000 - December 2000

Component (ng/m2 day)	W. mean	Min	Max	Num bel	Num sampl	QA flag	Samp flag
alpha_HCH	0.93	0.00	3.97	0	12	8	M
gamma_HCH	4.32	0.00	25.15	0	12	8	M
PCB_101	0.060	0.010	0.130	0	12	8	M
PCB_118	0.052	0.010	0.160	0	12	8	M
PCB_138	0.079	0.010	0.150	0	12	8	M
PCB_153	0.076	0.020	0.180	0	12	8	M
PCB_180	0.086	0.010	0.510	0	12	8	M
PCB_28	0.163	0.010	0.460	0	12	8	M
PCB_52	0.087	0.010	0.260	0	12	8	M

Annex D2

Annual statistics on POPs in air+aerosols

CZ0003R Kosetice Czech Republic

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	0.169	0.287	0.058	4.747	0.005	0.059	1.757	13.7	11	50		W
gamma_HCH	41.66	35.46	29.99	2.33	7.00	41.00	199.00	13.7	0	50		W
PCB_101	24.560	8.550	23.270	1.385	13.000	22.000	48.000	13.7	0	50		W
PCB_118	5.080	1.455	4.880	1.333	3.000	5.000	8.000	13.7	0	50		W
PCB_138	14.680	5.850	13.550	1.509	5.000	14.000	28.000	13.7	0	50		W
PCB_153	21.480	5.581	20.779	1.298	13.000	21.000	32.000	13.7	0	50		W
PCB_180	5.880	3.623	5.145	1.624	3.000	4.000	17.000	13.7	0	50		W
PCB_28	30.800	9.298	29.446	1.356	16.000	28.000	52.000	13.7	0	50		W
PCB_52	38.620	19.841	35.035	1.521	17.000	31.000	106.000	13.7	0	50		W

FI0096R Pallas Finland

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	0.006	0.005	0.004	3.036	0.001	0.003	0.017	23.0	2	12		M
gamma_HCH	10.08	8.11	7.58	2.22	2.00	7.00	29.00	23.0	0	12		M
PCB_101	0.958	0.416	0.865	1.641	0.376	0.934	1.697	23.0	0	12		M
PCB_118	0.314	0.141	0.280	1.695	0.100	0.304	0.534	23.0	0	12		M
PCB_138	0.364	0.173	0.323	1.724	0.113	0.324	0.724	23.0	0	12		M
PCB_153	0.391	0.160	0.357	1.614	0.153	0.397	0.636	23.0	0	12		M
PCB_180	0.079	0.052	0.059	2.460	0.015	0.063	0.172	23.0	3	12		M
PCB_28	1.969	1.084	1.721	1.738	0.644	1.608	4.664	23.0	0	12		M
PCB_52	2.320	1.473	1.930	1.910	0.666	1.934	5.774	23.0	0	12		M

IS0091R Storhofdi Iceland

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
gamma_HCH	3.86	1.50	3.55	1.50	1.82	3.67	7.16	99.3	0	24		W2
PCB_101	0.301	0.157	0.266	1.662	0.090	0.280	0.890	99.3	16	24		W2
PCB_105	0.10	0.05	0.09	1.74	0.04	0.12	0.17	99.3	23	24		W2
PCB_118	0.156	0.061	0.147	1.489	0.075	0.180	0.320	99.3	22	24		W2
PCB_138	0.240	0.100	0.222	1.550	0.115	0.180	0.385	99.3	24	24		W2
PCB_153	0.275	0.097	0.256	1.521	0.125	0.320	0.385	99.3	20	24		W2
PCB_156	0.10	0.01	0.10	1.12	0.07	0.10	0.12	99.3	24	24		W2
PCB_180	0.171	0.075	0.156	1.605	0.075	0.162	0.275	95.0	23	23		W2
PCB_28	1.705	0.917	1.505	1.732	0.680	1.975	4.280	99.3	23	24		W2
PCB_31	1.390	0.765	1.249	1.649	0.605	1.525	4.040	99.3	23	24		W2
PCB_52	0.706	0.473	0.623	1.678	0.305	0.740	2.610	99.3	23	24		W2

LT0015R Preila Lithuania

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	0.857	0.411	0.772	1.619	0.410	0.710	1.540	100.0	0	12		M

NO0042G Zeppelin, Spitsbergen Norway

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	0.011	0.010	0.008	2.780	0.000	0.009	0.054	29.5	1	54		W
gamma_HCH	5.90	2.25	5.39	1.64	0.43	5.43	10.80	28.4	0	52		W
PCB_101	0.807	0.945	0.618	1.903	0.080	0.610	5.630	28.4	0	52		W
PCB_105	0.10	0.11	0.07	1.90	0.03	0.07	0.68	28.4	0	52		W
PCB_118	0.294	0.351	0.226	1.838	0.090	0.200	2.330	28.4	0	52		W
PCB_138	0.308	0.376	0.215	2.202	0.020	0.180	2.360	28.4	0	52		W
PCB_153	0.398	0.514	0.295	1.927	0.100	0.260	3.290	28.4	0	52		W
PCB_156	0.03	0.03	0.03	1.90	0.01	0.02	0.19	28.4	0	52		W
PCB_180	0.103	0.092	0.083	1.889	0.010	0.080	0.550	28.4	1	52		W
PCB_28	5.152	4.001	4.197	1.867	0.810	3.800	24.800	28.4	0	52		W
PCB_31	4.865	3.786	3.958	1.875	0.690	3.620	23.600	28.4	0	52		W
PCB_52	1.733	1.119	1.502	1.691	0.240	1.400	7.110	28.4	0	52		W

NO0099R Lista Norway

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
gamma_HCH	24.52	24.09	17.23	2.27	3.25	16.70	110.00	14.2	0	52		W

SE0002R Rorvik Sweden

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	0.078	0.081	0.036	4.671	0.005	0.059	0.267	23.0	4	12		M
gamma_HCH	23.92	21.61	17.01	2.34	5.00	17.00	69.00	23.0	0	12		M
PCB_101	2.752	1.832	2.368	1.727	1.056	2.349	7.951	23.0	0	12		M
PCB_118	0.903	0.539	0.794	1.668	0.357	0.825	2.397	23.0	0	12		M
PCB_138	1.507	0.734	1.368	1.575	0.616	1.201	3.280	23.0	0	12		M
PCB_153	1.673	0.858	1.504	1.604	0.692	1.360	3.776	23.0	0	12		M
PCB_180	0.560	0.288	0.503	1.609	0.242	0.461	1.228	23.0	0	12		M
PCB_28	2.518	1.417	2.224	1.661	0.962	2.014	5.980	23.0	0	12		M
PCB_52	3.499	2.744	2.870	1.846	1.291	2.340	11.175	23.0	0	12		M

SE0012R Aspvreten Sweden

January 2000 - December 2000

Component	Arit mean	Arit sd	Geom mean	Geom sd	Min	50%	Max	% anal	Num bel	Num sampl	QA flag	Samp flag
benzo_a_pyrene	0.048	0.075	0.015	5.111	0.002	0.008	0.262	23.0	5	12		M
gamma_HCH	74.08	63.74	47.26	2.89	12.00	35.00	199.00	23.0	0	12		M
PCB_101	2.174	0.746	2.037	1.489	0.890	1.950	3.017	23.0	0	12		M
PCB_118	0.816	0.311	0.761	1.491	0.382	0.793	1.432	23.0	0	12		M
PCB_138	1.173	0.479	1.079	1.555	0.509	1.182	2.132	23.0	0	12		M
PCB_153	1.410	0.550	1.303	1.536	0.602	1.313	2.354	23.0	0	12		M
PCB_180	0.384	0.167	0.354	1.534	0.168	0.354	0.792	23.0	0	12		M
PCB_28	3.492	1.784	3.165	1.563	1.451	2.826	7.567	23.0	0	12		M
PCB_52	3.440	1.150	3.241	1.459	1.441	3.298	4.993	23.0	0	12		M

Annex D3

Monthly means for POPs in precipitation/deposition

BE0004R gamma_HCH	6.062	5.202	29.947	62.614	52.307	14.929	6.394	7.964	6.000	6.705	8.000	3.000
DE0001R benzo_a_pyrene	1.300	2.167	1.251	3.950	3.206	3.390	3.231	11.396	3.289	1.948	3.229	4.400
DE0001R gamma_HCH	1.800	4.123	5.244	28.230	16.441	8.083	4.810	5.488	5.219	4.174	3.478	2.600
FI0096R benzo_a_pyrene	0.500	3.000	0.500	3.000	2.000	0.500	0.500	5.000	0.500	5.000	0.500	0.500
FI0096R gamma_HCH	0.040	0.040	0.050	0.180	5.460	4.670	0.120	0.200	0.210	3.930	0.430	0.070
FI0096R PCB_101	0.020	0.030	0.005	0.040	0.005	0.100	0.040	0.040	0.050	0.080	0.100	0.050
FI0096R PCB_118	0.010	0.005	0.005	0.005	0.005	0.050	0.010	0.005	0.005	0.020	0.030	0.005
FI0096R PCB_138	0.030	0.030	0.020	0.030	0.050	0.130	0.060	0.050	0.040	0.050	0.070	0.050
FI0096R PCB_153	0.030	0.050	0.020	0.040	0.120	0.150	0.070	0.050	0.030	0.080	0.100	0.060
FI0096R PCB_180	0.010	0.010	0.005	0.010	0.005	0.080	0.030	0.040	0.010	0.020	0.040	0.020
FI0096R PCB_28	0.030	0.030	0.010	0.050	0.820	0.100	0.040	0.070	0.110	0.140	0.140	0.030
FI0096R PCB_52	0.080	0.120	0.015	0.015	0.500	0.190	0.120	0.130	0.130	0.120	0.210	0.090
IE0002R gamma_HCH	7.000	3.000	8.500	-	-	1.000	2.000	2.500	0.500	0.500	1.000	3.500
IE0002R PCB_52	7.000	3.000	8.500	-	-	2.000	1.000	1.500	1.000	0.500	0.500	1.000
IE0002R PCB_101	14.500	6.000	8.500	-	-	2.000	1.000	1.500	1.000	1.000	0.500	1.000
IE0002R PCB_118	14.500	6.000	8.500	-	-	4.500	1.000	1.500	1.000	0.500	0.500	1.000
IE0002R PCB_153	7.000	6.000	16.500	-	-	2.000	1.000	4.000	1.000	0.500	0.500	1.000
IE0002R PCB_138	7.000	6.000	16.500	-	-	2.000	1.000	4.000	1.000	0.500	0.500	1.000
IE0002R PCB_180	7.000	6.000	8.500	-	-	2.000	1.000	1.500	1.000	0.500	0.500	1.000
IS0091R gamma_HCH	0.076	0.091	0.079	0.130	0.224	0.249	0.090	0.060	0.072	0.086	0.122	0.078
IS0091R PCB_101	0.017	0.011	0.008	0.049	0.009	0.032	0.004	0.004	0.004	0.003	0.041	0.005
IS0091R PCB_118	0.012	0.007	0.005	0.049	0.006	0.022	0.004	0.004	0.003	0.003	0.046	0.008
IS0091R PCB_138	0.020	0.012	0.009	0.086	0.011	0.038	0.006	0.006	0.007	0.004	0.041	0.013
IS0091R PCB_153	0.020	0.012	0.009	0.107	0.011	0.038	0.013	0.007	0.008	0.004	0.053	0.012
IS0091R PCB_180	0.014	0.009	0.007	0.113	0.008	0.027	0.008	0.004	0.004	0.003	0.014	0.005
IS0091R PCB_28	0.138	0.085	0.064	0.175	0.073	0.259	0.037	0.035	0.043	0.026	0.083	0.130
IS0091R PCB_52	0.052	0.032	0.024	0.065	0.027	0.097	0.028	0.016	0.016	0.011	0.037	0.039
LT0015R benzo_a_pyrene	42.300	24.300	18.300	34.700	18.000	15.000	12.300	16.700	16.700	10.700	33.300	50.300
NL0091R gamma_HCH	5.000	5.000	16.825	66.836	70.000	5.000	5.000	5.000	5.000	5.000	5.000	5.000
NO0099R gamma_HCH	0.635	0.820	0.679	14.116	9.216	2.884	1.527	1.795	2.090	2.553	1.693	1.443
SE0002R benzo_a_pyrene	2.000	16.000	11.000	13.000	15.000	3.000	0.500	0.500	0.500	10.000	100.000	26.000
SE0002R gamma_HCH	0.035	0.470	0.035	7.120	18.910	0.035	0.070	0.035	0.080	0.470	0.100	0.080
SE0002R PCB_101	0.060	0.190	0.120	0.080	0.290	0.110	0.015	0.070	0.140	0.160	0.070	0.260
SE0002R PCB_118	0.015	0.070	0.090	0.060	0.110	0.050	0.015	0.015	0.320	0.090	0.050	0.090
SE0002R PCB_138	0.090	0.240	0.410	0.160	0.370	0.160	0.090	0.100	0.360	0.230	0.200	0.310
SE0002R PCB_153	0.140	0.280	0.380	0.160	0.380	0.240	0.110	0.130	0.500	0.280	0.160	0.350
SE0002R PCB_180	0.060	0.180	0.220	0.110	0.250	0.100	0.060	0.110	0.180	0.190	0.190	0.210
SE0002R PCB_28	0.035	0.110	0.060	0.060	0.200	0.120	0.035	0.130	0.670	0.110	0.035	0.090
SE0002R PCB_52	0.070	0.120	0.060	0.010	0.160	0.020	0.020	0.100	0.430	0.150	0.080	0.100
SE0012R benzo_a_pyrene	3.000	20.000	0.000	22.000	0.000	0.500	1.500	1.000	0.500	2.000	186.000	0.500
SE0012R gamma_HCH	0.000	0.000	0.000	0.090	25.150	5.930	3.640	0.430	0.000	2.960	12.490	1.900
SE0012R PCB_101	0.120	0.110	0.090	0.130	0.060	0.050	0.050	0.020	0.020	0.010	0.050	0.040
SE0012R PCB_118	0.160	0.070	0.070	0.100	0.050	0.030	0.040	0.010	0.020	0.010	0.070	0.020
SE0012R PCB_138	0.140	0.130	0.140	0.150	0.080	0.050	0.050	0.020	0.020	0.010	0.150	0.050
SE0012R PCB_153	0.180	0.110	0.100	0.130	0.090	0.060	0.060	0.020	0.030	0.020	0.090	0.060
SE0012R PCB_180	0.080	0.080	0.090	0.080	0.510	0.050	0.020	0.010	0.020	0.010	0.090	0.030
SE0012R PCB_28	0.170	0.230	0.140	0.260	0.360	0.460	0.110	0.200	0.020	0.010	0.060	0.020
SE0012R PCB_52	0.130	0.150	0.070	0.150	0.060	0.260	0.060	0.100	0.020	0.010	0.050	0.030

Annex D4

Monthly means for POPs in air+aerosols

CZ0003R benzo_a_pyrene	0.447	0.256	0.222	0.059	0.015	0.010	0.013	0.013	0.116	0.128	0.267	0.555
CZ0003R gamma_HCH	25.667	37.250	45.400	90.250	69.200	66.500	67.333	39.200	15.500	25.750	10.000	11.000
CZ0003R PCB_101	18.667	16.250	18.400	23.750	22.000	22.250	19.000	29.200	38.500	37.000	24.000	24.000
CZ0003R PCB_118	5.000	3.500	4.000	4.250	5.400	5.500	4.667	6.800	6.250	6.500	4.400	4.500
CZ0003R PCB_138	12.667	8.750	9.200	7.750	16.800	14.000	10.333	18.000	19.750	22.000	17.200	17.500
CZ0003R PCB_153	22.000	15.000	17.200	16.000	24.200	24.250	18.667	25.400	26.000	26.500	20.200	21.500
CZ0003R PCB_180	7.000	3.750	4.000	4.500	10.200	9.000	3.333	4.800	5.500	6.000	5.200	6.750
CZ0003R PCB_28	22.000	19.250	24.600	26.750	35.200	35.750	28.333	38.400	40.750	44.500	25.200	26.000
CZ0003R PCB_52	43.667	30.500	36.800	37.750	82.600	53.000	25.000	24.600	33.750	36.000	26.600	27.000
FI0096R gamma_HCH	2.000	3.000	5.000	8.000	29.000	18.000	12.000	7.000	4.000	19.000	9.000	5.000
FI0096R benzo_a_pyrene	0.005	0.007	0.002	0.006	0.003	0.003	0.001	0.003	0.006	0.017	0.014	0.001
FI0096R PCB_101	0.383	0.376	0.652	0.833	1.202	1.259	1.697	1.340	0.937	1.312	0.934	0.567
FI0096R PCB_138	0.134	0.113	0.268	0.301	0.456	0.532	0.724	0.409	0.324	0.484	0.379	0.244
FI0096R PCB_153	0.153	0.160	0.285	0.342	0.501	0.510	0.587	0.479	0.397	0.636	0.411	0.237
FI0096R PCB_180	0.049	0.015	0.015	0.063	0.120	0.172	0.015	0.141	0.090	0.117	0.091	0.056
FI0096R PCB_28	0.644	0.791	1.322	1.608	2.050	2.598	4.664	2.701	1.536	2.388	2.155	1.172
FI0096R PCB_52	0.666	0.809	1.228	1.665	2.457	3.468	5.774	3.847	2.444	2.435	1.934	1.116
FI0096R PCB_118	0.100	0.127	0.260	0.304	0.332	0.369	0.528	0.423	0.228	0.534	0.371	0.190
IS0091R gamma_HCH	1.883	1.959	3.046	4.637	6.464	4.610	3.308	2.860	5.255	3.645	4.950	3.524
IS0091R PCB_101	0.275	0.314	0.261	0.309	0.277	0.289	0.318	0.212	0.390	0.105	0.290	0.566
IS0091R PCB_105	0.140	0.156	0.132	0.154	0.137	0.144	0.047	0.049	0.082	0.050	0.053	0.049
IS0091R PCB_118	0.185	0.208	0.174	0.206	0.185	0.194	0.091	0.097	0.140	0.097	0.102	0.191
IS0091R PCB_138	0.320	0.366	0.306	0.361	0.322	0.338	0.138	0.146	0.140	0.149	0.152	0.150
IS0091R PCB_153	0.320	0.366	0.306	0.361	0.322	0.338	0.355	0.162	0.238	0.149	0.152	0.232
IS0091R PCB_156	0.090	0.104	0.087	0.104	0.092	0.097	0.091	0.097	0.093	0.097	0.102	0.101
IS0091R PCB_180	0.230	0.262	0.219	0.257	0.230	0.241	0.091	0.097	0.093	0.104	0.102	0.101
IS0091R PCB_28	2.203	2.508	2.097	2.475	2.205	2.309	0.813	0.875	0.840	0.881	0.920	2.356
IS0091R PCB_31	1.698	1.936	1.618	1.908	1.701	1.779	0.722	0.778	0.745	0.784	0.817	2.200
IS0091R PCB_52	0.825	0.941	0.785	0.928	0.828	0.866	0.364	0.390	0.375	0.390	0.407	1.367
LT0015R benzo_a_pyrene	1.540	1.400	0.710	1.050	0.520	0.450	0.410	0.440	0.730	0.990	0.650	1.410
NO0042G gamma_HCH	4.248	4.703	4.902	7.617	9.151	6.313	5.140	4.202	4.723	8.408	7.225	4.710
NO0042G benzo_a_pyrene	0.014	0.029	0.011	0.006	0.009	0.016	0.016	0.004	0.007	0.009	0.006	0.008
NO0099R gamma_HCH	6.810	8.075	7.400	47.575	40.494	45.640	18.428	23.380	32.775	32.065	21.840	10.439
NO0042G PCB_101	2.878	0.720	0.566	1.045	0.561	0.861	0.540	0.610	0.392	0.550	0.537	0.606
NO0042G PCB_153	1.512	0.515	0.276	0.347	0.279	0.503	0.355	0.310	0.170	0.212	0.178	0.222
NO0042G PCB_156	0.070	0.030	0.016	0.015	0.016	0.027	0.045	0.066	0.030	0.025	0.020	0.028
NO0042G PCB_180	0.258	0.110	0.054	0.078	0.068	0.103	0.135	0.144	0.087	0.090	0.062	0.066
NO0042G PCB_28	6.355	5.473	2.562	11.287	4.423	7.097	6.215	4.944	4.577	4.108	2.570	3.240
NO0042G PCB_31	5.823	5.188	2.424	10.550	4.220	6.776	5.980	4.656	4.355	3.897	2.410	3.054
NO0042G PCB_52	2.518	1.890	1.238	3.725	1.350	2.052	1.655	1.486	1.292	1.600	1.057	1.244
NO0042G PCB_105	0.300	0.110	0.072	0.067	0.059	0.114	0.103	0.138	0.045	0.060	0.042	0.062
NO0042G PCB_118	1.045	0.395	0.242	0.252	0.196	0.318	0.215	0.266	0.123	0.175	0.153	0.198
NO0042G PCB_138	1.055	0.403	0.198	0.220	0.349	0.362	0.335	0.318	0.138	0.118	0.115	0.140
SE0002R benzo_a_pyrene	0.037	0.101	0.111	0.060	0.005	0.005	0.005	0.005	0.059	0.102	0.267	0.181
SE0002R gamma_HCH	5.000	9.000	6.000	17.000	55.000	69.000	19.000	17.000	10.000	51.000	17.000	12.000
SE0002R inden_123cd_pyrene	0.035	0.137	0.136	0.065	0.035	0.035	0.035	0.035	0.074	0.154	0.370	0.274
SE0002R PCB_101	1.056	1.557	1.261	2.349	3.389	7.951	3.383	2.615	1.630	3.305	2.674	1.859
SE0002R PCB_118	0.357	0.551	0.455	1.086	0.935	2.397	1.132	0.997	0.567	0.975	0.825	0.562
SE0002R PCB_138	0.616	1.008	0.898	1.619	1.201	3.280	1.208	1.709	1.068	2.027	2.261	1.195
SE0002R PCB_153	0.692	1.092	0.898	1.568	1.420	3.776	1.239	2.055	1.190	2.341	2.443	1.360
SE0002R PCB_180	0.242	0.454	0.484	0.572	0.342	0.904	0.300	0.554	0.362	0.821	1.228	0.461
SE0002R PCB_28	0.962	1.508	1.448	2.471	2.709	5.980	2.477	1.543	1.719	4.355	3.029	2.014
SE0002R PCB_52	1.291	1.841	1.709	2.712	5.093	11.175	5.189	2.340	1.760	3.958	2.869	2.053

SE0012R	benzo_a_pyrene	0.072	0.068	0.007	0.038	0.008	0.002	0.003	0.003	0.003	0.094	0.262	0.010
SE0012R	gamma_HCH	12.000	15.000	13.000	101.000	133.000	98.000	93.000	19.000	35.000	199.000	149.000	22.000
SE0012R	PCB_101	1.421	1.235	0.890	1.793	2.910	2.860	2.889	3.017	1.950	2.891	2.304	1.929
SE0012R	PCB_118	0.483	0.456	0.382	0.704	1.145	0.842	0.793	1.432	0.824	1.095	0.981	0.653
SE0012R	PCB_138	0.632	0.617	0.509	0.828	1.556	1.498	1.467	2.132	1.201	1.454	1.182	1.004
SE0012R	PCB_153	0.765	0.790	0.602	1.087	1.817	1.938	1.976	2.354	1.222	1.677	1.374	1.313
SE0012R	PCB_180	0.203	0.251	0.168	0.271	0.463	0.454	0.445	0.792	0.354	0.499	0.381	0.333
SE0012R	PCB_28	2.826	2.272	1.451	2.946	4.382	2.998	2.763	2.831	2.642	7.567	6.454	2.774
SE0012R	PCB_52	2.656	2.163	1.441	2.786	4.803	4.993	3.868	4.443	3.298	4.636	3.706	2.490