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2000 work-plan for the implementation of the Convention on Long-Range Transboundary Air Pollution (CLRTAP)

PERSISTENT ORGANIC POLLUTANTS (POPs)

Description/objectives: Increase the provision of monitoring and modelling data on transboundary fluxes, concentrations and depositions of selected POPs over Europe. Develop the modelling bases of selected POPs (lindane, PAHs, PCBs (PCB-28, 52, 101, 118, 138, 153, 180) and some PCDD/Fs) and verify the functioning of the models. Study further the physical-chemical processes of POPs in different environmental compartments, taking also into account their transport within the EMEP region and on the hemispheric/global scale.

Main activities and time schedule:

(a) In 2000, the Parties, in co-operation with CCC will set up an EMEP network for POP measurements. As a first step, PAH, PCB, HCB, chlordane, lindane, α -HCH, DDT/DDE should be included in the EMEP measurement programme at five sampling sites: Scandinavia/Baltic, northern Atlantic region, continental Europe, Mediterranean region, south Atlantic region. To this end, financial support to laboratories able and willing to analyse samples from one or more countries may be necessary, or one central laboratory should be found. CCC will continue to collect the monitoring data on POPs available from other international programmes. The laboratory comparison initiated by CCC in 1999 will continue through most of 2000. The results will be discussed at a small workshop, to be organized in October/November 2000. CCC will also continue the work on the POP standard operating procedures and quality control routines for the manual for sampling and chemical analysis;

(b) MSC-E will study physical-chemical properties of selected POPs (see the list above). It will analyse and summarize scientific results obtained under EUROTRAC/MEPOP and other international programmes, such as in the hemispheric modelling under the Arctic Monitoring and Assessment Programme (AMAP), and under national programmes. It will pay specific attention to the improvement of the air-sea exchange module and the air-vegetation

exchange process to estimate concentrations, depositions and accumulation of the pollutants in air, soil, sea water and vegetation. Furthermore, MSC-E will study the model sensitivity, assess the seasonal and annual variations and check the calculated results against measurements, in co-operation with CCC. MSC-E will co-operate with MCS-W and the experts of the Task Force on Emission Inventories in the verification of POP emission data quality.

Executive summary

Nowadays problems of the environmental pollution by persistent organic substances are of great interest on national and international levels. Many of these pollutants are toxic or carcinogenic. Many of them enter the environment mainly as a result of economic activity. Persistent organic pollutants (POP) are transported over long distances, they are accumulated in different environmental compartments (soil, sea, biota) and inflict a negative effect on human health.

Investigations of emissions and transport of persistent organic pollutants recently became an integral part of a number of international programmes and organizations (WMO, WHO, UNEP, HELCOM, OSPAR, AMAP and others). At present the global binding treaty on POP is under development within the framework of UNEP.

In accordance with the POP Protocol (Article 9) Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutant in Europe (EMEP) submits information on POP long-range transport and depositions to the Executive Body for CLRTAP. This work is fulfilled by the Parties to CLRTAP and by international EMEP Centres: Chemical Co-ordinating Centre (CCC) and Meteorological Synthesizing Centres East (MSC-E) and West (MSC-W). CCC is responsible for measurements, MSC-W is responsible for the collection of official emission data, MSC-E - for research and modelling of POP long-range transport.

International scientific workshops make a significant contribution to developing of EMEP policy and strategy in the field of POP transboundary pollution. During last year the following workshops were held: Workshop on POPs and HMs (US EPA, October 1999); AMAP Workshop on techniques and associated uncertainties in quantifying the origin and long-range transport of contaminants in the Arctic (Bergen, Norway, June 1999); WMO/EMEP/UNEP Workshop on modelling of atmospheric transport and deposition of persistent organic pollutants and heavy metals (Geneva, Switzerland, November, 1999).

The Geneva Workshop was particularly beneficial for the identification of POP specific tasks and goals under EMEP. The following recommendations are noted among the priority tasks:

- investigations of POP accumulation in compartments along with their spatial distribution;
- trend analysis of accumulation in compartments;
- monitoring and the comparison of calculation results versus measurements not only for air but for other environmental compartments (soil, sea water, vegetation);
- assessment of emission data uncertainties;
- assessment of uncertainty of model results related to input information (sensitivity study) as a routinely part of modelling studies;
- a comprehensive assessment of dry deposition in particular to a forest;
- studies of pollutant behaviour in the marine environment including the transport by sea currents;
- investigations of the impact of pollutant distribution with particle sizes on deposition and concentration levels in different compartments;
- refinement of intercompartment exchange processes including re-emission.

The above recommendations and outcome of the previous Workshops formed the basis for integrated formulation of EMEP goals on POP problems in draft of “Strategy for EMEP 2000-2009”:

- quantification and harmonization of national emissions, quantifying and minimizing emission uncertainty for selected POPs (especially for pesticides);
- determination of source-exposure causal relationship on the basis of improvement of the understanding of exchange processes between atmosphere and other environmental compartments (soil, sea, and biota);
- preliminary assessment of transboundary fluxes as well as depositions and concentrations of selected POPs in different media (atmosphere, soil, sea, biota) including their spatial distribution for evaluation of possible harmful effect on ecosystems and human health;
- trend establishment for compliance;
- analysis of response of different environmental compartments to the emission reduction.

In accordance with the work-plan (ECE/EB.AIR/68, Annex IV, para 2,5) and taking into account the above mentioned recommendations a number of studies were carried out this year. The description of the progress and results is given in this report and Technical notes [Strukov *et al.*, 2000; Vulykh and Putilina, 2000] which are in fact an addition to this report. The report is divided into two parts. Part I summarizes the obtained results concerning the evaluation of European area pollution. Here the main attention is focused on model estimates of the media pollution in European countries, comparison of these estimates with monitoring data as well as on the consideration of differences in long-term trends within an individual region (country) from all-European trends. It is exemplified by Great Britain since for this country there is a most full set of measurements [Coleman *et al.*, 1998] and results of long-term accumulation trend studies [Harner *et al.*, 1995]. Some results of a technical character are only mentioned in Part I with reference Part II or technical notes where they are described in detail. Part II contains a description of the model modification and further development, general questions concerned with investigations of long-term trends of accumulation in compartments, investigations of concentration and deposition seasonal variations within the EMEP domain and the analysis of model sensitivity to different parameters and processes. The Technical note [Strukov *et al.*, 2000] describes the air-sea exchange module and POP transport by sea currents. The Technical note [Vulykh and Putilina, 2000] provides a comprehensive description of HCB physical-chemical properties and emissions. A detailed description of the modified scheme of POP particles-carriers dry deposition over forest is presented in [Tsyro and Erdman, 2000].

The scheme illustrating the line of investigations carried out in 2000 dealing with the model assessment of European pollution by persistent organic pollutants is given in fig. 1.

These investigations can be split in six parts:

1. The modification and further development of the POP long-range transport model for European region.
2. Investigations of physical-chemical properties and parametrization for some new POPs (four congeners of dioxins/furans and HCB) and its refinement for previously considered pollutants (PCB, B[a]P, γ -HCH).
3. Preparation of input data for modelling.
4. Collection and processing of measurement data.
5. Long-range modelling of selected POPs (first of all PCB, B[a]P, γ -HCH) and the analysis of the obtained results.

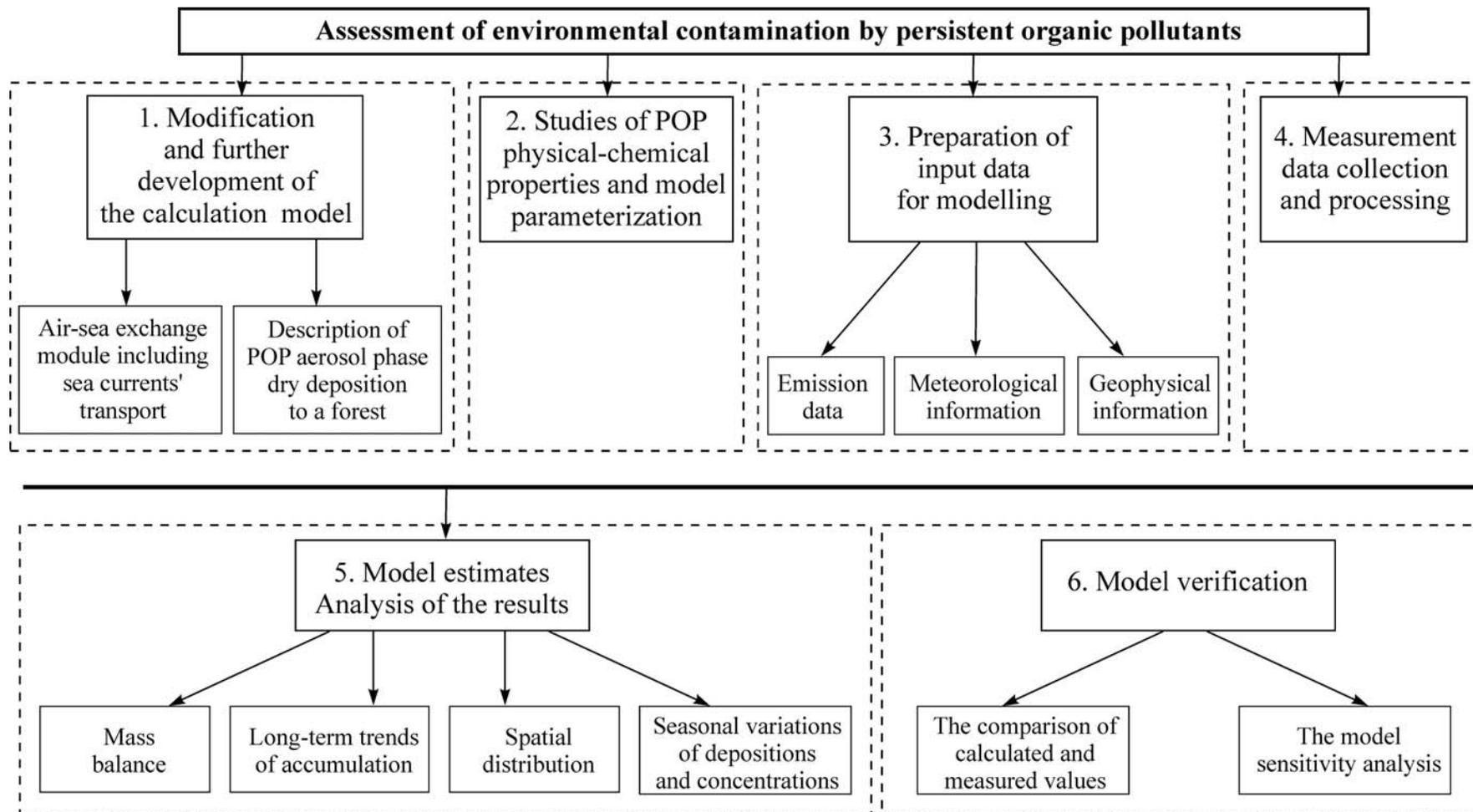


Figure 1. Main directions of activity aimed at model assessments of POP pollution in European region

6. Model verification: the comparison of calculated and observed values, model sensitivity analysis in regard to integration of new processes as well as to parametrization modifications.

Below the research under each item mentioned above is described.

1. *The modification and further development of the MSCE-POP model*

- *Air-sea exchange module.* The description of exchange processes in the marine environment and processes occurring in sea are modified as follows:
 - a) POP transport by sea currents and turbulent diffusion and primary description of the sedimentation process is introduced;
 - b) the description of air-sea exchange processes with regard for foam formation processes and choppiness is refined;
 - c) a multilayer ocean model is used.

The description of pollutant transport by sea currents made it possible to evaluate POP transport from regions located near powerful sources (the North Sea, English Channel) to remote regions of the EMEP domain. POP quantity absorbed by sea was refined. This amount appeared to be essentially larger in comparison with that obtained on the previous stage of model development. It is explained by the expanding of the sea reservoir due to integration of the multilayer ocean model and switching on the mechanism of POP transport to deep layers.

At the next stage of model development in order to improve the description of pollutant transport in the ocean it is necessary to refine the description of sedimentation process and partitioning between the dissolved and particulate phases. At present this work is under development.

- *Description of POP aerosol phase dry deposition.* This process exerts an essential effect on POP long-range transport. At this stage we concentrate our efforts on the refinement of processes of POP aerosol phase dry deposition to a forest on the basis of Ruijgrok model [Ruijgrok *et al.*, 1997]. Such modification results in air concentration change over the EMEP domain on the average by 5%, at the maximum decrease of these concentrations in some European regions by 50%. The obtained results agree with literature data [see e. g. Mclachlan and Horstmann, 1998; Beckett *et al.*, 1998].

2. Studies of POP physical-chemical properties and model parametrization

Physical-chemical properties of HCB and PCDD/F are being studied along with the refinement of some properties of previously considered pollutants (PCB, B[a]P, γ -HCH). These studies made the basis for model parametrization and provisional calculations of their long-range transport. In particular γ -HCH parameters defining air-vegetation exchange processes (octanol-air and organic carbon-water partition coefficients) were refined. Tests of HCB parametrization indicated that it is necessary to refine wet scavenging process since the description of this process on the basis of equilibrium distribution leads to an essential underestimation of washout ratio and, consequently, concentrations in precipitation. This effect can be conditioned by the increase of absorbing capacity of rain drops due to POP sorption on organic substances in them. We plan to report the results of HCB long-range transport modelling next year.

3. Preparation of input data for modelling

First of all model input data include emission data and meteorological and geophysical information.

- *Emission data.* The consideration of emission data is aimed at selecting for modelling the most stable and toxic representatives of POP groups (PCB, PAH, PCDD/F) which make the main contribution to emissions of the group and preparation of emission data for calculations. For this purpose during this year we examined the official data and expert estimates of POP studied. For the investigation of long-term trends of POP accumulation in different environmental compartments we used expert estimates obtained under POPCYCLING-Baltic project [*Pacyna et al.*, 1999] for a sufficiently long period of time – from 1970 to 1995.

For the calculated POPs (PCBs, B[a]P, γ -HCH) the emission information required for the long-range transport modelling was prepared. This information is: spatial distribution of emissions of the considered substances, emission distribution with height and emission seasonal variations.

- *Meteorological information.* In calculations we used meteorological data for 1970-97 jointly prepared with Hydrometeorological Centre of Russia.
- *Geophysical information.* This information contains: data on land use, information on leaf area index (LAI), the underlying surface roughness and distribution of organic carbon

content in soil. The geophysical data used in calculations is described in MSC-E/EMEP report [Pekar et al., 1999].

4. *Measurement data collection and processing*

On the basis of concentrations in air and precipitation measured at EMEP network mean monthly and mean annual concentrations of PCB, B[a]P and γ -HCH for 1998 were prepared. In comparison with 1997 the number of stations providing monitoring data on the listed pollutants increased: in particular B[a]P concentrations in precipitation are provided by station LT15 (Lithuania), the list of pollutants provided by station DE1 (Germany) is extended etc.

5. *Assessment of the selected POPs long-range transport*

On the basis of the prepared emission fields and model parametrization the long-range transport of PCB, B[a]P and γ -HCH for 1970-97 was calculated. The analysis of the obtained results was carried out in the following aspects:

- *POP partitioning between compartments (mass balance)*. In accordance with model assumptions all the emission of a considered pollutant enters the atmosphere, then is partially transported outside the EMEP grid and partially redistributed between compartments. PCB and γ -HCH export outside the grid is about 60% of the total emission and that of B[a]P - about 30%. The fraction of a substance absorbed by each compartment for different substances is different. About 70% of PCB absorbed by media due to redistribution is transported from atmosphere to soil, by 14% - to sea and vegetation each (including litterfall). 41% of B[a]P enters soil, 18% - sea and 7% - vegetation. For γ -HCH the corresponding values are: 48% enters soil, 33% - sea and 1% - vegetation. The content of pollutants in environmental compartments decreases due to degradation processes (scavenging processes and re-emission, i.e. the income of a substance to the atmosphere is taken into account when we consider the redistribution). After degradation the highest quantity of pollutants is remained: for PCB - in soil (about 40%), for B[a]P and γ -HCH - in sea (about 40% and 70%, respectively). The obtained results give a possibility to range the considered compartments in regard to their importance for the process of long-term accumulation.

The large calculated content of PCB and B[a]P in litterfall indicates that degradation processes in litterfall and exchange processes between litterfall and air as well as

between litterfall and soil should be refined. POP runoff from soil by melting and rain water and its riverine transport needs more profound investigation.

- *The analysis of POP long-term trends of accumulation in environmental compartments.* This analysis allows to evaluate the interval from the start of emission reduction to the start of concentration level decrease in media (response time) and to determine the extent to which the pollution level is affected by long-term accumulations. The analysis of calculated results obtained for PCB, B[a]P and γ -HCH points out that soil is the most inertial medium for PCB. The response time is about 10-15 years. By the end of calculated period re-emission occurs round the year. The marine environment is most inertial for B[a]P long-term accumulation. The response time for this pollutant is about 10 years. Contrary to PCB re-emission of B[a]P obviously is very weak. Sea is the most inertial media for γ -HCH with the characteristic response time about 5 – 6 years. Due to emission seasonal variation (it is assumed that γ -HCH emission takes place in the first half of a year) the re-emission process of this POP is mainly observed in the second half of a year. In some regions of the Mediterranean Sea the re-emission annual flux exceeds total annual deposition flux.

POP accumulation long-term dynamics in individual regions (countries) of Europe differs qualitatively from the dynamics of the whole European region. Such a difference was exemplified by Great Britain. In particular the response times for all the media for Great Britain is appreciably shorter than the mean response time for Europe because it is located close to major European emission sources. In future it is planned to carry out such an analysis for other European countries.

- *The analysis of deposition and concentration seasonal dynamics.* This analysis is concerned with investigations of POP deposition and concentration seasonal variations and their connection with variations of emissions, meteorological and some other parameters. For example, the role of temperature regime in the process of gaseous exchange of the atmosphere with the underlying surface is most pronounced in seasonal variations of gaseous exchange with land and sea. This process is most important for pollutants which have a substantial atmospheric gas-phase fraction (PCB and γ -HCH).
- *The analysis of spatial distribution.* This analysis was focused on studies of the pollution level of different environmental compartments in European countries and the comparison of these levels with emission levels in these countries. For this purpose the countries were ranged relative to the effect of own emission on:

- a) pollution level inside the country;
- b) pollution level of other countries.

Ranging according to the first criterion was made by the comparison of air concentration levels with emission density. If for some country both air concentration level and emission level exceeded the mean value for Europe, then it can be supposed that the main contribution to its pollution is made by country's own emission sources. If at high air concentrations the emission level is comparably low, then the environmental pollution in the country occurs essentially due to the transboundary transport.

For ranging by the second criterion we qualitatively assessed the export of pollutants emitted on one country and transported to other countries of European region. Certainly these estimates are very preliminary. For more precise assessment of the export value it is necessary to calculate "country-to-country" matrix.

6. Model verification

The model verification was made by the comparison of calculated and measured data and by the analysis of model sensitivity to new and refined processes integrated to the model.

- *The comparison of calculated and measured values.* This study was carried out in collaboration with national experts Dr. A.-R. Milukaitė (Lithuania) and Prof. I. Holoubek (the Czech Republic). The comparison results are:
 - Model results are in a satisfactory agreement with measurements. Monitored mean annual air concentrations differ from model estimates for PCB in 2 times (with scattering for individual countries up to 8 times), for B[a]P - on the average by 10 % (with scattering in 2-3 times for individual countries), for γ -HCH - in 5 times (with scattering for almost all the stations within one order of magnitude). The difference between measured and calculated values of concentrations in other compartments is within an order of magnitude. On the average the calculated and observed data differ: for PCB in 2 times, for B[a]P - in 1.5 times and for γ -HCH - in 2 times. Differences in concentrations in precipitation are within factors of 7.5, 2 and 2 for PCB, B[a]P and γ -HCH, respectively. The comparison of model estimates with monitoring data of Great Britain and Lithuania demonstrates their satisfactory consistency.
 - Model values of air concentrations for all the considered pollutants but B[a]P appeared to be lower than measurements. This can be explained in particular by the

fact that emission sources located outside the EMEP domain for such volatile pollutant as PCB and γ -HCH were not included into the model. Modelling on the hemispheric level can clarify this problem. At present the hemispheric model is under development.

- Since calculated concentrations in precipitation are underestimated for PCB and B[a]P more than air concentrations, the washout ratio used in the model appears to be underestimated. In future additional efforts should be made to refine washout ratio.

- *The analysis of model sensitivity* to POP transport by sea currents, dry deposition of POP aerosol phase to a forest and to variations of POP particle-carrier effective diameters, to variations of air-vegetation gas exchange parameters was made. It indicated the importance of the consideration of newly introduced processes for POP long-range transport. The sensitivity analysis helped to evaluate the uncertainty of pollution estimates introduced by the uncertainty of parametrization of the aerosol phase deposition to a forest and gaseous exchange with vegetation. More detailed information on investigations of the transport by sea currents and depositions to a forest can be found in relevant Technical notes [*Strukov et al.*, 2000; *Tsyro and Erdman*, 2000].

The results of this work reflecting the state of the art of knowledge in the field of European pollution by persistent organic pollutants are to be submitted to the Steering Body of EMEP thereby informing it of POP levels of pollution and deposition in European region. Besides these results can be used by European countries for familiarization with the state of art in the field of monitoring and estimation of POP emissions and qualitative assessment of POP transport and accumulation in different environmental compartments. These results can be also used by such international programmes and organizations as AMAP, OSPARCOM, HELCOM, WMO and various national programmes on the research of environmental pollution by persistent organic pollutants.

Introduction

The joint report of two Centers (MSC-E and CCC) includes the results of work on monitoring and modelling long-range transport of persistent organic pollutants in the EMEP region. For the sake of convenience and taking into account the interests of different groups of users - policy makers, scientists, and public - investigation results are presented in the form of the EMEP Report consisting of Part I and Part II and a number of Technical Notes.

In Part I of the report we present a summarized information on POP transport and accumulation (for PCB, B[a]P, and γ -HCH) in air, sea water, soil, and vegetation of European countries in 1997 obtained by the analysis of long-range transport modelling results for the above pollutants for the period from 1970 to 1997. An attempt to compare emissions, concentrations and depositions over different European countries was made. Model calculations are compared with available measurement data. Here we present also the results of investigations of long-term accumulation tendencies within one country exemplified by Great Britain.

Part II of the report describes further development and modification of the multicompartiment model and at the analysis of long-term POP accumulation and degradation tendencies in different media. Here the results of model sensitivity study with respect to a number of introduced processes and parameters are discussed.

The report is complemented by two Technical Notes [*Vulykh and Putilina, 2000*] and [*Strukov et al., 2000*]. First Note contains a detailed description of HCB physical-chemical properties, and the second one - the description of modified marine block integrated into MSC-E POP model.

The preparation of some sections was performed in collaboration with national experts as well as using result of investigations carried out under national programmes.

Chapter 1

The evaluation of POP transport and accumulation in European countries in 1997

This chapter describes the results of POP transport and accumulation in European region for 1997. For adequate description of contamination the calculations of POP transport and accumulation should be performed for decades. Consequently, to assess the contamination in 1997, the calculations were carried out for the period from 1970 to 1997. The adapted expert estimates of the POPCYCLING-Baltic project [*Pacyna et al.*, 1999] were used as the emission data.

The 3-D Eulerian multicompartment model (MSCE-POP) operates with the EMEP grid with spatial resolution 150x150 km. Along with the pollution transport in the atmosphere and ocean, the model includes the description of exchange processes between different environmental compartments. Figure 1.1 shows a scheme of the model structure with indication of the considered compartments and processes of exchange between them and of POP redistribution between different phases. The dashed arrows represent processes, which we plan to integrate to the model at future stages of its development.

The model considers the following compartments: air, soil, sea, vegetation and litterfall. It is assumed that initially emitted POP enters the atmospheric air. Then in the course of dry and wet depositions of the gaseous and aerosol phase a pollutant enters soil, sea and vegetation. Dry deposition process of the pollutant gas-phase is considered to be reversible allowing us directly in the modelling course to consider the re-emission process important for some pollutants. From vegetation with fallen leaves a pollutant enters the litterfall where from in some time finds its way to the soil. The accumulation in compartments decreases due to POP degradation in the course of chemical, biochemical and photochemical reactions.

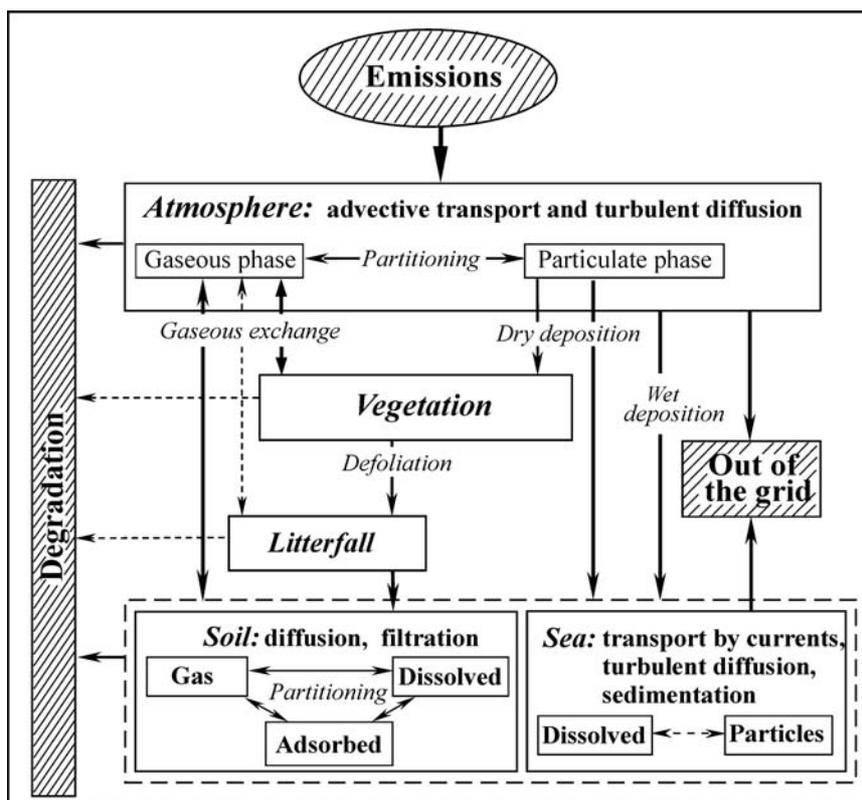


Figure 1.1. Structural model scheme

Due to advection and diffusion in the atmosphere and sea pollutants are transported at long distances from the emission source and partially are exported outside the calculation domain. The pollutant fraction transported outside the EMEP grid can be used as an estimate of the considered pollutant capability for the long-range transport.

The input information on POP modelling is emission fields, meteorological data, land use, properties of soil, vegetation and marine environment including sea currents and physical chemical properties of a pollutant in question.

In comparison with the previous model version described in [Pekar, 1996; Pekar et al., 1998, 1999] the model was modified as follows:

- The description of transport with sea currents and exchange processes in the ocean is made [Strukov et al., 2000].
- The scheme of dry deposition on vegetation is modified [Tsyro and Erdman, 2000].
- A preliminary parametrizations of the HCB [Vulykh and Putilina, 2000] and PCDD/Fs are prepared. The current parametrizations of γ -HCH, seven congeners of PCB and benzo[a]pyrene are refined.

The description of POP long-range transport model, its parametrization, and sensitivity study with respect to newly involved processes can be found in the second volume of the report. To take into account initial accumulation in all the media preliminary calculations (model spin up) were done. At present stage of investigations boundary air concentrations are taken as zero. For boundary concentrations in sea background concentrations found in the literature are used. This allowed to take into account the input of pollutants transported by sea currents from the EMEP grid border. The calculations used the meteorological data provided by Hydrometeorological Centre of Russia.

According to the POP Protocol, B[a]P, PCB, and γ -HCH were selected for the top-priority modelling. These substances can be characterized as persistent, toxic, and able to the long-range transport chemicals. From the viewpoint of model development these substances are interesting since they are differently distributed between the gaseous and aerosol phase in the air. Namely, the largest fraction of B[a]P is settled on particles, γ -HCH exists in the atmosphere mainly in the gaseous phase, and PCB is an intermediate substance.

In this chapter the analysis of calculated spatial distribution of selected POPs in 1997 is presented. The preliminary estimates of contamination levels in different media and deposition fluxes for each European country are considered. The qualitative estimates of transboundary transport of POPs between countries are given.

To evaluate the reliability of modelling results the comparison between calculated and measured values for the considered media is carried out. In the comparison the data from the EMEP monitoring network were used along with data found in the literature. The corresponding sections were written in collaboration with national experts (*Dr. A.-R. Milukaité* (Lithuania) and *Prof. I. Holoubek* (the Czech Republic)).

1.1. Polychlorinated biphenyls

Modelling of PCB transport within European region covered the period of 1970-97. To take into account PCB accumulation in soil and other media and to determine initial concentrations a preliminary calculation for 1940-69 has been carried out. According to the results obtained in [*Pekar et al.*, 1999] properties of *indicator congener* PCB-153 are used for physical-chemical properties of PCB mixture.

In calculations adapted PCB emission estimates for 1970-97 taken from POPCYCLING-Baltic project are used. The spatial emission distribution for the last year is demonstrated in figure 1.1. PCB emissions in European countries are presented in table 1.1 in the end of this section; see also figure 1.3 below.

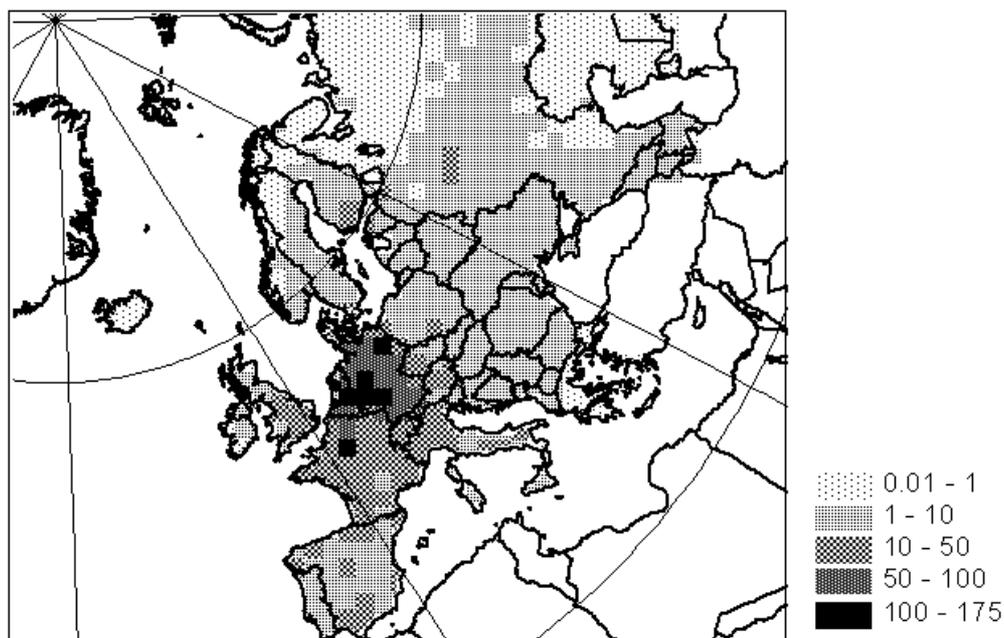


Figure 1.1. PCB emissions in 1997, g/km²/yr.

The analysis of long-term trends of the pollution of different environmental compartments (Volume 2) pointed out that PCBs are kept in soil for the longest period. Whereas in other compartments the emission reduction is reflected sufficiently rapidly in pollution level, PCB accumulations in soil can rest on high enough level even at emission reduction. As a consequence of emission decrease *re-emission from soil arises which can sustain pollution levels in other compartments during 10-15 years even when there is no emission.*

1.1.1. PCB concentrations and depositions in European countries in 1997

In this section we analyse mean calculated values of PCB depositions and concentrations in air, soil, sea water and vegetation in European countries in 1997. Data averaged over a year and territory of countries are summarized in table 1.1 in the end of this section. Figure 1.2 illustrates PCB air surface concentration distribution over European region.

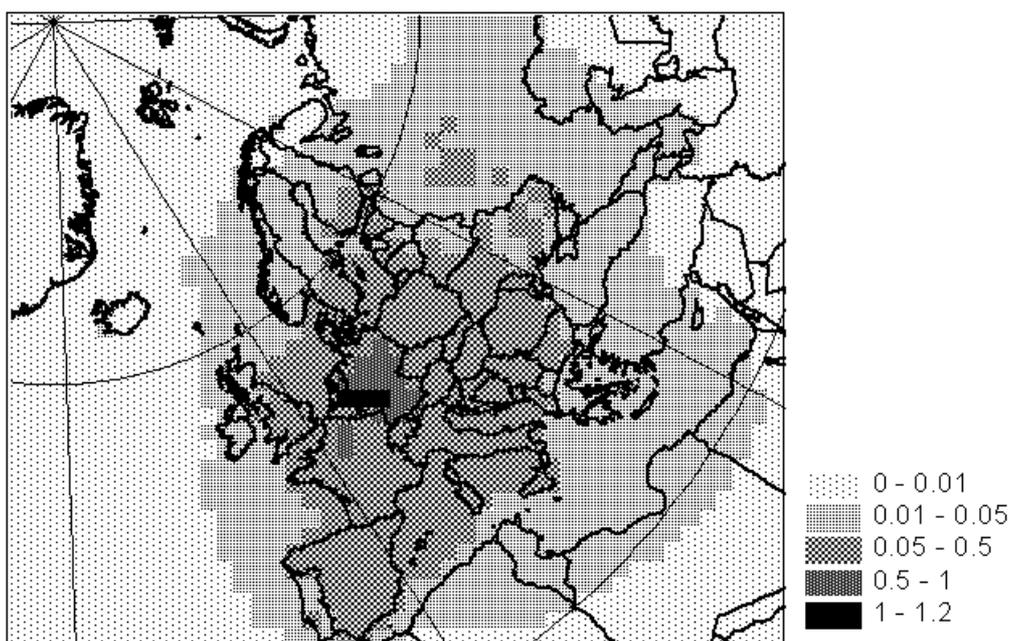


Figure 1.2. PCB air concentrations, ng/m³

The comparison of the emission distribution (fig. 1.1) and air concentration maps points out that emission levels define air concentrations to a great extent. This conclusion is verified by the analysis of seasonal variations of atmospheric concentrations made in volume 2 of this report. Therefore comparing air concentration levels with emission density it is possible to draw a qualitative conclusion on the extent of POP transboundary transport impact on the air pollution level. If both emission fluxes and air concentrations within a country are high enough then we assume that air pollution is defined mainly by sources inside a country. On the contrary if there are high air concentrations and a comparably low emission level then most probably POP transboundary transport plays a leading role in air pollution of the country.

Figures 1.3.a and b illustrate diagrams of the comparison of air concentrations in a number of European countries with their emission levels (countries with concentration levels above mean European ones are included to the comparison).

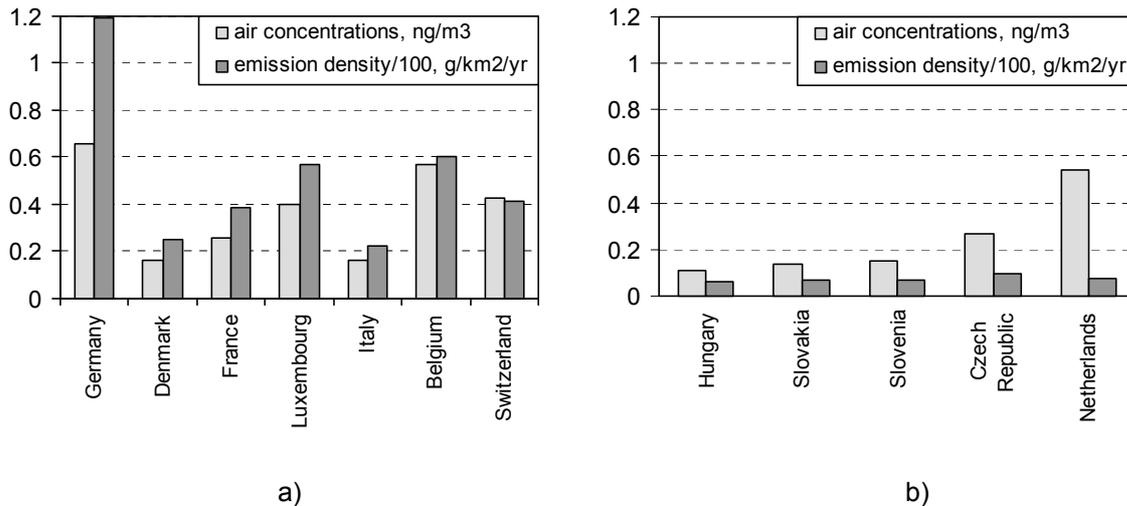


Figure 1.3. Mean air concentrations and emission density of PCB in a number of European countries in 1997

As evident from the diagrams in Germany, Denmark, France, Luxembourg, Italy, Belgium and Switzerland the air concentration levels are within 0.1 to 0.7 ng/m³ and emission densities are 20-120 g/km²/year (fig.1.3.a). At the same time in Hungary, Slovakia, Slovenia, the Czech Republic and the Netherlands the air concentration level is approximately the same but the emission density level is appreciably low (from 6 to 8 g/km²/year, figure 1.3.b). Hence it may be concluded, that air concentrations of the first group of countries can be explained by their own emissions, whereas the air concentrations of the second group of countries obviously are defined by PCB transport from other European countries.

The first group of countries is characterized by high PCB soil concentrations (30-120 ng/g). Figure 1.4 demonstrates the map of PCB soil concentration distribution. As shown in volume 2 of the report for PCB soil is the most *inertial* medium which is characterized by high accumulation, a delayed onset of accumulated content decrease compared with the beginning of emission reduction and by relatively slow removal at emission reduction. Therefore the soil pollution level in many respect defines long-term dynamics of the environmental pollution by polychlorobiphenyls.

Soil concentrations are highest in countries with high emission density (fig. 1.1) - on the territory of Belgium, France, Germany, Luxembourg and Switzerland and in neighbouring countries Austria, Croatia, the Czech Republic, Finland, Ireland, the Netherlands, Poland, Slovenia, Sweden and Great Britain.

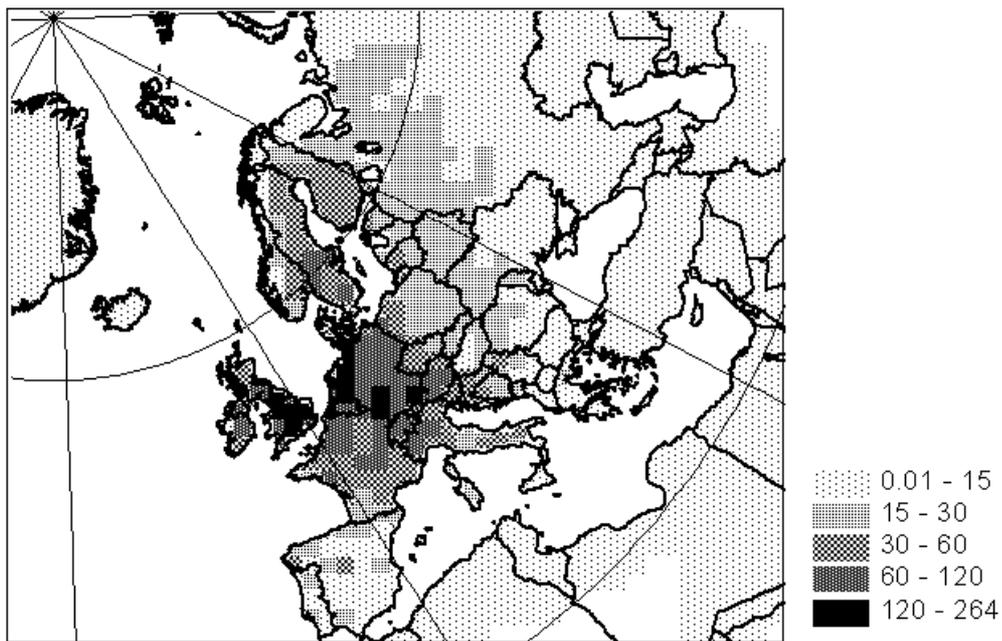


Figure 1.4. PCB soil concentrations for 1997, ng/g

High soil concentrations in European countries can give rise to PCB fluxes from soil to air (re-emission) round the year. This phenomenon is described in the literature (see, for example, [Harner *et al.*, 1995; Kaupp *et al.*, 1996]) and more fully analysed in volume 2 of this report. Preliminary estimates indicate that PCB essential accumulation in soil results in re-emission. The latter is capable to keep air concentrations for a long period of time (10-15 years) even when anthropogenic emissions are ceased.

The analysis of soil concentrations allows us to distinguish a separate group of countries such as Great Britain, Ireland, Sweden and Finland where air concentration level (1997) is relatively low (0.02 - 0.08 ng/m³) whereas soil concentrations resulted from long-term accumulation are appreciable (40 - 130 ng/g).

As seen from the above analysis soil is the most important reservoir where PCB is accumulated and stored. Soil content can support contamination levels in other media for sufficiently long time after emission reduction.

Now let us consider sea water pollution. Spatial distribution of marine concentrations is shown in figure 1.5.

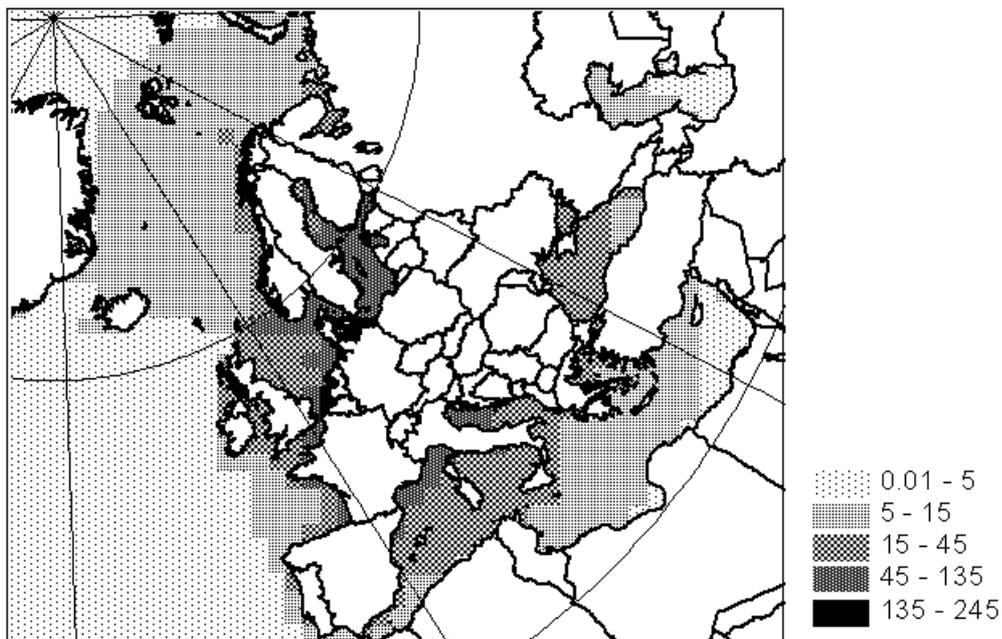


Figure 1.5. PCB concentrations in sea, ng/m³

High concentrations in sea water (maximum values are up to 245 ng/m³) are mainly observed in the North Sea near Belgium, the Netherlands and Germany. PCB high concentrations are also observed in the Baltic Sea washing Poland, Lithuania, Estonia and Latvia and in the northern part of the Mediterranean Sea. Modelling sea currents made it possible to consider PCB transport from regions with high concentrations north-eastward along Norwegian coast. PCB entering the marine environment from air (in marine regions close to major emission sources) and through the EMEP boundary (mainly in the southern and south-western Atlantic) then is transported by sea currents to the northern coast of Scandinavia. This leads to the appearance of re-emission fluxes over the sea to the north of Scandinavian Peninsula. Note that under model assumptions PCB income through the EMEP grid boundary is low due to low background marine concentrations.

Figure 1.6 shows spatial distribution of PCB concentration in vegetation. The highest concentrations (from 40 to 200 ng/g of dry weight) are observed in countries located near major emission sources (Luxembourg, Belgium, Germany and France) and in neighbouring countries (Austria, the Netherlands).

Figure 1.7 demonstrates the distribution of PCB total (dry + wet, gas + aerosol) deposition fluxes including particle dry deposition to a forest. Negative flux values indicate the availability of re-emission (relevant regions on the map are white).

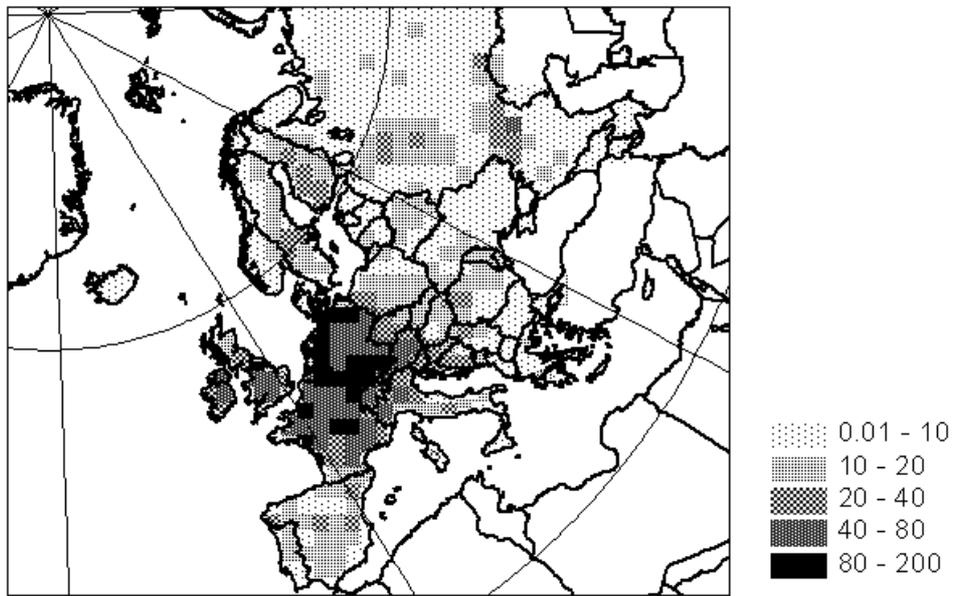


Figure 1.6. PCB concentrations in vegetation in 1997, ng/g d.w.

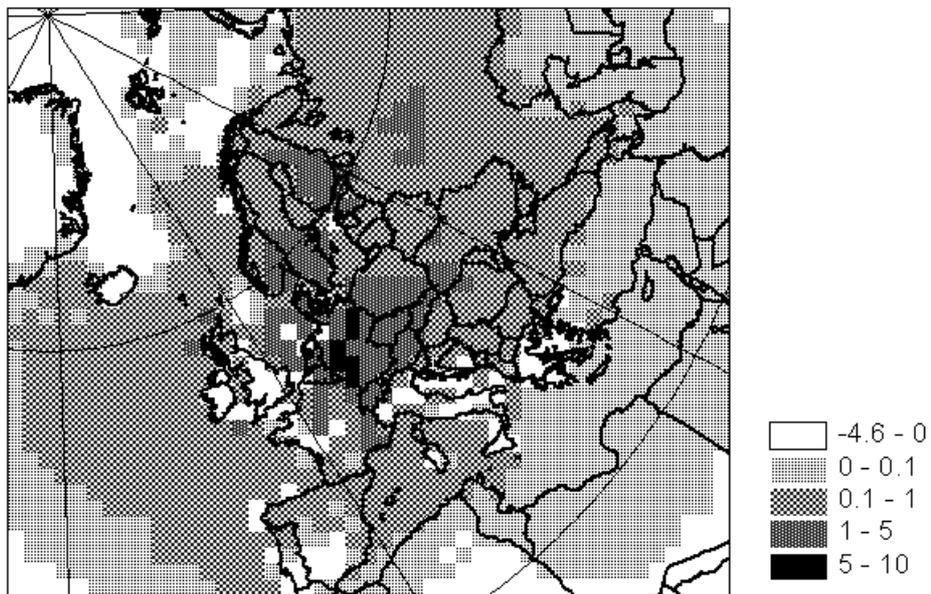


Figure 1.7. PCB total depositions in 1997, g/km²/yr. White colour indicates year-round re-emission process

The highest re-emission fluxes are in Great Britain, France and Germany reaching 2-4.6 g/km²/year that is comparable with emission fluxes in some European countries. According to preliminary estimates at the expense of re-emission process at present (in 1997) not less than 5 tonnes of PCB enter the atmosphere whereas total European emission is 113 tonnes.

At further emission reduction the re-emission value can be comparable with emission flux in individual European regions.

The comparison of PCB emission totals and deposition fluxes for each country makes it possible to evaluate approximately the quantity of emitted pollutant which falls out in other countries (export; for the computation procedure, see formula 1.1 below). According to these estimates the highest export is characteristic of Germany - 2.7 tonnes. Then in the order of decrease come France - 1300 kg, Spain - 600 kg, Italy - 400 kg, Great Britain - 200 kg, the Ukraine 200 kg, Poland - 150 kg and Sweden - 130 kg. The European part of Russian Federation exports 450 kg. PCB transport from Russia can affect such regions as Eastern Siberia and the Arctic. Certainly figures presented in this section are of a preliminary character. For more precise evaluation of the export it is necessary to calculate "country-to-country" matrices.

The qualitative analysis of the pollutant emitted by each country, which falls out in other countries is made by the formula:

$$Export = Emission \times \frac{TotDep}{TotEmis} \times (1 - DepFrac), \quad (1.1)$$

where *Export* is PCB amount emitted in a given country and deposited in other countries of the EMEP domain (export);

Emission is PCB total annual emission in a given country;

TotDep is PCB total annual deposition within the EMEP grid;

TotEmis is PCB total annual emission within the EMEP grid;

DepFrac is the ratio of PCB total depositions within a given country to total annual deposition within the EMEP grid.

The presented results of PCB content in the studied compartments of different European countries are summarized in table 1.1. The countries where air and soil concentrations exceed mean European level are shaded (0.13 ng/m³ and 34 ng/g respectively).

Table 1.1. PCB concentrations and depositions in European countries in 1997

Country	Air, ng/m ³	Soil, ng/g	Vegetation, ng/g d.w.	Sea, ng/m ³	Deposition density, g/km ² /yr.	Total deposition, t/yr.	Emission density, g/km ² /yr.	Total emission, t/yr.
Albania	0.06	9.8	5.1	32.9	0.14	0.004	5.74	0.15
Armenia	0.02	0.7	1.9	0.0	0.10	0.003	7.26	0.19
Austria	0.23	73.5	46.7	0.0	2.03	0.173	16.49	1.40
Azerbaijan	0.02	0.5	2.2	8.3	0.09	0.007	4.24	0.34
Belarus	0.05	22.6	10.9	0.0	0.62	0.126	2.54	0.52
Belgium	0.57	118.1	96.0	157.4	1.73	0.053	60.33	1.83
Bosnia&Herzegovina	0.08	25.3	17.3	46.8	-0.01	-0.001	4.60	0.23
Bulgaria	0.06	7.6	6.6	28.3	0.47	0.051	4.80	0.52
Croatia	0.09	31.5	21.2	54.9	0.07	0.004	4.44	0.24
Cyprus	0.01	0.2	0.0	5.0	0.03	0.000	0.00	0.00
Czech Republic	0.27	52.1	30.7	0.0	2.82	0.223	9.71	0.77
Denmark	0.16	28.9	10.8	122.7	0.65	0.026	25.28	1.03
Estonia	0.07	25.7	6.2	74.2	0.94	0.042	3.06	0.14
Finland	0.04	37.4	16.6	59.9	1.20	0.393	8.32	2.72
France	0.26	57.0	62.7	73.4	0.85	0.453	38.32	20.35
Georgia	0.03	1.6	3.3	24.4	0.26	0.017	4.75	0.31
Germany	0.66	110.9	90.2	181.9	4.49	1.600	119.1	42.46
Greece	0.04	9.2	7.6	14.6	-0.28	-0.034	1.82	0.22
Hungary	0.11	22.9	16.3	0.0	0.81	0.075	6.01	0.55
Iceland	0.01	1.3	1.9	8.3	-0.19	-0.018	0.52	0.05
Ireland	0.02	50.4	31.1	20.7	-1.38	-0.091	1.17	0.08
Italy	0.16	48.9	21.5	45.5	0.10	0.026	22.24	6.05
Kazakhstan	0.01	0.5	0.8	7.7	0.08	0.047	0.34	0.20
Latvia	0.05	25.5	5.9	66.3	0.74	0.045	2.05	0.13
Lithuania	0.06	29.0	9.3	74.8	0.71	0.046	2.72	0.18
Luxembourg	0.40	83.9	97.1	0.0	2.76	0.006	56.87	0.13
Malta	0.03	0.1	0.0	15.4	0.17	0.000	0.00	0.00
Netherlands	0.54	127.6	78.7	150.9	0.69	0.022	7.70	0.25
Norway	0.03	17.9	3.4	36.1	0.60	0.179	1.36	0.41
Poland	0.14	30.5	14.6	116.5	1.43	0.442	7.69	2.37
Portugal	0.05	14.0	9.1	15.3	0.04	0.004	6.64	0.56
Republic of Moldova	0.07	15.6	15.6	37.7	0.44	0.014	6.30	0.21
Romania	0.07	14.6	13.0	29.3	0.78	0.184	5.14	1.22
Russian Federation	0.03	12.4	8.1	30.9	0.58	2.145	1.91	7.04
Slovakia	0.14	25.5	14.6	0.0	1.30	0.064	6.70	0.33
Slovenia	0.15	54.6	35.7	98.0	0.83	0.015	6.76	0.12
Spain	0.10	14.6	14.4	27.5	0.13	0.060	18.37	8.72
Sweden	0.04	40.6	12.0	62.6	1.03	0.443	4.71	2.02
Switzerland	0.43	82.6	62.3	0.0	3.00	0.126	41.60	1.74
The FYR of Macedonia	0.06	10.9	7.1	34.4	0.26	0.007	3.81	0.10
Turkey	0.01	0.2	0.2	14.4	0.06	0.056	0.00	0.00
Ukraine	0.06	12.3	7.9	31.5	0.66	0.395	5.11	3.04
United Kingdom	0.08	123.9	37.8	43.7	-0.87	-0.202	15.96	3.71
Yugoslavia	0.08	20.5	13.8	39.0	0.60	0.061	5.66	0.57

* Negative values of deposition density and their total values indicate the presence of re-emission

1.1.2. Comparison of calculated PCB-153 values with measurements¹

This section is dedicated to the comparison of model results with measurements for PCB carried out at the EMEP monitoring network in 1989-97 [Berg and Hjellbrekke, 1998] and found in the literature. In particular data on Augsburg city (Germany) for 1992 (48.3⁰N, 10.9⁰W) [Kaupp et al., 1996] and on southern Great Britain [Harner et al., 1995] were used. For the comparison we use measurements of PCB-153 concentrations in different media (remember that the PCB transport was calculated using properties of this congener). To recalculate PCB-153 concentrations from that for all PCBs mixture it was assumed that PCB-153 emission is 4% of total PCB emissions [Pekar et al., 1999].

Air concentrations

Table 1.2 demonstrates measured and calculated mean annual concentrations and their ratios. The ratio between measured and calculated values on EMEP measurement sites varies from 0.5 to 8. On the average the measured values are 2.2 times higher than the calculated ones. The correlation between calculated and measured values is rather high.

Table 1.2. PCB-153 mean annual air concentration, pg/m³

Station	Country	Year	Measurement	Calculation	Meas./Calc.
SE97	Sweden	1991-94	2.90	1.16	2.5
CZ3	Czech Republic	1997	40.64	9.28	4.4
FI96	Finland	1996	1.52	0.64	2.4
IS91	Iceland	1995	0.69	0.24	2.9
IS91	Iceland	1996	1.77	0.30	5.9
IS91	Iceland	1997	0.27	0.28	1.0
NO42	Norway	1993	0.73	0.19	3.8
NO42	Norway	1994	0.63	0.16	4.0
NO42	Norway	1995	0.32	0.13	2.5
NO42	Norway	1997	0.70	0.12	5.8
SE2	Sweden	1994	5.32	4.08	1.3
SE2	Sweden	1995	4.45	4.17	1.1
SE2	Sweden	1996	2.15	4.32	0.5
DE9	Germany	1995	39.8	5.08	7.8
Augsburg	Germany	1992	3.84	20.40	0.2
Southern U.K.	UK	1990	15	3.48	4.3
Mean			7.55	3.38	2.2

¹ This section was written in the co-authorship with Prof. I.Holoubek (Czech Republic).

Measurement results of NO99 for 1993, 1994 and 1997 were not used in the comparison since measurement values of this station differ much from the stations located at similar geographical location. The comparison with air concentrations calculated earlier [Pekar *et al.*, 1999] shows that the model modification somewhat improved the agreement with observations. Mean ratio of measured to calculated data is 2.2 instead of 2.8 for calculations of the previous year.

Concentration in precipitation

Table 1.3 demonstrates calculated and measured mean annual concentration in precipitation and their ratio. The ratio varies from 1.6 to 20. In spite of the large underestimation of concentration in precipitation the correlation between measured and calculated data is rather high.

Table 1.3. PCB-153 mean annual concentration in precipitation, ng/l

Station	Country	Year	Measurement	Calculation	Meas./Calc.
IS91	Iceland	1995	0.037	0.003	12.6
IS91	Iceland	1996	0.070	0.004	19.3
IS91	Iceland	1997	0.009	0.003	3.2
DE9	Germany	1996	0.100	0.061	1.6
DE9	Germany	1997	0.428	0.041	10.5
FI96	Finland	1996	1.700	0.230	7.4
CZ3	Czech Republic	1996	0.278	0.030	9.3
Mean			0.37	0.05	7.4

The influence of emission sources located outside the EMEP domain can increase the calculated concentrations especially at stations near the grid boundary. In fact according to modelling results about 65% of the ejected PCB is transported outside the EMEP domain and, hence, a considerable amount can be imported from other regions. Such influence is to be examined by hemispheric modelling.

Besides, the difference in ratios of measured and calculated data on concentrations in air (2.2) and precipitation (7.4) indicates that washout ratio used in the model seems to be underestimated.

Concentrations in soil and vegetation

Table 1.4 presents measured and calculated mean annual concentrations in soil. The ratios of measured to calculated concentrations vary from 0.25 to 4.45. On the average the calculated concentrations in soil are 1.6 times lower than measured ones.

Table 1.4. PCB-153 soil concentrations, ng/g

Station	Country	Year	Measurement	Calculation	Meas./Calc.
SE97	Sweden	1991-94	4.8	2.235	2.15
Southern U.K.	UK	1990	1.5	6.05	0.25
CZ3	Czech Republic	1996	5.39	1.26	4.27
CZ3	Czech Republic	1997	4.61	1.21	4.45
Mean			4.08	2.69	1.58

Table 1.5 illustrates measured and calculated mean annual concentrations in needles of coniferous trees. For lack of the information on particle interaction with leaf area and needles the calculated values for the gas-phase only were used. Code FIN denoted Finnish stations which measurements for December 1992 were averaged: Kaskinen south, Kaskinen city, Kaskinen north, Vaasa south, Vaasa west island, Vaasa north, Kokkola south, Kokkola city [Sinkkonen *et al.*, 1995]. Corresponding measurements are systematized in POPCYCLING-Baltic project. The ratios of measured to calculated value are within 0.38-9.1. On the average measured values are about 1.8 times larger than calculated ones.

Table 1.5. PCB-153 concentrations in vegetation, ng/g d.w.

Station	Country	Year	Measurement	Calculation	Meas./Calc.
SE97	Sweden	1991-94	0.51	1.33	0.38
FIN	Finland	1992	1.52	0.17	9.06
CZ3	Czech Republic	1996	2.76	1.40	1.97
CZ3	Czech Republic	1997	2.85	1.44	1.92
Mean			1.91	1.08	1.77

Concentrations in sea water

Table 1.6 demonstrates PCB-153 calculated concentrations in the Baltic Sea. Measurement data were taken from POPCYCLING-Baltic project where all data on the Baltic Sea are systematized. These data are cited in [Axelmann *et al.* (in press Nov.99)] and [Schulz-Bull *et al.*, 1994].

Table 1.6. PCB-153 concentration in sea water, pg/l

Year	Measured	Calculated	Meas./Calc.
1989	4.92	10.93	0.45
1991	24.78	6.09	4.07
1992	19.57	5.07	3.86
1993	3.30	4.48	0.74
Mean	13.14	6.64	1.98

For the purpose of the comparison all the available measurements and the calculated values were averaged over the whole Baltic Sea basin. The data [Biziuk *et al.*, 1999] for 1996 (366.67 pg/l) and for 1997 (38300 pg/l) were excluded since they are as much as 4 orders of magnitude higher than other measurements. Mean calculated concentrations differ from mean measured ones within the factor of 2, the difference in measurement/calculations ratio is in the range of 0.5 - 4.1. Note that calculated values of concentrations averaged over different Baltic regions vary within 5 - 11 pg/l.

1.1.3. Conclusions

Using the multicompartiment MSCE-POP model the simulation of PCB transport within European region for the period of 1970-97 was made. Emission estimates for the indicated period obtained under POPCYCLING-Baltic project were used. On the basis of these computations mean depositions and concentrations in the studied compartments (air, soil, sea water, vegetation) in European countries for 1997 were presented. Preliminary estimates of PCB emitted within each country and deposited on other European countries were obtained (here we indicate countries with large enough export values only) as well as primary analysis of the relationship between emission levels in individual countries and their air concentrations is made. In order to verify the model measured and calculated values are compared.

1. The comparison of measured and calculated values pointed out that on the average calculation results for all studied compartments are lower than available measurement data. First of all it can be explained by the fact that the impact of emission sources outside the EMEP domain were neglected in the model. The mean ratio of measured to calculated air concentrations is 2.2. Most underestimated are concentrations in precipitation.
2. According to the ratio of air concentrations to emission densities European countries are divided into two groups. The first group involves countries where air pollution is defined

mainly by own emissions (Germany, Belgium, Luxembourg, Switzerland, France, Denmark and Italy). The second group consists of countries which pollution is defined by external sources (the Netherlands, the Czech Republic, Slovakia, Slovenia and Hungary).

3. According to preliminary calculations the countries with high export values are Germany, France, Spain, Italy, Great Britain, the Ukraine, Poland and Sweden.
4. Investigations of long-term trends of PCB accumulation in different media (see volume 2 of this report) indicated that even if all emissions were ceased the soil would be a "reservoir-emitter" keeping pollution levels in different environmental compartments during a long period of time (10-15 years). In future it is planned to investigate this process in more detail.

1.2. *Benzo[a]pyrene*

B[a]P transport modelling within the geographical scope of European regions is made for 1970-97. Adapted B[a]P emission data of POPCYCLING-Baltic project [*Pacyna et al.*, 1999] are used. For 1996 and 1997 emission is taken to be equal to that of 1995. To determine B[a]P initial concentrations preliminary model runs for 15 years on the basis of emission and meteorological data for 1970 are performed.

Emission spatial distribution for the last year is demonstrated in figure 1.8. B[a]P emission levels in European countries are presented in table 1.7 in the end of this section.

B[a]P deposition and concentration distribution in different compartments averaged over 1997 is discussed in this section. Mean annual values of concentrations and depositions are calculated for European countries. Preliminary ranging of countries relative to the relationship of the influence of internal and external sources is made. Calculated and measured data are compared. Note that B[a]P is present in the atmosphere in the aerosol and gaseous phase (the gas-phase fraction is about 10% of B[a]P total atmospheric content) therefore total (gas + aerosol) concentrations and depositions are considered.

Modelling results demonstrated that B[a]P concentration decrease in soil and sea is two times slower than emissions reduction. Therefore on B[a]P anthropogenic emission reduction the pollution level in these two compartments will be kept rather high during several decades (see volume 2 of this report). The comparison of calculated B[a]P

concentrations against measurements indicates that the discrepancy of concentrations in air and precipitation is within a factor of 2, for other compartments - within a factor of 5.

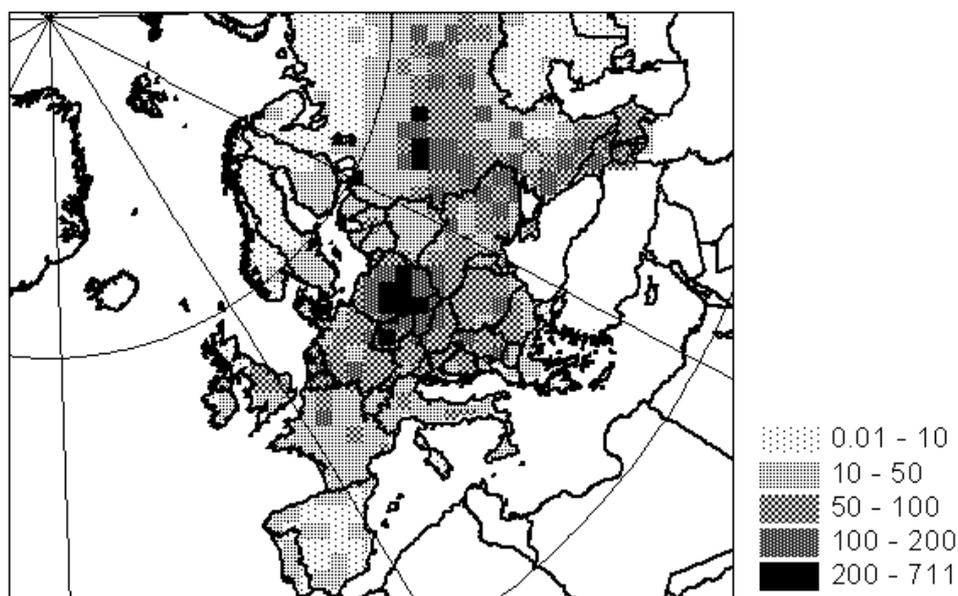


Figure 1.8. B[a]P emission in 1997, g/km²/yr.

1.2.1. B[a]P concentrations and depositions in European countries in 1997

This section deals with a preliminary analysis of calculated B[a]P depositions and concentrations in air, soil, sea water and vegetation in European countries. Corresponding data averaged over 1997 for countries are presented in table 1.7 in the end of this section. Below we present maps of the spatial distribution of B[a]P concentrations over media.

Figure 1.9 shows B[a]P air concentration spatial distribution. Comparison of B[a]P air concentrations and emission densities within European countries allows to distinguish the following two groups of countries. The first group includes Armenia, Azerbaijan, Slovenia, Georgia, Belgium, Yugoslavia, the Czech Republic and Poland. They have high B[a]P concentrations (0.4-1.2 ng/m³) which are mainly kept by relatively high emission densities in these countries (fig.1.10.a).

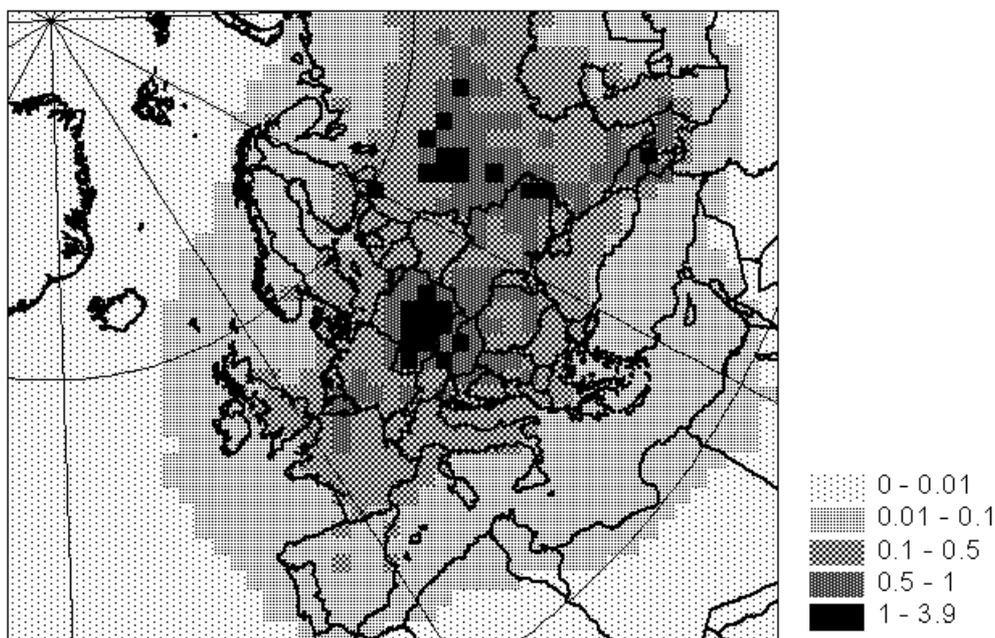


Figure 1.9. B[a]P air concentrations, ng/m³

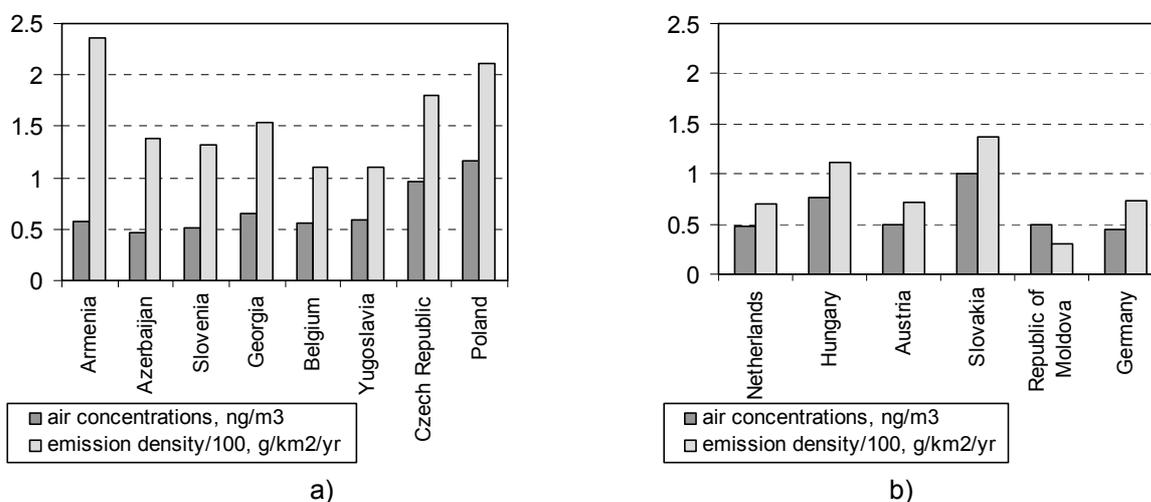


Figure 1.10. B[a]P air concentrations and the emission flux in a number of European countries in 1997

The other group contains countries with the same air concentrations level and relatively low emission fluxes (fig.1.10.b). They are the Netherlands, Hungary, Austria, Slovakia, Moldova, and Germany. Hungary and Slovakia are included in the second group since though emission density for these countries are higher than for some countries in the first group, air concentrations for them are much higher. Most probably the concentration levels in these countries in addition to their own emission is conditioned by a considerable effect of B[a]P transboundary transport from sources located in neighbouring countries.

The countries with high air concentrations are characterized by high B[a]P concentrations in soil which spatial distribution is demonstrated in figure 1.11. High soil concentrations are observed in Germany, Poland, the Czech Republic, Slovakia, Belgium, the Netherlands, Luxembourg and in a part of the Ukraine and Russia.

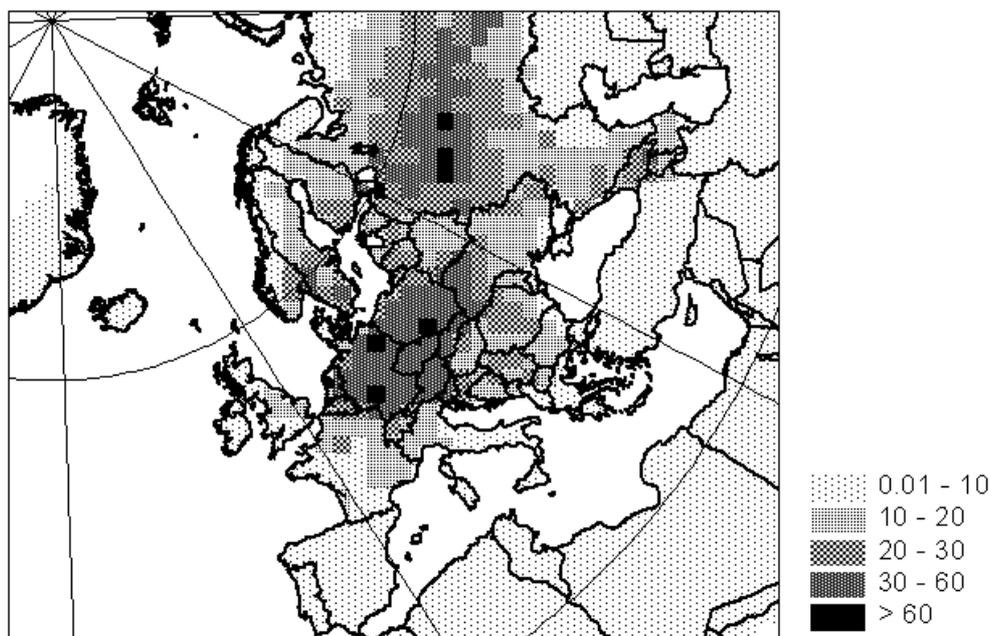


Figure 1.11. B[a]P soil concentrations, ng/g

B[a]P input to soil is supplied by dry and wet depositions. As evident from the comparison of maps of soil concentration distribution and total depositions (fig. 1.14 below) the regions of the highest soil concentrations approximately coincide with regions of high deposition densities, though they are "more uncertain" since they are formed as a result of multiannual accumulation. High soil concentrations (30-60 ng/g) are observed in the Czech Republic, Germany, Poland, Austria, Slovakia, Slovenia, and partially in the Ukraine and Russia.

Figure 1.12 shows the map of B[a]P concentration spatial distribution in sea water. High concentrations (up to $3 \mu\text{g}/\text{m}^3$) are observed in the Black Sea washing Georgia, Russia and the Ukraine, in the Baltic Sea washing Poland, the northern part of the Mediterranean Sea and in the North Sea washing Belgium and Germany. The absence of re-emission and low degradation rate in sea can be the reason for a considerable sea content level kept during 10 years even if emissions are ceased.

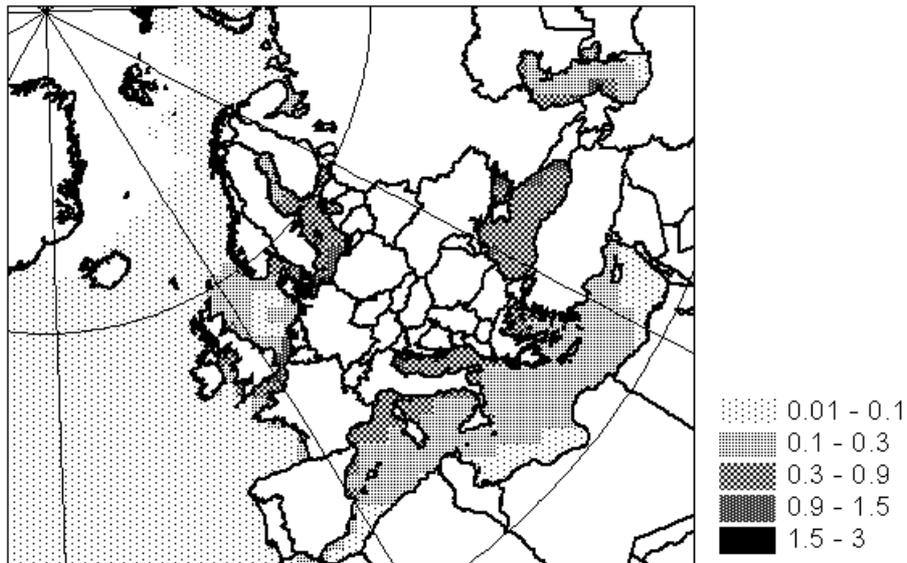


Figure 1.12. B[a]P concentrations in sea, $\mu\text{g}/\text{m}^3$

The consideration of concentration distribution in vegetation demonstrated in figure 1.13 indicated high concentrations of B[a]P gas-phase in vegetation (above 10 ng/g of dry weight) in Austria, Belgium, Bosnia and Herzegovina, Croatia, Georgia, Moldova, Romania, Slovenia, and Yugoslavia.

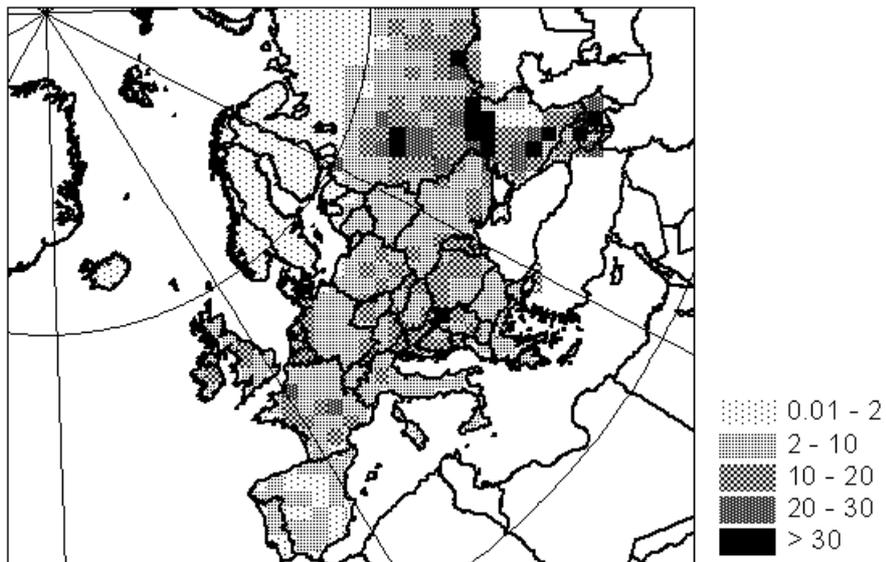


Figure 1.13. B[a]P concentrations in vegetation, ng/g d.w.

In conclusion let us consider the total deposition map (fig.1.14). High deposition densities comply with regions of maximum emissions located in Poland, the Czech Republic and

Russia. The highest fluxes (up to 145 g/km²/yr.) are observed in central and south part of Russian regions covered by forests. Deposition densities more than 20 g/km²/yr. are observed in Austria, Bosnia and Herzegovina, Germany, Hungary, Lithuania, Slovakia, Slovenia, Romania and Yugoslavia, in part of Belarus, the Ukraine and Moldova. Note that contrary to PCB for B[a]P re-emission from soil is negligible.

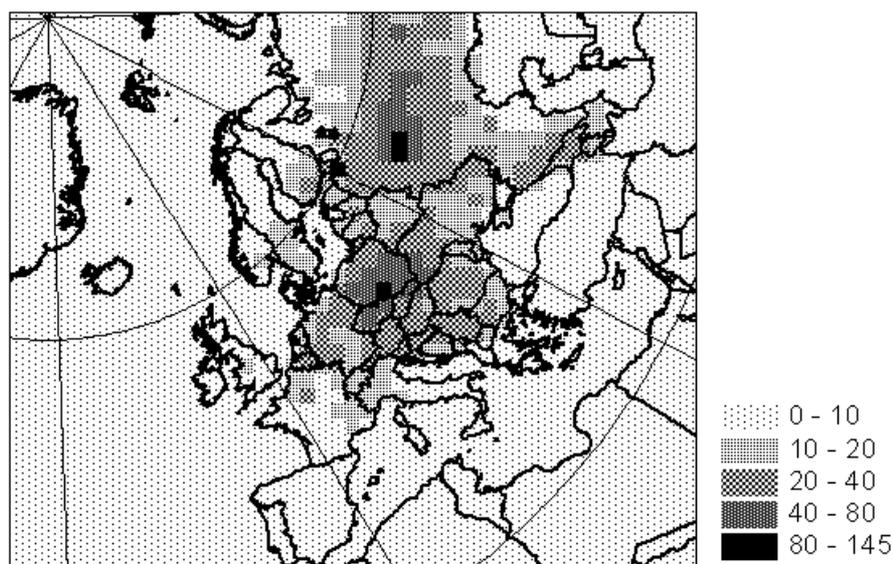


Figure 1.14. B[a]P depositions, g/km²/yr.

On the basis of the obtained emission and deposition densities for each country it is possible to evaluate (by formula 1.1 in section 1.1.1) the amount of the pollutant emitted by a country and exported to other countries.

The highest export is characteristic for Poland - 18 tonnes. Then in the order of export decrease come the Ukraine - 14.3 tonnes, France - 7.6 tonnes, Germany - 7.6 tonnes, Romania - 5.5 tonnes, the Czech Republic - 4.2 tonnes, Italy - 4.1 tonnes, Great Britain - 3.6 tonnes and Yugoslavia - 3.3 tonnes. For the European part of the Russian Federation the export is 36.7 tonnes. This export can essentially affect such regions as Eastern Siberia and the Arctic. For more precise description of B[a]P export it is planned to calculate “country-to-country” matrix in 2001.

Results of the analysis of B[a]P spatial distribution and its concentrations in compartments are summarized in table 1.7 (countries with higher than average European concentrations in air or soil respectively 0.36 ng/m³ and 17.6 ng/g are shaded).

Table 1.7. B[a]P concentrations and depositions in European countries in 1997

Country	Air, ng/m ³	Soil, ng/g	Vegetation, ng/g d.w.	Sea, ng/m ³	Deposition density, g/km ² /yr.	Total deposition, t/yr.	Emission density, g/km ² /yr.	Total emission, t/yr.
Albania	0.18	8.9	3.8	574.7	8.6	0.22	8.7	0.2
Armenia	0.57	17.0	12.3	0.0	18.6	0.48	235.3	6.0
Austria	0.5	36.2	10.1	0.0	28.2	2.40	71.9	6.1
Azerbaijan	0.47	10.1	13.4	907.2	12.5	0.99	137.5	10.9
Belarus	0.35	18.8	5.4	0.0	19.3	3.91	25.9	5.2
Belgium	0.56	26.1	11.3	1228.8	20.0	0.61	110.6	3.4
Bosnia&Herzegovina	0.43	19.3	15.0	1075.2	20.2	1.01	90.1	4.5
Bulgaria	0.39	12.7	7.2	714.8	17.3	1.87	62.2	6.7
Croatia	0.46	19.5	15.2	870.4	20.4	1.09	87.0	4.7
Cyprus	0.02	0.5	0.0	101.7	0.6	0.01	0.0	0.0
Czech Republic	0.97	48.2	8.8	0.0	45.1	3.55	180.8	14.3
Denmark	0.19	9.0	0.9	738.3	6.0	0.24	35.4	1.4
Estonia	0.23	19.2	1.0	627.6	13.4	0.59	8.3	0.4
Finland	0.1	16.3	1.2	418.3	10.4	3.39	21.1	6.9
France	0.24	13.1	8.6	472.4	10.8	5.74	49.7	26.4
Georgia	0.65	22.9	15.6	1332.4	26.0	1.71	154.5	10.1
Germany	0.45	45.3	6.7	1149.1	21.2	7.57	74.0	26.4
Greece	0.14	6.2	8.2	333.9	6.3	0.77	23.7	2.9
Hungary	0.76	21.3	10.7	0.0	25.7	2.37	111.2	10.3
Iceland	0.01	0.4	0.1	24.5	0.4	0.04	0.0	0.0
Ireland	0.06	4.3	6.1	110.0	3.3	0.22	18.9	1.2
Italy	0.23	9.5	5.7	462.2	9.1	2.46	51.1	13.9
Kazakhstan	0.11	2.9	1.1	296.3	3.7	2.14	11.1	6.4
Latvia	0.27	22.2	2.3	693.8	16.8	1.03	48.9	3.0
Lithuania	0.32	24.5	4.4	985.7	20.3	1.32	33.7	2.2
Luxembourg	0.31	25.0	9.3	0.0	16.3	0.04	109.3	0.2
Malta	0.03	0.6	0.0	137.2	1.3	0.00	0.0	0.0
Netherlands	0.47	25.4	8.1	891.9	14.4	0.47	70.5	2.3
Norway	0.04	9.7	0.3	147.3	3.9	1.16	7.5	2.2
Poland	1.17	41.8	8.8	1539.3	47.3	14.59	211.2	65.2
Portugal	0.06	3.4	2.0	121.2	3.3	0.27	19.5	1.6
Republic of Moldova	0.49	13.3	13.7	1191	16.1	0.53	31.0	1.0
Romania	0.52	19.4	10.7	773.0	23.4	5.54	80.1	19.0
Russian Federation	0.39	22.7	9.5	674.7	22.4	82.52	61.9	229
Slovakia	1.01	36.2	9.2	0.0	42.1	2.06	137.2	6.7
Slovenia	0.51	32.2	15.1	1004.1	29.6	0.54	132.4	2.4
Spain	0.07	3.8	2.6	205.3	3.4	1.59	20.2	9.6
Sweden	0.08	16.5	0.6	401.4	7.6	3.24	15.8	6.8
Switzerland	0.33	24.8	4.5	0.0	15.7	0.66	39.5	1.7
The FYR of Macedonia	0.37	15.0	8.8	651.0	17.0	0.44	74.5	1.9
Turkey	0.06	1.9	0.4	370.7	2.1	2.00	0.0	0.0
Ukraine	0.57	20.1	8.4	1182.4	22.4	13.32	87.1	51.8
United Kingdom	0.18	8.0	6.8	356.1	5.7	1.33	51.6	12.0
Yugoslavia	0.59	20.7	15.5	731.6	24.0	2.44	110.8	11.2

1.2.2. Comparison of B[a]P modelling results with measurements ²

Air concentrations

Table 1.8 presents measured and calculated mean annual values of concentrations and their ratios. The ratio of measured to calculated values are within the limits of 0.33-1.5 (except for a single value 18.7). On the average the calculated values are 1.2 times higher than the measured ones.

Table 1.8. B[a]P mean annual concentrations in air, ng/m³

Station	Country	Year	Measurement	Calculation	Meas./Calc.
SE97	Sweden	1991-94	0.08	0.237	0.34
CZ3 *	Czech Republic	1991	0.02	0.0448	0.45
CZ3 *	Czech Republic	1992	0.84	0.045	18.67
CZ3 *	Czech Republic	1993	0.03	0.0374	0.80
CZ3 *	Czech Republic	1994	0.03	0.0419	0.72
CZ3 *	Czech Republic	1995	0.04	0.0562	0.71
CZ3 *	Czech Republic	1996	0.06	0.043	1.40
CZ3	Czech Republic	1997	0.64	1.22	0.52
FI96	Finland	1996	0.02	0.03	0.67
NO42	Norway	1994	0.012	0.01	1.20
NO42	Norway	1995	0.012	0.025	0.48
NO42	Norway	1997	0.015	0.01	1.50
SE2	Sweden	1994	0.07	0.21	0.33
SE2	Sweden	1995	0.1	0.17	0.59
SE2	Sweden	1996	0.09	0.23	0.39
Mean			0.14	0.16	0.85

* gas-phase

The comparison of the calculated results with the data obtained in *M.Pekar et al.* [1999] indicates that the model modification improved the agreement with measurement data. Note that in contrast with PCBs calculated air concentrations of B[a]P on the average exceed measurements. This is in agreement with the fact that the transport outside the EMEP grid amounts to about 30% for this pollutant and the influence of external sources with respect to the EMEP grid seems to be not so significant.

Concentration in precipitation

Table 1.9 presents measured and mean annual calculated values of concentrations in precipitation and their ratios. The latter varies from 1.3 to 2.5. On the average the measured values are 2 times higher than the calculated ones.

² This section was written in the co-authorship with Dr. A.-R.Milukaité (Lithuania) and Prof. I.Holoubek (Czech Republic)

Table 1.9. B[a]P mean annual concentrations in precipitation, ng/l

Station	Country	Year	Measurement	Calculation	Meas./Calc.
DE1	Germany	1996	3.88	1.8	2.2
DE9	Germany	1995	3.84	2.89	1.3
DE9	Germany	1996	8.4	4.29	2.0
DE9	Germany	1997	6.26	2.78	2.3
SE2	Sweden	1996	10.58	4.29	2.5
Mean			6.6	3.2	2.0

The comparison of the calculated results with the data obtained by *M.Pekar et al.* [1999] indicates that the model modification improved the agreement with measurement data. Like in the case of PCB some underestimation of washout ratio seems to take place.

Concentrations in soil and vegetation

Table 1.10 demonstrates measured and calculated mean annual concentrations in the top soil layer at Czech station CZ3 (Kosetiče) and at Swedish station SE97 (Gardsjön). Calculations underestimate soil concentrations in 1.7 times on the average. Note that in calculations of soil concentrations the value of soil density was assumed to be 800 kg/m³ [Duyzer et al., 1997]. However, according to [Brorström-Lunden and Löfgren, 1998] forest humus density at Gardsjön station in the region of measurements is 262 kg/m³. Using the latter value we obtain the calculated concentration in soil in 1994 to be equal to 69.47 ng/g which is certainly closer to the measured value.

Table 1.10. B[a]P concentration in the soil, ng/g

Station	Country	Year	Measurement	Calculation	Meas./Calc.
SE97	Sweden	1994	98	22.75 (69.47)*	4.31 (1.41)*
CZ3	Czech Republic	1996	80.84	61.25	1.32
CZ3	Czech Republic	1997	58.23	59.75	0.97
Mean			79.02	47.92	1.65

* the values in the brackets are obtained with local soil density means

Table 1.11 presents measured and calculated mean annual concentrations of the gaseous component at Czech station CZ3 (Kosetiče) and at Swedish station SE97 (Gardsjön). The calculated values both overestimate (6 times) and underestimate (3.6 times) the measured values.

Table 1.11. B[a]P concentration in vegetation, ng/g d.w.

Station	Country	Year	Measurement	Calculation	Meas./Calc.
SE97	Sweden	1991-94	3.7	1.02	3.63
SE97	Sweden	1995	1.18	0.78	1.52
CZ3	Czech Republic	1996	2.07	13.78	0.15
CZ3	Czech Republic	1997	2.60	13.48	0.19
Mean			2.4	7.3	

Concentration in sea water

Table 1.12 presents calculated B[a]P concentrations in the Baltic Sea. Measured data were taken from POPCYCLING-Baltic project. These data are cited in [Axelmann *et al.* (in press Nov.99)] and [Biziuk *et al.*, 1999]. Besides the project materials contain unpublished data of *Eva Brorström-Lunden*.

Table 1.12. B[a]P concentrations in sea water, ng/l

Year	Measured	Calculated	Meas./Calc.
1988	0.0252	1.27	0.02
1989	0.0341	1.20	0.03
1991	0.0824	1.16	0.07
1992	0.1128	1.08	0.10
1993	0.0674	1.02	0.07
1996	4.8	0.82	5.85
1997	3.125	0.76	4.12
Mean	1.1781	1.04	1.13

Table 1.12 demonstrates strong scattering of measured data (from 0.03 to 4.8 ng/l). Such scattering can result from sampling at different depths. In particular data for 1988-93 refer to samples taken from the depths of 9 - 15 m. The depths at which samples of 1996-97 were taken are not indicated in project materials. Besides strong variability in space is peculiar to seawater concentrations. In particular, calculated data averaged over different Baltic regions vary considerably; e. g. for 1997 these mean values vary from 0.3 to 1.2 ng/l. Together with restricted number of available measurements, these facts make the comparison of water concentrations difficult. It can be stated only that the variation range of calculated and measured values is similar.

B[a]P concentrations in different environmental compartments in Lithuania

This section presents the comparison between measured and calculated B[a]P concentrations in different environmental compartments in Lithuania both averaged over the whole Lithuanian territory and obtained at specific monitoring stations. The data used here including air, soil and vegetation concentrations are partly published in [Milukaite *et al.*, 1995; Milukaite, 1998]. Air concentrations were measured at station Preila. The samples of soil and vegetation were taken different locations with different contamination levels.

Table 1.13 presents measured and calculated values of B[a]P air concentrations averaged within the year.

Table 1.13. Annual B[a]P air concentrations at station Preila (Lithuania), ng/m³

	1991	1992	1993	1994	1995	1996	1997
Measured	0.65	0.68	0.62	0.75	1.05	1.26	1.33
Calculated	0.68	0.62	0.50	0.51	0.59	0.58	0.51
Meas./Calc.	0.95	1.10	1.22	1.48	1.78	2.17	2.59

Except for 1991, the measured values exceed the calculated ones. According to measurement data B[a]P air content increased in the period 1994 – 1997 whereas the calculations indicate the tendency of their diminishing. During this period measurement to calculations ratio increases from 1.48 to 2.59. This increase can be explained by the increase of car number in Lithuania twice since 1990. Possibly, the refinement of emission data for Lithuania for the last two years will improve the agreement between measured and calculated values.

Table 1.14 presents soil concentrations of B[a]P in the soil top layer (0-5 cm) (averaged over different measurements) for regions located at different distances from emission sources.

Table 1.14. B[a]P soil concentrations in different regions of Lithuania, ng/g

Region type				Calculated
Industrial	Occasional sources	Background	Suburb	
59	8.2	2.8	23.3	24.50

Note that the measured values in the industrial regions are almost two times higher than calculated ones, but for suburban regions these values are almost equal. Calculated values is 8 times higher than measurements in background regions. As a whole one can draw a

preliminary conclusion that the model overestimates soil concentrations though for better results the comparison with higher spatial resolution (50x50 km) is preferable.

The data on B[a]P concentrations in vegetation are highly detailed with respect to vegetation types (table 1.15). Calculated values averaged over Lithuania for 1997 describe well concentrations in leaves of couch-grass and bent-grass being the most widely spread grass in Lithuania meadows. The measurements/calculation ratios for them equal to 1.45 and 1.02 respectively. On the other hand, measured concentrations of B[a]P in clover, moss, and needles at different distances from a road considerably exceed averaged calculated value (2.7, 8.1 and 10.8 times respectively).

Table 1.15. B[a]P concentrations in vegetation, ng/g d.w.

Leaves & stem	Couch grass		Bent-grass		Moss	Scots pine			Calculated
	Leaves	roots	Leaves	roots		Needles	Bark	wood	
12	6.4	86.8	4.5	116	35.7	47.5	87	7.1	4.4

As it is mentioned in [Milukaite, 1998], the accumulation of B[a]P in moss and needles occurs mainly due to deposition of the particulate phase.

Note that the analysis of measurement data shows that B[a]P can be accumulated not only in leaves but also in roots, bark, and wood of trees. The investigation of the B[a]P root uptake influence on accumulation in vegetation is planned in future.

Table 1.16 presents measured and calculated data on total B[a]P deposition fluxes averaged over Lithuania for a number of years. Measured values are growing in the period of 1990 – 1992 and are sharply diminished in 1993. The decrease of B[a]P flux in 1993 may be caused by the irregular sampling of bulk deposition during this year. As a whole calculated values appropriately reflect the level of measured fluxes. The ratio of measured to calculated value varies from 0.32 to 1.35 with the average 0.8. In general, we can assume that calculated values appropriately reflect the level of measured fluxes.

Table 1.16. Mean total deposition flux for B[a]P, g/km²/year

	1990	1991	1992	1993	1994	1995	1996	1997
Measured	12	20.4	22.8	9.6	25.2	30	22.4	18.6
Calculated	37.0	34.8	34.9	28.3	26.4	22.2	22.4	20.3
Meas./Calc.	0.32	0.59	0.65	0.34	0.95	1.35	1.00	0.92

1.2.3. Conclusions

Using MSCE-POP model the simulation of B[a]P transport within European region for the period of 1970-97 was made. Emission estimates obtained under POPCYCLING-Baltic project for the indicated time period were used in modelling. On the basis of calculations mean deposition and concentration values in environmental compartments (air, soil, sea water, vegetation) in European countries for 1997 were determined. Preliminary estimates of B[a]P emitted in each country and deposited in other European countries are estimated (here we indicate countries with sufficiently large export values only) and primary analysis of the relationship between emission and concentration levels in individual countries is made. To verify the model measured and calculated data for 1991-97 are compared.

1. The comparison of calculation and measurement results indicates that calculated air concentrations exceed measured ones on the average by 20% and measured concentrations in precipitation exceed calculated ones by 2 times. For concentrations in soil, vegetation and sea water the agreement of measured and calculated values is within a factor of 5. The number of measurement data for these media, however, is insufficient for model verification. For all the compartments but air the calculated results are somewhat underestimated.
2. Depending upon the relation between air concentrations and emission densities European countries are conventionally divided into two groups. The first group includes countries where air pollution is defined mainly by their own emissions (Armenia, Azerbaijan, Slovenia, Georgia, Belgium, Yugoslavia, the Czech Republic and Poland). The other group involves countries where air pollution is defined predominantly by external sources (the Netherlands, Hungary, Austria, Slovakia, Moldova, and Germany).
3. According to preliminary computation results the countries with high export values are Poland, the Ukraine, France, Germany, Romania, the Czech Republic, Italy, Great Britain and Yugoslavia. To more precisely quantify the transboundary B[a]P fluxes the calculation with 50x50 km grid of "country-to-country" matrix is planned in the next year.
4. For B[a]P sea is the most inertial media; soil and forest litter are less inertial than sea but more inertial for B[a]P than air and vegetation. Even if emissions are ceased a considerable sea content level can be supported during decades.
5. The B[a]P mass transported outside the EMEP grid amounts to about 30% of total emission quantity. This manifests that long-range transport modelling for this pollutant can be performed at the European scale as a first approximation.

1.3. γ -Hexachlorocyclohexane

Modelling of γ -HCH transport in European region is made for the period of 1970-97. Since it is important to take into account γ -HCH long-term accumulation in environmental compartments a preliminary calculation run covering 15-year period with emission and meteorological data for 1970 has been carried out. Like for other studied pollutants emission estimates obtained under POPCYCLING-Baltic project [Pacyna *et al.*, 1999] are used. Spatial emission distribution for the last year is presented in figure 1.15. The figure indicates that major emission sources (according to emission data used) are located in south-western Europe. γ -HCH emission data are demonstrated in table 1.17 in the end of this section.

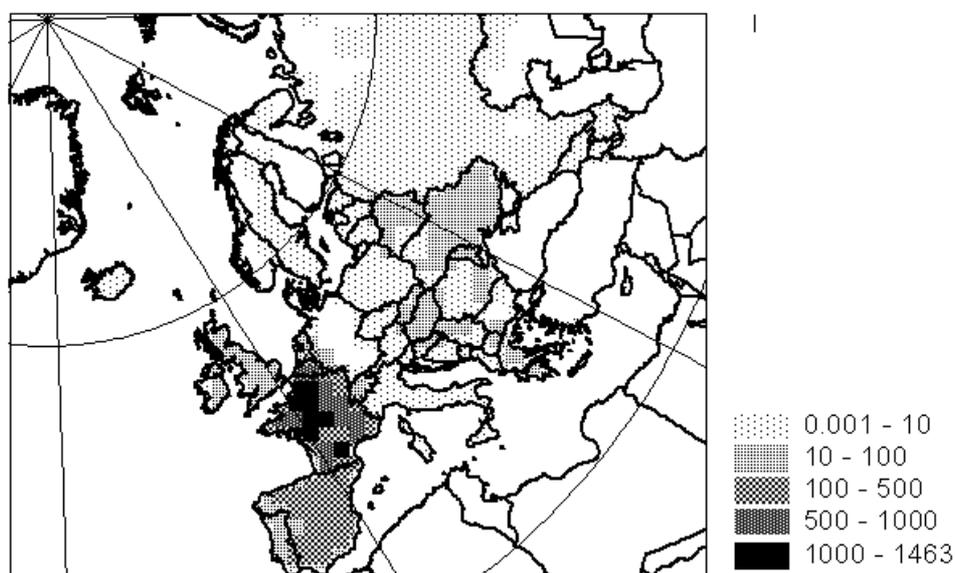


Figure 1.15. γ -HCH emission in 1997, g/km²/yr.

In this section we analyse γ -HCH depositions and concentrations in different environmental compartments averaged over individual countries. A provisional ranging of countries relative to the influence of internal and external sources on the air pollution level is made. It is assumed that γ -HCH in air is present in the gas-phase only.

The analysis of the calculation results pointed out that the marine environment exerts the largest influence on γ -HCH dispersion at long time scale (see volume 2 of this report). γ -HCH accumulated in sea can support its air concentrations level during 10-15 years. The comparison of calculated and measured data indicates that the model results for all the compartments are underestimated. To some extent it is explained by high volatility of this compound (according to calculations about 60% of its emission is transported outside the

EMEP grid). For this reason in order to obtain a correct assessment of the pollution level in European region γ -HCH modelling should be made on the hemispherical scale.

1.3.1. γ -HCH concentrations and depositions in European countries in 1997

This section deals with a provisional analysis of γ -HCH mean depositions and concentrations in air, soil, sea water and vegetation in European countries. Appropriate data averaged over 1997 and territories of individual countries are summarized in table 1.17 presented in the end of this section.

First air concentrations are considered. Figure 1.16 demonstrates mean air concentrations for 1997.

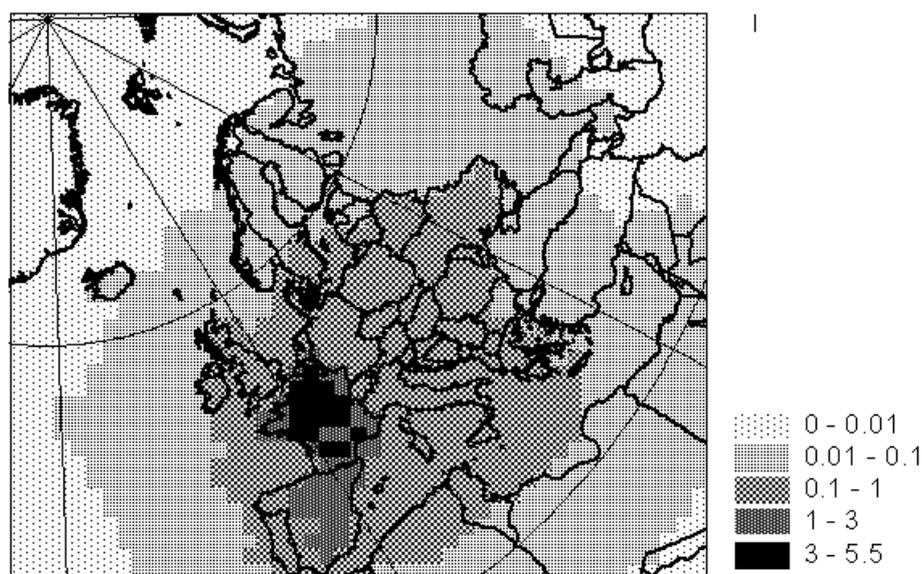


Figure 1.16. γ -HCH air concentrations in 1997, ng/m^3

The comparison of figures 1.15 and 1.16 shows that maximum air concentration region coincides with maximum emission region. The highest deposition densities ($1\text{-}4 \text{ ng}/\text{m}^3$) are observed in France, Belgium, Luxembourg and Spain, slightly lower densities ($0.5\text{-}1 \text{ ng}/\text{m}^3$) - in the Netherlands, Switzerland and Germany.

On the basis of the relationship between air concentrations and emission densities among countries in which γ -HCH air concentration level exceeds the average value ($0.39 \text{ ng}/\text{m}^3$) two groups of countries are distinguished (fig. 1.17).

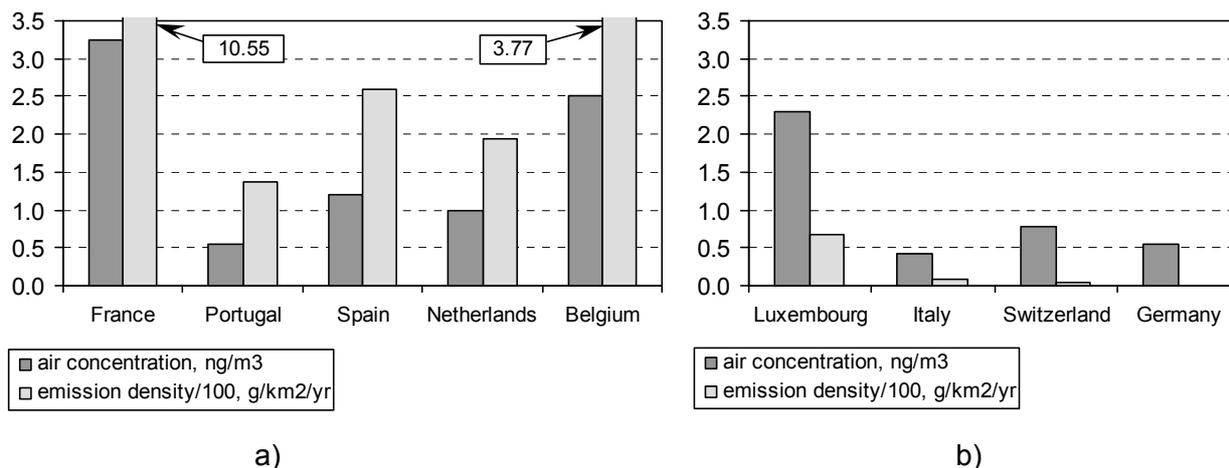


Figure 1.17. γ -HCH air concentrations and emission density in a number of European countries in 1997

The first group includes France, Portugal, Spain, the Netherlands and Belgium where high air concentrations are apparently explained by essential emission densities (350-1050 g/km²/yr.) in these countries (fig.1.17.a).

The second group includes countries where high air concentrations are accompanied by relatively low emission fluxes (less than 100 g/km²/yr.). They are Germany, Italy, Switzerland and Luxembourg (fig.1.17.b). Most probably a tangible level of air concentrations in these countries is mainly explained by the effect of γ -HCH transboundary transport from sources of other countries.

Separately stand Austria and Slovenia, which are characterized by low air concentrations while soil concentrations exceed mean European level.

Figure 1.18 presents soil concentration distribution map.

Like in the case of air concentrations maximum soil concentrations coincide with the maximum emission region. Maximum soil concentrations (15 - 25 ng/g) are observed in France, Luxembourg and Belgium, slightly lower (5 - 15 ng/g) - in Switzerland, the Netherlands, Germany, Spain, Italy and Austria. Note that in Luxembourg γ -HCH soil concentration is high (22 ng/g) whereas the emission level is relatively low (57.7 g/km²/yr.).

Concentration distribution in soil is more "uncertain" in comparison with air concentrations. In particular γ -HCH transport to the region of northern Scandinavian coast is made by sea currents (see the map of marine water concentrations).

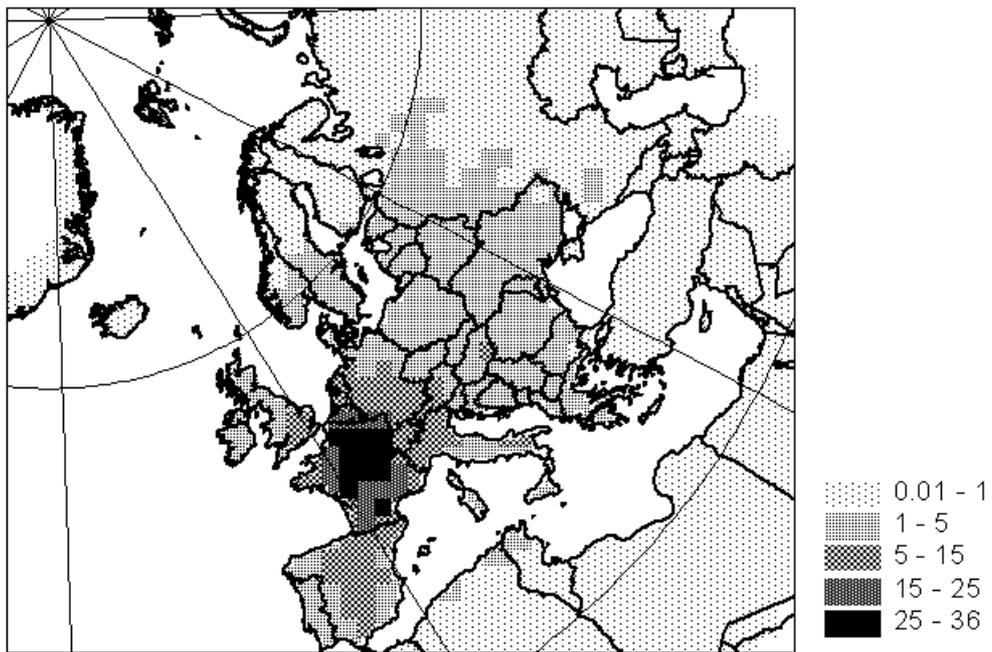


Figure 1.18. γ -HCH soil concentrations in 1997, ng/g

The map of mean marine concentration distribution in 1997 is demonstrated in figure 1.19.

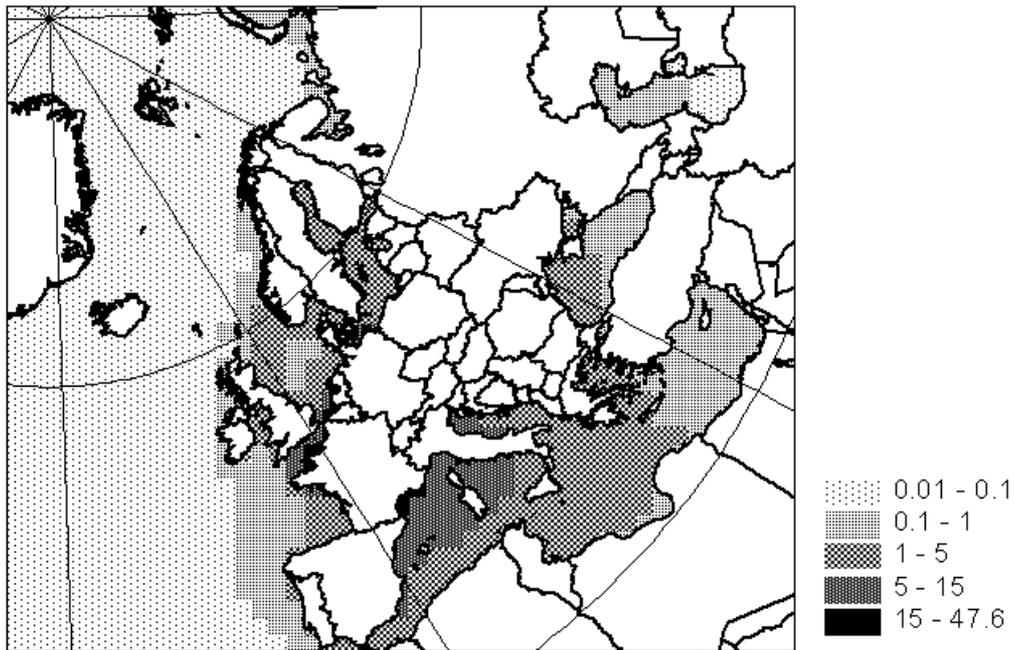


Figure 1.19. γ -HCH sea concentrations in 1997, $\mu\text{g}/\text{m}^3$

As evident from the map γ -HCH maximum concentrations are observed in sea regions located close to major emission sources. γ -HCH entering the sea near emission sources then is transported by sea currents up to the northern coast of Scandinavia. This is the reason for relatively high concentrations in soil of this region compared to other regions located approximately at the same distance from major sources.

Figure 1.20 illustrates the map of γ -HCH mean concentration distribution in vegetation in 1997.

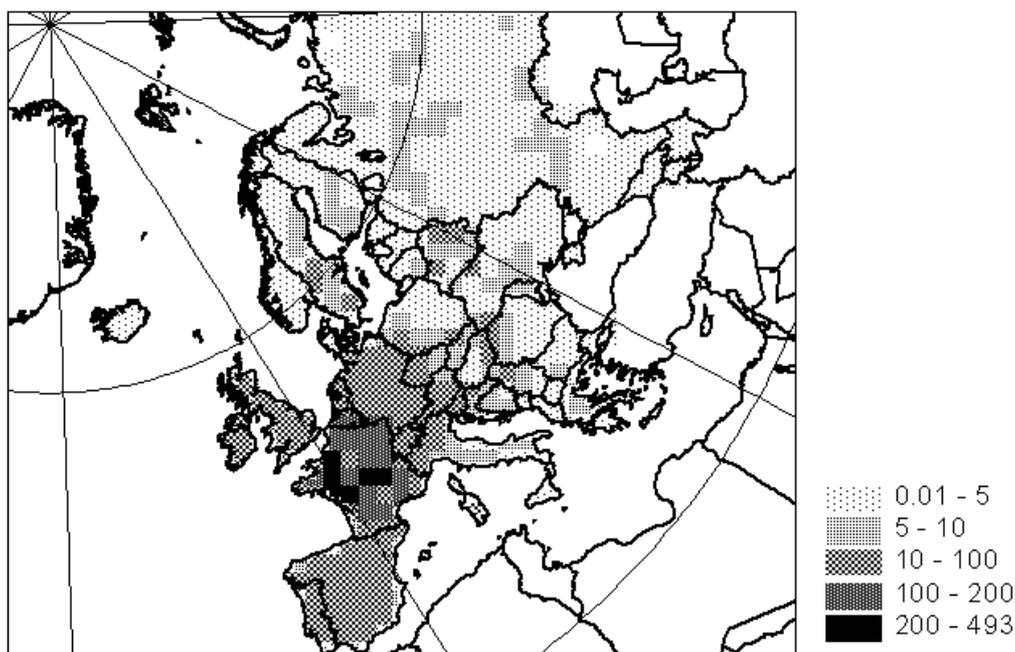


Figure 1.20. γ -HCH concentration in vegetation in 1997, ng/g d.w.

As seen from the map calculated vegetation concentrations correlate well with emission values. Maximum concentrations (100-170 ng/g of dry weight) are observed in France, Luxembourg and Belgium. Concentrations from 20 to 100 ng/g of dry weight are observed in Switzerland, the Netherlands, Spain, Ireland, Germany, Great Britain and Austria.

We now turn our attention to the analysis of deposition spatial distribution. Figure 1.21 demonstrates the map of γ -HCH mean annual total (wet + dry) depositions.

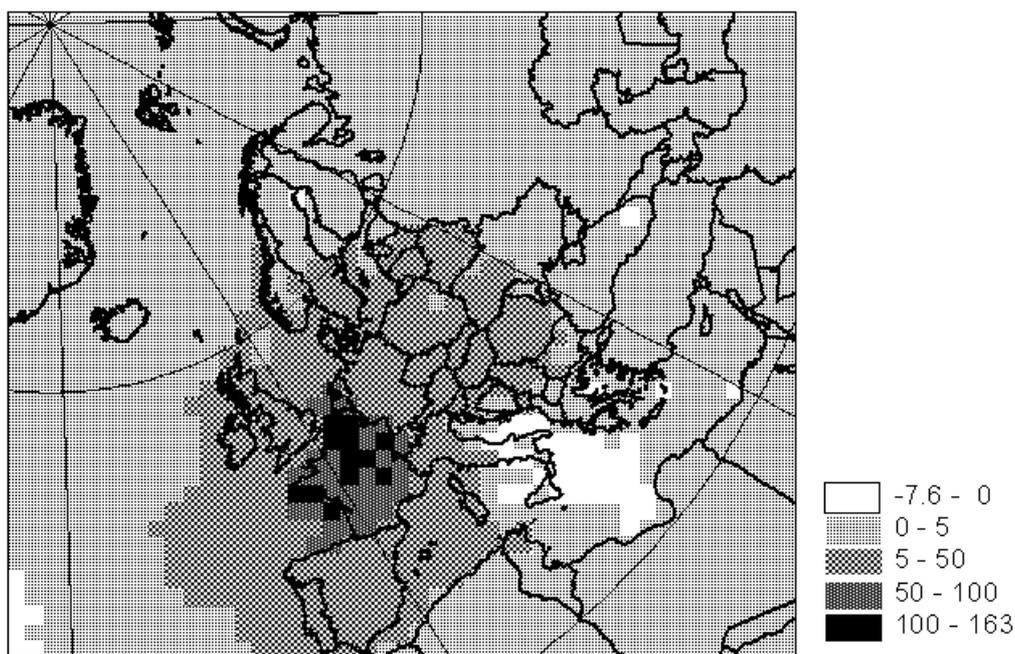


Figure 1.21. γ -HCH total depositions in 1997, g/km²

It is evident that re-emission takes place in some regions of the Mediterranean Sea. It is conditioned by the temperature regime and the relationship of air and sea concentrations in this region: air concentrations are relatively low whereas sea concentrations have here one of their maxima.

In addition to the qualitative analysis of the effect of emitted γ -HCH on air concentrations, data on deposition densities allow us to estimate the quantity of γ -HCH emissions of one country, which is deposited in other countries (export). This evaluation can be made by formula (1.1) from section 1.1.1. According to calculations the highest export is characteristic of France (80 tonnes). Then in the order of export decrease come Spain (18 tonnes), Great Britain (2 tonnes), Belgium and Portugal (1.5 tonne each), the Ukraine (1 tonne), Greece (800 kg) and Belarus (600 kg). Certainly these figures are of a preliminary character.

All the presented results on γ -HCH content in different media of different European countries are summarized in table 1.17. The shaded countries have air and/or soil concentrations exceeding mean European levels (0.39 ng/m³ and 3.88 ng/g, respectively).

Table 1.17. γ -HCH concentrations and depositions in European countries in 1997

Country	Air, ng/m ³	Soil, ng/g	Vegetation, ng/g d.w.	Sea, ng/m ³	Deposition density, g/km ² /yr.	Total depositions, t/yr.	Emission density, g/km ² /yr	Emissions, t/yr.
Albania	0.15	1.50	2.37	2668.5	2.45	0.062	18.2	0.463
Armenia	0.01	0.09	0.1	0	0.21	0.005	1.2	0.030
Austria	0.3	5.44	20.02	0	20.42	1.737	0.0	0.000
Azerbaijan	0.01	0.10	0.2	123.8	0.32	0.025	1.4	0.113
Belarus	0.15	2.19	7.86	0	6.92	1.405	22.9	4.645
Belgium	2.51	20.85	100.51	24220	92.16	2.794	377.4	11.441
Bosnia&Herzegovina	0.2	2.74	6.8	3024.1	5.86	0.294	10.3	0.515
Bulgaria	0.11	1.34	2.54	1565.8	4.78	0.516	0.0	0.000
Croatia	0.24	3.58	8.35	4621	7.06	0.379	12.1	0.651
Cyprus	0.04	0.19	0	389.41	0.61	0.008	7.2	0.091
Czech Republic	0.29	3.84	12.06	0	14.39	1.135	4.0	0.319
Denmark	0.19	2.48	2.65	3931.5	15.72	0.638	0.0	0.000
Estonia	0.07	1.26	2.4	2139.3	4.22	0.187	0.1	0.005
Finland	0.03	0.73	3.81	1497.7	1.92	0.627	0.0	0.000
France	3.25	22.30	165.63	18485	81.30	43.172	1054.6	560.00
Georgia	0.02	0.26	0.39	368.75	0.94	0.062	1.4	0.095
Germany	0.54	6.80	28.67	5577.7	26.99	9.627	0.0	0.000
Greece	0.17	1.25	3.74	1405.3	3.31	0.403	48.2	5.863
Hungary	0.26	3.79	8.03	0	9.89	0.912	30.2	2.781
Iceland	0.01	0.20	0.32	132.6	1.52	0.143	1.2	0.109
Ireland	0.17	1.91	31.03	1251.7	14.86	0.973	33.1	2.167
Italy	0.42	5.79	11.27	6080.4	12.27	3.339	8.2	2.230
Kazakhstan	0.02	0.14	0.26	241.64	0.59	0.338	0.8	0.446
Latvia	0.09	1.58	3.45	2487.7	6.07	0.372	0.0	0.002
Lithuania	0.11	1.66	4.68	2742.7	5.89	0.385	0.0	0.003
Luxembourg	2.3	21.59	115.61	0	89.20	0.199	67.7	0.151
The FYR of Macedonia	0.14	1.61	3.74	2535.4	4.07	0.104	2.5	0.063
Malta	0.18	0.99	0	2275.8	2.23	0.001	0.0	0.000
Netherlands	0.99	9.05	38.73	12927	55.36	1.799	194.1	6.307
Norway	0.04	0.89	1.06	906.27	3.90	1.167	0.0	0.000
Poland	0.18	2.19	5.85	3868.4	8.49	2.619	0.9	0.283
Portugal	0.55	3.08	16.62	2239.7	13.18	1.105	136.4	11.441
Republic of Moldova	0.14	1.71	6.18	2101.1	4.87	0.159	11.1	0.364
Romania	0.14	1.86	5	1646.3	6.76	1.602	9.7	2.308
Russian Federation	0.03	0.76	3.14	853.32	2.14	7.909	1.6	5.941
Slovakia	0.26	3.14	8.14	0	10.68	0.522	32.7	1.598
Slovenia	0.26	4.44	12.78	8372.5	12.19	0.222	10.0	0.182
Spain	1.2	5.98	31.27	5587.2	21.77	10.337	258.8	122.91
Sweden	0.06	1.21	5.39	2183.8	4.81	2.062	3.2	1.355
Switzerland	0.77	12.54	39.26	0	48.20	2.018	3.8	0.159
Turkey	0.02	0.21	0.04	680.76	0.87	0.833	0.0	0.000
Ukraine	0.13	1.61	4.24	1459.3	5.02	2.985	15.2	9.074
United Kingdom	0.31	3.20	28.18	4605	16.71	3.88	72.5	16.843
Yugoslavia	0.18	2.78	6.85	2673.9	7.01	0.712	15.2	1.538

1.3.2. Comparison of γ -HCH modelling results with measurements³

Air concentrations

Table 1.18 presents mean annual concentrations calculated and observed at EMEP stations and their ratios. These ratios vary from 0.11 to 20.5. However, if we exclude from the comparison station NO42 located near the EMEP grid boundary, the mean ratios will vary from 0.11 to 1.3. The underestimation of the calculated concentration at station NO42 can result from the fact that the calculation is made on the European scale and real concentrations can be affected by sources located outside the EMEP region. The considerations of these sources can be made by means of hemispherical modelling.

Table 1.18. γ -HCH mean annual air concentrations, ng/m³

Station	Country	Year	Measurement	Calculation	Meas./Calc.
FI96	Finland	1996	0.011	0.022	0.49
NO42	Norway	1993	0.014	0.001	20.5
NO42	Norway	1994	0.016	0.001	16.3
NO42	Norway	1995	0.013	0.001	20.3
NO42	Norway	1996	0.013	0.002	6.57
NO42	Norway	1997	0.015	0.002	8.75
NO99	Norway	1992	0.086	0.067	1.29
NO99	Norway	1993	0.059	0.101	0.58
NO99	Norway	1994	0.123	0.097	1.27
NO99	Norway	1995	0.065	0.076	0.85
NO99	Norway	1996	0.061	0.093	0.65
NO99	Norway	1997	0.062	0.085	0.73
SE2	Sweden	1994	0.051	0.149	0.34
SE2	Sweden	1995	0.027	0.117	0.23
SE2	Sweden	1996	0.025	0.172	0.14
IS91	Iceland	1995	0.014	0.011	1.31
IS91	Iceland	1997	0.006	0.016	0.42
CZ3	Czech Republic	1997	0.034	0.326	0.11
Mean			0.039	0.074	

The comparison of the calculated results with that obtained in *M.Pekar et al.* [1999] indicates that the model modification improved the agreement with measurement data excluding station NO42. On the other hand, modelling of γ -HCH transport on the hemispherical scale can result in some increase of calculated concentrations, since the export outside the EMEP grid for this pollutant is 60%.

³ This section was written in the co-authorship with Prof. I.Holoubek (Czech Republic)

Concentration in precipitation

Table 1.19 presents calculated and measured mean annual concentrations of γ -HCH in precipitation and their ratios. The ratio of measured to calculated values (except for two extreme values) varies from 0.54 to 6.2. On the average measured values are 2 times higher than the calculated ones. The correlation between measured and calculated values is rather high alongside total underestimation of concentrations in precipitation.

Table 1.19. Mean annual γ -HCH concentrations in precipitation, ng/l

Station	Country	Year	Measurement	Calculation	Meas./Calc.
DE1	Germany	1990	4.54	4.25	1.07
DE1	Germany	1992	18.22	3.54	5.15
DE1	Germany	1993	9.28	3.30	2.81
DE1	Germany	1995	6.32	6.86	0.92
DE1	Germany	1996	10.38	9.70	1.07
DE9	Germany	1995	6.52	8.17	0.80
DE9	Germany	1997	22.06	13.03	1.69
FI96	Finland	1996	0.34	6.07	0.06
IS91	Iceland	1995	0.44	0.36	1.24
IS91	Iceland	1996	0.27	0.50	0.54
IS91	Iceland	1997	0.42	0.35	1.20
NO99	Norway	1991	4.05	3.47	1.17
NO99	Norway	1992	5.02	1.59	3.16
NO99	Norway	1993	8.45	3.54	2.38
NO99	Norway	1994	9.98	5.00	2.00
NO99	Norway	1995	5.54	3.81	1.45
NO99	Norway	1996	8.01	7.56	1.06
NO99	Norway	1997	4.89	2.89	1.69
SE2	Sweden	1996	17.94	6.47	2.77
DK31	Denmark	1990	16.98	2.72	6.24
DK31	Denmark	1991	11.91	3.12	3.81
DK31	Denmark	1992	15.82	1.55	10.20
BE4	Belgium	1997	94.28	40.75	2.31
Mean			12.25	6.03	2.03

The decrease of calculated concentrations in precipitation compared to modelling results obtained in [Pekar *et al.*, 1999] is explained by the decrease of atmospheric concentrations. As it was mentioned above γ -HCH modelling on the hemispherical level can result in an increase of concentration values in precipitation.

Concentrations in soil and vegetation

Table 1.20 shows measured (at station CZ3 the Czech Republic) and calculated mean annual concentrations in soil. The mean ratio of measurements to calculations is 0.16. The overestimation of soil concentrations can be conditioned by the fact that while describing processes in the soil the model does not consider the run-off by melting and rain water and γ -HCH transport by river currents.

Table 1.20. γ -HCH concentration in the soil at station CZ3, ng/g

Year	Measurements	Calculation	Meas./Calc.
1996	0.099	3.46	0.03
1997	1.01	3.58	0.28
Mean	0.55	3.52	0.16

Table 1.21 demonstrates measured (at station CZ3) and calculated mean annual concentrations in needles of coniferous trees. The mean ratio of measured to calculated values is about 1.

Table 1.21. γ -HCH concentrations in vegetation at station CZ3, ng/g d.w.

Year	Measurement	Calculation	Meas./Calc.
1996	0.39	2.36	0.16
1997	4.84	2.92	1.65
Mean	2.61	2.64	0.99

It should be mentioned that at present the number of measurements in such compartments as sea water, soil and vegetation is insufficient for model verification.

Concentrations in sea water

Table 1.22 contains γ -HCH calculated concentrations in the Baltic Sea. Measurement data are taken from POPCYCLING-Baltic project where different data on the Baltic Sea are systematized. These data are cited in [Gaul, 1992; Biziuk et al., 1999; Harner et al., 1995].

Table 1.22. γ -HCH concentrations in sea water, pg/l

Year	Measured	Calculated	Meas./Calc.
1982	7600	9330	0.81
1983	4700	8876	0.53
1985	4550	7780	0.58
1986	3900	7164	0.54
1987	3500	6546	0.53
1988	2925	5862	0.50
1989	2850	5260	0.54
1990	2536	4665	0.54
1991	2250	4358	0.52
1996	10054	2897	3.47
1997	23000	2884	7.98
Mean	7269	4321	1.68
Mean 1982 – 1991	3868	6649	0.58

The table data indicate a large scattering of measured concentrations in the sea for 1982-91 and 1996-97. When the data for 1982-91 are used mean calculated concentrations differ from mean measured ones by a factor of 2 varying within the range of 0.5-0.8. Note that calculated concentrations averaged over different Baltic regions vary from 1521 to 4874 pg/l.

1.3.3. Conclusions

Using MSCE-POP model the simulation of γ -HCH transport within European region for the period of 1970-97 was made. Emission estimates obtained under POPCYCLING-Baltic project for the indicated time period were used in modelling. On the basis of calculations mean deposition and concentration values in environmental compartments (air, soil, sea water, vegetation) in European countries for 1997 were determined. Preliminary estimates of γ -HCH emitted in each country and deposited in other European countries are estimated and primary analysis of the relation between emission and air concentration levels in individual countries is made (here we indicate countries with sufficiently large export values only). To verify the model measured and calculated data for 1991-97 are compared.

1. The comparison of calculated versus measured values indicated that concentrations in the marine environment coincide within a factor of 2, in soil - within a factor of 6. The calculated content in air and precipitation is underestimated by a factor of 5 and 2,

respectively. As to other compartments the calculations give overestimated results. On the whole the consistency between calculations and measurements is improved compared to the results of the previous year. It should be mentioned that the comparison base for soil, sea and vegetation is insufficient for the reliable model verification.

2. γ -HCH high air concentrations ($0.5-3.5 \text{ ng/m}^3$) in France, Portugal, Spain, the Netherlands and Belgium can be explained by essential emission densities in these countries. On the contrary Germany, Italy, Switzerland, and Luxembourg have relatively high air concentrations ($0.5-2.5 \text{ ng/m}^3$) and low emission densities. Obviously air concentration levels in these countries is explained by the impact of γ -HCH transboundary transport from sources of other European countries.
3. According to preliminary calculations the countries with high export values are France (80 tonnes), Spain (18 tonnes), Great Britain (2 tonnes), Belgium and Portugal (1.5 tonne each), the Ukraine (1 tonne), Greece (800 kg) and Belarus (600 kg).
4. The investigation of γ -HCH accumulation in different compartments (see volume 2 of this report) pointed out that even if there was no emission sea plays the role of a "reservoir" keeping pollution levels in different environmental compartments for a long time (about 10 years). In future more thorough study of this process is planned.

Chapter 2

The analysis of POP long-term (1970-97) accumulation in environmental compartments of a particular country exemplified by Great Britain

In Chapter 1 a detailed analysis of POP contamination levels in the studied compartments for different countries by the last year (1997) has been made. These levels in particular countries result from a long-term history of POP accumulation in different media. That is why the investigation of POP accumulation dynamics in each European country is of great importance.

In this chapter we consider long-term trends of POP accumulation in a particular country - Great Britain which is selected because we possess a detailed long-term accumulation for this country [*Harner et al.*, 1995] as well as sufficient measurement data [*Coleman et al.*, 1998]. In future such investigations are planned to be carried out for other European countries.

2.1. The analysis of PCB accumulation

The analysis of PCB accumulation in air, soil, sea water and vegetation in Great Britain is made on the basis of monitoring and modelling the pollutant transport in European region for the period from 1970 to 1997.

Figure 2.1 gives plots of PCB depositions and concentrations in environmental compartments in Great Britain during 1970-97 in comparison with emission dynamics. Figure 2.1.a shows the emission dynamics. As follows from the plot a considerable emission reduction took place in Great Britain from 1970 to 1997.

Figures 2.1.b,c demonstrate plots of mean annual concentration variations in air, vegetation, and sea water washing Great Britain coasts. The analysis of the plots allows us to conclude that these concentrations reflect emission variations and they are well correlated. The concentrations in vegetation and marine environment are less subject to variations conditioned by meteorological situations than atmospheric concentrations.

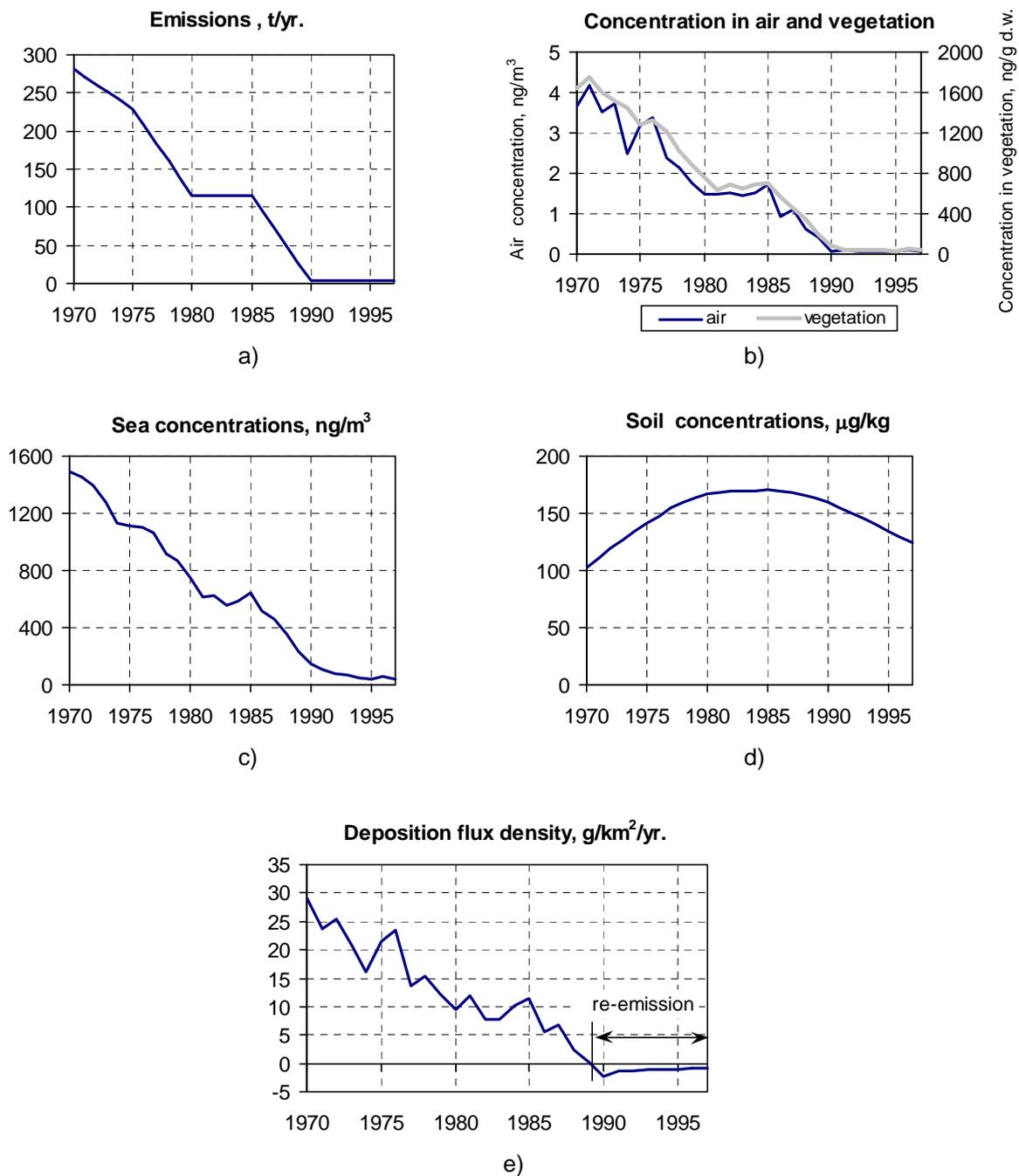


Figure 2.1. Dynamics of concentration variations in environmental compartments and PCB deposition densities in Great Britain in comparison with emission reduction during 1970-97

Figure 2.1.d shows the plot of PCB soil concentration variation. For PCB soil is an inertial medium and in spite of an abrupt emission reduction from 1970 to 1985 soil concentrations continue to grow and the maximum concentration is reached after 15 years (by 1985). Air concentrations, however, decrease more intensively. In 1989 mean annual re-emission flux starts to exceed depositions and by the end of simulation period PCB gaseous flux from soil is observed round the year. In figure 2.1.e a negative value of total depositions indicates that re-emission process is strong enough.

Comparing the dynamics of content variations in different compartments and deposition fluxes with mean European values it is necessary to mention some peculiarities of Great Britain. In particular, the re-emission process is set up by some years later than on the average in Europe. This fact can explain some delay of soil concentration decrease by the last year. Sea concentration dynamics better correlates with emission dynamics in Great Britain than in Europe on the whole. It is conditioned by water closeness (the Atlantic coast, the North Sea, English Channel) to the main sources of emission and its high mobility compared to internal seas.

Now we dwell on the comparison of modelling results with measurement data for Great Britain (table 2.1). Averaged over a year measurement data on air concentrations [Coleman *et al.*, 1998] for Great Britain are presented at 6 stations for 1991 and 1996. The ratio of measured concentrations to calculated ones vary from 0.8 for Haselrigg (1993) to 8.1 for London (1991). On the average measurement data exceed the calculated ones in 3 times. The greatest deviation of measured and calculated values are characteristic of big cities (Cardiff, London, Manchester). The correlation coefficient of measured and calculated data is 0.55.

Table 2.1. Comparison of PCB measured and calculated air concentrations for Great Britain

Location	Year	Measured, ng/m ³	Calculated, ng/m ³	meas./calc.
Cardiff	1991	0.49	0.09	5.2
Cardiff	1992	0.60	0.08	7.7
Cardiff	1993	0.19	0.08	2.5
Hazelrigg	1992	0.18	0.12	1.6
Hazelrigg	1993	0.08	0.11	0.8
Hazelrigg	1994	0.14	0.10	1.4
Hazelrigg	1995	0.10	0.11	0.9
Hazelrigg	1996	0.05	0.11	0.4
London	1991	1.43	0.18	8.1
London	1992	1.14	0.17	6.7
London	1993	1.31	0.17	7.9
London	1994	1.09	0.16	7.0
London	1995	1.25	0.16	7.7
London	1996	0.45	0.18	2.5
Manchester	1991	0.44	0.18	2.4
Manchester	1992	0.41	0.16	2.6
Manchester	1993	0.45	0.15	3.1
Manchester	1994	0.34	0.13	2.6
Manchester	1995	0.34	0.15	2.2
Manchester	1996	0.21	0.15	1.3
Middlesbrough	1992	0.25	0.12	2.1
Middlesbrough	1993	0.22	0.11	2.1
Middlesbrough	1994	0.09	0.10	1.0
Middlesbrough	1995	0.19	0.11	1.8
Middlesbrough	1996	0.18	0.11	1.5
Stevenage	1991	0.35	0.18	1.9
Stevenage	1992	0.19	0.16	1.2
Mean		0.45	0.13	3.2
Correlation				0.55

The analysis of table 2.1 data indicates that calculated results are in a satisfactory agreement with observations at stations located at a sufficiently long distance from powerful emission sources and appeared to be 5 times lower in big industrial cities (Cardiff, London, Manchester). It is in line with the fact that measurements in Cardiff, London and Manchester (urban areas), on the one hand, and for Hazelrigg (semi-rural) on the other hand differ on the average in 3.6-10 times. The comparison of calculations versus measurements in Hazelrigg is illustrated in figure 2.2. It is clear that calculated data agree with measurements within a factor of 2. The exceedance of calculated data above measured ones in 1996 can be

conditioned by the fact that Great Britain emission in 1996 is assumed to be equal to that of 1995.

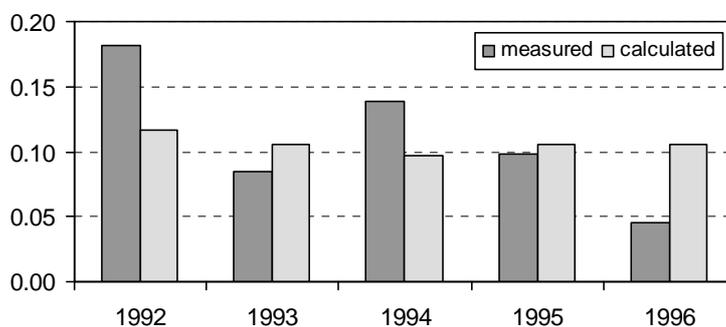


Figure 2.2. Mean calculated and measured PCB air concentrations for Hazelrigg, ng/m³

2.2. The analysis of B[a]P accumulation

Figure 2.3 demonstrates the dynamics of B[a]P emission, concentration and deposition variations in Great Britain during 1970-97. In particular figure 2.3.a illustrates B[a]P emission variations [Pacyna et al., 1999]. As follows from the plot during 1970-97 the emission is continuously lowering down. By 1997 the reduction is as much as 12 times.

Figures 2.3.b-d illustrate plots of B[a]P mean annual concentrations in air, vegetation, soil, and sea water for Great Britain. Concentration variations in these compartments agree with emission variations. During the considered period concentrations decrease in soil in 5 times, in sea in 5.7 times, in vegetation in 9.1 times and in air in 9.2 times. Consequently most inertial media are soil and sea water and least inertial are air and vegetation. Some annual variations in these compartments are conditioned by meteorological situation (annual variations of mean temperature, precipitation amount, direction of wind and its speed, and atmospheric stability). As evident from figure 2.3.b concentrations in air and vegetation correlate with each other though the latter is less subject to variation conditioned by meteorology.

A distinctive feature of sea concentration dynamics (fig.2.3.c) is its better correlation with emission dynamics compared to Europe as a whole. It is conditioned by the closeness of water washing Great Britain (the Atlantic, North Sea and English Channel) to major European emission sources.

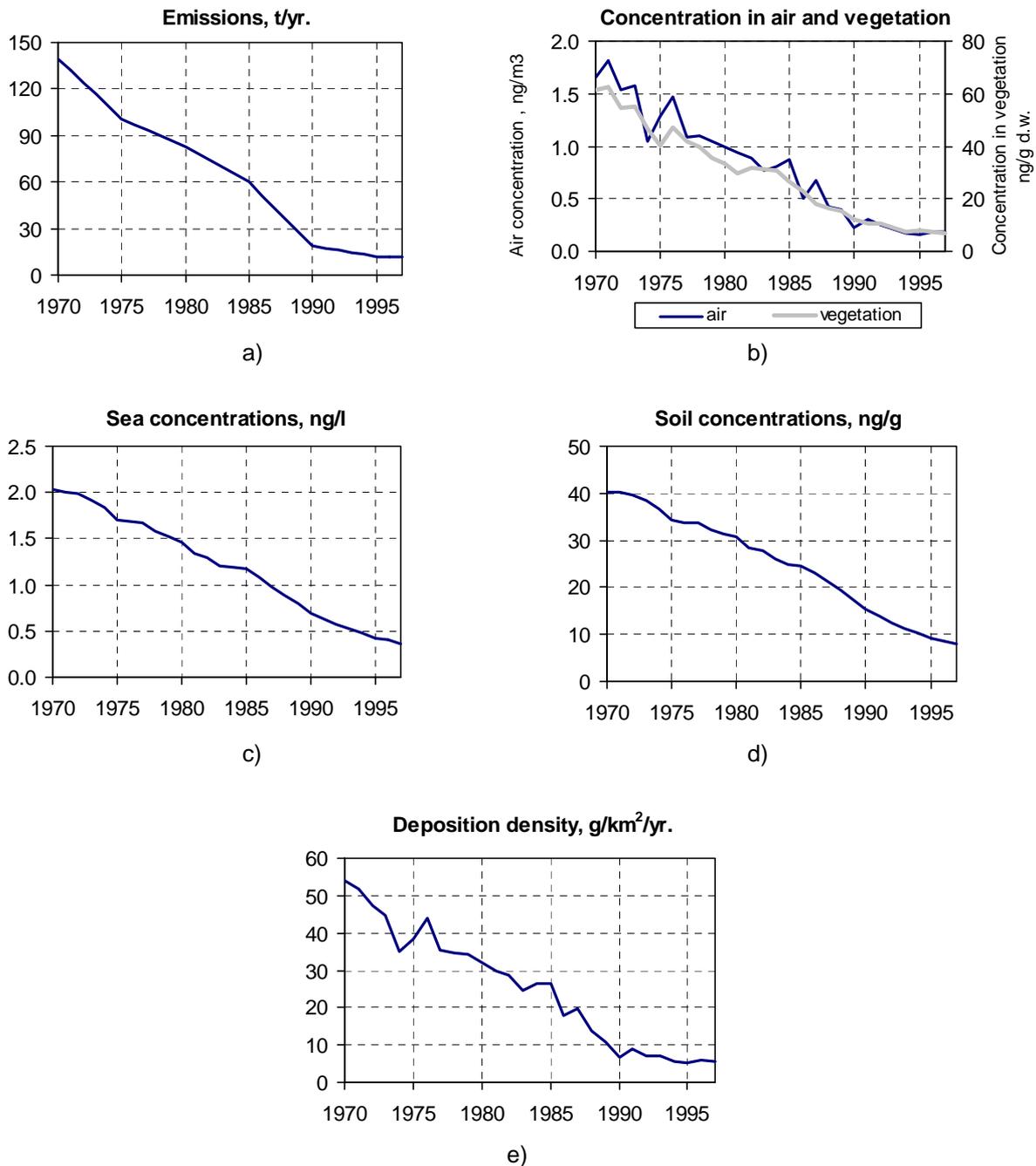


Figure 2.3. Dynamics of emission and concentration variations in compartments and B[a]P deposition densities in Great Britain in comparison with emission reduction during 1970-97

Table 2.2 illustrates the comparison of measured and calculated values of B[a]P air concentrations for Great Britain. Averaged over a year monitoring data [Coleman *et al.*, 1998] of 7 stations are given for the period of 1991-97. The ratio of measured to calculated values varies from 0.4 for Hazelrigg (1995) to 7.7 for Cardiff (1991). On the average measurement results exceed the calculated ones in 1.7 times. Like in the case of PCB the highest discrepancy between measured and calculated values are observed for big cities

(Cardiff, Manchester). The correlation coefficient between measured and calculated data is 0.47.

Table 2.2. Comparison of measured and calculated B[a]P air concentrations for Great Britain

Location	Year	Measured, ng/m ³	Calculated, ng/m ³	meas/calc
Cardiff	1991	2.26	0.29	7.7
Cardiff	1992	0.94	0.23	4.2
Cardiff	1993	0.34	0.20	1.7
Hazelrigg	1992	0.26	0.38	0.7
Hazelrigg	1993	0.36	0.31	1.1
Hazelrigg	1994	0.35	0.26	1.3
Hazelrigg	1995	0.11	0.25	0.4
Hazelrigg	1996	0.21	0.25	0.8
Hazelrigg	1997	0.16	0.28	0.6
High Muffles	1997	0.13	0.28	0.5
London	1991	1.11	0.57	1.9
London	1992	0.57	0.53	1.1
London	1993	0.75	0.43	1.7
London	1994	0.27	0.37	0.7
London	1995	0.40	0.34	1.2
London	1996	0.33	0.34	1.0
London	1997	0.29	0.33	0.9
Manchester	1991	1.92	0.62	3.1
Manchester	1992	1.56	0.51	3.1
Manchester	1993	0.78	0.43	1.8
Manchester	1994	1.04	0.34	3.1
Manchester	1995	0.37	0.37	1.0
Manchester	1996	0.47	0.36	1.3
Manchester	1997	0.82	0.36	2.2
Middlesbrough	1992	0.68	0.38	1.8
Middlesbrough	1993	0.57	0.31	1.8
Middlesbrough	1994	0.40	0.26	1.5
Middlesbrough	1995	0.52	0.25	2.1
Middlesbrough	1996	0.42	0.25	1.6
Middlesbrough	1997	0.31	0.28	1.1
Stevenage	1991	0.69	0.62	1.1
Stevenage	1992	0.70	0.51	1.4
Stoke Ferry	1997	0.14	0.28	0.5
Mean		0.61	0.36	1.7
Correlation				0.47

The analysis of table 2.2 indicates that calculated values for B[a]P agree well enough with measurements carried out at stations located sufficiently far from powerful emission sources

and they are 3 times lower than measurements in big industrial cities (Cardiff, London, Manchester). On the other hand, measurement results in industrial centers are also 2-5 times higher than in rural regions. The comparison of calculated and measured values for Hazelrigg is illustrated in figure 2.4. It is evident that in the most part calculated values agree well with measurements (within a factor of 2). The discrepancy can be explained by inaccurate emission data for Great Britain.

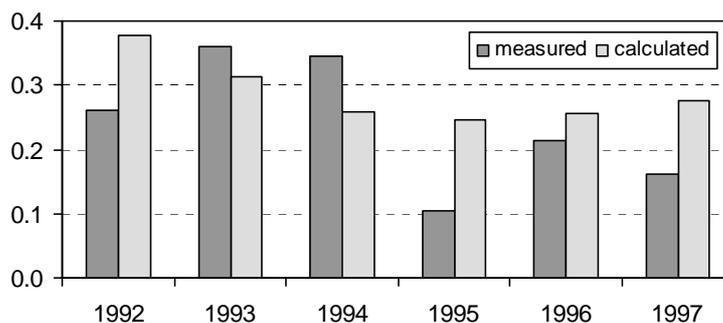


Figure 2.4. Mean calculated and measured B[a]P air concentrations for Hazelrigg, ng/m³

2.3. Analysis of γ -HCH accumulation

Figure 2.5 demonstrates plots of γ -HCH deposition densities in Great Britain and its concentrations in different compartments during 1970-97 in comparison with emission dynamics. Again emission data are taken from POPCYCLING-Baltic project [Pacyna et al., 1999] and emissions in 1996 and 1997 is taken as in 1995. Figure 2.5.a demonstrates the emission dynamics in Great Britain during this period. As seen from the plot, from 1970 to 1997 emission has no tendency to steady reduction as it was observed for the above considered pollutants. By the end of the calculation period it practically reaches the initial level.

Figure 2.5.b illustrates plots of γ -HCH mean annual concentration variations in air and vegetation for Great Britain. As evident from the plot like in the case of values averaged over the whole EMEP grid these concentrations repeat emission variations. Besides the plots demonstrate that concentrations in air and vegetation are well correlated with each other. It means that the equilibrium between air and vegetation is set up quite quickly.

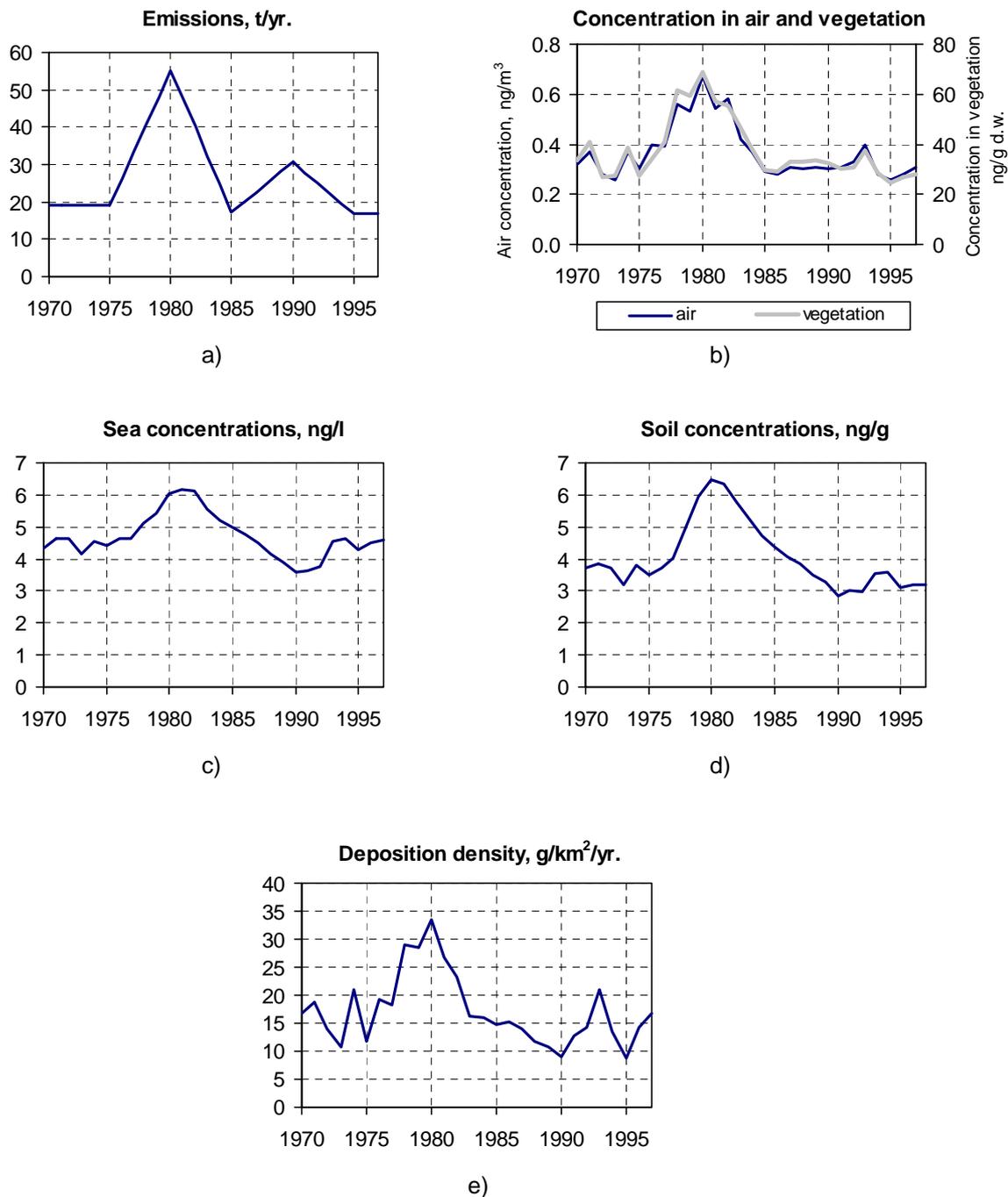


Figure 2.5. Dynamics of concentration variations in compartments and γ -HCH deposition density in Great Britain in comparison with emission dynamics during 1970-97

Figures 2.5.c,d show plots of γ -HCH concentration variations in soil and sea water. Like atmospheric concentrations the dynamics of concentration variations in soil and sea water in Great Britain repeat the dynamics of these variations for the whole EMEP domain. The marine environment is more inertial than soil. It is confirmed by the fact that maximum concentrations are reached with some delay compared to maximum emissions. Concentration variations in waters washing Great Britain, however, better correlate with

emission variations than for Europe on the whole. It is explained by the closeness of waters washing Great Britain (the Atlantic coast, North Sea, English Channel) to major European sources.

Figure 2.5.e presents plots of γ -HCH total deposition density over Great Britain. As follows from the plot deposition densities repeat emission variations although they are subjected to variations of meteorological conditions. The 10-times decrease of deposition density by the end of the calculated period is in agreement with the emission decrease. The analysis of dry deposition seasonal variations shows γ -HCH re-emission in the second half of the year (see volume 2 of the report).

Table 2.3 illustrates the comparison of γ -HCH calculated and measured data at 2 stations in Great Britain [Coleman *et al.*, 1998]. The data are averaged over 1997. The agreement between them is within a factor of 2.

Table 2.3. Comparison of measured and calculated γ -HCH air concentrations in Great Britain in 1997

Station	Measured, ng/m ³	Calculated, ng/m ³	Meas./Calc.
Stoke Ferry	1.37	0.68	2.02
Hazelrigg	0.34	0.28	1.23

In conclusion it should be mentioned that the obtained results in the most part are consistent with data of [Harner *et al.*, 1995]. In future it is planned to carry out similar analysis for other European countries in collaboration with interested national experts.

Chapter 3

Analysis of available POP measurement data for 1998¹

Introduction

Persistent organic components (POPs) were not a part of EMEP's monitoring program before 1999. Co-operation concerning POPs between EMEP and other international programs was, however, extended from 1996. As a first step CCC developed a database and the participants were asked to deliver data from existing POP networks (e.g. OSPAR, HELCOM, AMAP). The following POPs are included in this report: PCBs, γ -HCH and benzo[a]pyrene.

3.1. The measurement sites

The location of the measurement sites for which there are data reported for POPs for 1998 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.	
Czech Rep.	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
Iceland	IS91	Stórhöfði	63°24'N	20°17'W	118
Ireland	IE2	Turlough Hill	53°02'N	6°24'W	420
Norway	NO42	Spitzbergen, Zeppelinfjell	78°54'N	11°53'E	474
	NO99	Lista	58°06'N	6°34'E	13

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.

¹ This section was written by Torunn Berg and Rita Larsen (CCC)

3.2. *Summaries of the data*

Annual summaries of POPs in precipitation and air are given in Annex A and B, 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 (further details in 3.3)
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 *T.Berg and A.-G.Hjellbrekke [1998]*.

Monthly averages of POPs are given in Annex C and D. 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

3.3. 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: $\mu\text{g}/\text{m}^2$ 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. A few data with extremely high detection limits are not included in the report (Precipitation data from Ireland).

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 Islandic 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.

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 (fig. 3.1).

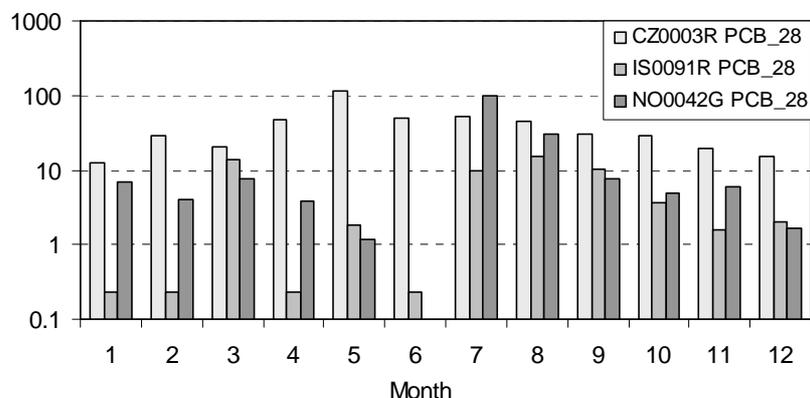


Figure 3.1. Concentrations of PCB-28 in air + aerosol at three EMEP stations, pg/m³

Benzo[a]pyrene (also other PAHs) is rapidly destroyed by UV. In the absence of local sources, therefore, a pronounced seasonal trend is to be expected, which is seen especially for CZ03. Different methods are used for the different stations, and the results from LT15 are e.g. given as deposition rates, $\mu\text{g}/\text{m}^2$ 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 in 2001. An Expert Meeting on measurements of POPs in air and precipitation was, however, held at Lillehammer, Norway in November 1997. The Expert Meeting gave technical recommendations on measurements of POP in air and precipitation, and on the quality assurance of the POP measurements. A summary from this meeting is published in *A.Lükewille* [1998].

Chapter 4

Emissions¹

In accordance with the Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on persistent organic pollutants the pollutants are divided into three groups on each the Parties take certain obligations:

1. To eliminate the production and use of aldrin, chlordane, chlordecone, DDT, dieldrin, endrin, heptachlor, hexabromobiphenyl, hexachlorobenzene, mirex, PCB, toxaphene.
2. To restrict use of DDT, HCH, PCB.
3. To reduce annual emissions of PAH (for the purpose of emission inventories four indicator compounds shall be used: benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene and indeno[1,2,3-cd]pyrene), dioxins/furans, hexachlorobenzene.

According to the Protocol each Party shall develop and maintain emission inventories for the substances listed in item 3 and shall collect available information on the production and sales of substances listed in items 1 and 2.

Official data on POP emissions and expert estimates

Persistent organic pollutants are capable to be accumulated in different environmental compartments and inflict a negative impact on nature and man. In this connection it is desirable to track their income to the atmosphere and their fate for a long period of time. At present it became possible due to estimates of selected POP emissions for 1970, 1975, 1980, 1985, 1990 and 1995 made under POPCYCLING-Baltic project [*Pacyna et al.*, 1999]. In this work in addition to emission estimates used emission factors in their historical evolution are presented, i.e. their variations in different sectors of human activity during 25

¹ This section was written by Mrs. O.Afinogenova and Ms. O.Rozovskaya (MSC-EAST)

years are shown. Contributions of each sector (Selected Nomenclature for sources of Air Pollution, level 1) to total emissions during the period considered are also estimated.

Since the official emission data do not cover such a long period of time and they are not available for all European countries model calculations of polychlorinated biphenyls, polyaromatic hydrocarbons (on the example of benzo[a]pyrene) and γ -hexachlorocyclohexane were carried out on the basis of expert estimates [*Pacyna et al.*, 1999] (between basic years a linear interpolation of emissions was used). For 1996 and 1997 we used the emission of 1995.

It should be noted that the submission of official emission data is improved. Further we present both official data submitted by countries and available in MSC-W emission database as of early April 2000 and expert estimates used for modelling.

Polychlorinated biphenyls (PCB)

Polychlorinated biphenyls are derivatives of biphenyl in which one or more hydrogen atoms are substituted for chlorine. There are 209 isomers of PCB which have different physical-chemical properties and different toxicity. PCB mixtures with different isomer content are widely used in industry. These mixtures are chemically highly inertial, fire-resistant, have dielectric properties and low viscosity. The commercial production and use of PCB began in 1929 and continued to the beginning of the 70s. They were used both in open systems (lubricants, softeners, impregnating agents, etc.) and in closed systems (capacitors, transformers, hydraulic systems, heat exchangers).

According to estimates of a number of experts [*Berdowski et al.*, 1997; *Pacyna et al.*, 1999] early in the 90s main emission of polychlorinated biphenyls (more than 90% for Europe as a whole) is caused by the leakage from electrical equipment where PCB mixtures are used. Besides PCB emissions take place in high temperature processes (combustion of fuel, municipal and industrial wastes, production of sinter, secondary non-ferrous metallurgy etc.). Re-emission from previously polluted compartments is also a source of PCB.

Official data on PCB emissions at least for one year from 1985 to 1998 were submitted by 11 countries (table 4.1).

Table 4.1. Official data of PCB emissions, kg/yr.

Country	UN/ECE reported official emission data									
	1985	1990	1991	1992	1993	1994	1995	1996	1997	1998
Bulgaria		258					382	262	282	253
Finland					5300	1100	15800			
Czech Rep.		773	772	741	644	630	623	555	448	458
France		56	63	63	64	62	58	58	56	58
Germany		43579				30894				
Hungary	169	135	120	108	106	105	101	97.7	97.6	92.2
Lithuania									12.5	14.2
Netherlands		0				0.283	0.015			
Poland		2373					2338	2342	2386	2312
Slovakia		161					139		137	
UK		6976	6400	5901	5407	4845	4290	3749	3247	2748

As it was indicated above according to expert estimates by the early 90s main contribution to PCB emission is leakage from electrical equipment whereas among countries which submitted official data only Germany and Poland accounted this constituent. It is shown in splitting of total emissions over sectors submitted by countries.

Expert estimates of PCB emissions for 1970, 1975, 1980, 1985 1990 and 1995 used in model calculations are illustrated in table 4.2 [*Pacyna et al.*, 1999].

According to expert estimates during 1970-95 PCB total emission in Europe decreased in 15 times.

The comparison of available official data and expert estimates indicate that for the majority of countries expert estimates are higher than official data and in some cases this excess is essential (the Netherlands, France). As it was already mentioned the discrepancy is mainly connected with leakage from electrical equipment in which PCB mixtures are used.

Gridded emission distribution over the EMEP domain with spatial resolution 150x150 km² for 1997 is illustrated in figure 4.1.

Table 4.2. Expert estimates of PCB emissions in European countries, kg/yr.

Country	1970	1975	1980	1985	1990	1995
Albania	2	2	7	5	143	146
Armenia*	48	56	58	59	204	186
Austria	38010	30893	15465	15384	1368	1403
Azerbaijan*	87	101	104	106	369	336
Belarus	58	71	77	83	566	516
Belgium	49507	40307	20414	20223	1835	1829
Bosnia&Herzegovina *	26	33	36	53	240	231
Bulgaria	186	204	214	216	578	518
Croatia *	27	34	37	54	248	238
Czech Republic	393*	436*	488*	488*	886	766
Denmark	24971	20546	10522	10387	1011	1026
Estonia	86	118	127	113	175	135
Finland	23451	19283	9852	9983	2670	2721
France	258091	214783	110217	111732	19958	20347
Georgia*	81	94	97	99	344	312
Germany	402160	323809	164226	160185	43256	42462
Greece	17936	18589	19732	20238	309	221
Hungary	180	185	195	182	592	554
Iceland	1025	872	456	486	47	49
Ireland	6013	6477	6922	7152	86	77
Italy	270757	224464	114592	114619	6165	6054
Kazakhstan * **	51	59	61	62	215	197
Latvia	28	25	23	21	135	126
Lithuania	24	26	43	42	200	178
Luxembourg	2060	1810	896	858	130	127
Netherlands	65823	55127	28618	29231	257	250
Norway	19598	16203	8259	8365	402	408
Poland	637	791	978	1007	2488	2374
Portugal	18171	17903	19731	20103	563	557
Republic of Moldova	22	36	41	44	225	206
Romania	194	258	328	355	1290	1218
Russian Federation *	1826	2126	2185	2223	7738	7042
Slovakia	114*	133*	140*	141*	346	327
Slovenia *	14	18	19	28	128	123
Spain	170541	143581	75919	77692	8705	8721
Sweden	41163	33743	17029	16960	1991	2022
Switzerland	31662	25916	12966	13212	1686	1742
The FYR of Macedonia *	11	14	15	22	101	97
Ukraine	1181	1181	1210	1372	3551	3043
United Kingdom	282443	228002	114801	114675	3831	3706
Yugoslavia *	66	82	89	131	597	574
Total, t/y	1729	1428	757	758	116	113

* estimated on the basis of spatial emission distribution [*Pacyna et al.*, 1999] in former Czechoslovakia, USSR and Socialist Federal Republic of Yugoslavia

** part of Kazakhstan, Uzbekistan and Turkmenistan covered by the EMEP grid

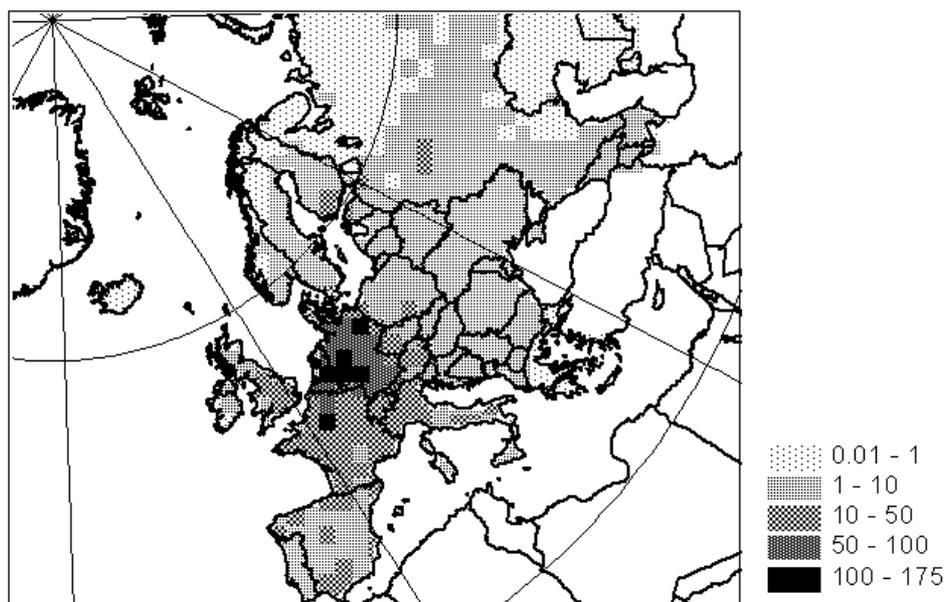


Figure 4.1. PCB emission spatial distribution for 1997, g/km²/yr.

As seen from figure 4.1 in 1997 the highest emission density takes place in Germany. The total emission in Germany amounts to 40% of that in Europe.

Polycyclic aromatic hydrocarbons (PAH)

In this report we present modelling results for one of PAH indicators - benzo[a]pyrene (B[a]P). Benzo[a]pyrene is mainly formed at incomplete combustion of fuel and at some technological processes.

Official data on polycyclic aromatic hydrocarbon emissions at least for one year from 1985 to 1998 were submitted by 21 countries. However, only three of them - Denmark, Lithuania and Poland - submitted emission data separately for each of the four indicator compounds. Russia submitted data on benzo[a]pyrene only. The official data of the 4 countries are illustrated in table 4.3.

Table 4.3. Official data on PAH emissions, t/yr.

Country	UN/ECE reported official emission data								
	1990	1991	1992	1993	1994	1995	1996	1997	1998
<i>Denmark</i> ¹					9.85	10.4	11.4	11.2	10.5
benzo[a]pyrene					2.659	2.801	3.101	3.044	2.859
benzo[b]fluoranthene					3.595	3.737	4.144	4.065	3.829
benzo[k]fluoranthene					1.347	1.372	1.519	1.492	1.419
indeno[1,2,3-cd]pyrene					2.247	2.45	2.611	2.55	2.383
<i>Lithuania</i> ¹								71.2	53.1
benzo[a]pyrene									14.155
benzo[b]fluoranthene									10.772
benzo[k]fluoranthene									10.350
indeno[1,2,3-cd]pyrene									17.859
<i>Poland</i>	372 ²					536 ²	523 ²	495 ²	181 ¹
benzo[a]pyrene									54.4
benzo[b]fluoranthene									55.4
benzo[k]fluoranthene									15.8
indeno[1,2,3-cd]pyrene									55.5
<i>Russia</i> (B[a]P only)	18.3	17.3	15.6	15.3	15.5	15.3	15	15	14.7

¹ - Total of 4 substances² - 6 of Borneff

According to the official data benzo[a]pyrene emission in Russia in 1998 is almost the same as in Lithuania and 3.7 times lower than in Poland. For the transparency of the submitted data it should be useful to compare calculation methodology.

Expert estimates of benzo[a]pyrene emissions [*Pacyna et al.*, 1999] used in model calculations are demonstrated in table 4.4.

According to expert estimates total benzo[a]pyrene emissions in Europe decrease in 2 times during 1970-95.

The comparison of benzo[a]pyrene emissions expert estimates with official data manifests that official data for Denmark are 2 times higher than expert estimates (1995), for Lithuania – 6 times (official data for 1998 were compared with estimates for 1995). On the opposite for Russia expert estimates exceed official data in 15 times. For Poland expert estimates essentially agree with official data.

Gridded benzo[a]pyrene emission distribution over the EMEP domain with spatial resolution 150x150 km² for 1997 is demonstrated in figure 4.2.

Table 4.4. Expert estimates of B[a]P emissions in European countries, t/yr.

Country	1970	1975	1980	1985	1990	1995
Albania	0.069	0.075	2.329	0.135	0.111	0.221
Armenia*	6.591	7.607	8.275	9.520	10.019	6.028
Austria	20.895	13.580	12.491	7.032	4.543	6.112
Azerbaijan*	11.906	13.741	14.947	17.196	18.097	10.888
Belarus	6.634	6.295	6.003	6.591	9.256	5.246
Belgium	29.215	16.744	11.829	11.623	4.361	3.352
Bosnia&Herzegovina *	3.391	4.848	5.436	8.217	8.809	4.520
Bulgaria	6.427	6.591	6.780	8.082	7.570	6.707
Croatia *	3.500	5.004	5.611	8.481	9.092	4.665
Czech Republic	17.305*	17.927*	22.340*	21.808*	18.669	14.258
Denmark	1.801	1.412	1.976	2.732	1.755	1.435
Estonia	0.453	0.435	0.439	0.466	0.432	0.366
Finland	6.516	5.979	6.262	6.106	6.496	6.881
France	112.422	76.235	64.949	45.470	30.175	26.397
Georgia*	11.098	12.808	13.933	16.029	16.868	10.149
Germany	224.376	198.570	199.143	170.937	191.692	26.397
Greece	13.500	10.665	9.627	5.576	5.690	2.889
Hungary	18.202	18.297	19.751	19.879	16.708	10.254
Ireland	4.342	2.401	5.202	6.409	1.766	1.237
Italy	30.356	26.530	26.508	18.389	15.203	13.904
Kazakhstan * **	6.966	8.039	8.745	10.061	10.587	6.370
Latvia	3.863	3.291	3.127	2.830	2.654	2.997
Lithuania	5.922	4.675	4.078	3.308	3.163	2.200
Luxembourg	0.349	0.295	0.262	0.257	0.211	0.244
Netherlands	23.774	16.596	13.355	6.599	4.737	2.291
Norway	43.937	31.638	28.166	12.773	10.614	2.235
Poland	68.107	73.787	98.974	88.358	57.867	65.159
Portugal	2.090	1.203	1.202	1.497	1.501	1.639
Republic of Moldova	0.969	1.527	1.671	1.813	1.653	1.014
Romania	22.111	31.329	38.189	40.791	27.903	18.982
Russian Federation*	249.965	288.473	313.809	361.027	379.932	228.594
Slovakia	9.566*	9.686*	10.103*	9.579*	9.811	6.705
Slovenia*	1.808	2.585	2.899	4.381	4.697	2.410
Spain	23.598	22.779	24.522	15.475	12.417	9.607
Sweden	13.429	12.004	11.259	11.322	8.740	6.777
Switzerland	9.880	6.014	5.466	3.084	2.357	1.653
The FYR of Macedonia*	1.428	2.041	2.289	3.459	3.708	1.903
Ukraine	87.241	87.241	91.898	94.184	91.241	51.824
United Kingdom	140.232	101.432	82.828	59.881	18.457	11.993
Yugoslavia*	8.434	12.058	13.521	20.437	21.908	11.242
Total	1253	1162	1200	1142	1051	598

* estimated on the basis of spatial emission distribution [*Pacyna et al.*, 1999] in former Czechoslovakia, USSR and Socialist Federal Republic of Yugoslavia

** part of Kazakhstan, Uzbekistan and Turkmenistan covered by the EMEP grid

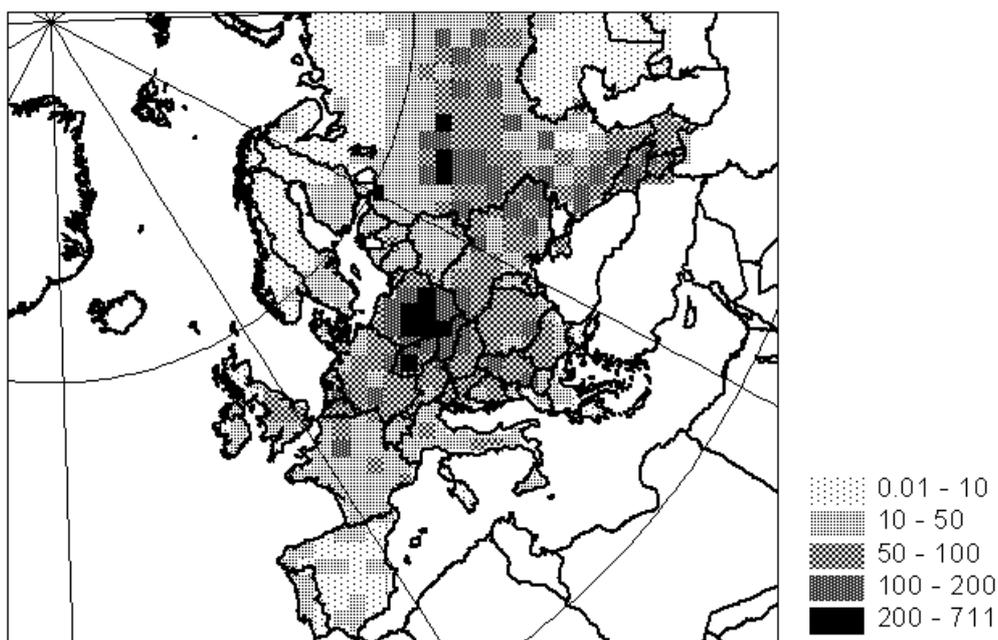


Figure 4.2. Spatial distribution of benzo[a]pyrene emission in 1997, g/km²/yr.

As seen from figure 4.2 the maximum benzo[a]pyrene emission density is characteristic of countries located in Central and Eastern Europe. The contribution of Russia to total European emission amounts to 38%, of Poland – 11%, and of the Ukraine – 9%.

Hexachlorocyclohexane (HCH)

Hexachlorocyclohexane is an insecticide widely used in the world since the 40s. There are eight isomers of HCH. γ -HCH has the best insecticide properties. Up to the end of the 1970s the main source of γ -HCH was use of technical HCH containing different isomers in these or those proportions. Later lindane (HCH containing at least 99% of γ -isomer) became the main source of this isomer [Breivik *et al.*, 1999].

Official data on HCH emissions at least for one year during the period from 1990 to 1998 were submitted by 7 countries (Austria and Moldova reported data on HCH use). These data are contained in table 4.5.

Table 4.5. Official data on HCH emissions, kg/yr.

Country	UN/ECE reported official emission data									
	1980	1990	1991	1992	1993	1994	1995	1996	1997	1998
Croatia		9400						12800	3100	5000
Denmark						61				0
Germany						15000				
Hungary		9281	60	12	462	798	1650	2400	31	22
Netherlands		0		0		0	0	0	0	
Russia		923000								
UK		99023	85335	74016	64604	56733	50114	44518	39761	35695
Austria (use)						12000	8056	8640	2324	0
Moldova (use)										
HCH lindane	5635000	73800 800	72000 150	50000	57000	36500				

The data testifies that in principal there is a tendency of HCH emission reduction.

Expert estimates of γ -HCH use and emissions in European countries for every year from 1970 to 1996 were made by *J.Pacyna et al.* [1999]. These estimates were used in model calculations. Table 4.6 illustrates emission data for 1970, 1975, 1980, 1985, 1990 and 1996.

According to expert estimates total γ -HCH emission in Europe decreases in 3.5 times during 1970-96.

Since the share of γ -isomer is unknown in official data on HCH emissions, in the comparison of them with expert estimates it is possible to indicate only extreme cases:

- according to expert estimates emission exists but according to official data it is absent (the Netherlands).
- according to expert estimates emission is absent but according to official data it is present (Germany, 1994).

Gridded γ -HCH emission distribution over the EMEP domain with spatial resolution 150x150 km² for 1997 is demonstrated in figure 4.3.

Table 4.6. Expert estimates of γ -HCH emissions in European countries, kg/yr.

Country	1970	1975	1980	1985	1990	1996
Albania	7661	7661	6097	4433	2228	463
Armenia*	2714	2714	2111	1037	69	30
Austria	2105	3741	5915	6213	3392	0
Azerbaijan*	10252	10252	7974	3916	259	113
Belarus	77182	77182	60030	35688	4645	4645
Belgium	21319	21319	19075	23049	15411	15890
Bosnia&Herzegovina	31703*	31703*	17355*	7332*	6260	567
Bulgaria	22226	22226	22141	17256	5152	0
Croatia	49914*	49914*	27324*	11544*	2582	472
Cyprus	543	543	476	272	91	91
Czech Republic	5608*	3681*	58*	58*	22	319
Denmark	3036	3290	4348	2924	2925	0
Estonia	169	169	164	156	7	3
Finland	818	1157	4970	1840	0	0
France	273780	273780	322228	294072	270456	560000
Georgia*	8667	8667	6741	3310	219	95
Germany	25868	52604	111447	49608	27662	0
Greece	47286	47286	67599	37694	2952	5863
Hungary	630106	91287	1828	11508	1435	2870
Iceland	0	40	35	39	73	109
Ireland	2132	2132	2279	2513	1773	2167
Italy	322113	426810	551635	568015	282218	2230
Kazakhstan **	40550	40550	31539	15488	1025	446
Latvia	615	615	319	104	13	3
Lithuania	1609	1609	1251	744	97	2
Luxembourg	273	273	317	191	135	151
Netherlands	38619	38619	32686	36589	3814	6307
Norway	380	380	1104	1178	130	0
Poland	22790	16817	7095	2773	446	280
Portugal	1875	1295	1610	2344	3390	13189
Republic of Moldova	13876	13876	10792	6416	835	364
Romania	46220	46220	20223	6351	3693	2035
Russian Federation*	540047	540047	420036	206267	13652	5941
Slovakia	2379*	1562*	25*	25*	304	581
Slovenia	9854*	9854*	5394*	2279*	294	169
Spain	98958	98958	221668	275160	180146	122909
Sweden	4589	1995	3115	1575	2668	1101
Switzerland	3772	3772	2753	1531	16	159
The FYR of Macedonia	19522*	19522*	10687*	4515*	2298	69
Ukraine	277142	277142	215555	105853	12877	9047
United Kingdom	19214	19214	56228	17240	31083	30039
Yugoslavia	87799*	87799*	48064*	20305*	18690	1694
Total, t/y	2775	2358	2332	1789	905	790

* estimated on the basis of spatial emission distribution [*Pacyna et al.*, 1999] in former Czechoslovakia, USSR and Socialist Federal Republic of Yugoslavia

** part of Kazakhstan, Uzbekistan and Turkmenistan covered by the EMEP grid

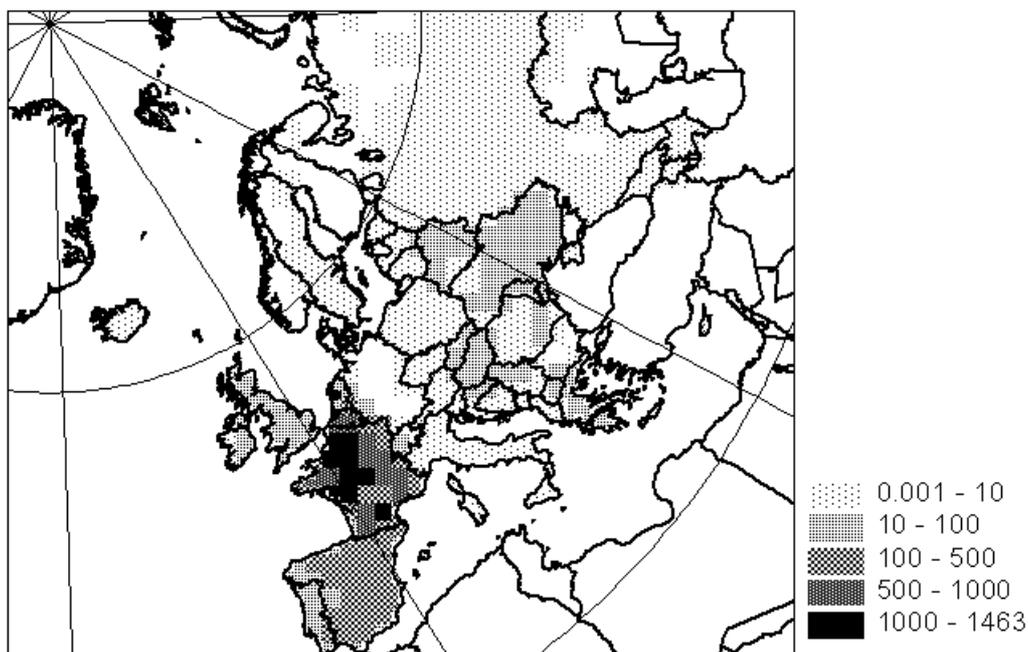


Figure 4.3. Spatial distribution of γ -HCH emission in 1997, g/km²/yr.

As seen from figure 4.3 the highest density takes place in France. The contribution of this country to total European emission is 71% in 1997.

Emission distribution with height

At present there are no official data on the emission distribution with height (below 100 m and above 100 m). In this report the same distribution with height as in MSC-E report for 1999 [Pekar *et al.*, 1999] based on expert estimates was used:

- PCB and γ -HCH – the whole emission enters the atmosphere below 100 m;
- B[a]P - 90% of emission enters the atmosphere below 100 m and 10% - above 100 m.

Emission seasonal variations

The emission input to the atmosphere was assumed as follows [Pekar *et al.*, 1999]:

- PCB - uniform round the year;
- B[a]P sinusoid with amplitude of 20% and with maximum in winter;
- γ -HCH - 10% of total emission - in February, 15% - in March and by 25% - in April, May and June each.

Uncertainty of emission estimates

The uncertainty of POP emission estimates are conditioned by a number of reasons: a wide range of emission factor values even for one and the same kind of activity, incomplete information on fuel consumption, product output, on the quantity and method of pesticide application in different countries, etc.

In MSC-E report for 1999 [Pekar *et al.*, 1999] model calculations of PCB, B[a]P and lindane transport were based on official data and estimates made by J.Berdowski *et al.* for 1990 [Berdowski *et al.*, 1997]. J.Berdowski *et al.* estimated the uncertainty for PCB, PAH and pesticide emission estimates within factors of 2-5 [Berdowski *et al.*, 1997]. Emission estimates of J.Pacyna *et al.* [1999] used in this report cover 25-year period and in this case the uncertainties can be higher.

Discrepancies (in some cases rather appreciable) between used emission estimates [Pacyna *et al.*, 1999] and official data testify to a necessity of reporting by countries more complete information on emissions (estimation methodology used, emission factors, total emission splitting by sectors, etc.). It would be beneficial for the analysis of the quality of both official data and expert estimates.

In EMEP persistent organic pollutant study is a new direction of activity and there are a great number of open questions concerning POP input to the atmosphere, transport, accumulation in different compartments, physical-chemical properties, etc. As far as emission inventory is concerned the collection and analysis of estimation methodology of POP emissions and emission factors considering peculiarities of technologies used in different countries are likely to be important. Further efforts aimed at improving and supplying additional information on POP to the Guidebook on Emission Inventory will be very beneficial.

Conclusions

1. In accordance with the EMEP working plan for 2000 and recommendations of scientific workshops the work on the development and modification of the POP long-range transport model was performed. In particular, the block of atmosphere/sea exchange was completed by description of POP transport with sea currents, and dry deposition scheme over forests was refined.
2. The examination of POP physical-chemical properties was continued. On the basis of these investigations the appropriate model parameterizations for PCB, B[a]P, γ -HCH were refined.
3. Collection and analysis of POP concentration measurements in different media obtained both at the EMEP monitoring network and in the framework of national measurement campaigns and programmes was performed.
4. The assessment of contamination levels in different environmental compartments was carried out for different European countries. On the basis of the obtained data most of the European countries can be divided into two groups depending on the relation between mean emission densities and air concentrations. Countries for which air concentration level is defined mainly by their own POP emissions form the first group. For countries of the second group air pollution is caused predominantly by the transboundary transport from emission sources located outside their territories. The comparison of emission and deposition data over countries made it possible to obtain preliminary estimates of the exported POP amount. More precise estimates can be obtained by calculation of "country-to-country" matrix.
5. A more detailed analysis of POP long-range transport and accumulation was carried out for some countries. Namely:
 - The analysis of POP long-term accumulation and behaviour in different media was carried out with Great Britain as an example and a number of features peculiar to a given country was revealed.

- The comparison of PCB, B[a]P, and γ -HCH calculation and measurement results obtained on the national basis was performed in collaboration with national experts. In particular the comparison manifested a good agreement between B[a]P calculation and measurement results in Lithuania.

Similar investigations are planned in future for other European countries.

6. The comparison of calculation results with available measurement data manifested that among the considered pollutants the model describes B[a]P long-range transport in the most reliable way. Calculated values for PCB and γ -HCH are as a rule less than measured ones. It is supposed that taking into account emission sources located outside the EMEP grid will improve the agreement between calculation and measurement data. More complete set of data on all the media is required for the detailed model verification.

7. At further stages of investigation it is supposed:

- To carry out model assessment of the transport and contamination levels for HCB and selected congeners of PCDD/Fs.
- To refine the model block describing exchange processes between the atmosphere, soil, and vegetation taking into account POP degradation in vegetation.
- To develop an operational version of B[a]P long-range transport model with spatial resolution 50x50 km and to assess transboundary fluxes (using “country-to-country” scheme).
- To clarify the influence of different emission scenarios on the contamination level dynamics for different media in the European region.
- To assess the influence of external sources of semi-volatile POPs upon European contamination level.

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Annex A

Annual statistics for POPs in air for 1998

CS0003R KOSETICE CZECH REPUBLIC

January 1998 - December 1998

Component	Arit	Arit mean	Geom sd	Geom mean	Min sd	50%	Max	Num bel	Num samp	Samp flag	QA flag
benzo[a]pyrene	0.320	0.566	0.085	5.945	0.01	0.09	3.14	16	52	W	
γ -HCH	127.235	154.569	59.457	4.164	2	65	699	0	51	W	
PCB-101	26.843	12.556	24.311	1.567	9	24	69	0	51	W	
PCB-118	2.961	1.766	2.456	1.899	1	3	8	14	51	W	
PCB-138	26.510	12.547	23.970	1.574	8	22.5	72	0	51	W	
PCB-180	15.706	8.864	13.885	1.633	4	13	55	0	51	W	
PCB-28	38.706	30.402	30.817	1.934	10	28	164	0	51	W	
PCB-52	36.490	19.884	31.877	1.694	12	32.5	107	0	51	W	

IS0091R STORHOFDI ICELAND

January 1998 - December 1998

Component	Arit	Arit mean	Geom sd	Geom mean	Min sd	50%	Max	Num bel	Num samp	Samp flag	QA flag
γ -HCH	5.258	3.057	3.986	2.545	0.334	4.392	10.187	2	22	W2	
PCB-101	0.223	0.190	0.156	2.371	0.067	0.101	0.586	13	22	W2	
PCB-118	0.112	0.166	0.069	2.255	0.047	0.047	0.712	21	22	W2	
PCB-138	0.073	0.133	0.050	1.790	0.043	0.043	0.667	22	22	W2	
PCB-153	0.135	0.154	0.094	2.171	0.050	0.082	0.667	22	22	W2	
PCB-180	0.194	0.702	0.053	2.593	0.041	0.041	3.337	21	22	W2	
PCB-28	5.278	6.861	1.791	5.453	0.234	2.315	25.907	16	22	W2	
PCB-52	0.318	0.312	0.246	1.877	0.160	0.177	1.430	21	22	W2	

NO0042G SPITZBERGEN NORWAY

January 1998 - December 1998

Component	Arit	Arit mean	Geom sd	Geom mean	Min sd	50%	Max	Num bel	Num samp	Samp flag	QA flag
benzo[a]pyrene	0.012	0.019	0.005	0.004	0.0001	0.005	0.107	22	38	D2	
γ -HCH	37.865	11.436	34.961	1.638	4.280	38.850	60.600	0	47	D2	
PCB-101	1.609	2.181	0.907	2.693	0.260	0.565	10.200	0	39	D2	
PCB-118	1.189	1.555	0.636	2.958	0.120	0.480	6.060	0	39	D2	
PCB-138	1.760	2.228	0.908	3.207	0.130	0.750	9.150	0	39	D2	
PCB-153	1.500	1.870	0.832	2.897	0.150	0.705	7.510	0	39	D2	
PCB-180	0.354	0.397	0.201	3.042	0.010	0.170	1.540	1	39	D2	
PCB-28	15.141	29.407	5.918	3.488	0.870	5.445	137.000	0	39	D2	
PCB-52	3.682	5.528	2.195	2.436	0.580	1.920	27.400	0	39	D2	

NO0099R LISTA NORWAY

January 1998 - December 1998

Component	Arit	Arit mean	Geom sd	Geom mean	Min sd	50%	Max	Num bel	Num samp	Samp flag	QA
γ -HCH	53.263	67.070	34.459	2.350	9.940	28.800	405.000	0	53	W	

Annex B

Annual statistics for POPs in precipitation for 1998

DE0001R WESTERLAND GERMANY

January 1998 - December 1998

Component	W.Mean	Min	Max	Num bel	Num day	Samp flag	QA flag
γ -HCH	6.389	0.95	36	0	12	M	
benzo[a]pyrene	5.799	0.67	62	2	11	M	
PCB-101	0.06	0.03	0.07	12	12	M	
PCB-118	0.07	0.00	0.07	9	12	M	
PCB-118	0.07	0.07	0.07	2	5	M	
PCB-138	0.115	0.07	0.60	10	12	M	
PCB-153	0.106	0.07	0.45	9	12	M	
PCB-28	0.054	0.01	0.07	12	12	M	
PCB-52	0.134	0.07	0.52	11	12	M	
Precip	-	7.5	124.4	0	12	M	
Precip	-	13.2	154.6	0	12	M	
Precip off	-	11	149	0	12	M	

DE0009R ZINGST GERMANY

January 1998 - December 1998

Component	W.Mean	Min	Max	Num bel	Num day	Samp flag	QA flag
benzo[a]pyrene	5.821	1.8	10.6	0	12	M	
PCB-101	0.128	0.03	0.84	8	12	M	
PCB-118	0.102	0.07	0.27	3	5	M	
PCB-138	0.181	0.07	0.86	7	12	M	
PCB-153	0.135	0.07	0.68	7	12	M	
PCB-180	0.125	0.07	0.59	8	12	M	
PCB-28	0.093	0.01	0.50	3	5	M	
PCB-52	0.163	0.07	0.75	9	12	M	
Precip	-	15.2	190	0	12	M	

IS0091R STORHOFDI ICELAND

January 1998 - December 1998

Component	W.Mean	Min	Max	Num bel	Num day	Samp flag	QA flag
γ -HCH	0.179	0.002	3.112	6	24	W2	
PCB-101	0.003	0.001	0.057	21	24	W2	
PCB-118	0.008	0.001	0.143	14	24	W2	
PCB-138	0.007	0.001	0.094	21	24	W2	
PCB-153	0.004	0.001	0.044	22	24	W2	
PCB-180	0.013	0.001	0.061	15	24	W2	
PCB-28	0.025	0.016	0.186	24	24	W2	
PCB-52	0.012	0.003	0.500	23	24	W2	
Precip off	-	5.0	167.0	0	24	W2	

LT0015R PREILA LITHUANIA

January 1998 - December 1998

Component	W.Mean	Min	Max	Num bel	Num day	Samp flag	QA flag
γ-HCH	1.751	1.08	3.55	0	11	M	8

NO0099R LISTA NORWAY

January 1998 - December 1998

Component	W.Mean	Min	Max	Num bel	Num day	Samp flag	QA flag
γ-HCH	5.26	0.5	53.3	0	56	W	
Precip	-	0	43.8	3	56	W	

Annex C

Monthly mean values on data for POP in air for 1998

Station	Component	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	QA
CS0003R	benzo[a]pyrene	1.21	0.54	0.493	0.09	0.022	0.01	0.01	0.01	0.084	0.102	0.353	0.954	
CS0003R	γ-HCH	3.25	40.25	98.25	443.4	200.75	241.5	135.4	93.25	69.4	80	37	17.75	
CS0003R	PCB-101	12	28	19.75	42.4	32.5	28.25	32.4	41.75	23.6	22.75	17	17.25	
CS0003R	PCB-118	1.5	5.25	2.75	4	2.75	2.5	4.6	4.25	2.2	2.75	1	1.5	
CS0003R	PCB-138	11.25	42.5	33.5	39.8	19.75	17.25	35.8	31.5	20.8	23	18	20.75	
CS0003R	PCB-180	7.25	28	21.25	25.2	14	10.75	20.2	13.5	11	11.25	9.5	14.25	
CS0003R	PCB-28	12.25	28.75	20	46.6	114	49.75	52	45	30	28.75	19.25	15	
CS0003R	PCB-52	18	48.75	36.5	53.8	77	44	37	40.25	26.2	23	16.25	15.25	
IS0091R	γ-HCH	2.696	0.345	7.638	7.017	9.695	3.603	4.508	6.761	7.278	7.867	5.008	2.58	
IS0091R	PCB-101	0.133	0.212	0.076	0.067	0.181	0.067	0.353	0.162	0.481	0.579	0.12	0.088	
IS0091R	PCB-118	0.219	0.38	0.068	0.047	0.047	0.199	0.047	0.047	0.073	0.073	0.049	0.047	
IS0091R	PCB-138	0.043	0.355	0.043	0.043	0.043	0.043	0.044	0.043	0.049	0.048	0.043	0.047	
IS0091R	PCB-153	0.303	0.512	0.06	0.05	0.05	0.05	0.133	0.083	0.098	0.096	0.052	0.05	
IS0091R	PCB-180	0.041	1.689	0.041	0.041	0.041	0.041	0.044	0.041	0.07	0.041	0.041	0.041	
IS0091R	PCB-28	0.234	0.234	13.485	0.234	1.844	0.234	9.722	15.092	10.412	3.705	1.57	1.976	
IS0091R	PCB-52	0.177	0.177	0.491	0.177	0.231	0.177	0.177	0.177	0.22	1.045	0.21	0.442	
NO0042G	benzo[a]pyrene	0.041	0.008	0.009	0.005	0.004	0.007	0.022	-999.99	0.004	0.004	0.0003	0.015	
NO0042G	γ-HCH	8.241	7.112	11.410	11.670	2.22	-999.99	17.800	9.797	7.658	10.163	9.602	5.842	
NO0042G	PCB-101	0.825	0.775	1.747	2.338	0.55	-999.99	8.06	2.45	0.654	0.508	0.428	0.43	
NO0042G	PCB-118	0.367	0.373	0.757	1.038	0.29	-999.99	5.587	2.84	0.765	0.438	0.198	0.316	
NO0042G	PCB-138	0.438	0.442	0.74	1.082	0.3	-999.99	7.223	4.78	1.455	0.824	0.257	0.596	
NO0042G	PCB-153	0.535	0.505	0.795	1.195	0.34	-999.99	6.443	3.812	1.092	0.635	0.27	0.458	
NO0042G	PCB-180	0.255	0.223	0.148	0.207	0.07	-999.99	1.203	0.985	0.293	0.208	0.075	0.168	
NO0042G	PCB-28	7.035	4.108	7.642	3.9	1.2	-999.99	100.067	30.75	7.554	4.831	5.92	1.646	
NO0042G	PCB-52	2.003	1.592	2.903	2	0.58	-999.99	19.633	6.512	2.114	1.567	1.87	1.064	
NO0099R	γ-HCH	25.1	30.6	110.535	88.9	93.7	52.775	57.225	36.1	87.067	42.5	22.1	23.871	

Annex D

Monthly mean values on data for POP in precipitation for 1998

Station	Component	Jan	Feb	Mar	Apr	May	Jun	July	Aug	Sep	Oct	Nov	Dec	QA
DE0001R	benzo[a]pyrene	26	62	2.61	0.7	999	0.67	0.67	1.2	1.5	1.7	3	2.8	
DE0001R	γ -HCH	3	36	14	13.9	12.4	8.8	4.5	0.95	2.9	5.3	2.2	2.4	
DE0001R	PCB-101	0.03	0.03	0.03	0.03	0.03	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0001R	PCB-118	0.07	0.07	0.07	0.07	0.07	-999	-999	-999	-999	-999	-999	-999	
DE0001R	PCB-118	0.07	0.07	-999	-999	-999	-999	-999	-999	-999	-999	-999	-999	
DE0001R	PCB-138	0.07	0.07	0.07	0.6	0.25	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0001R	PCB-153	0.07	0.07	0.42	0.26	0.45	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0001R	PCB-28	0.01	0.01	0.01	0.01	0.01	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0001R	PCB-52	0.07	0.07	0.52	0.07	0.07	0.13	0.13	0.13	0.13	0.13	0.13	0.13	
DE0001R	precipitation-amount	42.8	26.1	24.6	70	7.5	45	56.4	109.2	71.2	124.4	100.6	61.8	
DE0001R	precipitation-amount	74	30.3	43.9	70.2	13.2	56.9	101.1	66.5	85.6	154.6	65	50.5	
DE0001R	precipitation-amount	61.9	23.5	30.1	56.1	11	46.4	92	52.6	75.2	149	57.7	47.2	
DE0009R	benzo[a]pyrene	3.1	3.5	2.9	6.7	3.9	9.5	2.1	1.9	1.8	3	10.6	9	
DE0009R	PCB-101	0.03	0.71	0.31	0.11	0.84	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0009R	PCB-118	0.14	0.07	0.07	0.07	0.27	-999	-999	-999	-999	-999	-999	-999	
DE0009R	PCB-138	0.86	0.5	0.42	0.12	0.54	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0009R	PCB-153	0.68	0.16	0.22	0.13	0.16	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0009R	PCB-180	0.59	0.26	0.2	0.07	0.1	0.07	0.07	0.07	0.07	0.07	0.07	0.07	
DE0009R	PCB-28	0.01	0.5	0.01	0.01	0.03	-999	-999	-999	-999	-999	-999	-999	
DE0009R	PCB-52	0.07	0.75	0.07	0.07	0.52	0.13	0.13	0.13	0.13	0.26	0.13	0.13	
DE0009R	precipitation-amount	58.5	36.6	37.8	72	15.2	190	83.2	55.6	53.8	51.6	84	34	
IS0091R	γ -HCH	0.133	0.057	0.341	0.619	0.46	0.344	0.11	0.141	1.111	0.038	0.043	0.003	
IS0091R	PCB-101	0.015	0.001	0.001	0.002	0.002	0.019	0.004	0.002	0.001	0.002	0.002	0.001	
IS0091R	PCB-118	0.044	0.001	0.011	0.005	0.014	0.022	0.021	0.005	0.001	0.002	0.007	0.003	
IS0091R	PCB-138	0.084	0.002	0.01	0.008	0.001	0.01	0.004	0.002	0.001	0.002	0.003	0.009	
IS0091R	PCB-153	0.039	0.002	0.002	0.007	0.001	0.005	0.004	0.002	0.001	0.003	0.001	0.003	
IS0091R	PCB-156	0.025	0.004	0.006	0.001	0.011	0.001	0.001	0.01	0.003	0.001	0.006	0.001	
IS0091R	PCB-180	0.054	0.007	0.004	0.002	0.004	0.001	0.005	0.011	0.007	0.008	0.021	0.017	
IS0091R	PCB-28	0.066	0.016	0.016	0.126	0.041	0.016	0.019	0.026	0.016	0.023	0.016	0.016	
IS0091R	PCB-52	0.02	0.003	0.003	0.168	0.01	0.003	0.009	0.005	0.003	0.003	0.01	0.003	
IS0091R	precipitation-amount	41.385	70.882	47.689	28.24	68.714	41	46.133	135.881	31.571	49.114	123.701	140.531	
LT0015R	benzo[a]pyrene	1.76	1.56	1.57	1.9	1.08	1.39	-999	1.14	1.29	3.55	2.37	1.65	
NO0099R	γ -HCH	3.968	1.677	14.708	4.833	15.051	8.697	1.927	2.541	5.476	1.999	2.566	2.516	
NO0099R	precipitation-amount	30.574	25.541	82.198	39.044	18.247	40.381	36.306	75.191	72.707	84.681	81.147	51.688	