Chapter 2

DESCRIPTION OF CALCULATION DOMAIN AND INPUT DATA USED FOR MODELING

This chapter is devoted to a brief description of model input information used by the participating models at Stage II of the intercomparison study. This description covers two types of model input information: input data common for all models and data used by each particular model individually.

To describe the European calculation domain including its total fractions of land, water and vegetation areas, several input characteristics: land cover data, leaf area index, and organic matter content in the soil are agreed to be used by all models in Stage II experiments. Besides, for the sake of comparison, calculation of mass balance estimates requires other harmonised input information among which emissions data and/or initial concentrations of PCBs in the main environmental media, and physical-chemical properties and degradation rates of the considered POPs are the most important. The common data sets used in the calculations are presented below.

In addition to them, the participating models, which differ from each other by their type, resolution, overall modelling approaches, processes description and number of environmental media considered, require also a lot of another input data, e.g. on meteorological parameters, geophysical characteristics, concentrations of different reactants, data on specific aerosol surface in air, data on sea currents, the particles settling rates in seawater and so on. Due to a large diversity, such input data used by the participating models are described very briefly in this chapter.

A brief description of measurement data on air concentrations and depositions of PCBs used for the comparison with model estimates is given in Chapter 3 (Section 3.7).

2.1. Description of mass balance approaches used in participating models

Evaluation of the mass balance for PCBs within the specified calculation domain is based on the calculations of mass balances for main environmental compartments, which are considered by the particular model. ADEPT, CAN/POPs, CliMoChem, DEHM-POP, EVN-BETR and UK-MODEL, G-CIEMS, SimpleBox, and MSCE-POP considered the dispersion of PCB emissions between the atmosphere and the underlying surfaces. Further redistribution and accumulation of PCBs in the main environmental compartments other than atmosphere (soil and water) is also included in these models, but ADEPT model is purely atmospheric. Vegetation is taken into account in CliMoChem, EVN-BETR and UK-MODEL, G-CIEMS, SimpleBox, and MSCE-POP models. EVN-BETR and UK-MODEL, SimpleBox and MSCE-POP models imply also sediments and MSCE-POP considers the exchange between the atmosphere and the cryosphere (sea ice and snow).

Most of the models describe the same main processes: gas/aerosol partitioning, degradation in various environmental media, wet and dry deposition processes, and gaseous exchange between different environmental compartments. Along with degradation and outflow from the calculation domain, CliMoChem consider also deposition of the pollutants to the deep sea as additional loss process. Model parameterizations and comparison of the results of Stage I were discussed in detail in [*Shatalov et al.,* 2004]. Sensitivity study with respect to descriptions of basic processes is given in Chapter 4 of this report.

As it was agreed at the third EMEP expert meeting on intercomparison of POP models, this Intermediate Technical Report summarizes results on calculations of PCB mass balance estimates and sensitivity studies of process description and mass balance estimates. The included mass balance estimates for 2000 were obtained by participants on the basis of one-year calculation with zero initial concentrations (ADEPT, CliMoChem, DEHM-POP, EVN-BETR and UK-MODEL, G-CIEMS, MSCE-POP, SimpleBox) and with initial concentrations in media given as input data (CAN/POPs, EVN-BETR and UK-MODEL, G-CIEMS, DEHM-POP, MSCE-POP, SimpleBox). Calculations of CliMoChem, EVN-BETR and UK-MODEL and SimpleBox carried out for 20-year period (from 1981 to 2000) with zero initial data with historical emissions are also included in this note. Calculations were performed with the use of two different data sets: “own or alternative” and “reference» and for the three considered congeners – PCB-153, PCB-28 and PCB-180 (See Table 2.1). In Table 2.1 main comments and descriptions of model approaches to the calculation of mass balance provided by the participants when they submitted their calculation results on Stage II experiments can be also found.

Table 2.1. *Calculations of the mass balance and comments to these estimates*

Models	Physical-chemical data set	PCBs	Calculations and comments
ADEPT	Reference	PCB-153	One-year calculations are made with zero initial concentrations
EVN-BETR and UK-MODEL	Reference	PCB-153, PCB-28, PCB-180	One-year calculations are carried out using both a single box version of European model and the spatially resolved version. Results are calculated on the basis of initial concentrations given as input data, zero initial concentrations for one-year period (2000) and with historical emissions for 20-year period (from 1981 to 2000). The only exception was that the initial conditions for the vegetation were set to zero.
SimpleBox	Alternative and Reference	PCB-153, PCB-28, PCB-180	One-year calculations are made with zero initial concentrations and with initial concentrations in media given as input data; calculations are also carried out for 20-year period (from 1981 to 2000) with zero initial data with historical emissions. Calculations are done with 2 versions of the SimpleBox model, SimpleBox 3.0 and SimpleBox 3.12 The difference between the models is in the definition of the soil compartment. In the old Simplebox 3.0 model we assume that chemicals are distributed equally thoughout the soil compartment (the model calculates average soil concentrations). In the new SimpleBox 3.12 model it is assumed that concentrations of chemicals in the soil compartment decline exponentially (based on the chemical specific soil penetration depth). With this model we can calculate the chemical concentration at each depth of the soil compartment, and we can calculate average concentrations over, for example, the upper 5 cm of the soil based on the concentration profile. We did this for the MSC-E calculations. **General Mass balance equation for SimpleBox** Vi * dCi/dt = Emissioni + Importi - Exporti - Degradationi + Advectionij + Diffusionij -**Mass balance equation for the air compartment**: Vair * dCair/dt = EmissionAir + ImportAir - ExportAir - Vair * DegAir * Cair - SUM(DepWater)* Cair - SUM(DepSoil)* Cair - SUM(DepVeg)* Cair - SUM(XCHairwater) * Cair - SUM(XCHairsoil)* Cair - SUM(XCHairveg)* Cair + SUM(XCHwaterair) * Cwater + SUM(XCH soilair) * Csoil + SUM(XCHvegair)* Cveg Vair = volume of air compartment (m^3) Cair = total concentration in air (mol/m^3) t = time (s) EmissionAir = Emission mass flow into air compartment (mol/s) ImportAir = Import mass flow into air (mol/s) ExportAir = Export mass flow out of air (mol/s) DegAir = pseudo first order transformation rate constant in air (s^{-1}) DepWater/DepSoil/DepVeg = transport coefficient for atmospheric deposition (wet and dry) to water, soil, veg resp. (m^3/s) XCHair-water/soil/veg = transport coefficient gas absorption to water, soil, vegetation resp. (m^3/s) XCHwater/soil/veg-air = transport coefficient volatilization from water, soil, vegetion resp. (m^3/s) Cwater = concentration in water (dissolved) (mol/m^3) Csoil = concentration in soil (mol/m^3) Cveg = concentration in vegetation (mol/m^3)

SimpleBox	Alternative and Reference	PCB-153, PCB-28, PCB-180	**SimpleBox settings:** Height air = 1 km Soil depth = 5 cm (for forest and urban soil), 20 cm for agricultural soil Water depth = 200 m in continental level of SimpleBox. For regional level only 3 and 10 m water depths exist So, we only calculated the mass and degraded mass in air for the first 1 km, in soil for a depth of 5 cm and in water for a depth of 200 m. For water we could only calculate values on a continental level. **Comments**: SimpleBox is not a spatially explicit multimedia fate model. The only possibility for incorporating some spatial variability is by nesting regional and local model scales in the larger continental and global scales. So, the regional and the continental scale together form the calculation domain. The regional level covers the cells: 3W-10E/55-57N; 2W-11E/53-54N; 1W/52N; 1E-11E/51N; 3E-11E/50N; 5E-10E/49N; 6E-10E/48N. According to SimpleBox model layout it was pointed out which cells of the calculation domain belong to the regional level and which belong to the continental level. For this study we have taken the calculation domain specified by MSC-E as the total model domain (10W-30E/35N-70N). Within this area we have distinguished a regional level that covers the river basins that flow into the Southern North Sea (North-Western Europe). The rest of the model domain belongs to the continental model scale.
CAN/POPs	Own	PCB-153; PCB-180	One-year calculations are made with initial concentrations in media given as input data.
CliMoChem	Own and Reference	PCB-153, PCB-28, PCB-180	Calculations are carried out for 20-year period (from 1981 to 2000) with zero initial data with historical emissions and for one-year of 2000 on zero initial concentrations. **Comments:** CliMoChem is a climate zone model and does not provide longitudinal resolution. It is not possible to compute masses and mass fluxes for sub-zonal regions like the EMEP region (35°N – 70°N; 10°W – 30°E). Therefore, masses and mass fluxes were converted as followed. First, calculations were done for 10 latitudinal zones of equal latitudinal width, but varying longitudinal length (zone 1: North Pole, zone 10 South Pole). The EMEP region is covered by zone 2 (72°N – 54°N) and zone 3 (55°N – 36°N). Second, the masses and mass fluxes in zone 2 and zone 3 were summarized. Third, based on CliMoChem's assumption that the mass within a zone is uniformly distributed, the summarized masses and mass fluxes were divided by 9 (360°/40°) to obtain masses and mass fluxes per EMEP region. CliMoChem does calculate masses and mass fluxes per season (three months, starting in January) and monthly values are not available. Therefore, we suggest summarizing monthly outputs calculated by other models to obtain seasonal values that can be compared to CliMoChem's results.
MSCE-POP	Own and Reference	PCB-153, PCB-28, PCB-180	One-year calculations are made with zero initial concentrations and with initial concentrations in media as input data. **Comments:** The model used is a hemispheric model spatially resolved with resolution 2.5°x2.5°. It includes the following main environmental compartments: atmosphere, soil, seawater, sea ice/snow and vegetation. Horizontal spatial resolution in the oceanic compartment is two times finer. Vertical structure of the atmosphere consists of 12 layers with bounds defined in σ-coordinates. The overall coverage is about 12 km. Soil is divided into 7 vertical layers with thickness 0.1, 0.3, 0.6, 1, 2, 5,11 cm. In the ocean depth is given by special batimetry file.
G-CIEMS	Alternative and Reference	PCB-153	One-year calculations are made with initial concentrations in media given as input data and with zero initial conditions. **Comments:** All target region was treated as one-box environment. Land, emission and relevant information are developed by average and/or sum of the provided data-set. Generic moderate, tropic and polar regions are assumed to calculate the target region. The net in/out flux of the target region between surrounding generic region are presented in the "inflor-outflow" section.
DEHM-POP	Own and Reference	PCB-153, PCB-28, PCB-180	One-year calculations are made with zero initial concentrations and with initial concentrations in media given as input data. **Comments:** Results on calculations of mass contained and degraded in different environmental compartments are given for 15 km layer of the atmosphere; for 15 cm layer of soil; for 75 m layer of water.

2.2. Description of calculation domain

The calculation domain agreed to be used in Stage II experiments covers the most part of the European region occupying also some small parts of the North Atlantic and the Arctic oceans and Northern Africa (see Fig. 2.1). In the horizontal dimensions it is confined by the surface grid (i,j) with latitude: 35° N – 70° N; and longitude:10° W – 30° E. It has spatial resolution 1°×1°, which is arranged such that i = 1 is centered at 179.5°W and I increases eastward; and j = 1 is centered at 89.5°N and J increases southward.

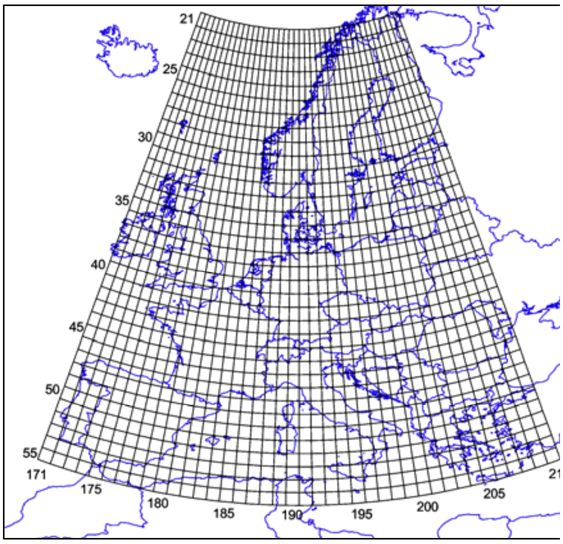

Fig. 2.1. The European calculation domain (1°×1° grid)

Information on land cover data, leaf area index, organic matter content in the soil and total fractions of land, water and vegetation areas in the European calculation domain were given as common input data for Stage II experiments and are described below. In addition, comments to these input data provided by the participants are also given in the appropriate subsections.

2.2.1. Land cover data

Land cover information used for the description of the European calculation domain within Stage II experiments and presented as input data is based on 25-category USGS Land Use/Land Cover dataset obtained from NCAR Mesoscale Modeling System (MM5) [*Guo and Chen*, 1994]. The original data set includes the detailed information on underlying surface types with high spatial resolution (10'x10'). Land cover data for model intercomparison study are presented in 1°×1° surface grid system. Each grid cell is characterized by several types of surface proportional their area. The USGS Land Use/Land Cover System Legend is presented in Table 2.2.

Total area of the European calculation domain is equal to 1.05E+07 km^2. Data on total fractions of land, water and vegetation areas in this calculation domain calculated by MSC-E on the basis of 25-category USGS Land Use/Land Cover dataset from [*Guo and Chen*, 1994] and presented as input data are given in Table 2.3.

***Table 2.2.** USGS Land Use/Land Cover System Legend*

No	Description
1	Urban and Built-Up Land
2	Dryland Cropland and Pasture
3	Irrigated Cropland and Pasture
4	Mixed Dryland/Irrigated Cropland and Pasture
5	Cropland/Grassland Mosaic
6	Cropland/Woodland Mosaic
7	Grassland
8	Shrubland
9	Mixed Shrubland/Grassland
10	Savanna
11	Deciduous Broadleaf Forest
12	Deciduous Needleleaf Forest
13	Evergreen Broadleaf Forest
14	Evergreen Needleleaf Forest
15	Mixed Forest
16	Water Bodies
17	Herbaceous Wetland
18	Wooded Wetland
19	Barren or Sparsely Vegetated
20	Herbaceous Tundra
21	Wooded Tundra
22	Mixed Tundra
23	Bare Ground Tundra
24	Snow or Ice
25	Unidentified

Table 2.3. *Total percentage of land, water and vegetation areas in the European calculation domain*

Land Cover Categories	Fractions (%):
Water	46.9
Soil	53.1
Vegetation	52.6

Comments of CliMoChem:

Currently, land cover data in *CliMoChem* are taken from [*DeFries and Townshend*, 1994]. The resulting surface type distribution summarized for zone 2 and zone 3 can be found in Table 2.4. Table 2.4 also shows surface type distribution based on [*Guo and Chen*, 1994].

Table 2.4. *Surface fraction based on [DeFries and Townshend, 1994][1] and [Guo and Chen, 1994] [2]*

Environmental Media	Surface Fraction	
	CliMoChem zone '2+3'	European region
Bare soil	0.0166[1]	0.0120[2]
Sea	0.3952[1]	0.4687[2]
Vegetation-Covered Soil	0.5882[1]	0.5193[2]

The spatial dimensions of the specific compartments directly influences mass fluxes and bulk compartment specific parameters (i.e. bulk vegetation compartment). Thus, the use of different land cover data results in different masses in the compartments.

Therefore, along with calculations carried out on the basis of land cover data from [*DeFries and Townshend*, 1994] additional calculations were performed where the default surface type distribution in zone 2 and zone 3 was overwritten with the surface type distribution from [*Guo and Chen*, 1994] for the European region. The data for the 25 land use categories summarized in the file LandUse11_domain.xls were regrouped in the following surface types: bare soil, grassland, coniferous forest, deciduous forest and water.

Comments of MSCE-POP:

Since the formulation of MSCE-POP model does not require detailed specification of data on the underlying surface, the original 25-categories of land cover from [*Guo and Chen*, 1994] were reduced to six general categories (deciduous forests, coniferous forests, grassland, urban and built-up land, bare land and glaciers, water bodies) and redistributed over the model grid. As an example we present here the spatial distribution of two land cover categories from reduced set: deciduous forests and grassland within the Northern Hemisphere (Fig. 2.2).

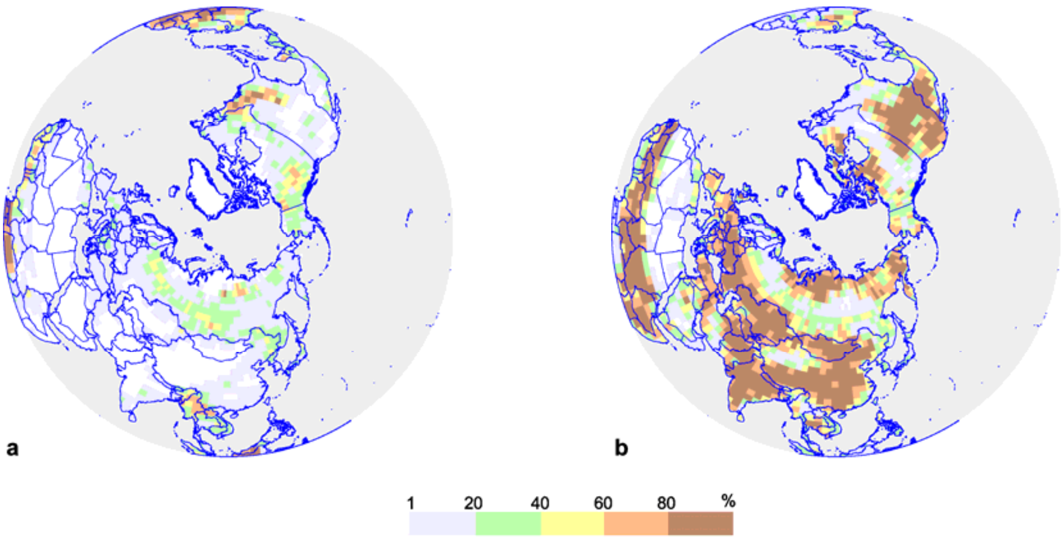

Fig. 2.2. *Fraction of the area covered by deciduous forests (a) and grassland (b) in the Northern Hemisphere used by MSCE-POP model*

2.2.2. Leaf Area Index

Leaf Area Index (LAI) data set used for the description of the European calculation domain within Stage II experiments and presented as input data includes the geographically resolved data for 1987 with monthly resolution. The Leaf Area Index for a given grid cell implies the ratio between the area of leaves in this cell to its total area (m^2/m^2). The geographically resolved leaf area index data with monthly resolution was adopted from CD-ROM of NASA Goddard Space Flight Center [*Sellers et al*., 1994, 1995] and converted to 1°×1° surface grid system. Consistency of these data in relation to the land cover information was investigated by correlation analysis.

2.2.3. Organic matter content in the soil

Data on organic matter content in the soil are taken from http://www.giss.nasa.gov/ and presented in 1°×1° surface grid system. The organic matter content in the soil for a given grid cell implies the share of organic matter in this cell relative to its total content (%).

Comments of CliMoChem:

CliMoChem's default model parameters were used to describe organic matter content in soil, as well as LAI. The reason for not using the data proposed by MSC-E is the fact that they are area specific and not – as required in *CliMoChem* – vegetation type specific. Table 2.5 gives an overview of the annually averaged organic matter content in soil and LAI for the different soil cover types. References can be found in the supporting information to [*Wegmann et al*., 2004].

Table 2.5. *Leaf area index (m^3/m^2) and fraction of organic carbon for different soil cover (%)*

Soil cover	LAI	Organic carbon content
Bare soil	none	2
Grassland	$2.77*10^{-4}$	8.5
Coniferous forest	$1.68*10^{-3}$	35.5
Deciduous forest	$7.81*10^{-4}$	22.5

2.2.4. Parameters of the environmental compartments

For calculations of mass balance estimates for the specified domain, such input parameters as volume of each environmental compartment including air height, soil depth and water depth were used by the participants in accordance with their modelling approaches. However, calculated values of mass contained and degraded in the environmental media are agreed to be submitted for the atmospheric layers of 1 km, 5 km and 10 km height, for soil layers of 5 cm and 10 cm depth; and seawater layer of 200 m depth.

Other particular geophysical parameters of environmental compartments used in the calculations such as properties of atmospheric aerosol (e.g. fraction of organic carbon content in the aerosol, specific aerosol surface, etc), soil, water and vegetation are also chosen for each model individually. Geophysical parameters of environmental compartments used by the participating models for the calculations of mass balance of PCBs in the specified calculation domain are presented in Table 2.6.

2.2.5. Other input data

The participants in accordance with their modelling approaches also use some other inputs parameters. They can include meteorological data, concentrations of OH-radical in the atmosphere, data on sea currents, concentrations of suspended particles, the particles settling rates in seawater and the content of organic carbon (OC) in these particles and so on. For example, in MSCE-POP model meteorological data provided by The System of Diagnosis of the Lower Atmosphere (SDA) developed by Hydrometeorological Centre of Russia [*Frolov et al.*, 1994; *Rubinstein et al*., 1997,1998; *Frolov et al.*, 1997 a,b,c] are used. A large set of meteorological parameters among which are data on wind velocity; precipitation rates; surface pressure; air temperature and a number of parameters of the atmospheric boundary layer (e.g. friction velocity, Monin-Obukhov length etc.) is implemented in the parameterization of the main processes of POP behavior. For the description of POP degradation in the atmosphere in MSCE-POP a spatial distribution of monthly mean concentrations of hydroxyl radical from [*Spivakovsky et al*., 2000] is used. The original data were interpolated to the calculation grid.

References to the input parameters used individually can be also found in the detailed descriptions of the participating models presented in the report on Stage I [*Shatalov et al.,* 2004].

Table 2.6. *Geophysical parameters of environmental compartments in the calculation domain used by participating models*

Media	Parameter	Dimension	DEHM-POP		ADEPT		CAN/POPs		CliMoChem		G-CIEMS		EVN-BETR and UK-MODEL		SimpleBox		MSCE-POP	
			Value	Reference	Value	Reference	Value	Reference	Value	Reference	Value	Reference	Value	Reference	Value	Reference	Value	Reference
Atmosphere	Air volume	km^3			3.85E+06		3.40E+08	own model	5.57E+08 (height: 6km)		1.05E+08		1.42E+07		9.60E+06		1.26E+08 (height: 12km)	Own
	Total Suspended Matter, TSP	$\mu g/m^3$	Variable	Own			Variable		86	*Wegmann et al.*, 2004	30		30				30	
	Fraction of organic carbon content in the aerosol, foc	%	20				Variable		not used		20		20				20	
	Specific aerosol surface, θ	m^2/m^3	Not used		1.50E-04		Variable		not used						1.50E-04		1.50E-04	*Whitby*, 1978
	Others …..														1.72E-01*		[OH] spatial distribution	*Spivakovsky et al.*, 2000
Soil	Soil volume	km^3					5.95E+03	own model	4.94E+03 (depth 0.1m)		5.57E+02		8.39E+02		7.21E+02**		1.11E+03 (depth 0.2m)	Own
	Bulk soil density, ρ	kg/m^3	1.35E+03	*Jury et al.*, 1983			1.35E+03	MSC-E	2.4	*Scheringer*, 2003	1.50E+03		2.40E+03		1.70E+03		1.35E+03	
	Volumetric water content in soil	%	30	*Jury et al.*, 1983			30	MSC-E	30	*Wegmann et al.*, 2004	30		0.3		20		30	
	Volumetric air content in soil	%	20	*Jury et al.*, 1983			20	MSC-E	20		20		0.3		20		20	
	Others …..																	
Water	Water volume	km^3					1.07E+06	own model	8.71E+06 (depth: 200m)		3.35E+06		8.66E+05		8.71E+05		9.83E+05 (depth 200m)	
	Freshwater	-											1.27E+05		3.39E-02		-	
	Seawater	-							8.71E+06		1		5.78E+06		9.66E-01		9.83E+05 (depth 200m)	
	Inclusion of suspended particulate matter	-	no				N/A		yes		yes		yes				yes	
	Others…..																	
Vegetation	Specific surface area of vegetation, *aV*	m^2/m^3													1.275E+03		8.00E+03	*Duyzer and van Oss*, 1997
	grassland	m^2/m^3							2.77E-04***	*Wegmann et al.*, 2004								
	coniferous forest	m^2/m^3							1.68E-03***	*Wegmann et al.*, 2004								
	decideous forest	m^2/m^3							7.81E-04***	*Wegmann et al.*, 2004								
	Others…..																	

* - Junge constant, *Pa·m;*
** - the total value of soil volume; for natural soil it is equal to 2.42E+02 km^3; for agricultural soil it is equal to 4.76E+02km^3; for other soil it is equal to 2.90E+00 km^3;
***-annual average values.

2.3. Physical-chemical properties and degradation rate constants

The data sets of basic physical-chemical properties and degradation rates of PCBs in the main environmental media are one of the most important input data used for model calculations. The key characteristics required for POP modelling are the following:

- air-water Henry's law constant (K_H);
- subcooled liquid vapour pressure (p_{0L});
- coefficients of partitioning between different media (octanol-water partition coefficient (K_{OW}), octanol-air partition coefficient (K_{OA}), organic carbon-water partition coefficient (K_{OC}));
- water solubility;
- degradation rate constants for different environmental compartments;

Temperature dependencies of Henry's law constant, subcooled liquid-vapour pressure, octanol-air partition coefficient and degradation rate constants are also of great importance for the reliable description and parameterization of POP behaviour in the environment.

The participating models perform calculations of mass balance estimates at Stage II using individual data sets of these physical-chemical properties and degradation rates. Descriptions of individual data sets for the considered congeners of PCBs ("own data sets") presented by CAN/POPs, DEHM-POP, G-CIEMS, EVN-BETR and UK-MODEL, CliMoChem and MSCE-POP are given in the MSC-E Technical Report 1/2004 [*Shatalov et al.,* 2004]. Comparison of base values of key physical-chemical parameters and coefficients of their temperature dependencies made within Stage I of POP model intercomparison study are also presented in [*Shatalov et al.,* 2004].

To study sensitivity of participating models with respect to variability of PCB physical-chemical parameter values, Stage II experiments and partly experiments proposed in the course of Stage I are repeated with the help of common "reference data sets". The latter are internally consistent data sets of key physical-chemical properties and degradation rates of PCBs taken mostly from [*Li et al*., 2003; *Mackay et al.,* 1992] and proposed for model testing. These data sets are also presented in the report on Stage I [*Shatalov et al.,* 2004]. For models using "reference data sets" as own physical-chemical properties, "alternative data sets" based on individual data of CliMoChem and DEHM-POP models and based mainly on data taken from [*Beyer et al*., 2002] are proposed for this sensitivity study. The "alternative data sets" used for modelling of PCB-153, PCB-28 and PCB-180 are presented below in Annex A.

2.4. Description of PCB emissions

Although reliable emission data for persistent organic pollutants PCBs is essential both to understand and control the large-scale environmental distribution of these contaminants, surprisingly little is still known about their emissions on a regional and global scale. Official emission data for PCBs submitted by Parties to CLRTAP tend to have an incomplete spatial, temporal and congeneric distribution which obstructs the possibility to use them for regional / hemispheric / global modelling activities. For this reason, the model calculations presented herein rely on global emission estimates derived from a European Union research project (Global SOC ENV4 – CT97 – 0638). These estimates were developed, using a dynamic mass balance approach, aiming to quantify the global historical production, consumption and emissions of selected PCBs [*Breivik et al*. 2002a,b].

The production of PCBs was addressed by collecting data from the literature on production of total PCB as well as of technical mixtures [*Breivik et al.*, 2002a]. These data were coupled with the chemical composition of the mixtures in order to estimate the production of individual homologues and congeners. Whenever there were gaps in the data, assumptions were made to come up with quantitative estimates for the production. More than 70% of the total PCB produced was in the form of Tri-CB, Tetra-CB and Penta-CB. The estimated total production of the selected 22 congeners (ΣPCB22) adds up to 0.566 million tonnes.

The global historical consumption pattern was estimated from collected data on the import, export, usage, and restrictions of PCB import on a regional/national level, along with assumptions regarding the PCB trade between countries and regions. The homologue and congener consumption was approached by treating groups of countries as closed markets. The results indicate that near 97% of the total historical consumption of PCBs occurred in the Northern Hemisphere (Figure 2.3). The main uncertainty related to the spatial pattern, is the estimated consumption for countries where no information is available on imports, exports and consumption. However, these countries contribute very little to the overall consumption and hence the associated uncertainty on a global scale is considered relatively small.

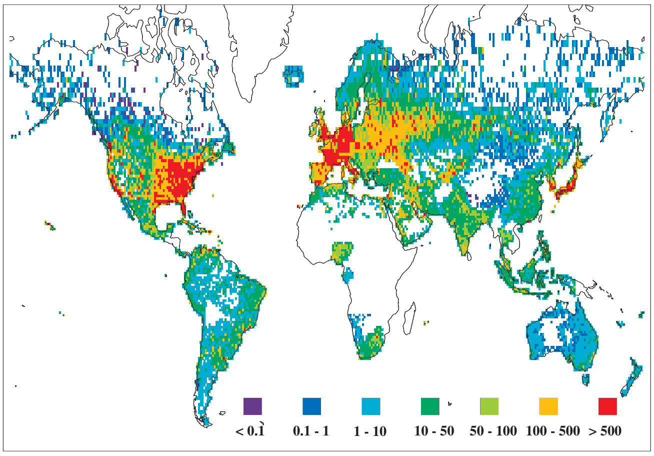

Fig. 2.3. *Estimated cumulative consumption of total PCBs on a 1˚ by 1˚ basis, using population distribution for spatial allocation [Breivik et al., 2002a]*

The diversity of PCB consumption and disposal pathways of PCB products makes the true emission pathways equally complex [*Breivik et al*., 2002b]. A lumped approach has been chosen to bridge the gap between consumption and emissions, thus clustering categories of usage, accidental releases and disposal pathways together. The crucial simplification in this approach is that any potential usage and loss process to the atmosphere is included in the selected categorisation of emission sources. Usage and disposal were further divided into four lumped sub-categories each and two accidental release pathways were addressed. Emissions were estimated through a mass balance approach.

The uncertainties in the emission estimates are predicted to be much higher than the uncertainty in the production estimates. For this reason, three different emission scenarios were elaborated (Minimum/Default/Maximum). These date are considered crucial as a research tool for mass balance studies and regional and global fate modelling. The results could also assist in the evaluation and interpretation of atmospheric levels of PCBs.

Mass balance studies as well as model studies utilising these data in combination with monitoring results have already facilitated some interpretation of the uncertainties/reliability of these estimates. *Wania and Daly* [2002] used the Globo-POP model to evaluate the global fate of PCBs. They found the maximum emission estimate to be the more reliable when comparing predicted and observed concentrations. Secondly, *Meijer et al*., [2003] studied the global occurrence of PCBs in soil. They suggested that the surface soil burden account for about 1.6% of the known historical production, and reasoned that it is likely that the true cumulative emission is toward – or above – the upper emission scenario presented by *Breivik et al*. [2002b]. For these reasons, the higher emission scenario has been used as common model input in the model calculations presented in this report.

Spatial distribution of PCB emissions in 2000 and in all years of the preceding period from 1980 up to the base year over the grid were prepared by S. Gong and P. Huang on the basis of this emission scenario in compliance with global distribution of population density. As an example, a map of gridded data of PCB-153 emissions in the Northern Hemisphere used for modeling at Stage II is presented in Fig. 2.4.

Within Stage II calculation experiments all amount of PCB emissions is assumed to be released into the atmosphere and for each year of the period the total annual emission value is continually distributed among the year.

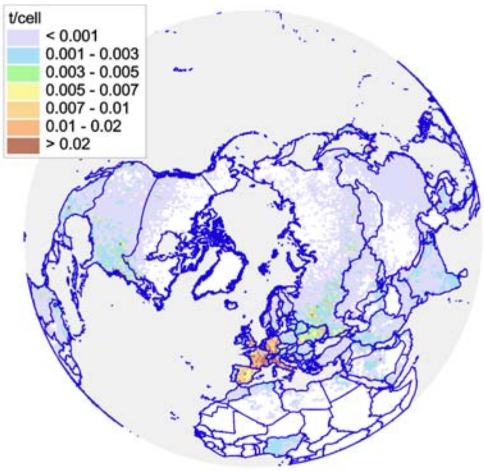

Fig. 2.4. *Spatial distribution of PCB-153 emissions in the Northern Hemisphere in 2000 prepared on the basis of data from [Breivik et al., 2002b]*

2.5. Initial concentrations of pollutants in media

For the sake of comparison of the model results vs measurements in 2000, one-year calculations of pollution levels of such persistent pollutants with significant long-range potential as PCBs requires appropriate information on their mass accumulated in the main environmental media of the European calculation domain up to the base year within rather long period of time.

Therefore, according to the agreed programme of Stage II modeling of PCB mass balance made on the basis of zero initial concentrations for one year are supplemented by the calculations for the same year but with the initial conditions of PCBs in the environmental compartments. So, calculations of PCB mass balance for one-year period (2000) with initial concentrations given as input data and for 20-year period (from 1981 to 2000) with zero initial data with historical emissions are performed. In the first case PCB initial concentrations as common input data for all participating models are calculated using hemispheric version of MSCE-POP model. In the latter case, initial conditions for PCBs in main environmental compartments are calculated by each participating model in their model runs using historical emissions from 1981 to 1999.

Input data on initial concentrations of PCB-153, 28 and 180 in the main environmental media (atmosphere, soil, water and vegetation) obtained by MSC-E for the end of 1999 and as it was agreed delivered for other models calculations are based on the data on global emission inventory from [*Breivik et al*., 2002b] taking into account long-term calculations from 1970 to 1999. These MSC-E calculations were performed under the joined EMEP, AMAP and WMO project "Persistent toxic substances, food safety and indigenous people of the Russian North" [*Dutchak et al*., 2002]. Data for modeling within Stage II are given in the 1°×1° surface grid system. As an example, spatial distribution of initial concentrations of PCB-153 in the atmosphere (a), soil (b), seawater (c) and vegetation (d) in the Northern Hemisphere in 1999 calculated by MSC-E on the basis of emission historical scenario of 1970-1999 from [*Breivik et al*., 2002b] are shown in Fig. 2.5.

These input data on initial concentrations of the considered congeners of PCBs were used in the calculations of CAN/POPs, DEHM-POP, EVN-BETR and UK-MODEL, G-CIEMS and SimleBox model. Of note, in the case of EVN-BETR and UK-MODEL model run the initial conditions for the vegetation are set to zero.

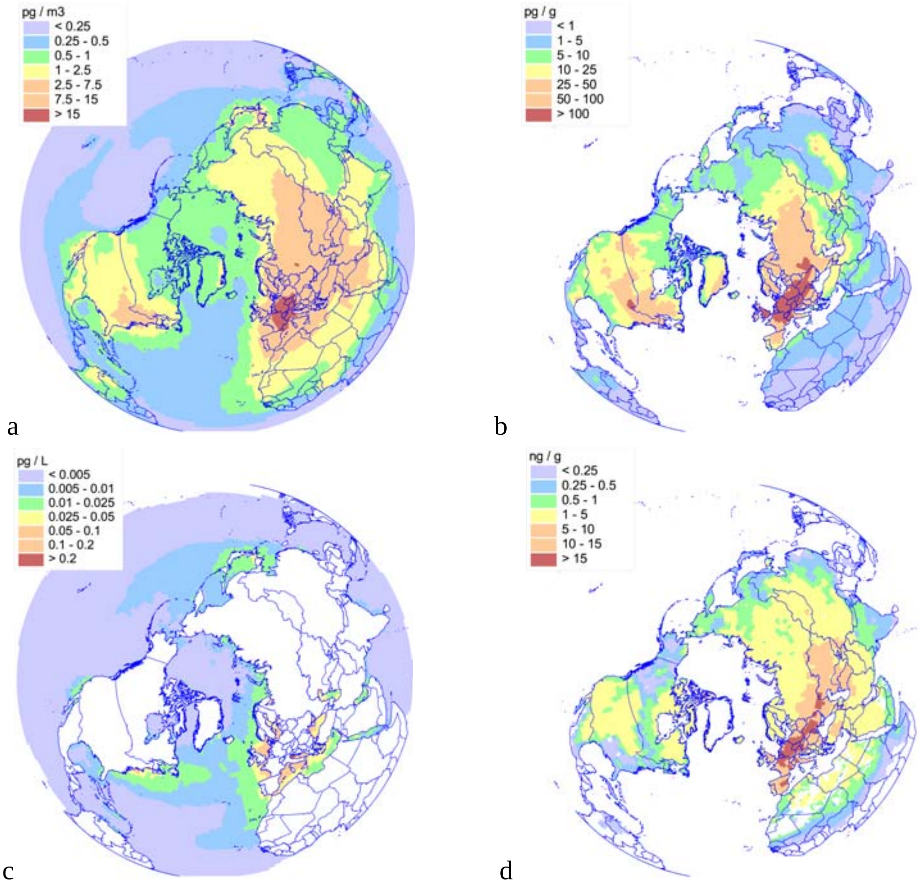

Fig. 2.5. *Spatial distribution of initial concentrations of PCB-153 in the atmosphere, (a), soil (b), seawater (c) and vegetation (d) in the Northern Hemisphere in 1999 calculated by MSC-E on the basis of historical data 1970-1999 from [Breivik et al., 2002b]*

Up to the moment of this report preparation, simulations of mass balance estimates of PCBs for 20-year period (from 1981 to 2000) with zero initial data based on a historical emission scenario run from 1981 to 1999 have been made and presented by the following models: CliMoChem, EVN-BETR and UK-MODEL, SimpleBox and MSCE-POP. Initial conditions on PCB mass contained in 1, 5 and 10 km layers of the atmosphere; 5 and 10 cm soil layers, 200m water layer and vegetation (kg) by the end of 1999 calculated by CliMoChem, SimpleBox and MSCE-POP models on the basis of "reference" and "own/alternative" physical-chemical data sets are presented in Table 2.7.

Table 2.7. *Values of PCBs mass contained in specified sub-domains in air, soil, seawater and vegetation compartments (kg) by the end of 1999*

Models	With "reference" data set							With "own/alternative" data set						
	Atmosphere			Soil		Water	Vege-tation	Atmosphere			Soil		Water	Vege-tation
	1 km	5 km	10 km	5cm	10cm	200m		1 km	5 km	10 km	5cm	10cm	200m	
PCB-153														
CliMoChem	0.466	2.330	4.660	4788.559	9577.118	162.328	111.377	1.242	6.208	12.415	7125.757	14251.513	286.554	45.659
SimpleBox	13.026	-	-	0.139	-	2.060	0.060	14.178	-	-	0.293	-	4.662	0.036
MSCE-POP	10.505	26.993	33.011	22907.554	23157.949	2.041	34.787	11.002	28.271	34.574	22930.800	23181.449	2.241	42.141
PCB-28														
CliMoChem	4.220	21.098	42.196	5239.535	10479.071	961.232	202.713	2.431	12.156	24.311	1845.345	3690.690	176.171	8.517
SimpleBox	27.195	-	-	0.052	-	1.561	0.068	23.409	-	-	0.019	-	0.292	0.018
MSCE-POP	44.296	113.819	139.194	22699.220	22947.338	10.197	97.119	40.335	103.643	126.749	22735.852	22984.370	11.596	104.608
PCB-180														
CliMoChem	0.035	0.175	0.350	1379.516	2759.032	38.735	21.214	0.243	1.214	2.428	3327.713	6655.425	60.341	16.047
SimpleBox	11.006	-	-	0.153	-	2.059	0.026	11.916	-	-	0.369	-	5.394	0.022
MSCE-POP	3.244	8.337	10.195	22829.235	23078.774	0.458	7.740	3.450	8.864	10.840	22879.179	23129.264	0.489	8.681