A.2.
Central Eastern European and Central Asia Countries Information

Hexachlorocyclohexanes in the Central and Eastern European Countries in the comparison with other parts of the world - The results from European measurements and modelling programmes -

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Introduction

In the last decades, PBT compounds and especially polyhalogenated aromatics have become a major issue of research in order to investigate their ubiquitous environmental occurrence, biochemical and toxic effects, human exposure and health risk assessment. Some of these pollutants such as polychlorinated biphenyls, phenols, benzenes and DDT have been produced intentionally in a wide variety of commercial applications because of their excellent technological or pesticide properties. Other persistent and very toxic pollutants such as polychlorinated dibenzo-p-dioxins and dibenzofurans have been formed as undesirable by-products, e.g., during the manufacture of the previously mentioned chemicals, waste combustion, the chlorine bleaching of pulp and paper, some metallurgical processes, etc (Wallack *et al.*, 1998).

International activities concerning to PBTs

UN/ECE

Environmental pollution by heavy metals (HMs) and persistent organic pollutants (POPs) more and more attracts attention on national and international levels (Dutchak, 2000). A range of projects related to the long-range atmospheric transport of these substances are being carried out by several international organisations and programmes such as WMO, UNEP, HELCOM, OSPAR, WHO, EUROTRAC, MEDPOL, EEA, AMAP.

An essential progress in the field of HMs and POPs air pollution was achieved within the framework of the UN ECE Convention on Long-Range Transboundary Air Pollution (LRTAP). In June 1998 in Arhus (Denmark) 36 Parties to the Convention signed the Protocols on POPs and HMs. In addition to the fulfilment of the basic obligations on the control of emissions, production and use Parties to the Protocols shall encourage research, monitoring and co-operation, in particular, in the fields of emissions, long-range transport, deposition levels, etc (UN/ECE, 1998).

In accordance with the Protocols EMEP shall provide the Executive Body with information on the long-range transport and deposition of HMs and POPs. EMEP results should promote further evaluating international abatement strategies and reviewing the implementation of the Protocols and compliance with Parties obligations.

At the first stage, EMEP plans to concentrate its efforts on emission inventory, monitoring and modelling of the following substances: cadmium, lead, mercury and selected POPs (PAH, PCB, HCB, chlordane, HCH, DDT/DDE, dioxins/furans). Data on measurements and emissions and their geographical distribution is prerequisite for a successful long-range transport modelling.

Chemical Coordinating Centre (CCC) coordinates and develops measurements and their quality assurance, data reporting and the monitoring database and processes, evaluates and reports data. Taking into account that HMs and POPs have not been a part of the EMEP monitoring programme before 1999, the Steering Body of EMEP requested CCC to collect already available data on these pollutants from national and international programmes (HELCOM, AMAP, OSPAR, MEDPOL).

UNEP

Based on a mandate given at UNEP GC 18/32, Nairobi, May 1995 and at UNEP GC 19/13C, Nairobi, Jan-Feb. 1997, UNEP has convened an Intergovernmental Negotiating Committee (INC) to develop an international legally binding instrument for implementing international action on certain persistent organic pollutants (POPs) (Wahlström, 2000). Other important activities preceding the negotiations were the UNEP meeting in Washington in October 1995,

which initiated a Global Programme of Action for the Protection of the Marine Environment from Land-Based Activities and the meeting of the Intergovernmental Forum on Chemical Safety (IFCS) *ad hoc* Working Group on Persistent Organic Pollutants in Manilla in June 1996.

UNEP Chemicals has recently taken steps towards consolidating and expanding its activities in the field of chemicals assessment, with a strong emphasis on water related effects and impacts. Starting in late Spring 2000, UNEP Chemicals will execute a two-year US$ 5 million global project on Regionally Based Assessment of Persistent Toxic Substances. The project will address Persistent Toxic Substances (PTS), which is a broader group than the POPs in the negotiations. The project will focus on the aquatic environment and will be executed in twelve regions around the globe.

Criteria for including substances in the project were suggested at the 1st technical expert workshop of the PDF-B phase as follows:

- persistence, including continuous release of moderately persistent substances.
- bio-accumulation.
- toxicity.
- organometallic substances should be included.
- regional and sub-regional transport scales.

Expected outputs of the project are:

- Overall global assessment of the issues and problems with PTS.
- Assessment of priorities in regions.
- Priorities for future GEF intervention.

Emission inventories

Detailed and accurate emission inventories for organic and inorganic toxic compounds are needed for several reasons (Breivik and Pacyna, 1999, 2000). First, one of the key users of emission inventories are dispersion modellers that use these data to address source-receptor relationships at various scales. Further, emission inventories are also needed as useful documentation between states that have agreed to reduce the emissions of these toxic compounds. While strong efforts have been made to improve inventories needed for research on ozone depletion (CFCs), climate change (CO_2, CH_4, N_2O) and acid deposition (SO_2, NO_x), inventories for inorganic and organic toxic compounds are still unreliable and inaccurate (Graedel *et al*. 1993, Pacyna and Graedel, 1995).

The relative poor quality of the current data on toxic compounds is obviously mitigating the confidence in model outputs, e.g. the reliability concerning impact of current or altered emissions on environmental levels. However, improvements have been made in recent years, and the objective of this document is to highlight the status of current data related to emission inventories for inorganic and organic toxic compounds in Europe.

The approach on how emission inventories are being established varies depending on the compound of interest. However, there are a few general aspects of the methodology. The ideal approach for estimating the actual emissions from a given plant is through monitoring and measurements of the chemical composition of the exhaust gas and the accompanying flow. If such site-specific data are not available, preferably emission data reported from national experts are used as indicated previously. In other cases, emissions of a given pollutant are usually estimated by the use of activity data and emission factors. Activity data are usually found in national and international statistical yearbooks, and emission factors can be found in emission inventory guidebooks (Holoubek *et al*., 2000a; Parma *et al*. 1995; UN/ECE, 1996).

The uncertainties, whether due to a general lack of proper data or methodological errors, result in varying quality of the emission data. In inventories, each estimate is therefore often accompanied by a simple quality code based on the assessment of the quality of the collected or generated emission data. These codes could then be transferred to quantify the uncertainty, e.g. of the total emissions in a country.

One of the first European inventories for POPs was presented by Duiser and Veldt (1989) for the Dornier-report (Axenfeld *et al*. 1989). It covered emissions of selected PAHs, PCBs, gamma-HCH (Lindane) and HCB in Europe for the reference year 1982. European emissions of Lindane (gamma-HCH) and Benzo[a]Pyrene (B[a]P) for the year 1990 were later included as a part of the previously mentioned ESQUAD-study (Berdowski *et al*. 1994).

Recently however, the study by Berdowski *et al*. (1997) has gained most interest. This study for 1990 covers several POPs, namely:

- Polyaromatic Hydrocarbons (PAHs)
- Polychlorinated Biphenyls (PCBs)
- Endosulfan

- Fenthione
- Quintozene
- Lindane (gamma-HCH)
- Hexachlorobenzene (HCB)
- Pentachlorophenol (PCP)
- Dioxins and Furans (PCDD/Fs) [In g I-TEQ/year]
- Tetrachloroethene (C_2Cl_4)
- Trichlorethene (C_2Cl_3H)
- Trichlorobenzene
- 1,1,1-Trichloroethane
- Tetrachloromethane, and
- Xylene

Very recently, a new inventory for POPs in Europe was completed (Pacyna *et al*. 1999). In addition to these European inventories, there have also been studies conducted with global resolution for selected pesticides as HCHs (Li, 1999), Toxaphene and DDT (Voldner and Li, 1995). Many of these data are available on 1°x1° resolution on request from GEIA (the IGBP Global Emission Inventory Activities programme). A simple global inventory for PCDD/Fs, was presented by Brzuzy and Hites (1996). Very recently an inventory of national and regional PCDD/Fs data became available from the UN Environment Programme (UNEP, 1999).

Example of pesticide POPs - HCHs

HCH (Hexachlorocyclohexane) is an insecticide that was heavily used within Europe (Breivik *et al*. 1999) and other parts of the world (Li, 1999). Principally, two formulations have been made for the use as an insecticide. One is technical HCH that is the unpurified technical mixture of various isomers (about 55-80% alpha-HCH, 5-14% beta-HCH, 8-15% gamma-HCH as well as minor quantities of delta-HCH and epsilon-HCH). Within this "raw" product, only gamma-HCH has any significant insecticidal properties. The other HCH-insecticide is lindane or essentially pure gamma-HCH.

Unfortunately, only a few countries report their annual usage of each of these formulations based on active ingredient with isomeric resolution. Thus, we first collected all data that could be found on national annual consumption of lindane or technical HCH. Next, we collected data on insecticide usage in various countries, or data on usage of organochlorinated pesticides. In order to fill out both temporal and spatial gaps, we developed regional ratios (West, former Eastern, Mediterranean and Nordic regions) as well as grouping the ratios from 1970-1979, 1980-1985, 1986-1990 and 1991-1996. Finally we estimated the consumption of various isomers based on information on the relative consumption of technical HCH versus lindane as well as regional/national measures against the usage of each of the two principal HCH-formulations. The intention was to assess the level and trend of the consumption over Europe. The result is given in Figure 1.

With this extrapolation method, the risk of significantly over- or underestimating the consumption at the national level for a given year is high. As can be seen the uncertainty bands that are depicted do also indicate a high uncertainty in the estimated historical consumption of total HCH. A more detailed description of these estimates can be found in Breivik *et al*. (1999).

The uncertainties in the usage estimates are further amplified when we utilise these data to estimate the interannual emissions of HCHs to the air, e.g. for use in regional atmospheric transport models. To estimate the actual losses of HCHs to air within a year, based on the annual consumption is difficult for several reasons, e.g.:

1) When is the insecticide used?
2) How is the insecticide applied?
3) Where is the insecticide used in a country?

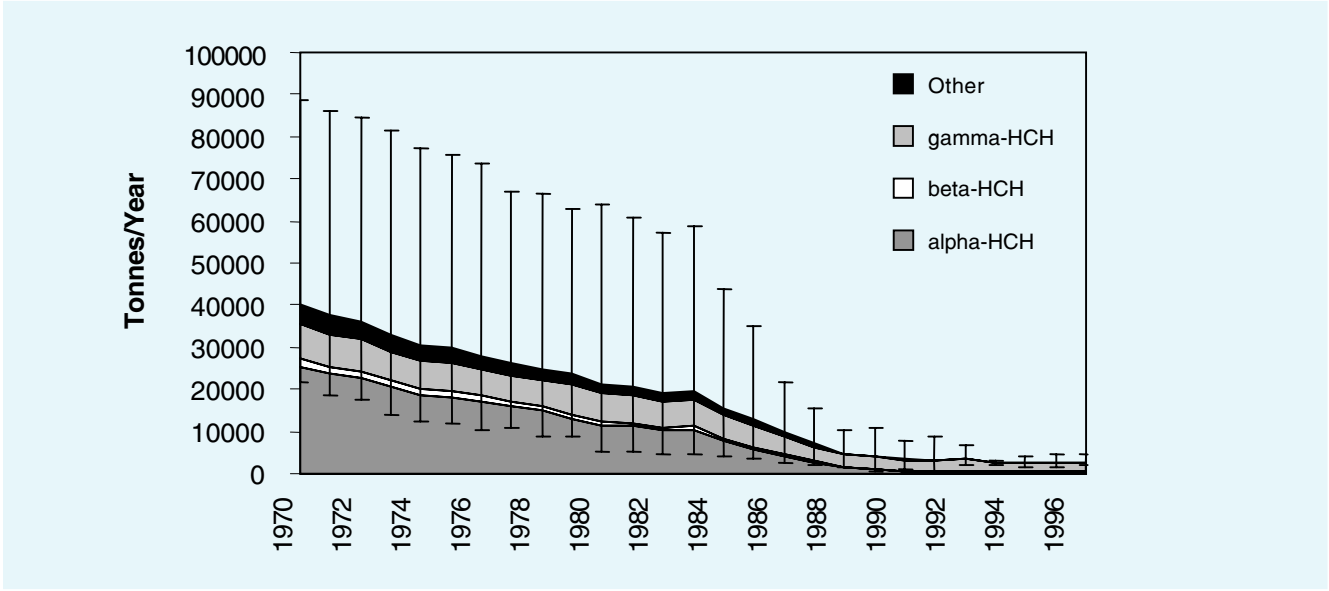


Figure 1. Estimated annual usage of various isomers in Europe, 1970-1996 (Breivik and Pacyna, 2000)

A rigorous treatment would require that each individual farmer using the insecticide would need to be localised and answer how he used HCHs (direct incorporation into the soil, seed treatment, spray application?) as well as the amounts that were applied at a given point of time. Further, the losses to air e.g. from spray application would be significantly influenced by e.g. the wind speed, the humidity as well as the temperature at the time of application. This would need consideration as well. Obviously this information is impossible to obtain at a European scale for a 25 years period, and we have problems with parameterisation similar as modellers. Therefore, all we can do is to give some estimates on losses to air and/or other compartments and document our assumptions in hope that the estimates can be improved in the future as knowledge and methods are improved.

If we simply neglect losses/emissions of HCHs during the actual production and manufacturing process as well as any potential losses to the environment from handling, storage and disposal, the major loss process to air will be from application in the field. Our principal tasks within the project concerning HCH emissions were to:

a) Estimate fractions of annual national consumption used for various applications (soil, seed and other treatment)

b) Estimate losses to air and other compartments from these applications, based on literature research on field and chamber studies

c) Estimate spatial distribution of consumption / emissions by using crop area distribution as surrogate parameter for national usage/emissions (50x50 km resolution)

d) Estimate time of application by including an annual distribution function for annual emissions.

Emission of hexachlorocyclohexane (HCH)

Hexachlorocyclohexane is an insecticide widely used in the world since the 40s (Shatalov *et al.*, 2000). There are eight isomers of HCH. Gamma-HCH has the best insecticide properties. Up to the end of the 1970s the main source of gamma-HCH was use of technical HCH containing different isomers in these or those proportions. Later lindane (HCH containing at least 99% of gamma-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 1.

Table 1. Official data on HCH emissions [kg.yr^{-1}]

Country	UN/ECE reported official emission data									
	1980	1990	1991	1992	1993	1994	1995	1996	1997	1998
Croatia		9,400						12,800	3,100	5,000
Denmark						61				0
Germany						15,000				
Hungary		9,281	60	12	462	798	1,650	2,400	31	22
Netherlands		0		0		0	0	0	0	
Russia		923,000								
UK		99,023	85,335	74,016	64,604	56,733	50,114	44,518	39,761	35,695
Austria (use)						12,000	8,056	8,640	2,324	0
HCH Lindane	5,635,000	73,800 800	72,000 150	50,000	57,000	36,500				

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

Expert estimates of gamma-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 2 illustrates emission data for 1970, 1975, 1980, 1985, 1990 and 1996.

According to expert estimates total gamma-HCH emission in Europe decreases by 3.5 folds during 1970-96.

Since the share of gamma-isomer is unknown in official data on HCH emissions, in the comparison among 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 gamma-HCH emission distribution over the EMEP domain with spatial resolution 150x150 km^2 for 1997 is demonstrated in Figure 2.

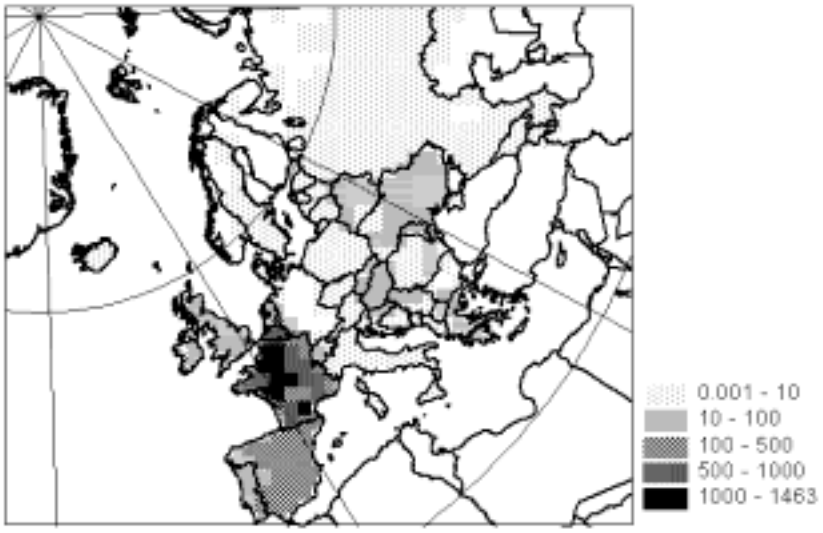


Figure 2. Spatial distribution of gamma-HCH emission in 1997 [g.km^{-2}.yr^{-1}]

As seen from Figure 2 the highest density takes place in France. The contribution of this country to total European emission is 71% during 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 gamma-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;
- gamma-HCH - 10% of total emission - in February, 15% - in March and by 25% - in April, May and June each.

Table 2. Expert estimates of gamma-HCH emissions in European countries [kg.yr^{-1}].

Country	1970	1975	1980	1985	1990	1996
Albania	7,661	7,661	6,097	4,433	2,228	463
Armenia*	2,714	2,714	2,111	1,037	69	30
Austria	2,105	3,741	5,915	6,213	3,392	0
Azerbaijan*	10,252	10,252	7,974	3,916	259	113
Belarus	77,182	77,182	60,030	35,688	4,645	4,645
Belgium	21,319	21,319	19,075	23,049	15,411	15,890
Bosnia Herzegovina	31,703*	31,703*	17,355*	7,332*	6,260	567
Bulgaria	22,226	22,226	22,141	17,256	5,152	0
Croatia	49,914*	49,914*	27,324*	11,544*	2,582	472
Cyprus	543	543	476	272	91	91
Czech Republic	5,608*	3,681*	58*	58*	22	319
Denmark	3,036	3,290	4,348	2,924	2,925	0
Estonia	169	169	164	156	7	3
Finland	818	1,157	4,970	1,840	0	0
France	273,780	273,780	322,228	294,072	270,456	560,000
Georgia*	8,667	8,667	6,741	3,310	219	95
Germany	25,868	52,604	111,447	49,608	27,662	0
Greece	47,286	47,286	67,599	37,694	2,952	5,863
Hungary	630,106	91,287	1,828	11,508	1,435	2,870
Iceland	0	40	35	39	73	109
Ireland	2,132	2,132	2,279	2,513	1,773	2,167
Italy	322,113	426,810	551,635	568,015	282,218	2,230

Kazakhstan *, **	40,550	40,550	31,539	15,488	1,025	446
Latvia	615	615	319	104	13	3
Lithuania	1,609	1,609	1,251	744	97	2
Luxembourg	273	273	317	191	135	151
Netherlands	38,619	38,619	32,686	36,589	3,814	6,307
Norway	380	380	1,104	1,178	130	0
Poland	22,790	16,817	7,095	2,773	446	280
Portugal	1,875	1,295	1,610	2,344	3,390	13,189
Republic of Moldova	13,876	13,876	10,792	6,416	835	364
Romania	46,220	46,220	20,223	6,351	3,693	2,035
Russian Federation*	540,047	540,047	420,036	206,267	13,652	5,941
Slovakia	2,379*	1,562*	25*	25*	304	581
Slovenia	9,854*	9,854*	5,394*	2,279*	294	169
Spain	98,958	98,958	221,668	275,160	180,146	122,909
Sweden	4,589	1,995	3,115	1,575	2,668	1,101
Switzerland	3,772	3,772	2,753	1,531	16	159
The FYR of Macedonia	19,522*	19,522*	10,687*	4,515*	2,298	69
Ukraine	277,142	277,142	215,555	105,853	12,877	9,047
United Kingdom	19,214	19,214	56,228	17,240	31,083	30,039
Yugoslavia	87,799*	87,799*	48,064*	20,305*	18,690	1,694
Total, t/y	2,775	2,358	2,332	1,789	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

Uncertainty of emission estimates

The uncertainty of POPs 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, 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 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 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 there is a new direction of activity and there are a great number of open questions concerning POPs 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 POPs 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 POPs to the Guidebook on Emission Inventory will be very beneficial.

Environmental levels of HCHs and lindane

Concentrations in the atmosphere

Lindane concentrations in air along Europe range from 0.01 to 1.4 ng.m^{-3}. Concentrations in precipitation can also vary in a wide range from 5 to 125 ng.l^{-1}. Lindane concentrations over the North Sea amount to 4-5 ng.m^{-3}.

Concentrations in fresh water

Typical concentrations are 10 - 40 ng.l^{-1} of lindane. Its concentration in bottom sediments can be 100 fold higher (1,800 - 2,400 ng.kg^{-1}) (Yufit and Klyuev, 1997).

Table 3 presents the list of EMEPsites used here and further in the report. Below measurement data on some POPs in regions of the North and Baltic Seas are presented (Tables 4 and 5). Tables 6 and 7 contain measurement data of EMEP monitoring network.

Table 3. List of EMEP monitoring stations submitted data on concentrations of some POPs (Berg *et al.*, 1996)

Country	Station codes	Station name	Location		Height above sea, m
			Lat.	Long.	
Czech Republic	CZ3	Kosetice	49°35'N	15°05'E	633
Denmark	DK31	Ulborg	56°17'N	8°26'E	10
Finland	FI96	Pallas	67°58'N	24°7'E	566
Germany	DE1	Westerland Zingst	54°55'N 54°26'N	8°18'E 12°44'E	12 1
Iceland	IS91	Stórhöfdi	63°24'N	20°17'W	118
Ireland	IE2	Turlough Hill	53°02'N	6°24'W	420
Norway	NO42	Spitsbergen, Zeppelinfjell	78°54'N	11°53'E	474
Sweden	SE2	Rörvik	57°25'N	11°56'E	10

Table 4. Air concentrations of some POPs 1987, 1988 (Baltic and North Seas) [pg.m^{-3}]

	alpha-HCH	gamma-HCH
Kiel Bight	80 - 3,000	50 - 10,000
Ship measurements, various campaigns 88 - 89	(53) (82) (170) (184) (46)	(69) (216) (864) (2,780) (176)

Table 5. Concentrations in precipitation of some POPs (North Sea)* [ng.l^{-1}]

	Volume [l]	alpha-HCH	gamma-HCH
Collafirth	29.3	1.74	0.88
Rattray Hd.	19.4	2.64	2.14
Carnbee	19.5	3.81	3.06
Tantallon	12.3	4.78	5.25
Lindisfarne	15.9	4.55	4.91
Flamborough	15.3	4.82	6.49
Burnham	9.5	3.96	8.96
Average	**17**	**3.76**	**4.53**

* from [Wells and Johnstone, 1987]

Table 6. EMEP data on air concentrations of some POPs [pg.m^{-3}] (Berg *et al.*, 1996)

	IS91/95*	NO42/93	NO42/94	NO42/95	NO99/92	NO99/93	NO99/94	NO99/95
alpha-HCH	17/12	77/69	61/58	63/60	93/82	74/67	65/56	52/50
gamma-HCH	14/11	14/13	16/14	13/12	86/59	59/43	123/55	65/39

* site code / year
** arithmetic mean/geometric mean

Table 7. EMEP data on concentrations of some POPs in precipitation (1990-1995) [ng.l^{-1}] (Berg *et al.*, 1996]

	DE1			DK31		NO99				
	90	**92**	**93**	**91**	**92**	**91**	**92**	**93**	**94**	**95**
alpha-HCH	0.85	1.89	1.01	1.71	1.2	2.69	2.07	2.07	2.14	2.00
gamma-HCH	4.54	18.22	9.28	11.91	15.8	4.05	5.02	8.45	9.98	5.54

Concentrations of HCHs in Europe

Figures 3 and 4 show temporal trends for alpha-HCH and gamma-HCH in air at 5 stations. The concentration level of alpha-HCH at the Norwegian stations is relatively high compared to the other stations, but decreasing. This is probably due to higher input of technical HCH at high latitudes. Almost 80% of the remaining use of alpha-HCH in Europe in 1996 was assigned to the new states of the former Soviet Union (422 t of technical HCH) (Breivik *et al.*, 1999). The other 20% were attributed as usage in some former eastern European countries (Breivik *et al.*, 1999). Iceland is influenced by westerly air masses, which explains the lower concentrations seen at IS0091R.

Lista (NO0099R) at the southern coast of Norway shows the highest concentrations of gamma-HCH in air, which may be due to long-range transport from southern parts of Europe. According to Centre International d'Etudes du Lindane (CIEL, 1998), the average annual lindane consumption in Europe was 2,130 t during the period from 1992 to 1997. France was the major user of lindane in Europe during this period, with an annual average consumption of 1,600 t (CIEL, 1998).

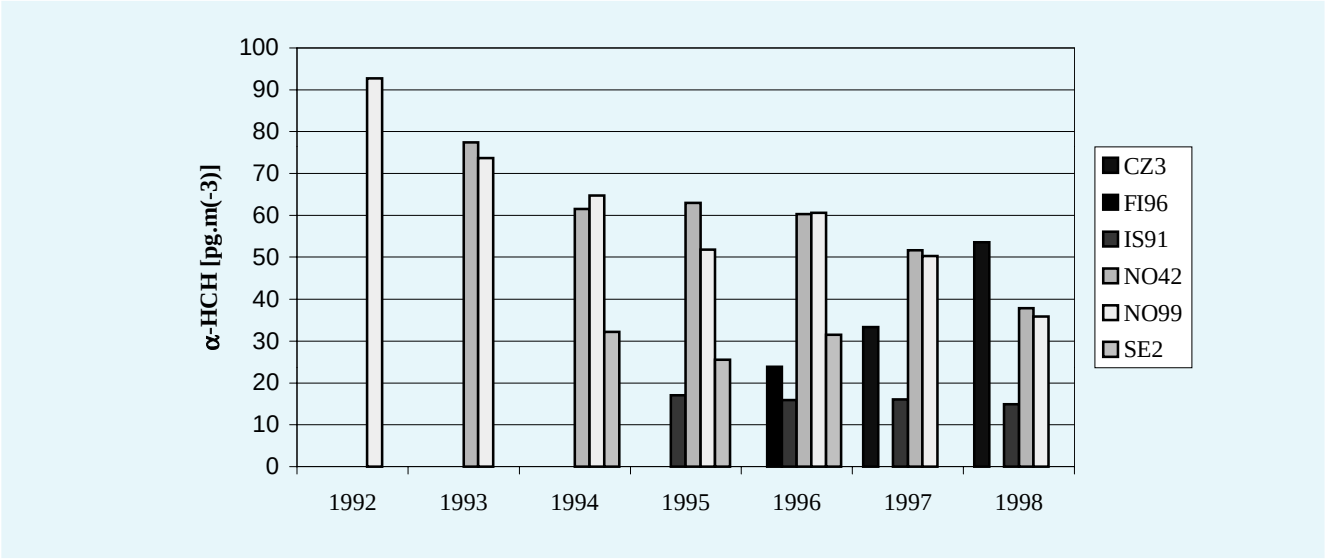


Figure 3. Annual weighted means for alpha-HCH during 1992-1998.

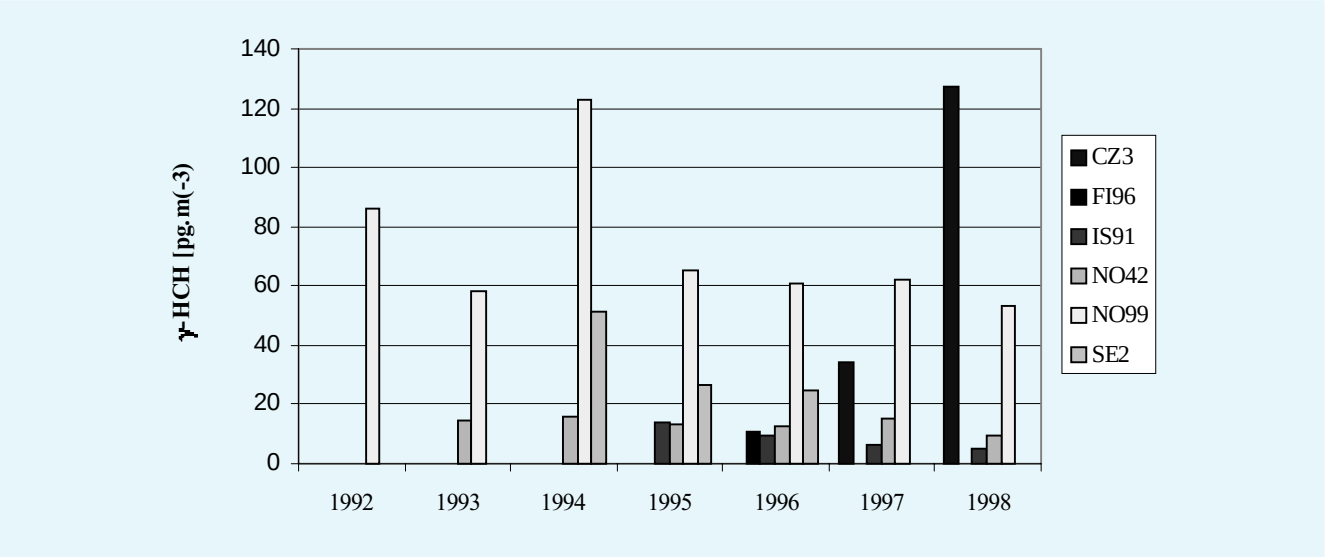


Figure 4. Annual weighted means for gamma-HCH during 1992-1998.

The situation in the Central and Eastern European countries

The region of "Central and Eastern Europe" is in this paper located in the area from the Baltic Sea to south part of Adriatic Sea and from The Czech Republic to Baltic countries (Holoubek *et al*., 1999, 2000b). In general, there are not many data on PBT compounds levels in many CEE countries of CEE region. The better situation in concerning data on the industrial and pesticide chemicals in Croatia, Czech Republic, Poland and Slovakia. In some others, such as Bulgaria, Hungary, Slovenia, satisfactory information concerning the contamination with pesticides exist. In the rest of CEE countries, only limited data on PBT sources and levels are available (UNEP/IFCS, 1998).

The first complex description of state of environmental pollution with PBT types of compounds in Central and Eastern European countries was prepared by Prof. Heinisch (Heinisch *et al*., 1994). Prof. Heinisch and his co-workers continued in this work and published many other reports, for example three volumes study concerning comparison of situation in Germany, mainly in Bavaria and new countries and Czech Republic (Heinisch *et al*., 1997a, b,c).

We have not to forget the fact that this region has very specific problems of environmental pollution, which are the results of the recent wars. Destruction of industrial facilities and spilling of chemicals have the worst effect for

the environment (Bosnia and Herzegovina, Croatia, Serbia and Montenegro). But the bizarre situations like usage of transformer oil, as a diesel fuel and antilace shampoo containing lindane against pests in the gardens could not be bypassed (Knezevic and Sober, 1998). One potential result of the pollution of the food, water and whole environment is dramatic increasing in the digestive system carcinoma, particularly large intestine carcinoma, which has been observed last two years in Bosnia and Herzegovina.

Most of the countries in the region produce and/or formulate pesticides. The pesticide registration is a primary requirement for import, production and distribution. During the period of centralised economy in this region, the import was monopolised by the relevant state organisation. By the end of the 80s, many private companies and minor distributors were involved in import and distribution of pesticides (PHARE 1997; Bratanova *et al*., 1998), many of which lack the required experience, a large part of the farming population has insufficient education and training in plant protection (Tasheva, 1998).

Pesticide concentrations in Danube River and its tributaries show significant differences between countries in the number and the types of pesticides analysed. The cumulative number of analysed pesticides was 76. Residues of only 36 pesticides and metabolites have been detected. The most frequently detected pesticides are organochlorine compounds and triazines. Only DDT and metabolites, HCH and its isomers and atrazine and its metabolites were found in more than 50 % of samples.

The special attention must be given to unwanted pesticides. The problem of unwanted and expired pesticides pose the greatest danger to the natural environment and people, which is brought about by chemisation of agriculture in CEE countries. This problem results from many years of errors in pesticide management and especially in their distribution. In Poland, the amount of unwanted pesticides is estimated at about 60,000 tonnes - about 10,000 tonnes in the tombs, another 25,000 tonnes at stores and about 25,000 tonnes at individual farmers (Stobiecki, 1998). The inventory of banned organochlorine pesticides in stocks in Bulgaria in 1996 showed about 35 tonnes, which is relative high quantity for the small territory of Bulgaria (Tasheva, 1998). Historical changes in these countries caused that together with the advent of market economy the problem of storing expired pesticides ceased to exist. Countries still have not solved the problem of safety storage for pesticides and other chemicals classified as poisons and they have no information concerning the quantity of pesticide and chemical (for example PCBs) waste. Many CEE countries have no special sites for dangerous materials or incinerators in which these types of chemicals could be safety burnt.

Potential risk is linked with storage of unused PBT pesticides. In this respect, for example, Poland has evidence of storage more than 10,000 tonnes of unused pesticides (mixture of different pesticides including other PBT compounds). The situation is probably very similar in many other CEE countries (UNEP, 1998). These were identified as possible hot spots in the region.

In some countries of the region (i.e. in Bulgaria) it was recognised that concentrations of PBT pesticides in environmental media decreased on such levels that no further systematic monitoring is needed (Tasheva, 1998). Different situation could be found in Albania (Koci, 1998) or Rumania (PHARE 1997; Bratanova *et al*., 1998; Toader and Chitimea, 1998) where still relatively high concentrations are measured in water and sediments (DDT and other chlorinated pesticides). In the Slovak Republic measured data show high exposure to HCB from an unknown source, which resulted in fact that critical, high concentrations could be found in human tissues (UNEP, 1998; Kocan *et al*. 1996, 1998, 1999).

Many of pesticides from UN-ECE list of POPs, never been used in many countries from this region (aldrin, chlordane, mirex, heptachlor, toxaphene). These were banned, restricted in several countries from the region similar to some other countries of the world. Hungary was among the first countries in the world that ban or severely restricted chlorinated pesticides still in 1966 (UNEP, 1998).

The soil contamination by hexachlorocyclohexane and consequently the HCH accumulation in agricultural products were of great interest due to their long persistence in our ecosystem (Schlosserova, 1993). In the Slovak Republic hexachlorocyclohexane was synthesised in the former CHZJD factory in Bratislava. During the years 1956-1966, more than 13,000 tonnes of gamma-HCH was produced. Some additional raw HCH was imported from the former Yugoslavia. After isolation of the gamma isomer it was formulated into different pesticides in same factory. Naturally, the produced pesticides were intended partly for domestic utilisation. A very rough estimate of annual consumption of Lindane containing pesticides in the Slovak Republic was about 1 tonne per year. This amount has been gradually decreased. The variety utilised Lindane based formulations has been as well descended: e.g. in the List of Permitted Pesticides for 1972 were listed 11 pesticides on the basis of Lindane. These were insecticides applied to the most important crops (potatoes, beet roots, rapeseeds, hops), staining agents for treating seeds of cereals, maize, legumes, sugar beet, cucumber, cotton, hemp, rapeseed, water melon, soil desinsectants for growing beet roots, sugar beet, cereals, maize, tobacco, hops, young fruit, trees and vines, and fumigant.

The application of all these pesticides was possible only with some restrictive precautions. The wide and systematic utilisation of Lindane containing insecticides was banned in 1974. The year 1992 was the last one when Lindane seed treating agents were permitted as formulations Lindane WP 80 for rapeseed and Lindane 50/35 WP for flax and hemp seeds. A new Lindane containing pesticide was given in the List of Permitted Pesticides for 1993. Emdenit was intended for controlling pine insects but only for two years period.

Modelling of HCHs and lindane

Modelling of gamma-HCH transport in European region is made for the period of 1970-97. Since it is important to take into account gamma-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 5. The figure indicates that major emission sources (according to emission data used) are located in South-western Europe. gamma-HCH emission data are demonstrated in Table 8 in the end of this section.

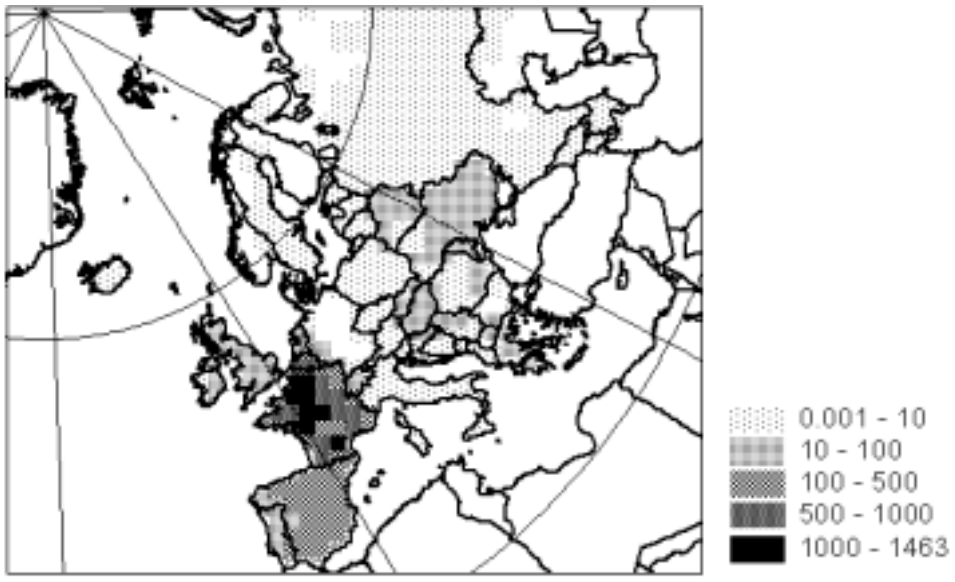


Figure 5. Gamma-HCH emission in 1997 [g.km-2.yr^{-1}]

In this section we analyse gamma-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 gamma-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 gamma-HCH dispersion at long time scale (see volume 2 of this report). gamma-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 gamma-HCH modelling should be made on the hemispherical scale.

Gamma-HCH concentrations and depositions in European countries in 1997

This part deals with a provisional analysis of gamma-HCH mean depositions and concentrations in air, soil, seawater and vegetation in European countries. Appropriate data averaged over 1997 and territories of individual countries are summarised in Table 8 presented in the end of this section.

First air concentrations are considered. Figure 6 demonstrates mean air concentrations for 1997.

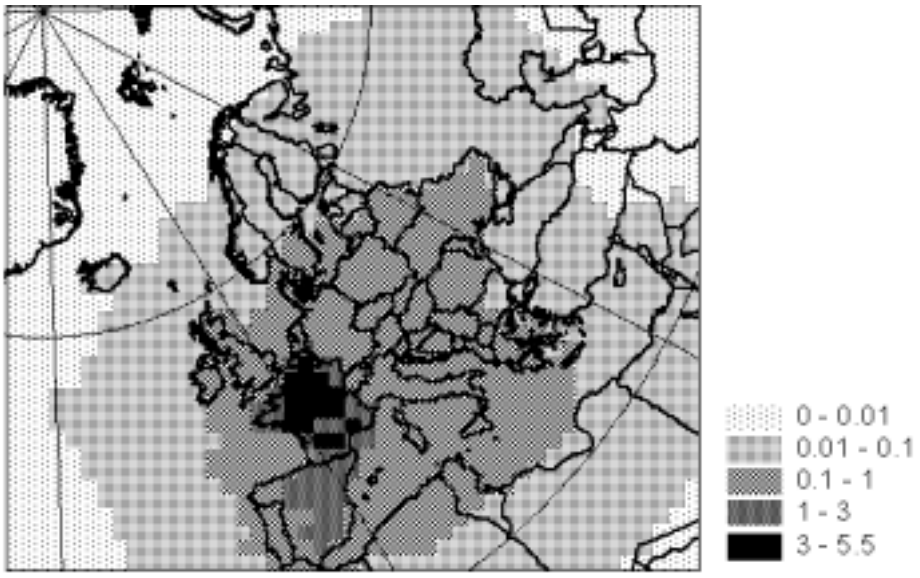
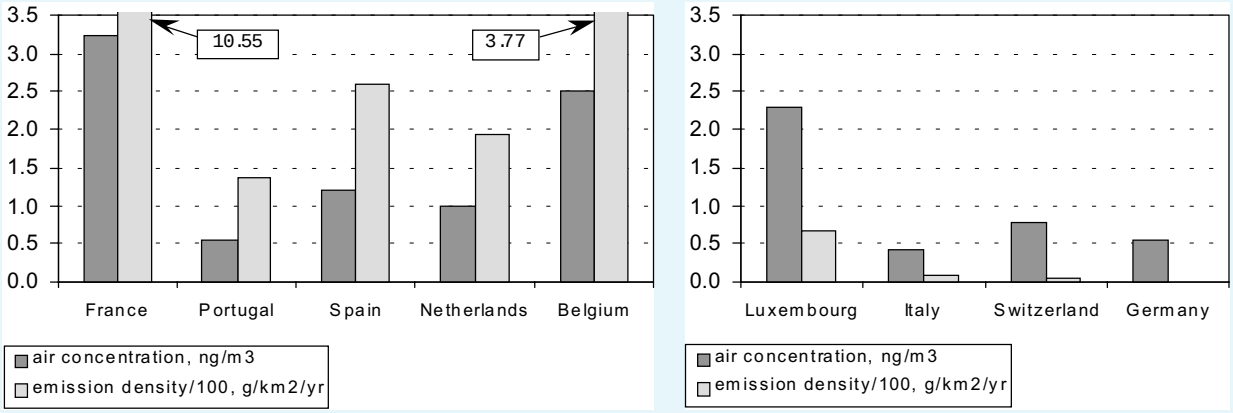


Figure 6. gamma-HCH air concentrations in 1997 [ng.m^{-3}]

The comparison of Figures 5 and 6 shows that maximum air concentration region coincides with maximum emission region. The highest deposition densities (1-4 ng.m^{-3}) are observed in France, Belgium, Luxembourg and Spain, slightly lower densities (0.5-1 ng.m^{-3}) - were recorded in the Netherlands, Switzerland and Germany.

On the basis of the relationship between air concentrations and emission densities among countries in which gamma-HCH air concentration level exceeds the average value (0.39 ng.m^{-3}) two groups of countries are distinguished (Figure 7).



a) b)

Figure 7. Gamma-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.km^{-2}.yr^{-1}) in these countries (Figure 7a).

The second group includes countries where high air concentrations are accompanied by relatively low emission fluxes (less than 100 g.km-2.yr^{-1}). These countries are Germany, Italy, Switzerland and Luxembourg (Figure 7b). Most probably a tangible level of air concentrations in these countries is mainly explained by the effect of gamma-HCH transboundary transport from sources of other countries.

Separately stand Austria and Slovenia, which are characterised by low air concentrations while soil concentrations exceed mean European level. Figure 8 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^{-1}) are observed in France, Luxembourg and Belgium, slightly lower (5-15 ng.g^{-1}) in Switzerland, the Netherlands, Germany, Spain, Italy and Austria. Note that in Luxembourg gamma-HCH soil concentration is high (22 ng.g^{-1}) whereas the emission level is relatively low (57.7 g.km^{-2}.yr^{-1}).

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

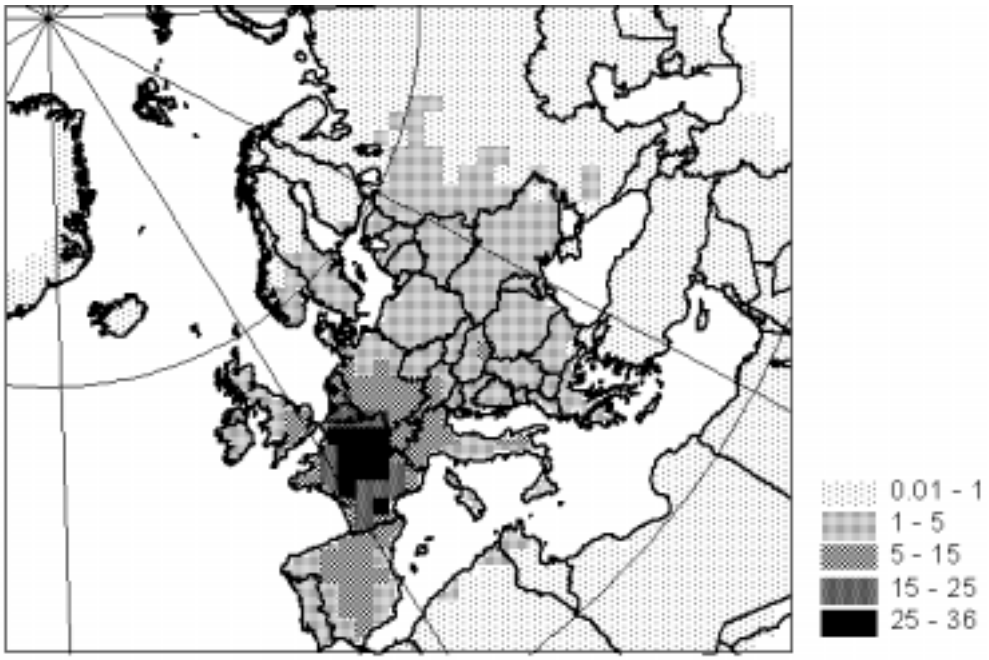


Figure 8. Gamma-HCH soil concentrations in 1997 [ng.g^{-1}]

The map of mean marine concentration distribution during1997 is demonstrated in Figure 9.

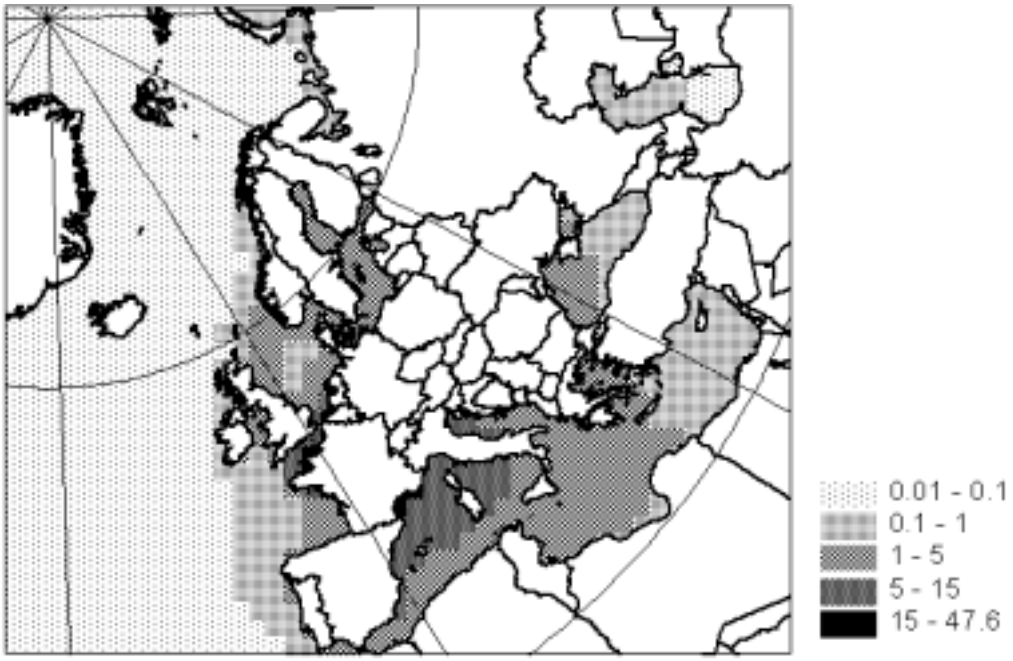


Figure 9. Gamma-HCH sea concentrations in 1997 [mg.m^{-3}]

As evident from the map gamma-HCH maximum concentrations are observed in sea regions located close to major emission sources. gamma-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 10 illustrates the map of gamma-HCH mean concentration distribution in vegetation in 1997.

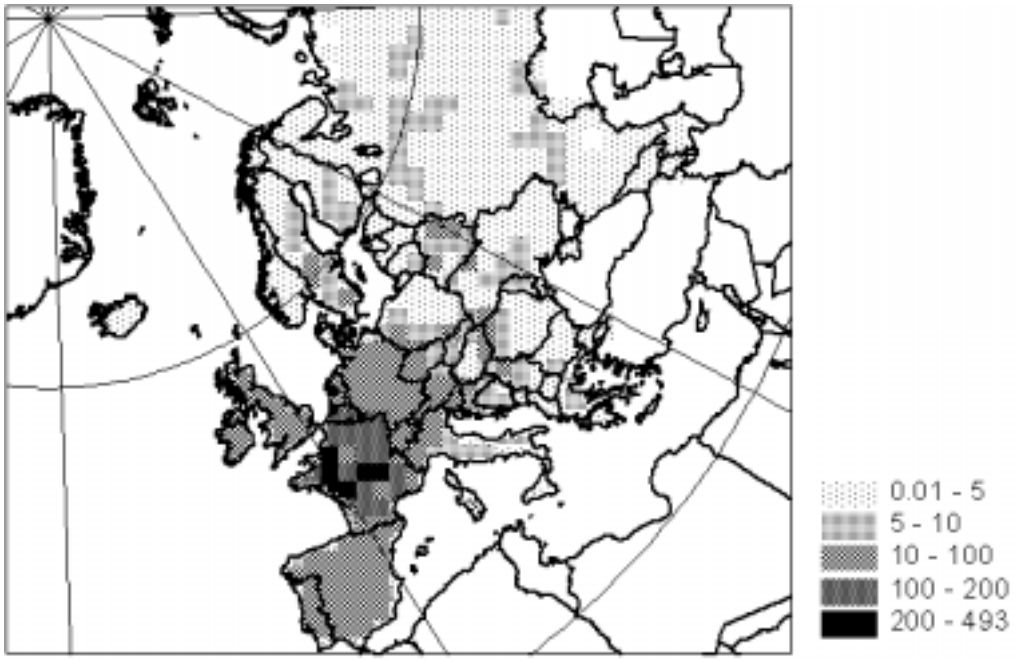


Figure 10. Gamma-HCH concentration in vegetation in 1997 [ng.g^{-1} d.w.]

As seen from the map, calculated vegetation concentrations correlate well with emission values. Maximum concentrations (100-170 ng.g^{-1} of dry weight) are observed in France, Luxembourg and Belgium. Concentrations from 20 to 100 ng.g^{-1} of dry weight are observed in Switzerland, the Netherlands, Spain, Ireland, Germany, Great Britain and Austria.

All the presented results on gamma-HCH content in different media of different European countries are summarized in Table 8*. The shaded countries have air and/or soil concentrations exceeding mean European levels (0.39 ng.m^{-3} and 3.88 ng.g^{-1}, respectively).

Table 8. Gamma-HCH concentrations and depositions in European countries in 1997

Country	Air [ng.m^{3}]	Soil [ng.g^{-1}]	Vegetation [ng.g^{-1} d.w.]	Sea [ng.m^{-3}]	Deposition density [g.km^{-2}.yr^{-1}]	Total depositions [t.yr^{-1}]	Emission density [g.km^{-2}.yr^{-1}]	Emissions [t.yr^{-1}]
Albania	0.15	1.50	2.37	2,668.5	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	2,422	92.16	2.794	377.4	11.441
Bosnia Herzegovina	0.2	2.74	6.8	3,024.1	5.86	0.294	10.3	0.515
Bulgaria	0.11	1.34	2.54	1,565.8	4.78	0.516	0.0	0.000
Croatia	0.24	3.58	8.35	4,621	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	3,931.5	15.72	0.638	0.0	0.000
Estonia	0.07	1.26	2.4	2,139.3	4.22	0.187	0.1	0.005
Finland	0.03	0.73	3.81	1,497.7	1.92	0.627	0.0	0.000
France	3.25	22.30	165.63	1,848.5	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	5,577.7	26.99	9.627	0.0	0.000
Greece	0.17	1.25	3.74	1,405.3	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	1,251.7	14.86	0.973	33.1	2.167
Italy	0.42	5.79	11.27	6,080.4	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	2,487.7	6.07	0.372	0.0	0.002
Lithuania	0.11	1.66	4.68	2,742.7	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	2,535.4	4.07	0.104	2.5	0.063
Malta	0.18	0.99	0	2,275.8	2.23	0.001	0.0	0.000
Netherlands	0.99	9.05	38.73	1,292.7	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	3,868.4	8.49	2.619	0.9	0.283
Portugal	0.55	3.08	16.62	2,239.7	13.18	1.105	136.4	11.441
Republic of Moldova	0.14	1.71	6.18	2,101.1	4.87	0.159	11.1	0.364
Romania	0.14	1.86	5	1,646.3	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	8,372.5	12.19	0.222	10.0	0.182
Spain	1.2	5.98	31.27	5,587.2	21.77	10.337	258.8	122.91
Sweden	0.06	1.21	5.39	2,183.8	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	1,459.3	5.02	2.985	15.2	9.074
United Kingdom	0.31	3.20	28.18	4,605	16.71	3.88	72.5	16.843
Yugoslavia	0.18	2.78	6.85	2,673.9	7.01	0.712	15.2	1.538

Comparison of gamma-HCH modelling results with measurements

Air concentrations

Table 9 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 9. Gamma-HCH mean annual air concentrations [ng.m^{-3}]

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 gamma-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%.

PBT compounds in ambient air are monitored in the area of Kosetice observatory (professional observatory of Czech Hydrometeorological Institute located in south Bohemia). Observatory Kosetice was established as a regional background station of international monitoring (EMEP, GAW, GEMS) and national monitoring programmes (monitoring of Czech MOE, Project TOCOEN, GEOMON) (Holoubek *et al.*, 2000c; Vana *et al.*, 1997).

Table 10. Chlorinated pesticides in ambient air in Kosetice

Concentrations [pg.m^{-3}]	alpha-HCH	beta-HCH	gamma-HCH
Average	193	130.6	416.2
Stand. Deviation	126.5	54.2	190.2
Minimum	100	100	100
Maximum	670	245	720
Median	150	100	400

Table 11 shows the results of measurements from 1996 to 1999. Figure 11 depicts profiles of observed concentrations of HCHs (Holoubek *et al.*, 2000b).

Table 11. Observed concentrations of PCBs and OCPs, Kosetice observatory, 1996 - 1999

OCPs	Minimum	Maximum	Arithmetic mean	Geometric mean	Median
Gas phase [ng.m^{-3}]					
alpha-HCH	0.0003	0.2120	0.0274	0.0000	0.0225
beta-HCH	0.0003	0.0700	0.0049	0.0000	0.0003
gamma-HCH	0.0004	0.6410	0.0503	0.0000	0.0120
delta-HCH	0.0003	0.0100	0.0004	0.0000	0.0003
Σ-HCHs	0.0000	0.7710	0.0827	0.0000	0.0435
Aerosol [ng.m^{-3}]					
alpha-HCH	0.0003	0.0210	0.0042	0.0000	0.0030
beta-HCH	0.0003	0.0200	0.0009	0.0000	0.0003
gamma-HCH	0.0004	0.0890	0.0058	0.0000	0.0020
delta-HCH	0.0003	0.0020	0.0003	0.0000	0.0003
Σ-HCHs	0.0000	0.1040	0.0108	0.0000	0.0050
Gas phase + aerosol [ng.m^{-3}]					
alpha-HCH	0.0006	0.2130	0.0315	0.0000	0.0270
beta-HCH	0.0005	0.0740	0.0059	0.0000	0.0005
gamma-HCH	0.0008	0.6990	0.0561	0.0000	0.0170
delta-HCH	0.0005	0.0100	0.0006	0.0000	0.0005
Σ-HCHs	0.0020	0.8410	0.0935	0.0000	0.0510

HCHs and some other chlorinated PBTs are present in the atmosphere today due to their volatilisation from soil and sediments, i.e. as secondary inputs from old deposits, and also due to a long-range atmospheric transport from regions in which they are still used. These results reflect the global trends. HCHs occurrence remains at the level of the European background. From the HCHs pictures we can see the decreasing tendency during this period (See Figure 11).

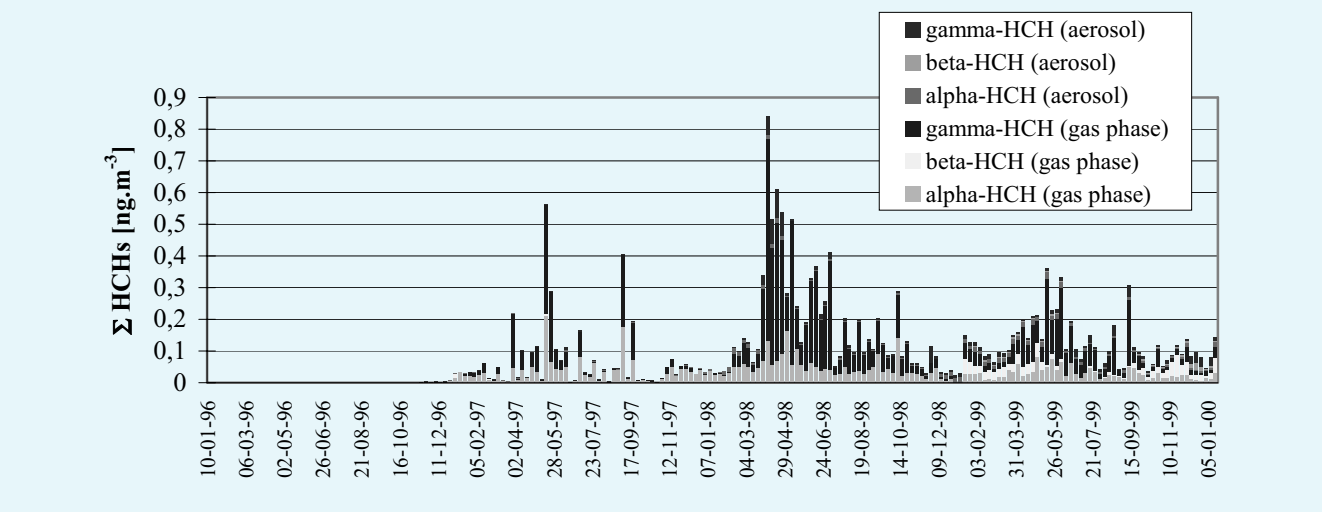


Figure 11. The middle European background levels of HCHs isomers in ambient air - observatory Košetice, south Bohemia, 1996-1999

Concentrations in precipitation

Table 12 presents calculated and measured mean annual concentrations of gamma-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 fold higher than the calculated ones. The correlation between measured and calculated values is rather high alongside total underestimation of concentrations in precipitation.

Table 12. Mean annual gamma-HCH concentrations in precipitation [ng.l^{-1}]

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 previously mentioned, gamma-HCH modelling on the hemispherical level can result in an increase of concentration values in precipitation.

Concentrations in soil and vegetation

Table 13 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 rainwater and gamma-HCH transport by river currents.

Table 13. Gamma-HCH concentration in the soil at station CZ3 [ng.g^{-1}]

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

Table 14 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 14. Gamma-HCH concentrations in vegetation at station CZ3 [ng.g^{-1} 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 seawater, soil and vegetation is insufficient for model verification.

Concentrations in seawater

Table 15 contains gamma-HCH calculated concentrations in the Baltic Sea. Measurement data are taken from POPCYCLING-Baltic project where different data on the Baltic Sea are systematised. These data are cited in (Gaul, 1992; Biziuk *et al.*, 1999; Harner *et al.*, 1995).

Table 15. Gamma-HCH concentrations in sea water [pg.l^{-1}]

Year	Measured	Calculated	Meas./Calc.
1982	7,600	9,330	0.81
1983	4,700	8,876	0.53
1985	4,550	7,780	0.58
1986	3,900	7,164	0.54
1987	3,500	6,546	0.53
1988	2,925	5,862	0.50
1989	2,850	5,260	0.54
1990	2,536	4,665	0.54
1991	2,250	4,358	0.52
1996	10,054	2,897	3.47
1997	23,000	2,884	7.98
Mean	7,269	4,321	1.68
Mean 1982 - 1991	3,868	6,649	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}.

Gamma-Hexachlorocyclohexane - long-term trends

This part is dedicated to the analysis of long-term trends in gamma-HCH accumulation in various compartments (the atmosphere, sea water, vegetation, forest litter) within the EMEP grid. The calculations were performed by MSCE-POP model including the description of pollutant transport by sea currents. Due to high vapour pressure over subcooled liquid gamma-HCH fraction sorbed by aerosol does not exceed 1%, the aerosol phase was neglected in calculations (Shatalov *et al.*, 2000).

In modelling we used the emission inventory made under POPCYCLING-Baltic project (Pacyna *et al.*, 1999), which allowed us to study long-term trends of the content and concentrations in different compartments (the atmosphere, sea water, vegetation, forest litter); emission data are available for 1970-95. gamma-HCH emissions in 1996 and 1997 were fixed at the level of 1995. In the previous report (Pekar *et al.*, 1999) gamma-HCH transport has been calculated for a long period of time. However, the same emission distribution determined for 1990 (Berdowski *et al.*, 1997) was employed. The application of long-term trends gives a possibility to refine the long-term dynamics of gamma-HCH accumulation in different environmental compartments.

In order to take into account primary gamma-HCH accumulation in the soil and seawater the model was spun-up covering 15 years on the basis of the emission and meteorological data of 1970.

Figure 12 shows the graphs of gamma-HCH content dynamics in different environmental compartments. This dynamics is essentially determined by emission variations. The graph of emissions over the whole EMEP region from year to year is shown in Figure 12a. As seen from this graph an essential decrease of gamma-HCH emission begins with 1980 and by late 1997 it amounts to about 30% of the initial value.

In accordance with the emission dynamics the total content of gamma-HCH in the atmosphere also tends to decrease (Figure 12b).

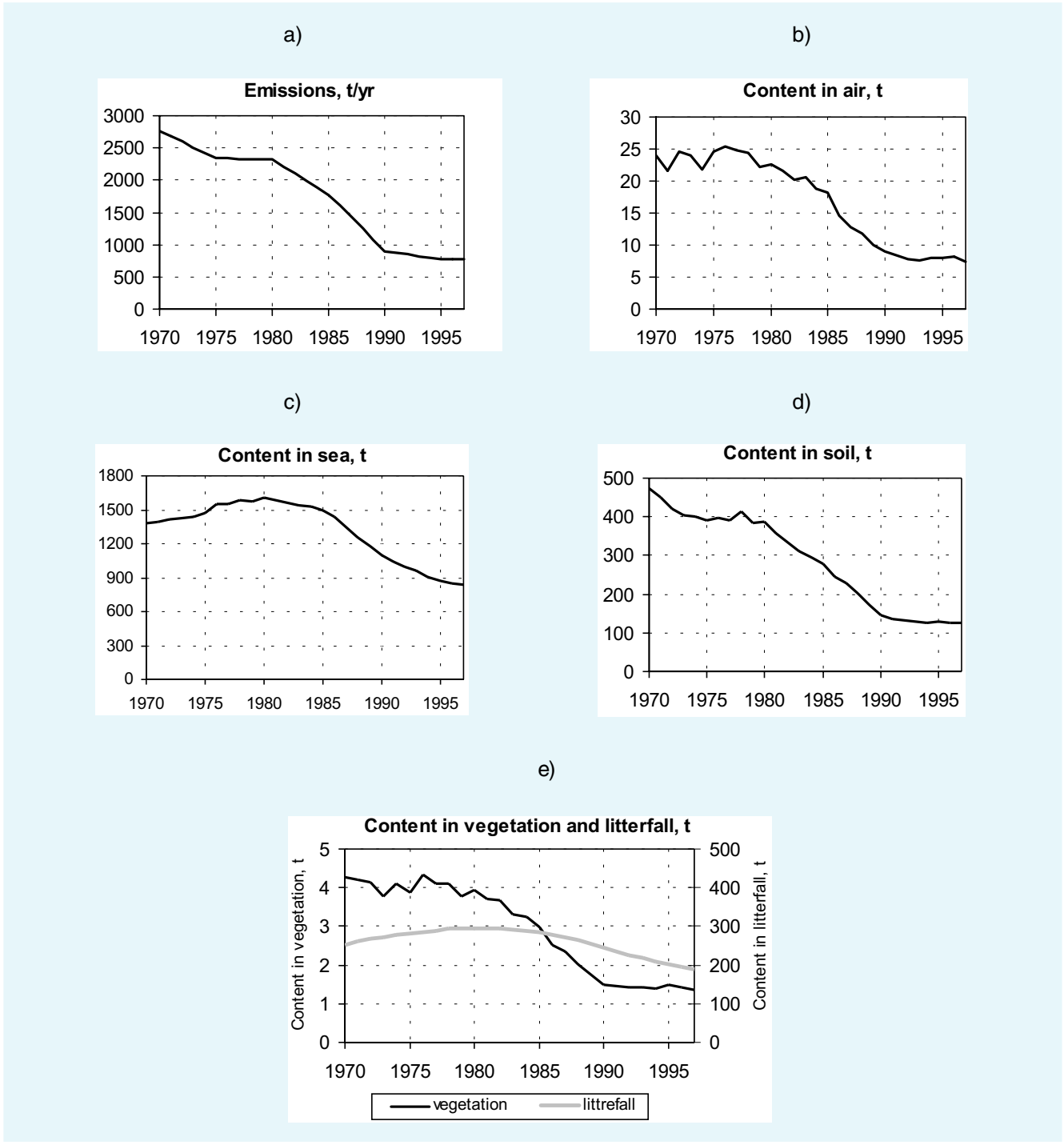


Figure 12. Gamma-HCH content in compartments for 1970-97 obtained with model spin-up (1940 - 1969) in comparison with emission dynamics

Obviously the atmosphere is most non-inertial among other environmental components. Atmospheric concentrations respond immediately to emission variations because the characteristic airborne transport time within the EMEP domain is estimated by days.

Figure 12 shows the plot of gamma-HCH content in vegetation and forest litter. As evident from it the vegetation content repeats on the average emission variations, i.e. the vegetation sluggishness is not high. The content in the forest litter, however, responds to the emission reduction with some delay (as it was mentioned above the intensive emission decrease began in 1980 whereas the maximum content in the forest litter was in 1982-83). It happens because the pollutant transport from the forest litter to soil is slow enough (the characteristic time is about 10 years).

It should be remembered that due to lack of reliable data on degradation rates in vegetation at present this process is neglected in the model. The investigation of a possible effect of the degradation process on pollutant accumulation in vegetation has been made in the previous report (Pekar *et al*., 1999).

Figure 12 illustrates the dependence of gamma-HCH soil content on time. As evident from this graph gamma-HCH soil content is correlated with the emission values. In other words, the soil is a weakly inertial media for gamma-HCH at least from the viewpoint of long-term dynamics (on annual time scale).

Finally Figure 12 demonstrates gamma-HCH content in the seawater. It is evident that the response of seawater content lags behind the emission variation (the delay is about 5-6 years). It is conditioned by the fact that the degradation rate in the sea is considerably lower than in the soil. Gamma-HCH half-life in the sea is 5 years whereas for soil this value is not more than a year (Strand and Hov, 1996). Consequently for sea during the period covered by calculation (28 years) the total degradation amount exceeds maximum content by 3.6 fold whereas in the soil the total degradation for the same period exceeds maximum content almost in 20 times.

Conclusions

Using MSCE-POP model the simulation of gamma-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 gamma-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. gamma-HCH high air concentrations (0.5-3.5 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 ng.m^{-3}) and low emission densities. Obviously air concentration levels in these countries are explained by the impact of gamma-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 gamma-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.

The mass balance in the compartments in question by 1997 is presented in the Figure 13. This scheme demonstrates gamma-HCH total mass redistribution (both emitted and accumulated) over basic processes and compartments for the simulation period (Shatalov *et al*., 2000). About 60% of the total emission is transported outside the EMEP grid and the rest is redistributed between the media. Of the latter amount 48% comes to soil, 33% to sea, 18% to air and 1% to vegetation. The degradation process diminishes media contents. The analysis of the final content in media and contributions of the media to the degradation process follows.

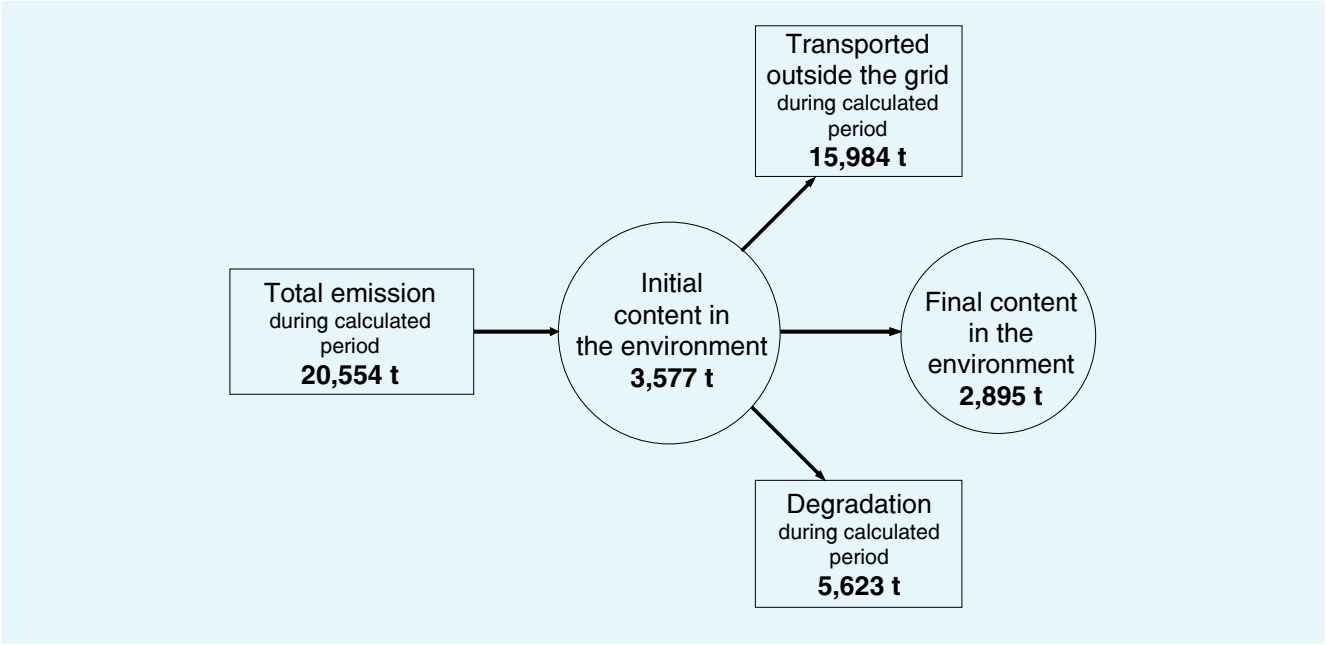


Figure 13. Gamma-HCH redistribution over main processes and compartments in the period covered by calculations

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