Annex A

EMISSIONS

The objective of the Protocol on POPs "is to control, reduce or eliminate discharges, emissions and losses of persistent organic pollutants" [*ECE/EB.AIR/60*]. The Protocol envisages that Parties shall submit official information about POP emissions to the UN ECE Secretariat. Official data on the emission totals of PAHs, PCDD/Fs, HCB and PCBs were reported by 36 countries for the period from 1990 to 2003 (for at least one year). The officially reported emission data are available from WEBDAB: *http://webdab.emep.int.* The number of countries reporting data on POP emissions increased considerably during last years. Analysis of the official data was carried out in this Annex. Nevertheless, to estimate total emission values and their spatial distribution in the European region expert estimates still have to be used for modelling. For hemispheric modelling of HCB and PCBs, only expert estimates of their emissions were used.

A.1. Polycyclic aromatic hydrocarbons

Major sources of PAH emissions are identified in the Protocol on POPs [*ECE/EB.AIR/60*]. They are divided into two groups: stationary and mobile. Stationary sources: domestic wood and coal heating; open fires (refuse burning, forest fires and after-crop burning); coke and anode production; aluminium production (via Soederberg process); wood preservation, except for the countries for which this category does not make a significant contribution to its total emissions of PAH (as defined in Annex III). Mobile sources: exhaust from diesel and gasoline vehicles.

According to the Protocol on POPs for the purposes of emission inventories four indicator compounds of PAHs should be used. They are benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F) and indeno[1,2,3-cd]pyrene (I_P) [*ECE/EB.AIR/60, Annex III*). "The inclusion of these PAHs in the list of controlled priority pollutants is mainly based on their carcinogenicity, spread and persistence in different environmental compartments" [*Mantseva et al.*, 2002]. The official information on total emission data of these four PAHs is available for 22 European countries for 1990-2003 (for at least one year). 17 countries of them are Belarus, Croatia, the Czech Republic, Denmark, Estonia, France, Germany, Hungary, Iceland, Ireland, Lithuania, Luxembourg, Monaco, Poland, Republic of Moldova, Slovakia and the United Kingdom submitted the emissions of each of four indicator compounds for the indicated period (for at least one year). For the first time, Croatia, France, Germany and Iceland reported these emissions. The Russian Federation submitted only B[a]P emissions. The values of emissions in Armenia, Azerbaijan, Georgia, Latvia and Ukraine are taken from the expert estimates prepared by *V.Tsibulsky et al.* [2001]. The value of emissions in Cyprus is based on the data prepared by *J.Berdowski et al.* [1997]. For the rest countries the expert estimates of B[a]P emissions [*Pacyna et al.*, 1999] are used.

In addition there are the emission totals of PAHs from 9 countries. The PAH emissions from Bulgaria (the total emissions of 10 individual PAH compounds), Belgium (the total emissions of 14 individual PAH compounds), as well as from the Netherlands, Slovenia, Spain and the Ukraine (the composition of PAHs is unknown) were not used in the calculations. The chemical composition of PAH national emissions in these countries does not allow to determine the values of B[a]P, B[b]F, B[k]F and I_P emissions. The PAH emissions from Canada, Kyrgyzstan and the United States were not used as they are outside the EMEP area.

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

Benzo[a]pyrene

Official data on B[a]P emissions were submitted by 18 European countries for the period from 1990 to 2003 (for at least one year). For Austria, Finland, Hungary, Italy, Luxembourg, Monaco, Norway and Sweden the B[a]P emissions were estimated on the basis of the fractions of B[a]P in officially submitted PAH emission totals. These fractions were calculated on the basis of the data from [*Baart et al.*, 1995]. According to this paper, the mean B[a]P contribution to the total emissions of the 4 indicator is around 29%, and to the total emission of the 6 Borneff is 8%. For Croatia, Lithuania, Poland, Republic of Moldova and Slovakia fractions were calculated on the basis of their official data.

The annual total B[a]P emissions (1990-2003) used for modelling this year are displayed in Table A.1. It includes official data on B[a]P emissions (grey background & in bold); B[a]P emissions estimated on the basis of the official total PAH emission data (in bold) and expert estimates.

Table A.1. *B[a]P emissions (official data & expert estimates) for the period from 1990 to 2003, t/y*

Country	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003
Albania	0.11	0.13	0.16	0.18	0.20	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22
Armenia	0.24	0.24	0.25	0.25	0.26	0.26	0.27	0.27	0.27	0.27	0.27	0.27	0.27	0.27
Austria	**5.24**	**5.40**	**4.05**	**3.04**	**2.78**	**2.88**	**3.21**	**2.78**	**2.66**	**2.57**	**2.38**	**2.64**	**2.53**	**2.74**
Azerbaijan	1.19	1.34	1.50	1.66	1.81	1.97	2.02	2.06	2.06	2.06	2.06	2.06	2.06	2.06
Belarus	10.23	10.23	10.23	10.23	10.23	10.23	10.23	10.23	10.23	10.23	10.23	**10.23**	**10.46**	**9.51**
Belgium	4.36	4.16	3.96	3.76	3.55	3.35	3.35	3.35	3.35	3.35	3.35	3.35	3.35	3.35
Bosnia& Herzegovina	8.81	7.95	7.09	6.24	5.38	4.52	4.52	4.52	4.52	4.52	4.52	4.52	4.52	4.52
Bulgaria	7.57	7.38	7.23	7.05	6.88	6.71	6.71	6.71	6.71	6.71	6.71	6.71	6.71	6.71
Croatia	**4.38**	4.10	3.82	3.54	3.26	2.98	**2.70**	**2.66**	**2.49**	**2.30**	**2.63**	**2.03**	**2.06**	2.06
Cyprus	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015	0.015
Czech Republic	18.67	17.79	16.91	16.02	15.14	14.26	13.32	12.39	11.45	10.51	9.58	8.64	**7.70**	**6.40**
Denmark	**1.96**	**2.26**	**2.26**	**2.43**	**2.37**	**2.36**	**2.50**	**2.48**	**2.24**	**2.33**	**2.72**	**2.99**	**2.89**	**2.97**
Estonia	3.28	3.28	3.28	3.28	3.28	3.28	3.28	3.28	3.28	3.28	3.28	3.28	**3.28**	**3.95**
Finland	**4.41**	**4.29**	**4.33**	**4.40**	**4.38**	**4.74**	**4.44**	**4.50**	**4.55**	**4.45**	**4.25**	**4.57**	**4.72**	**3.92**
France	**12.41**	**14.71**	**13.42**	**12.96**	**10.82**	**10.76**	**11.36**	**10.17**	**10.62**	**10.24**	**9.78**	**10.10**	**9.17**	**10.02**
Georgia	1.59	3.55	5.51	7.47	9.43	11.39	11.38	11.38	11.38	11.38	11.38	11.38	11.38	11.38
Germany	14.04	14.04	14.04	14.04	**14.04**	14.04	14.04	14.04	14.04	14.04	14.04	14.04	14.04	14.04
Greece	5.69	5.13	4.57	4.01	3.45	2.89	2.89	2.89	2.89	2.89	2.89	2.89	2.89	2.89
Hungary	**10.56**	**9.73**	**6.95**	**6.46**	**5.87**	**5.41**	**5.06**	**4.84**	**4.28**	**4.37**	**4.45**	**4.44**	**5.96**	**6.70**
Iceland	**0.02**	**0.02**	**0.02**	**0.03**	**0.03**	**0.03**	**0.03**	**0.03**	**0.03**	**0.04**	**0.05**	**0.05**	**0.06**	**0.06**
Ireland	3.30	3.30	3.30	3.30	3.30	3.30	3.30	3.30	3.30	3.30	3.30	3.30	**3.30**	**3.14**
Italy	**26.65**	**30.68**	**29.54**	**30.15**	**31.20**	**32.20**	**29.49**	**32.91**	**31.86**	**34.64**	**34.59**	**34.94**	**31.98**	31.98
Kazakhstan	8.92	8.21	7.50	6.79	6.08	5.37	5.37	5.37	5.37	5.37	5.37	5.37	5.37	5.37
Latvia	1.86	2.01	2.17	2.32	2.47	2.62	4.74	6.85	6.85	6.85	6.85	6.85	6.85	6.85
Lithuania	19.23	19.23	19.23	19.23	19.23	19.23	19.23	**19.23**	**14.16**	**13.12**	**10.89**	**30.05**	**16.71**	**18.00**
Luxembourg	0.03	0.03	0.03	0.03	**0.03**	**0.02**	**0.02**	**0.01**	**0.01**	**0**	**0**	0	0	0
Monaco	**0.002**	**0.002**	**0.003**	**0.003**	**0.003**	**0.003**	**0.003**	**0.004**	**0.003**	**0.004**	**0.004**	**0**	**0**	**0**
Netherlands	4.74	4.25	3.76	3.27	2.78	2.29	2.29	2.29	2.29	2.29	2.29	2.29	2.29	2.29
Norway	**4.63**	**4.45**	**4.30**	**4.49**	**4.45**	**4.49**	**4.66**	**4.64**	**4.58**	**4.20**	**4.45**	**4.85**	**5.47**	**4.40**
Poland	**46.17**	**50.55**	**49.79**	**73.43**	**67.11**	**68.82**	**65.22**	**56.61**	**51.10**	**51.01**	**48.30**	**46.78**	**46.00**	46.00
Portugal	1.50	1.53	1.56	1.58	1.61	1.64	1.64	1.64	1.64	1.64	1.64	1.64	1.64	1.64
Republic of Moldova	**1.17**	**0.93**	**0.76**	**0.62**	**0.59**	**0.81**	**0.68**	**0.96**	**0.90**	**0.83**	**0.45**	**0.44**	**0.52**	0.52
Romania	27.90	26.12	24.34	22.55	20.77	18.98	18.98	18.98	18.98	18.98	18.98	18.98	18.98	18.98
Russian Federation	**18.26**	**17.30**	**15.60**	**15.29**	**15.45**	**15.28**	**15.02**	**14.95**	**14.71**	**15.32**	**15.43**	16.67	**17.90**	17.90
Serbia& Montenegro	21.91	19.78	17.64	15.51	13.38	11.24	11.24	11.24	11.24	11.24	11.24	11.24	11.24	11.24
Slovakia	**14.28**	12.74	11.21	9.67	8.13	**6.60**	6.44	**6.29**	**5.44**	**5.68**	**6.06**	**5.92**	**5.95**	**6.22**
Slovenia	4.70	4.24	3.78	3.33	2.87	2.41	2.41	2.41	2.41	2.41	2.41	2.41	2.41	2.41
Spain	12.42	11.86	11.29	10.73	10.17	9.61	9.61	9.61	9.61	9.61	9.61	9.61	9.61	9.61
Sweden	**10.85**	**10.60**	**9.39**	**8.71**	**7.94**	**7.91**	**7.72**	**6.32**	**6.26**	**5.98**	**4.90**	**5.36**	**5.29**	**5.83**
Switzerland	2.36	2.22	2.08	1.94	1.79	1.65	1.65	1.65	1.65	1.65	1.65	1.65	1.65	1.65
The FYR of Macedonia	3.71	3.35	2.99	2.63	2.26	1.90	1.90	1.90	1.90	1.90	1.90	1.90	1.90	1.90
Ukraine	27.48	25.04	22.61	20.17	17.73	15.29	15.70	16.10	16.10	16.10	16.10	16.10	16.10	16.10
United Kingdom	**66.39**	**60.79**	**52.08**	**32.91**	**29.63**	**23.41**	**12.06**	**10.26**	**8.76**	**7.70**	**5.74**	**6.39**	**5.45**	**4.03**
Total	443	435	404	396	372	357	341	332	316	316	307	327	311	310

Benzo[b]fluoranthene, benzo[k]fluoranthene, indeno(1,2,3-c,d)pyrene

Official data on B[b]F, B[k]F and I_P (3 PAHs) emissions were submitted by 16 European countries for the period from 1990 to 2003 (for at least one year). At present, expert estimates of the 3 PAH emissions in the European region are available only for 1990 [*Baart et al*., 1995]. For modelling the 3 PAH emission values are estimated on the basis of the ratio of each from 3 PAHs to B[a]P emission values in European countries, calculated using official information and data available in [*Baart et al.*, 1995]. Similarly to B[a]P, for countries that officially submitted PAH emission totals, 3 PAH emissions are estimated on the basis of their fractions in the PAH totals. According to *A.Baart et al.*, 1995 the mean values of ratios of the 3 PAHs to the B[a]P emissions and their contributions to the PAH emission totals are given in Table A.2.

Table A.2. *Mean values of ratios and contributions for the 3 PAHs*

	B[b]F	B[k]F	I_P
Ratio to B[a]P	0.96	0.67	0.86
Contribution to the 4 total PAHs, (%)	27	19	24
Contribution to the 6 Borneff, (%)	8	5	7

The annual total B[b]F, B[k]F and I_P emissions (1990&2003) used for modelling this year are shown in Table A.3. It includes official emission data (grey background & in bold); emissions estimated on the basis of the official total PAH emission data (in bold) and expert estimates.

Table A.3. *B[b]F, B[k]F and I_P emissions(official data & expert estimates) for 1990 and 2003, t/y*

Country	B[b]F		B[k]F		I_P	
	1990	2003	1990	2003	1990	2003
Albania	0.11	0.22	0.11	0.22	0.11	0.22
Armenia	0.32	0.34	0.23	0.12	0.40	0.69
Austria	**5.42**	**2.83**	**2.80**	**1.46**	**3.84**	**2.01**
Azerbaijan	6.09	13.50	1.67	3.52	0.59	0.74
Belarus	20.33	**19.17**	5.52	**5.22**	5.67	**5.38**
Belgium	3.27	2.51	2.53	1.94	3.62	2.78
Bosnia& Herzegovina	8.81	4.52	5.90	3.03	7.14	3.66
Bulgaria	7.57	6.71	5.30	4.69	6.06	5.37
Croatia	**5.74**	2.72	**1.93**	0.92	**3.12**	1.48
Cyprus	0.015	0.015	0.009	0.009	0.013	0.013
Czech Republic	17.55	**6.89**	8.03	**2.63**	14.94	**5.36**
Denmark	**2.56**	**3.95**	**0.87**	**1.34**	**1.64**	**2.22**
Estonia	5.02	**4.42**	2.38	**2.37**	2.37	**2.48**
Finland	**4.89**	**4.34**	**2.68**	**2.38**	**3.78**	**3.36**
France	**14.41**	**11.58**	**9.28**	**7.72**	**7.48**	**6.18**
Georgia	1.83	13.58	0.67	4.57	3.92	33.96
Germany	14.46	14.46	9.41	9.41	13.90	13.90
Greece	6.26	3.18	3.98	2.02	5.12	2.60
Hungary	**11.88**	**7.37**	**6.60**	**3.79**	**10.56**	**4.30**
Iceland*	**0.005**	**0.005**	**0.04**	**0.10**	**0.01**	**0.02**
Ireland	4.49	**4.20**	2.29	**2.14**	2.09	**1.94**
Italy	**23.90**	28.67	**20.22**	24.26	**21.14**	25.36
Kazakhstan	8.74	5.26	4.64	2.79	9.45	5.69
Latvia	2.27	8.24	0.85	2.78	5.24	20.38
Lithuania	14.24	**10.62**	13.53	**7.63**	24.21	**10.39**
Luxembourg	0.03	0	0.03	0	0.03	0
Monaco	**0.002**	0	**0.001**	0	**0.002**	0
Netherlands	2.70	1.31	3.03	1.47	3.36	1.63
Norway	**4.19**	**3.99**	**3.62**	**3.44**	**2.02**	**1.93**
Poland	**50.31**	49.75	**15.28**	14.99	**47.60**	49.35
Portugal	1.50	1.64	1.25	1.15	1.20	1.31
Republic of Moldova	**1.37**	**0.71**	**0.60**	**0.31**	**3.04**	**1.65**
Romania	23.73	16.13	18.70	12.72	14.51	9.87
Russian Federation	17.89	17.54	9.50	9.31	19.36	18.97
Serbia& Montenegro	21.91	11.24	14.68	7.53	17.75	9.11
Slovakia	**5.88**	**1.89**	**17.18**	**7.78**	**4.61**	**1.47**
Slovenia	4.70	2.41	3.15	1.61	3.80	1.95
Spain	11.55	8.93	9.81	7.59	11.05	8.55
Sweden	**10.85**	**5.83**	**7.75**	**4.16**	**9.30**	**5.00**
Switzerland	1.86	1.31	1.67	1.17	0.59	0.41
The FYR of Macedonia	3.71	1.90	2.48	1.28	3.00	1.54
Ukraine	42.12	34.63	17.60	12.28	23.63	15.00
United Kingdom	**81.58**	**4.77**	**42.24**	**2.92**	**33.87**	**2.89**
Total	476	343	280	187	355	291

** National Total for the EMEP grid domain*

Dynamics of PAH emissions in Europe for the period from 1990 to 2003

According to the official data and expert estimates, the PAH emissions have decreased in Europe since 1990. The decrease of the PAH emissions is mainly due to the reduction in emissions in the United Kingdom, the Ukraine, the Czech Republic and Serbia and Montenegro. Time-series for PAH emissions are demonstrated in Fig. A.1. The decrease of each PAH emissions since 1990 is displayed in Table A.4. The reductions in PAH emissions vary from 18% (I_P) to 33% (B[k]F).

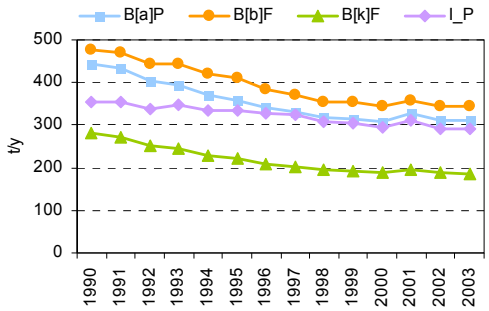


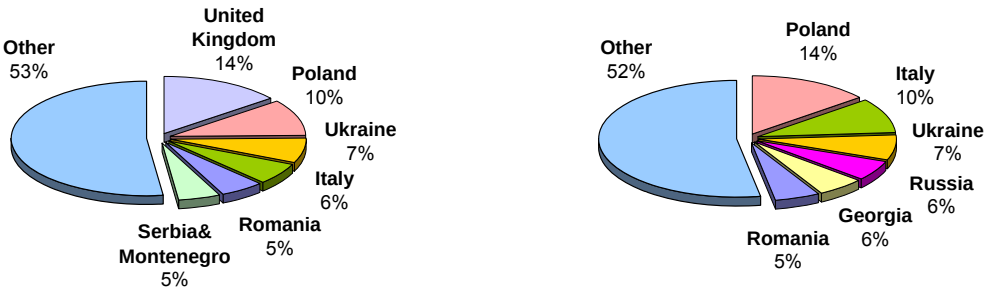
Fig. A.1. *Time-series for PAHs*

Table A.4. *Decrease of PAH emissions 1990-2003*

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

According to the official information, emissions of PAHs in the United Kingdom, Slovakia, Austria and Sweden between 1990 and 2003 dropped by 15, 2.4, 2 and 2 times, respectively. In Iceland and Denmark emissions have increased by 2.4 and 1.5 times, respectively since 1990. The slight increase of emissions in 2001 is the result of increase of Lithuanian emission data.

The contribution of individual countries to the total emissions of B[a]P, B[b]F, B[k]F and I_P in Europe in 1990 and 2003 is presented in Fig. A.2. The contributors to the whole European emissions were Italy, Poland, Romania and the Ukraine for both 1990 and 2003.



a b

Fig.A.2*. The contribution of individual countries to the total emissions of PAHs in Europe in 1990 (a) and 2003 (b)*

Spatial distribution of PAH emissions

The spatial distribution of B[a]P, B[b]F, B[k]F and I_P emissions in the period from 1990 to 2003 was prepared for modelling on the basis of the official data on spatial distribution of PAH emissions submitted by thirteen countries (Table A.5). For other countries, expert estimates were used [*Pacyna et al*., 1999; *Berdowski et al*., 1997]. Spatial distribution of PAH emissions in 1990 and 2003 is given in Fig. A.3.

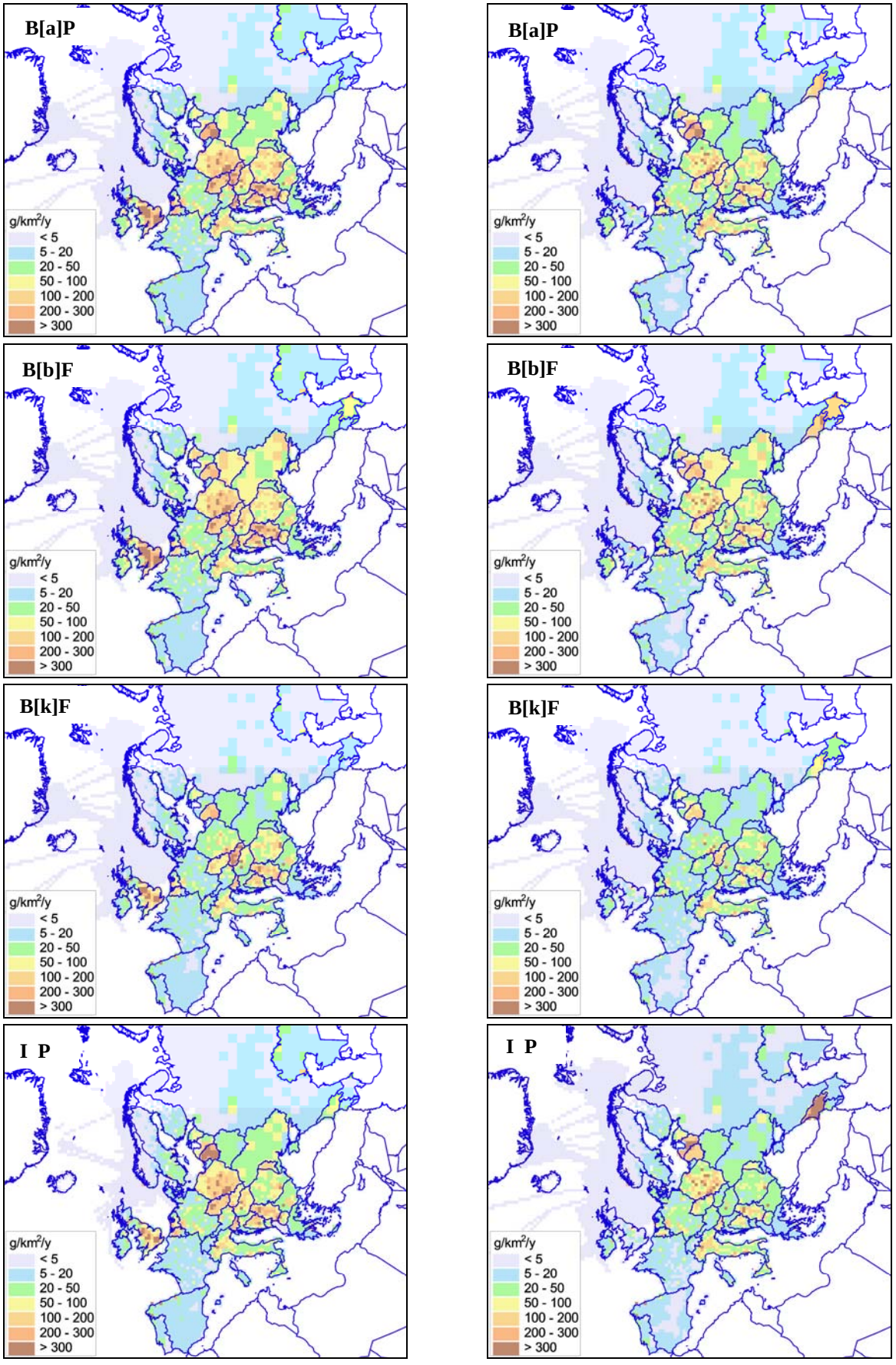


Fig. A.3. *Spatial distribution of PAH emissions in 1990 (a) and 2003 (b)*

Table A.5. *The list of countries submitted official data on spatial distribution of PAH emissions (1990-2003)*

Country	Year
Austria	1990, 1995, 2000
Belgium	
Bulgaria	
Denmark	
Hungary	
Iceland	
Norway	
Netherlands	1990, 1995
Finland	1990-2003
Spain	
United Kingdom	1999, 2000
France	2000
Poland	

High PAH emission intensity was characteristic of Bosnia and Herzegovina, the Czech Republic (except B[k]F), Lithuania, Poland (except B[k]F), Serbia and Montenegro, Slovakia (except B[b]F and I_P), Slovenia, and the United Kingdom in 1990. In 2003 high emission intensity was characteristic of Georgia, Latvia, Lithuania and some regions of Poland, as well as Azerbaijan (only B[b]F) and Slovakia (only B[k]F).

PAH emissions by sectors

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

The sector split for the national total emissions of PAHs is presented in Fig. A.4. Graphs show that the Residential sector is the major contributor. Its share varies from 19% in Republic of Moldova to 90% in the Czech Republic and on average it amounts to 58%. The predominant source in the Residential sector is combustion of wood. The second most important sector is the Metal Production. This sector is the largest source of PAHs for Iceland, Republic of Moldova and Norway.

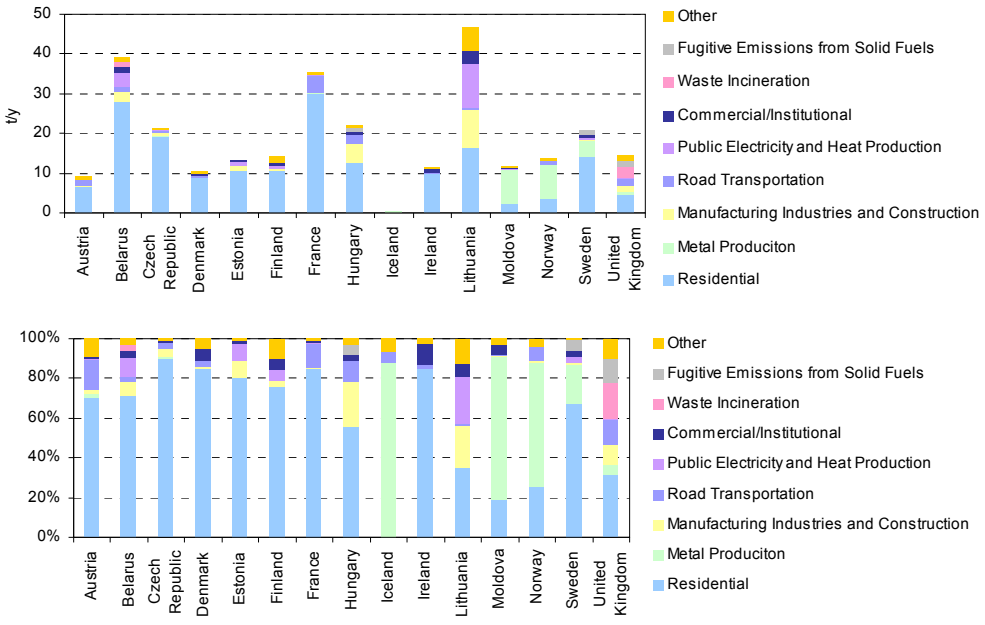


Fig. A.4*. Sector split for PAHs in Europe in 2003*

Official data on PAH emissions by sectors in Austria, Denmark, France, Iceland, Norway, Sweden and the United Kingdom are available for each year of the period from 1990 to 2003.

As an example, PAH emission time-series for the United Kingdom is demonstrated in Fig. A.5. The decreasing PAH emission trend is the result of the sharp drop in the Metal Production sector (by 99%) and the drop to zero in 1994 in the Field Burning of Agricultural Wastes sector.

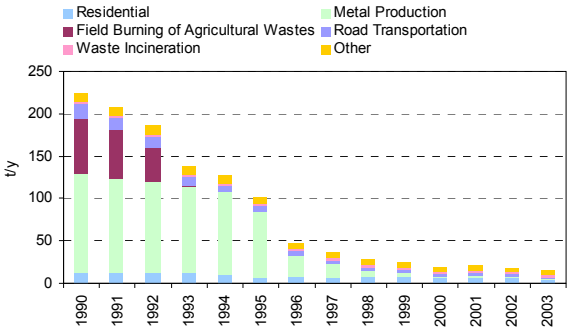
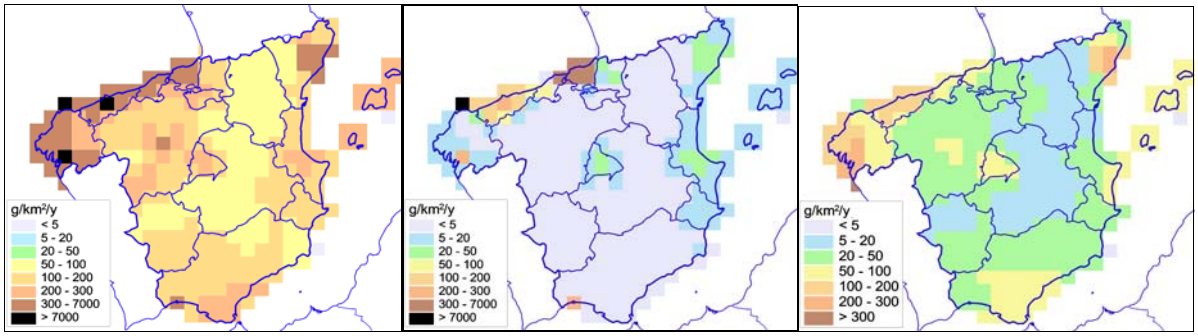


***Fig. A.5**. PAH emission time-series for the United Kingdom*

PAH gridded sector data

The official information on PAH gridded sector data for each of the relevant aggregated NFR (Nomenclature For Reporting) sectors [*EB.AIR/GE.1/2002/7, Table III B*] is submitted by Finland (2001-2003), France (2000), Iceland (1990/1995/2000) and Spain (1990-2003). As an example, the spatial distribution of the total PAHs and some of sector emissions for Spain in 2003 is given in Fig. A.6. As can be seen from the map 'Total PAHs', there are three hot spots. One of them, located in the north-east of province Galicia, belongs to the sector 01 'Combustion in Power Plants and Industry', which amounts to 30% from Total PAHs. The rest of them, located in the south-west of province Galicia and in the centre of province Asturias, belong to the sector 05 'Industrial Processes', which amounts to 27% from Total PAHs. The sectors 03 'Commercial Residential and Other Stationary Combustion', 07 'Agriculture' and 08 'Waste' approximately equal and on the whole amount to about 40% from Total PAHs.

Total PAHs ***NFR 01*** ***NFR 03***



NFR 05 ***NFR 07*** ***NFR 08***

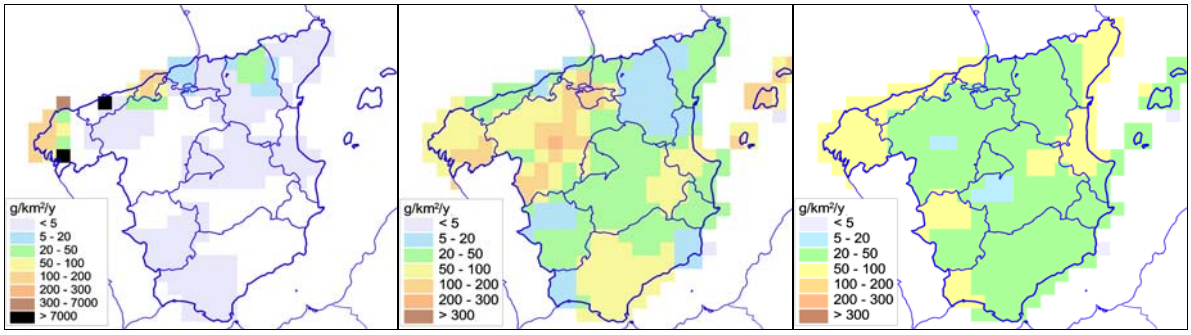


***Fig. A.6.** Spatial distribution of the total PAHs and some of sector emissions for Spain in 2003*

Seasonal PAH emission variations

The considerable contribution of the Residential sector to the total PAH emissions determines pronounced seasonal emission variation. In the model calculations data on the seasonal distribution of PAH emissions (Table A.6) taken from [*Baart et al.*, 1995] were used. According to [*Baart et al.*, 1995] the level of PAH emissions in winter is higher than in summer. In the model calculations the same seasonal distribution of PAH emissions was used for all the countries. It causes additional uncertainties to the calculation results.

Table A.6. *Seasonal distribution (%) of PAH emissions in Europe in 1990*

PAH	1st quarter	2nd quarter	3rd quarter	4th quarter
B[a]P	30	20	20	30
B[b]F	31	19	19	31
B[k]F	28	22	22	28
I_P	33	17	17	33

Uncertainties in PAH emission estimates

The most significant uncertainties in PAH emission estimates used in the model calculation result from the compilation of official data and various expert estimates, which in their turn are uncertain to a large extent. Qualitative and quantitative characteristics of uncertainties in national totals of PAH emissions are not available at present, except Denmark (uncertainty of the Danish PAH total emissions for 2003 is over 900% [*Illerup et al.*, 2005]).

According to [*Pacyna et al*., 1999] and [*Berdowski et al.*, 1994; *Berdowski et al*., 1997 и *Slooff et al*., 1989] “uncertainties were assessed within the factor of ±2-5. In [*Parma et al.*, 1995 and *Slooff et al*., 1989] it was emphasized that the time variation of emissions measured at an individual factory can be significant for different sampling intervals”.

A significant source of uncertainties in PAH emissions estimates is associated with the fractions used to convert the values of the total PAH emissions that include the 4 indicator PAHs or 6 Borneff PAHs to the values of B[a]P, B[b]F, B[k]F and I_P emissions.

The major uncertainty of estimates of B[b]F, B[k]F and I_P emissions used in the model calculations stems from the utilization of the ratio of B[b]F, B[k]F and I_P emissions to B[a]P emissions calculated on the basis of both official data and expert estimates.

Another source of uncertainties in PAH emissions estimates used in the model calculations is associated with the utilization of fairly rough estimates of the PAH emission seasonal variations. As it is seen from comparison of measured and calculated values of B[a]P contamination (see below), the refinement of emission seasonal variations is essential for evaluation of the pollution in the EMEP region.

A.2. Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) contaminate the environment as by-products of various chemical processes and in the course of combustion of chlorinated substances. The major sources of PCDD/F emissions are specified in the Protocol on Persistent Organic Pollutants [*ECE/EB.AIR/60*]. Stationary and mobile sources are distinguished. Stationary sources are related to waste incineration; thermal metallurgical processes (production of aluminium and other non-

ferrous metals, iron and steel); combustion plants providing energy; residential combustion and specific chemical production processes releasing intermediates and by-products. Mobile sources are related to combustion process in the engine (halogenated additives in fuels and lubricants).

Official data on total emissions of PCDD/Fs were reported by 34 countries for the period from 1990 to 2003 (for at least one year). For the first time, Iceland, Switzerland and the FYR of Macedonia reported emission data. The information about the spatial distribution of PCDD/F emissions was submitted by 13 countries (Austria, Belarus, Belgium, Bulgaria, Finland, France, Hungary, Iceland, Netherlands, Norway, Poland, Spain, Sweden). For the remaining countries, we used expert estimates of PCDD/F total emissions and their spatial distribution [*Pacyna et al*., 1999].

The annual total PCDD/F emissions (1990-2003) used for modelling this year are displayed in Table A.7. It includes official data on PCDD/F emissions (in bold) and expert estimates.

Table A.7. *PCDD/F emissions (official data & expert estimates) for the period from 1990 to 2003, g TEQ/y*

Country	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003
Albania	3	3	3	3	3	3	3	3	3	3	3	3	3	3
Armenia	43	40	37	34	31	28	28	28	28	28	28	28	28	28
Austria	**161**	**135**	**76**	**67**	**56**	**58**	**60**	**60**	**56**	**53**	**50**	**53**	**41**	**43**
Azerbaijan	77	71	66	61	55	50	50	50	50	50	50	50	50	50
Belarus	107	94	81	68	55	42	29	**16**	**16**	**15**	**18**	**23**	**25**	**26**
Belgium	**624**	590	557	523	489	**455**	396	336	276	217	**157**	127	**97**	**96**
Bosnia&Herzegovina	19	19	20	20	21	22	22	22	22	22	22	22	22	22
Bulgaria	**554**	535	515	495	476	**456**	**341**	**310**	**288**	**245**	**233**	**201**	**219**	**255**
Croatia	**179**	165	152	138	124	111	**97**	**95**	**111**	**98**	**109**	**76**	**75**	75
Cyprus	13	13	13	13	13	13	13	13	13	13	**13**	10	8	**5**
Czech Republic	**1252**	**1220**	**1220**	**1140**	**1135**	**1135**	**922**	**830**	**767**	**643**	**744**	**620**	**177**	**114**
Denmark	93	93	93	93	93	93	93	93	**93**	88	83	**78**	**78**	**78**
Estonia	15	14	14	13	13	12	11	10	8	7	5	4	**2**	**4**
Finland	**30**	**33**	**31**	**32**	**33**	**34**	**32**	**32**	**32**	**32**	**31**	**31**	**32**	**34**
France	**1765**	**1814**	**1834**	**1892**	**1890**	**1692**	**1477**	**1041**	**938**	**614**	**524**	**392**	**365**	**247**
Georgia	72	67	62	56	51	46	46	46	46	46	46	46	46	46
Germany	**1196**	981	765	550	**335**	**307**	307	307	307	307	307	307	307	307
Greece	155	148	142	135	129	122	122	122	122	122	122	122	122	122
Hungary	**157**	**151**	**126**	**122**	**104**	**116**	**108**	**103**	**94**	**93**	**99**	**104**	**73**	**76**
Iceland*	**9**	**9**	**9**	**8**	**7**	**6**	**5**	**5**	**4**	**3**	**3**	**3**	**3**	**2**
Ireland	27	27	27	27	27	27	27	27	27	27	27	27	**27**	**26**
Italy	**443**	**447**	**422**	**411**	**410**	**450**	**383**	**384**	**376**	**353**	**319**	**295**	**286**	286
Kazakhstan	38	35	33	30	27	25	25	25	25	25	25	25	25	25
Latvia	19	19	19	19	19	19	19	19	19	19	19	19	**19**	19
Lithuania	24	21	19	16	13	11	8	**6**	**6**	**5**	**4**	**13**	**12**	**12**
Luxembourg	**40**	36	32	27	**23**	**24**	**16**	**16**	**8**	8	8	8	8	8
Monaco	**2**	**2**	**3**	**3**	**3**	**3**	**3**	**4**	**4**	**4**	**4**	**4**	**4**	**3**
Netherlands	**742**	607	472	337	201	**66**	61	56	51	47	**42**	**41**	**41**	**40**
Norway	**130**	**98**	**96**	**95**	**94**	**70**	**50**	**41**	**35**	**39**	**34**	**34**	**31**	**29**
Poland	**529**	**535**	**517**	**592**	**520**	**515**	**484**	**440**	**381**	**381**	**333**	**447**	**433**	433
Portugal	41	40	38	37	36	34	34	34	34	34	34	34	34	34
Republic of Moldova	18	16	15	13	11	10	9	8	6	5	**4**	**3**	**4**	**6**
Romania	129	119	110	100	91	81	81	81	81	81	81	81	81	81
Russian Federation	**991**	**947**	**901**	**878**	**825**	**769**	**637**	**614**	**606**	**625**	**631**	643	**655**	655
Serbia&Montenegro	46	48	49	51	52	54	54	54	54	54	54	54	54	54
Slovakia	**189**	183	176	170	163	**157**	141	**125**	**138**	**127**	**145**	**130**	**184**	**135**
Slovenia	**9**	8	7	6	**6**	**5**	**5**	**4**	**4**	**4**	**3**	**3**	**3**	**3**
Spain*	**178**	**187**	**197**	**193**	**185**	**157**	**154**	**125**	**127**	**134**	**141**	**137**	**137**	**135**
Sweden	**53**	**47**	**43**	**42**	**41**	**36**	**35**	**34**	**33**	**31**	**30**	**32**	**31**	**33**
Switzerland	**242**	222	202	182	162	142	122	102	82	62	**42**	42	42	**42**
The FYR of Macedonia	166	166	166	166	166	166	166	166	166	166	166	166	**166**	**163**
Ukraine	925	855	784	713	642	571	571	571	571	571	571	571	571	571
United Kingdom	**1232**	**1212**	**1181**	**964**	**767**	**794**	**533**	**442**	**349**	**327**	**294**	**296**	**285**	**259**
Total, kg TEQ/y	**12.7**	**12.1**	**11.3**	**10.5**	**9.6**	**9.0**	**7.8**	**6.9**	**6.5**	**5.8**	**5.7**	**5.4**	**4.9**	**4.7**

* National Total for the EMEP grid domain

Dynamics of PCDD/F emissions in Europe for the period from 1990 to 2003

According to the official data and expert estimates, the European emissions of PCDD/Fs dropped by 63% (or 2.7 times) between 1990 and 2003 (Fig. A.7). The total PCDD/F emissions in the EMEP region amounted to 4.7 kg TEQ in 2003. The decrease of the European emission of PCDD/Fs is mainly due to reduction in emissions in France, the Czech Republic, the United Kingdom, Germany, the Netherlands and Belgium.

According to the official data, PCDD/F emissions in most countries tend to decrease (1990-2003). The overall PCDD/F emissions in eighteen countries reported data for both years 1990 and 2003 decreased by 78% (or 4.6 times). The maximum decrease of the PCDD/F emissions was reported by the Netherlands (19 times), and the maximum increase - by Monaco (1.2 times) (Fig. A.8).

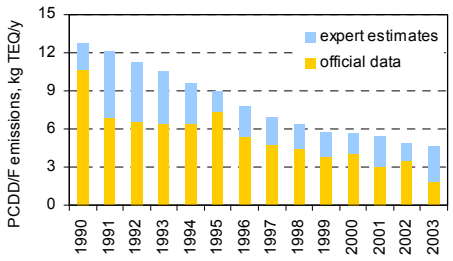


Fig. A.7. *PCDD/F emissions in Europe for the period from 1990 to 2003*

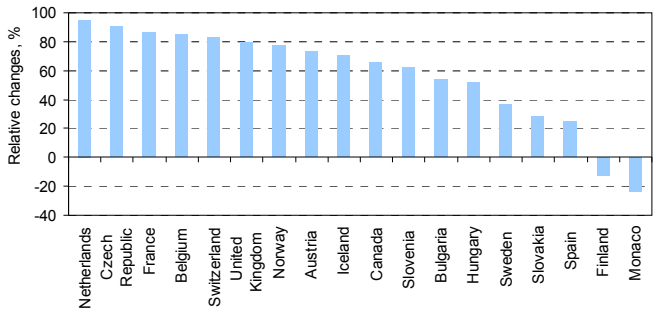


Fig. A.8. *Relative changes of PCDD/F emissions for 1990-2003 in 18 countries*

The contribution of individual countries to the total emissions of PCDD/Fs in Europe in 1990 and 2003 is presented in Fig. A.9. The contributors to the whole European emissions were Germany, the Russian Federation, the Ukraine and the United Kingdom for both 1990 and 2003.

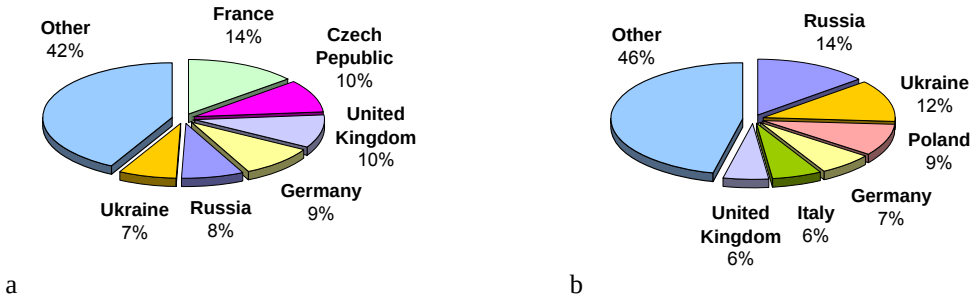


Fig. A.9*. The contribution of individual countries to the total emissions of PCDD/Fs in Europe in 1990 (a) and 2003 (b)*

Spatial distribution of PCDD/F emissions

Official information on the spatial distribution of PCDD/F emissions in the period from 1990 to 2003 is available for 13 countries (Table A.8). For the rest countries, expert estimates of spatial distribution of PCDD/F emissions [*Pacyna et al*., 1999] were used. Spatial distribution of the emissions of 17 toxic congeners of PCDD/Fs in 1990 and 2003 is given in Fig. A.10. High PCDD/F emission intensity was characteristic of Belgium, Bulgaria, the Czech Republic, Switzerland, the FYR of Macedonia, and the United Kingdom in 1990, while the same intensity is characteristic of the FYR of Macedonia only in 2003.

Table A.8. *The list of countries submitted official data on spatial distribution of PCDD/F emissions (1990-2003)*

Country	Year
Austria	1990, 1995, 2000
Bulgaria	
Hungary	
Iceland	
Norway	
Netherlands	1990, 1995
Finland	1990-2003
Spain	
Belarus	2000
Belgium	
France	
Poland	
Sweden	

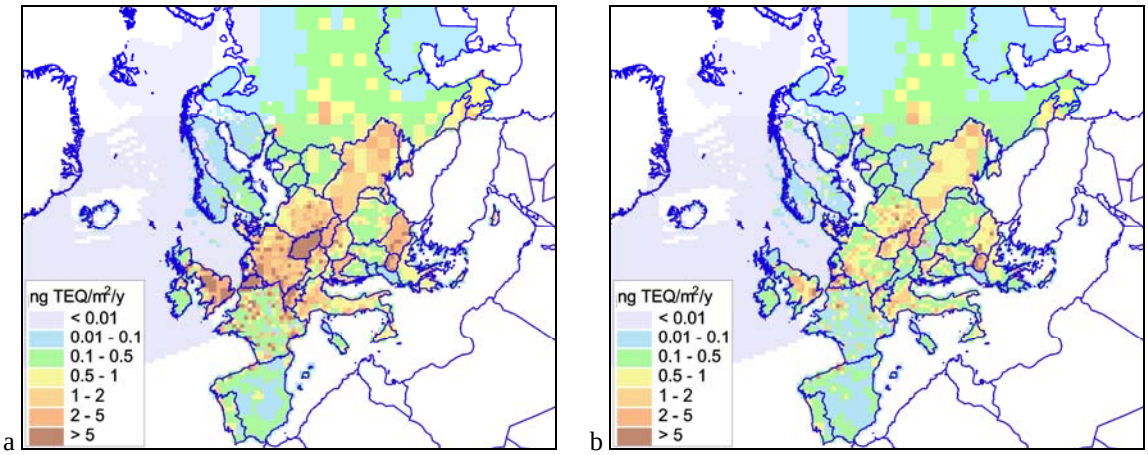


Fig. A.10*. Spatial distribution of the total emission of 17 toxic congeners of PCDD/Fs in 1990 (a) and 2003 (b)*

PCDD/F emissions by sectors

Official information on emissions of PCDD/Fs by sectors in 2003 is available for 23 countries. The sector split for the national total emissions of PCDD/Fs is presented in Fig. A.11. The largest contribution to the total PCDD/F emissions is made by the Residential sector (25%). The next most important sector is the Waste Incineration. This sector is the largest source PCDD/Fs for Belarus, Cyprus, Iceland, Republic of Moldova, Switzerland and the United Kingdom.

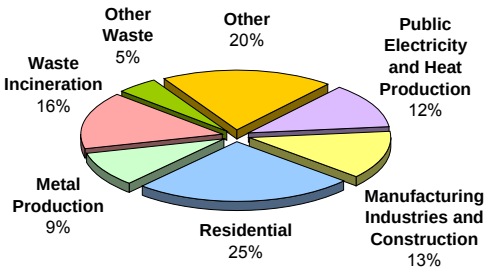
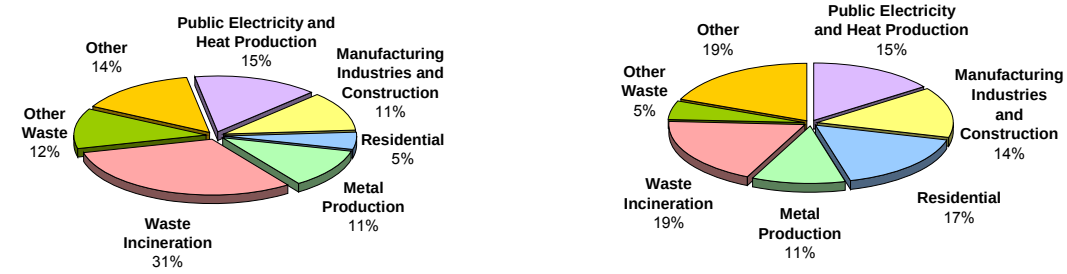


Fig. A.11*. Sector split for PCDD/Fs in Europe (23 countries) in 2003*

Official information on emissions of PCDD/Fs by sectors in both 1990 and 2003 is available for 8 countries (Austria, France, Iceland, Netherlands, Norway, Sweden, Switzerland, United Kingdom). The maximum contribution to the total PCDD/F emissions in both 1990 and 2003 is made by the Waste Incineration sector (31% and 19%, respectively) (Fig. A.12). The contribution of the Residential

sector to the total PCDD/F emissions during 1990-2003 increased by 3.4 times, while the contributions of the Other Waste and the Waste Incineration sectors decreased by 2.4 and 1.7 times, respectively.



a b

***Fig.A.12**. Sector split for PCDD/Fs in Europe (8 countries) in 1990 (a) and 2003 (b)*

Official data on PCDD/F emissions by sectors in Austria, France, Iceland, Norway, Sweden and the United Kingdom are available for each year of the period from 1990 to 2003. As an example, PCDD/F emission time-series for France is demonstrated in Fig. A.13. The most decrease of PCDD/F emissions is observed in the Public Electricity and Heat Production sector (by 90%), as well as in the Waste Incineration and the Manufacturing Industries and Construction sectors.

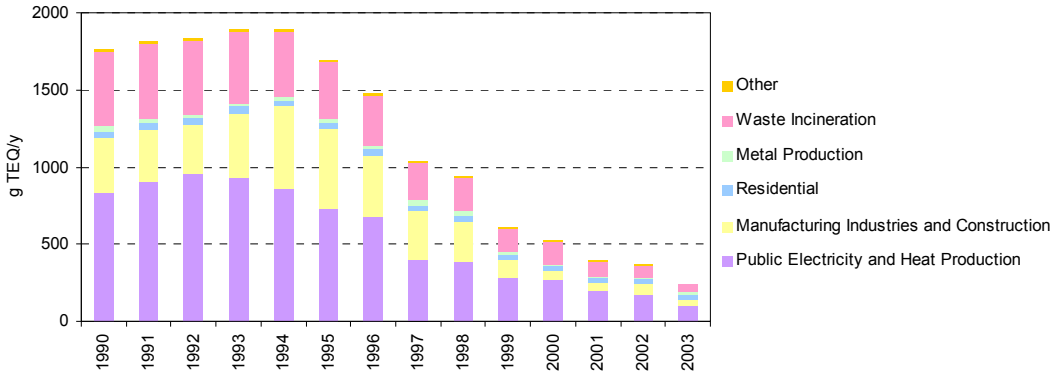


***Fig. A.13**. PCDD/F emission time-series for France*

PCDD/F gridded sector data

The official information on PCDD/F gridded sector data for each of the relevant aggregated NFR sectors [*EB.AIR/GE.1/2002/7, Table III B*] is submitted by Finland (2001-2003), France (2000), Iceland (1990/1995/2000) and Spain (1990-2003). As an example, spatial distribution of the total PCDD/Fs and some of sector emissions for France in 2000 is given in Fig. A.14. The sector 01 'Combustion in Power Plants and Industry' amounts to approximately 60% from Total PCDD/Fs. The second important sector is the sector 08 'Waste'. It amounts to about 30% from Total PCDD/Fs. Other contributions come from the sectors 03 'Commercial Residential and Other Stationary Combustion' (7%) and 05 'Industrial Processes' (2%).

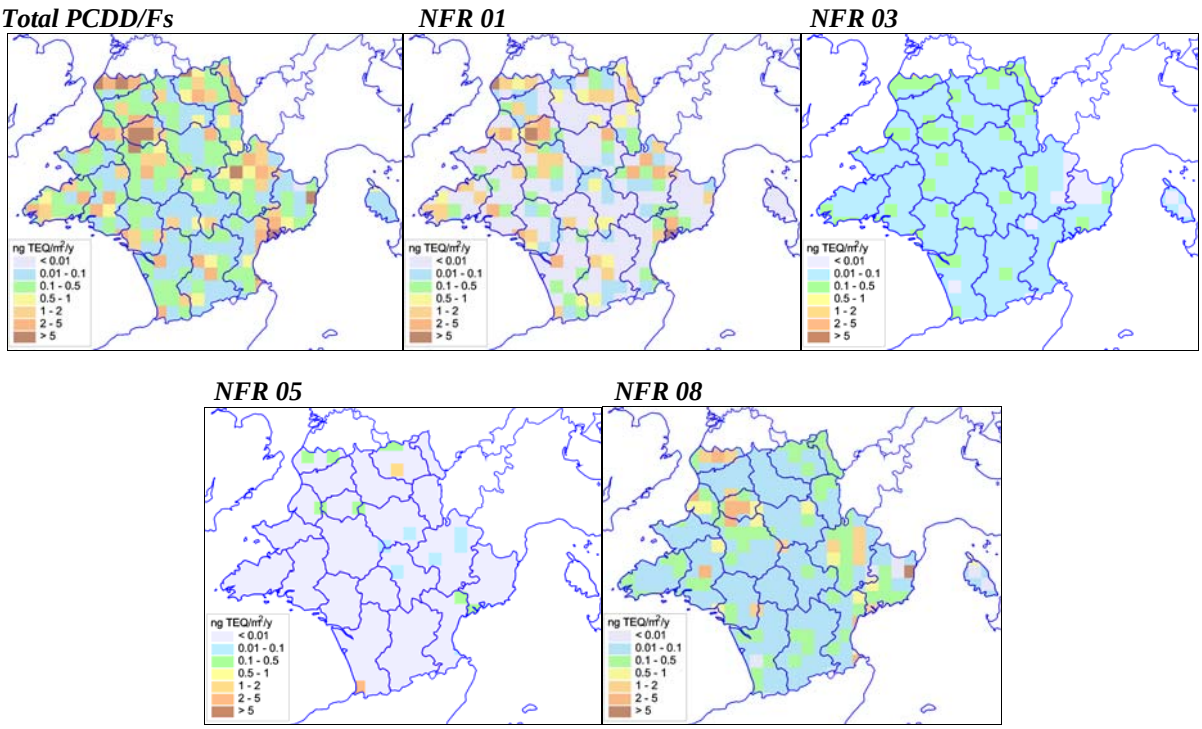


Fig. A.14. *Spatial distribution of the total PCDD/Fs and some of sector emissions for France in 2000*

Toxic congeners of PCDD/Fs

On the basis of the given above data (Table A.7) and using congener compositions of PCDD/F mixture in various European countries [*Pacyna et al.,* 1999] spatial distributions for all 17 toxic congeners of PCDD/Fs for the period from 1990 to 2003 were prepared for modelling. Emission contributions of each of the 17 toxic congeners to the total toxicity of PCDD/Fs mixture are shown in Fig. A.15. The main contributor to the toxicity of the mixture of toxic congeners is 2,3,4,7,8-PeCDF (37% in 1990 and 38% in 2003). At this stage, evaluation of overall toxicity of PCDD/F mixture was performed on the basis of simulations of 2,3,4,7,8-PeCDF congener (feasibility of such evaluation and uncertainty analysis see [*Shatalov et al.,* 2003]). Spatial distribution of the emissions of 2,3,4,7,8-PeCDF in 1990 and 2003 are displayed in Fig. A.16.

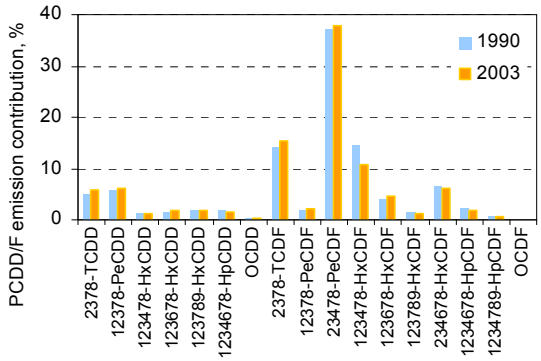


Fig. A.15. *Emission contributions of each of the 17 toxic congeners to the overall toxicity of PCDD/Fs mixture in 1990 and 2003*

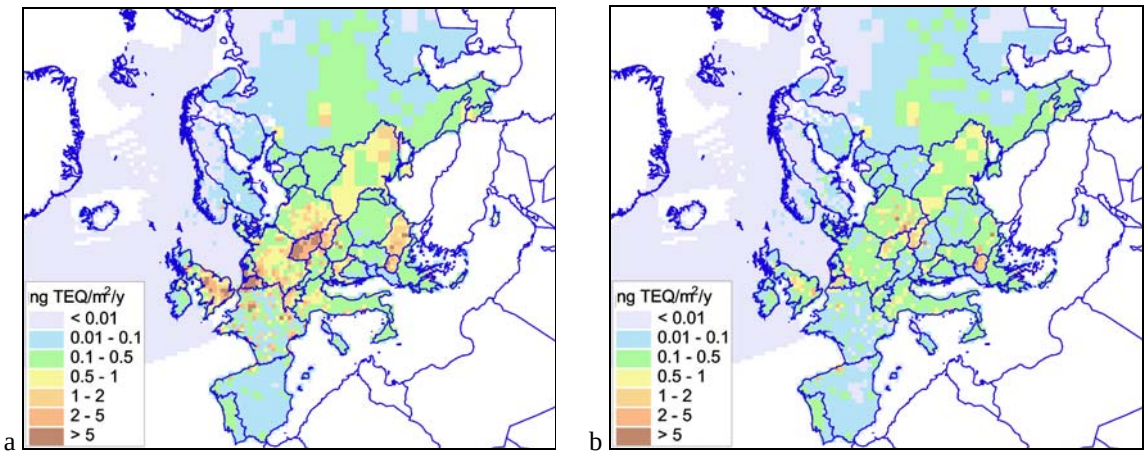


***Fig. A.16**. Spatial distribution of 2,3,4,7,8-PeCDF emissions in 1990 (a) and 2003 (b)*

Uncertainties in PCDD/F emission estimates

The most important uncertainties in the PCDD/F emission estimates used in the calculations are inherent due to the uncertainties introduced both in official data and expert estimates. According to [*Pacyna et al*., 1999], major uncertainties in PCDD/F emissions in the expert estimates arise from the following reasons:

- many sources of these pollutants are not well characterized, thus the actual emissions are supposed to be underestimated;
- the variability at a single plant can be of an order of magnitude between different sampling periods [*Parma et al*., 1995];
- "allocation" of available emission factors between countries.

Thus, with the limited information available, the uncertainty of the TEQ emissions should be considered as an order-of-magnitude estimate [*Pacyna et al*., 1999]. Besides, usage of data on particular congener in mass units from [*Pacyna et al*., 1999] for evaluation of emission congener composition introduces additional uncertainties. Official information submitted by countries is somewhat uncertain. Finally, it should be noted that seasonal variations of PCDD/F emissions used in the computation are also rather uncertain.

A.3. Hexachlorobenzene

Emissions of hexachlorobenzene (HCB) stem from its usage in agriculture as pesticide (fungicide) as well as from the use of other pesticides containing HCB. This chemical is also an intermediate product or by-product in the production of other chlorinated substances. Besides, as stated in the Protocol on POPs [*ECE/EB.AIR/60*, 1999], major sources of HCB emissions from thermal and chemical processes, in which it is formed by a mechanism similar to that of PCDD/Fs, can be as follows: waste incineration plants, including co-incineration; thermal sources of metallurgical industries; and use of chlorinated fuels in furnace installations.

Official information about total emissions of HCB was reported by 24 countries for the period from 1990 to 2003 (for at least one year) (Table A.9). For the first time, Estonia and Finland reported emission data. The information about the spatial distribution of HCB emissions was submitted by Austria, Belgium, France, Poland and Spain.

Table A.9. *Official HCB emission data, kg/y*

Country	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003
Austria	93	85	69	64	52	53	56	52	49	47	43	47	44	47
Belarus													7.6	5.1
Belgium	18					30					55	55	61	61
Bulgaria	544					79	87	47	76	46	54	43	38	45
Canada	89	71	74	75	72	73	72	71	33	33	40	41	45	28
Croatia	0.3						0	0	0	0	0			
Czech Republic												0	4.1	0.07
Estonia														0.25
Finland														0.46
France	1650	1670	1696	1631	1789	1787	1698	1713	1693	1686	1786	1773	1780	1762
Germany	86				6									
Hungary													4.4	4.5
Ireland													0.04	0.04
Lithuania												0.02		
Netherlands	0					0					0	0	0	0
Norway												51	7.9	0.85
Poland	62	39	39	43	38	51	48	51	43	40	46	8	8.5	
Republic of Moldova											0.66	1	0.75	0.13
Russia *	1.6	1.6	1.6	1.7	1.6	1.3	1.1	0.98	0.95	0.98	1.1		0.96	
Slovenia	0				0	0	0	0	0	0	0			
Spain	6647	6204	5369	5108	5563	4894	5417	6070	6119	6072	6100	6098	6106	6145
Switzerland	0	0	0	0	0	0	0	0	0	0				
United Kingdom	3515	3361	3858	4169	4376	4549	4621	4621	4856	534	325	314	307	302
United States	1450						281			4835				

* European part within EMEP

According to the official data, the total HCB emissions in seven countries submitted data for both years 1990 and 2003 decreased by 19% (or 1.2 times). The maximum emission decrease was reported by Bulgaria (12 times), and the maximum increase - by Belgium (3.4 times) (Fig. A.17).

Official information on emissions of HCB by sectors in 2003 is available for 14 countries. The sector split for the national total emissions of HCB is presented in Fig. A.18. The maximum contribution to the total HCB emissions is made by the Manufacturing Industries and Construction sector (27%). This sector is the largest source of HCB for Estonia, France, Hungary, Ireland and Republic of Moldova. The second most important sector is the Metal Production. The next important sector is the Agriculture (Other). This sector is the key sector for Spain and the United Kingdom.

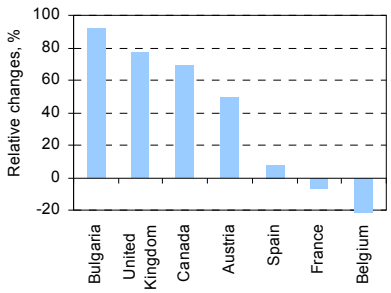


Fig. A.17. *Relative changes of HCB emissions for 1990-2003 in 7 countries*

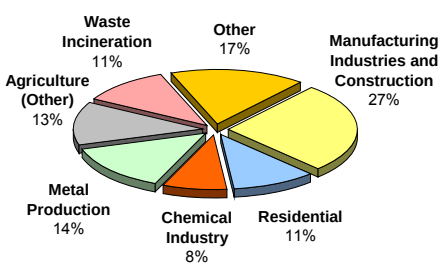


Fig. A.18*. Sector split for HCB in Europe (14 countries) in 2003*

For modelling of the HCB long-range transport within the Northern Hemisphere in 2003, a conventional emission scenario was prepared with the use of different expert estimates (Table A.10).

Table A.10. *HCB emission data used in calculation in 2003*

Country	Emission	Comments
USA	11791	*Bailey*, 2001
Canada	690	
Albania	55	*Pacyna et al.*, 1999
Austria	81	
Belarus	471	
Belgium	75	
Bulgaria	300	
Czechoslovakia	334	
Denmark	115	
Estonia	71	
Finland	126	
France	1285	
Germany	1555	
Greece	175	
Hungary	305	
Iceland	7	
Ireland	46	
Italy	795	
Latvia	128	
Lithuania	176	
Luxembourg	3	
Moldova	100	
Netherlands	92	
Norway	45	
Poland	930	
Portugal	145	
Romania	740	
Russian Federation (ETR)	10980	
Spain	1172	
Sweden	161	
Switzerland	56	
Ukraine	2095	
United Kingdom	511	
Yugoslavia	310	
Russian Federation (ATR)	3000	HCB emission values were estimated on the basis of the relationships of gross domestic product and official HCB emission data for countries with similar economic indexes
China	10607	
Republic of Korea	3548	
Japan	2530	
Pakistan	701	
India	35682	Difference between hemispheric emissions and sum of emissions from North-American continent, Europe and Asian countries mentioned above
Total	91988	
Global	91988	*Bailey*, 2001

The HCB emission values in the USA, Canada and emissions for the whole of the Northern Hemisphere are taken from the expert estimates by *R.Bailey* [2001]. For the European countries and the European part of Russia, the HCB expert estimates prepared by [*Pacyna et al.*, 1999] were used (Fig. A.19). The HCB emission values in China, Pakistan, Republic of Korea, the Asian part of Russia and Japan were estimated on the basis of the relationship of the gross domestic product and official HCB emission data taken for countries with similar economic indexes. HCB emission values in India are estimated as the difference between the total emissions of the Northern Hemisphere and the sum of emissions from North-American continent, Europe and Asian countries mentioned above.

For the evaluation of the transcontinental transport, HCB sources of the Northern Hemisphere were split into 5 groups (North America, Central Asia, Europe, Russia and South-eastern Asia). The spatial

distribution of emissions for each group was prepared (Fig. A.20). The most intensive emission sources are located in India.

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

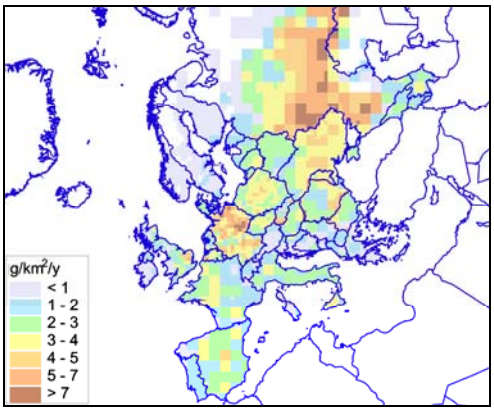


Fig. A.19. *Spatial distribution of HCB emissions in the EMEP region in 2003*

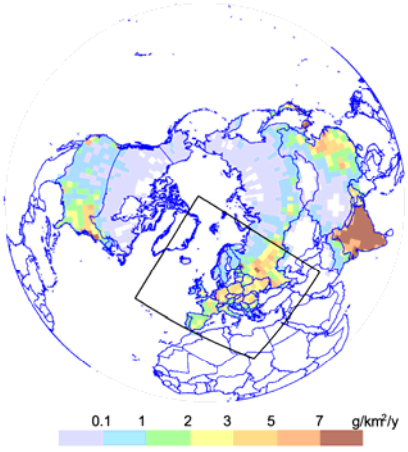


Fig. A.20. *Spatial distribution of HCB emissions in the Northern Hemisphere in 2003*

Uncertainties in HCB emission estimates

According to the expert estimates [*Bailey*, 2001], "the major uncertainties in HCB emissions are as follows: HCB concentrations in pesticides in developing countries; Emissions from chemical process industries in developing countries; Emissions of HCB from combustion processes; Emissions from metals industries; HCB concentrations in soil and volatilization rate from soil; HCB used or produced in military smokes; Processes for which no information was found and which may emit HCB". The uncertainty of HCB emission from combustion process is especially great. "HCB has been reported as a product of incomplete combustion in many studies where chlorine is present. The quantitative results on yield of HCB from combustion under different conditions are scattered so that it is impossible to assign factors with much confidence. Thus, a 100-fold range of HCB emissions from combustion processes is used" [*Bailey*, 2001].

According to the expert estimates [*Pacyna et al.*, 1999], the major uncertainty of HCB emissions is originated from its agricultural use (contribution of the agricultural sector to the overall emissions is 80%) since "there is a general lack of information in usage of pesticides on a compound-specific basis". The expert estimates of HCB emissions from other activity sectors (solvents and other products use, production processes, waste treatment and disposal) also include some uncertainties.

Another source of uncertainties in estimates of HCB emissions is associated with the approach based on calculation of relationship (of the gross domestic product and official HCB emission data taken for countries with similar economic indexes) used to evaluate HCB emissions in China, Pakistan, Republic of Korea, the Asian part of Russia and Japan.

A.4. Polychlorinated biphenyls

Polychlorinated biphenyls (PCBs) as technical mixtures with different sets of individual homologues and congeners have been widely used for a long period of time. Mixtures of these chemicals have been produced in many countries under various trade names: Aroclor (USA), Clophen (West Germany), Kanechlor (Japan), Delor (Czechoslovakia), Pyroclor (United Kingdom), Sovol, Sovtol (Russia) and others. The total global production of PCBs from 1930 to 1993 amounted to approximately 1.3 million tonnes [*Breivik et al*., 2002a]. PCB emissions to the environment occur from a great number of different categories of sources: open usage (plasticisers, carbonless copy paper, lubricating oils, etc.), small capacitor usage, nominally closed usage (hydraulic and heat transfer fluids, etc.) and closed usage (transformers and large capacitors), landfills, open burning (open pit burning, backyard barrel burning, domestic incineration in stoves, etc.), waste incineration and destruction, spillage to soil and fires [*Breivik et al*., 2002b].

Official information about total emissions of PCBs was submitted by 19 countries for the period from 1990 to 2003 (for at least one year) (Table A.11). According to the official data submitted for the both years, the total PCB emissions of Bulgaria, the Czech Republic, France, Hungary, Monaco, Slovakia, Slovenia, Spain and the United Kingdom decreased by 4.3 times. The information about the spatial distribution of PCB emissions was submitted by Finland and France.

Table A.11. *Official PCB emission data, kg/y*

Country	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003
Belarus													6.6	6.9
Belgium						0.0051					0	0		
Bulgaria	258					382	262	227	253	247	229	212	250	261
Czech Republic	773	772	741	644	630	623	555	448	458	485	474	407	82	3.2
Estonia													47	54
Finland				5300	1100	15800								0.2
France	88	96	97	101	97	96	97	94	93	91	93	95	95	98
Germany	43579				30868									
Hungary	135	120	108	106	104	101	99	96	92	93	89	89	99	103
Lithuania								12	14	13	11	15	13	13
Monaco	0.28	0.28	0.31	0.34	0.37	0.37	0.39	0.44	0.42	0.42	0.43	0.46	0.41	0.34
Netherlands	0					0					0	0	0	0
Poland	2425	2367	2322	2348	2330	2323	2348	2342	2353	2331	2265	2327	2282	
Republic of Moldova											196	192	232	265
Slovakia	164					138		137	139	136	129	135	136	129
Slovenia	357				265	235	214	194	184	105	143	122	92	81
Spain	0	0	0	0	0	0	0	0	0	0	0	0	0	0
United Kingdom	7138	6559	6059	5559	4996	4448	3909	3404	2838	2162	1641	1555	1446	1378
United States	102						195			34066				

Official information on emissions of PCB by sectors in 2003 is available for 12 countries. The sector split for the national total emissions of PCB is presented in Fig. A.21. The maximum contribution to the total PCB emissions is made by the Public Electricity and Heat Production sector (39%). This sector is the key sector for the Czech Republic, Estonia, France, Hungary and Monaco. The next important sector is the Residential. This sector is the largest PCB sources for Bulgaria and Lithuania. Emissions from The Solvent and Other Product Use (Other) are most important in Republic of Moldova and Slovenia.

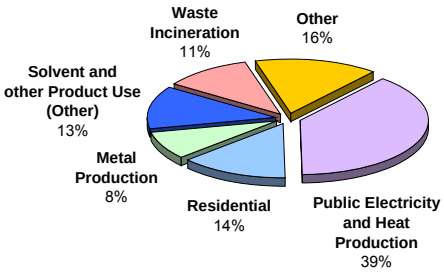


Fig. A.21*. Sector split for PCB in Europe (12 countries) in 2003*

The modelling of PCB long-range transport for the period from 1990 to 2003 was performed for an individual PCB congener (PCB-153). The sources located over the whole Northern Hemisphere were taken into account in the computations. Emission data of PCB-153 were taken from the use of a global emission inventory of 22 PCB congeners (High emission scenario) [*Breivik et al*., 2002 b]. The inventory was based on historical data on global production and consumption of these PCBs [*Breivik et al*., 2002a]. The emissions of 22 individual PCB congeners were evaluated for 114 countries for the period of 1930 to 2000, with allowance made for High, Middle and Low emission scenarios. The total historical global emissions of the 22 PCB congeners for the High scenario in 1930-2000 were 92 kt. In the period from 2001 to 2003, the annual values of emissions were accepted to be equal to those of 2000.

Fig. A.22. *Splitting of PCB-153 emissions into groups of sources*

For the evaluation of intercontinental transport, PCB sources of the Northern Hemisphere were split into 6 groups: Russia, North-western Europe, South-eastern Europe, America (both North and South), South-eastern Asia, and Central Asia and North Africa (Fig. A.22).

Emissions of PCB-153 from the selected regions-sources of the Northern Hemisphere in 1990 and 2003 used in calculations are given in the Table A.12 (High emission scenario).

Table A.12. *Emissions of PCB-153 congeners from the selected regions-sources of the Northern Hemisphere in 1990 and 2003 [Breivik et al., 2002b], t/y*

Year	Regions-sources						Total
	America	North-western Europe	South-eastern Europe	Africa and Central Asia	South-eastern Asia	Russia	
1990	9.71	9.50	11.60	8.81	3.81	1.98	45
1991	8.44	7.29	8.99	7.27	3.31	1.93	37
1992	6.62	8.85	7.77	5.21	2.33	1.89	33
1993	6.01	5.54	5.91	4.62	2.09	1.84	26
1994	4.49	5.28	3.86	2.82	1.29	1.79	20
1995	3.92	4.95	3.43	2.31	1.07	1.75	17
1996	3.57	4.46	3.25	2.16	1.00	1.70	16
1997	3.24	4.15	3.10	2.04	0.92	1.65	15
1998	2.91	3.86	2.95	1.92	0.86	1.61	14
1999	2.55	3.61	2.80	1.78	0.77	1.56	13
2000	2.24	3.29	2.65	1.66	0.67	1.51	12
2001	2.24	3.29	2.65	1.66	0.67	1.51	12
2002	2.24	3.29	2.65	1.66	0.67	1.51	12
2003	2.24	3.29	2.65	1.66	0.67	1.51	12

The contribution regions-sources to emissions of PCB-153 in the Northern Hemisphere in 1990 and 2003 is presented in Fig. A.23. In contrast with 1990, the contribution of North-western Europe increased, and the contribution of South-eastern Europe decreased in 2003. The contribution of Russia increased up to 13% and actually became equal to that of the Africa and Central Asia in 2003. As can be seen from these diagrams, the main contributors to the pollution of Northern Hemisphere were North-western Europe, South-eastern Europe and America (around 70%) for both 1990 and 2003.

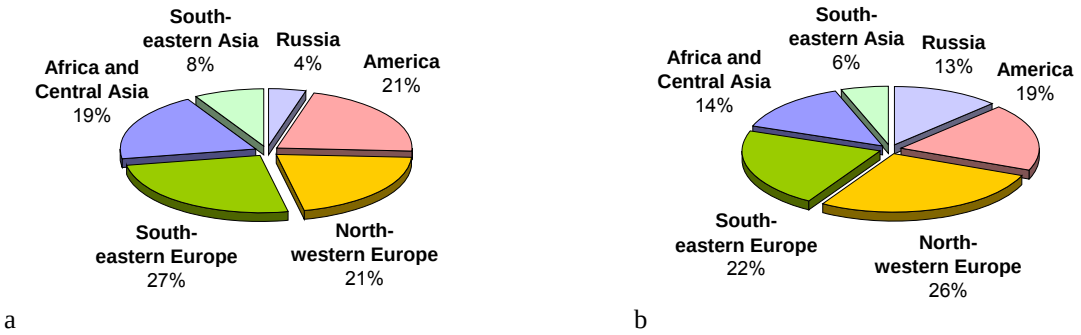


Fig. A.23. *The contributions of selected regions-sources to emissions of PCB-153 in the Northern Hemisphere in 1990 (a) and 2003 (b)*

Dynamics of PCB-153 emissions in the Northern Hemisphere and Europe in 1990-2003

Emission dynamics of PCB-153 in the Northern Hemisphere and European region in 1990-2003 estimated on the basis of data available in [*Breivik et al*., 2002a,b] is demonstrated in Fig. A.24. Total PCB emissions in the Northern Hemisphere and European region reduced 3.8 and 3.1 times, respectively in the period from 1990 to 2003.

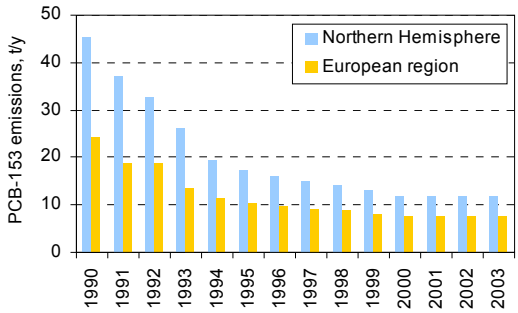


Fig. A.24. *PCB-153 emission dynamics in the Northern Hemisphere and European region in 1990-2003, t/y*

Spatial distribution of PCB-153 emissions

The spatial distribution of emissions of PCB-153 in the EMEP region in 1990 and 2003 was made on the basis of the spatial distribution of PCB emissions prepared by *J.Pacyna et al.* [1999] (Fig. A.25). The spatial distribution of emissions of PCB-153 in the Northern Hemisphere for the period from 1990 to 2003 was obtained on the basis of the high emission scenario available in [*Breivik et al*., 2002b] (Fig. A.26). For the evaluation of the emission spatial distribution over the 2.5^0x2.5^0 calculation grid, data on the population density (1990) available in the Canadian Global Emissions Interpretation Centre (http://www.ortech.ca/cgeic) were used. The most intensive emission sources are located in Belgium, France, Germany, Italy, Spain and the United Kingdom. Relatively high emission intensity is characteristic of the Czech Republic and Switzerland.

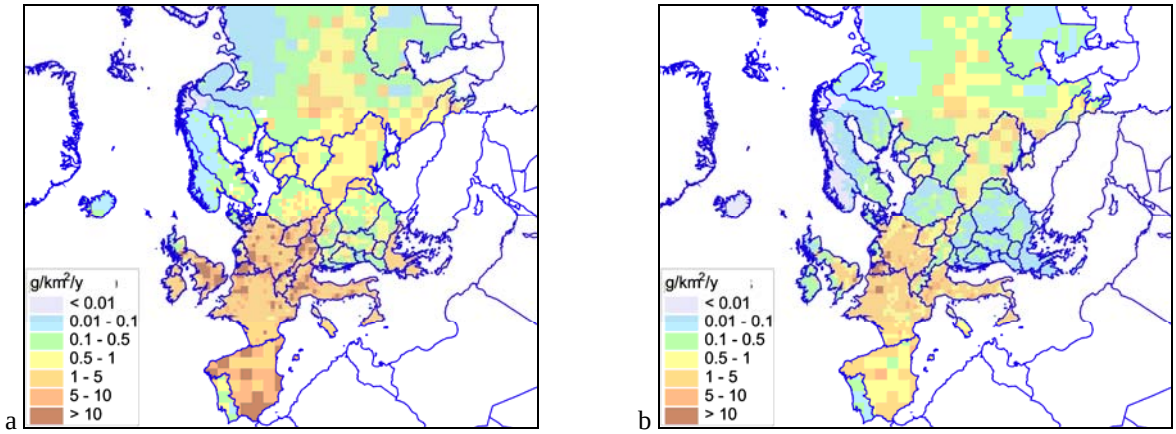


Fig. A.25. *Spatial distribution of emissions of PCB-153 in the EMEP region in 1990 (a) and 2003 (b)*

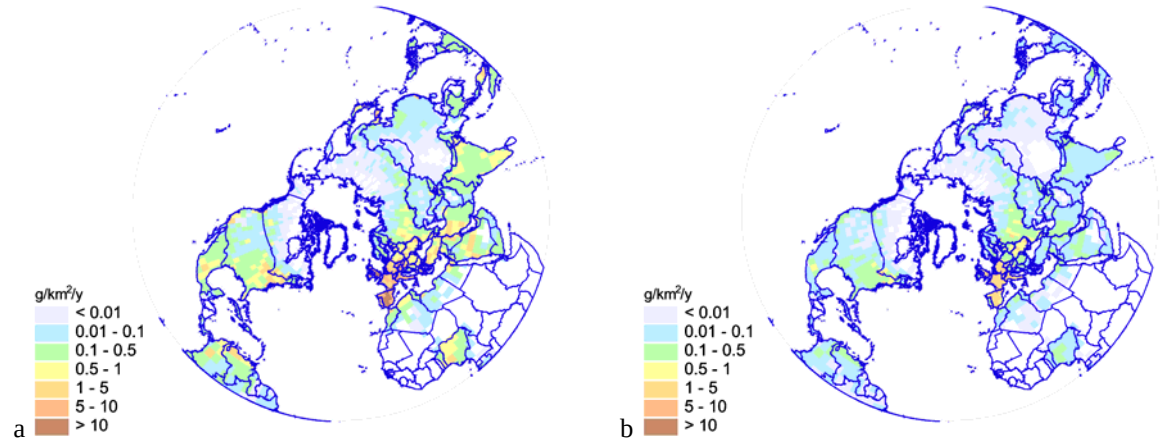


Fig. A.26. *Spatial distribution of emissions of PCB-153 in the Northern Hemisphere in 1990 (a) and 2003 (b)*

Uncertainties in PCB emission estimates

Due to uncertainties at the temporal and spatial scales involved in this emission inventory, according to *K. Breivik et al*. [2002 a,b], the actual emission values should be considered as order-of-magnitude estimates. Basic uncertainties involved in estimates of the global consumption and emissions of PCB individual congeners are:

1. Uncertainties in estimates of global production and consumption:

- Due to deficient information, the inventory of PCB global production did not include the amount of these substances produced in factories of Poland, East Germany and Austria.
- Data on production reported for a period in excess of one year (e.g. a 5-year period) were uniformly distributed over the indicated period.
- The highest uncertainty in the global production estimate lies in default homologue and congener composition (e.g. for France, Spain and Italy) and variability between characteristic technical mixture compositions.

- For individual congeners the uncertainty in the global production estimate increased for the period after the 70s due to the increased number of PCB producers. Nevertheless, recent data are more accurate than the data from the past.
- In realization of the method of global consumption estimation, a number of assumptions were made. Reliable information was available only for countries with traditionally high consumption of PCBs. For other countries, assumptions based on trade between different countries and regions were made using Gross Domestic Product as a surrogate parameter.
- Approximately 70 non-OECD countries are not involved in the estimate; otherwise, the probable consumption in them would be about 6% of the total export in countries of this category. It is presumed that it introduced minor uncertainties into the general inventory.

2. Uncertainty in the emission estimate:

- The representativeness and accuracy of emission factors are the major source of uncertainties in a given estimate of emissions.
- The global movement of PCB-containing products and wastes between countries was not considered in detail.
- In view of the deficiency of reasonable estimates of the detailed spatial patterns, potential "point sources" of PCB atmospheric emissions were not considered separately. For the same reason incidental or unwanted formation of PCBs and emissions from the combustion process (de novo synthesis) were also not considered.

Uncertainties in estimates of global consumption and emissions will grow with the increase of the specification level (e.g. more detailed usage and waste disposal categories). Clearly the supposed uncertainties in emission estimates appear to be higher than the uncertainty in production estimates.

Problems and recommendations

1. Due to a lack of European data sets, we still have to use expert estimates. Completeness of official data for the whole period (1990-2003) is about:

   35% of total PAH (4 indicators) emissions;

   50% of total PCDD/F emissions;

   25% of total HCB emissions.

2. The information on spatial distribution of POP emissions, as well as the LPS emissions are not fully complete.

3. Data on the POP emission height distribution, temporal variation and projections from the countries would be very helpful.

4. Quantitative characteristics of uncertainties in national totals are not available, except Denmark.