



DANISH EMISSION INVENTORY FOR HEXACHLOROBENZENE AND POLYCHLORINATED BIPHENYLS

Scientific Report from DCE – Danish Centre for Environment and Energy

No. 103

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Data sheet

Series title and no.:	Scientific Report from DCE – Danish Centre for Environment and Energy No. 103
Title:	Danish emission inventory for hexachlorobenzene and polychlorinated biphenyls
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Institution:	Aarhus University, Department of Environmental Science
Publisher:	Aarhus University, DCE – Danish Centre for Environment and Energy ©
URL:	http://dce.au.dk
Year of publication:	June 2014
Editing completed:	June 2014
Referee:	Stine Justesen, Danish Environmental Protection Agency
Financial support:	Danish Environmental Protection Agency
Please cite as:	Nielsen, O.-K., Plejdrup, M.S., Winther, M., Nielsen, M., Fauser, P., Mikkelsen, M.H., Albrektsen, R., Hjelgaard, K., Hoffmann, L., Thomsen, M., Bruun, H.G., 2013. Danish emission inventory for hexachlorobenzene and polychlorinated biphenyls. Aarhus University, DCE – Danish Centre for Environment and Energy, 65 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 103 http://www.dce2.au.dk/pub/SR103.pdf
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Abstract:	This report documents the improved air emission inventories for hexachlorobenzene (HCB) and polychlorinated biphenyls (PCBs). The emission inventories now include all sources covered by the EMEP/EEA Guidebook. The completeness and accuracy of the inventories for these pollutants have been significantly improved through the results of this project. The project also included a screening for pentachlorobenzene (PeCB); the result of this screening was that the current data available do not allow for the elaboration of an emission inventory.
Keywords:	Emission inventory, HCB, hexachlorobenzene, PCBs, polychlorinated biphenyls, PeCB, pentachlorobenzene, POPs, persistent organic pollutants, UNECE, LRTAP, Stockholm Convention
Layout:	Ann-Katrine Holme Christoffersen
Front page photo:	Ann-Katrine Holme Christoffersen
ISBN:	978-87-7156-074-9
ISSN (electronic):	2245-0203
Number of pages:	65
Internet version:	The report is available in electronic format (pdf) at http://www.dce2.au.dk/pub/SR103.pdf

Contents

Preface	5
Summary	6
Sammenfatning	8
1 Introduction	10
1.1 International obligations	10
1.2 Danish emission inventories	10
1.3 Relevant source categories	11
2 Emission inventory methodology for hexachlorobenzene	15
2.1 Stationary combustion	15
2.2 Mobile combustion	19
2.3 Fugitive emissions from fuels	21
2.4 Industrial processes	22
2.5 Solvent and other product use	24
2.6 Agriculture	29
2.7 Waste	31
3 Emission inventory methodology for polychlorinated biphenyls	32
3.1 Stationary combustion	32
3.2 Mobile combustion	39
3.3 Fugitive emissions from fuels	41
3.4 Industrial processes	41
3.5 Solvent and other product use	45
3.6 Agriculture	51
3.7 Waste	52
4 Screening for pentachlorobenzene	53
4.1 Stationary combustion	53
4.2 Mobile combustion	54
4.3 Fugitive emissions from fuels	54
4.4 Industrial processes and solvent and other product use	54
4.5 Agriculture	55
4.6 Waste	55
4.7 Conclusions	55
5 Conclusions	57
6 References	60

Preface

This report presents the updated Danish air emission inventory for hexachlorobenzene (HCB) and a new emission inventory for polychlorinated biphenyls (PCBs). The emission inventories have been established in accordance with the EMEP/EEA Guidebook and covers all sources for which the EMEP/EEA Guidebook contains methodologies and default emission factors.

In addition to the inventories for HCB and PCBs, a screening has been made regarding the possible emission sources and levels for pentachlorobenzene (PeCB).

The Department of Environmental Science and DCE - Danish Centre for Environment and Energy at Aarhus University has carried out the work. The project has been financed by the Danish Environmental Protection Agency (DEPA).

Summary

The Danish air emission inventories for hexachlorobenzene (HCB) and polychlorinated biphenyls (PCBs) have previously not been complete. In this project, the emission inventories have been improved using the latest guidance provided in the EMEP/EEA air pollutant emission inventory guidebook (the Guidebook). In addition to the Guidebook a literature survey has been carried out to complement and verify the emission factors provided in the Guidebook.

Based on the available data, it has been possible to substantially increase the completeness of the reporting of HCB and PCBs emissions. As a result there are now only nine categories where the notation key NE (not estimated) is used compared to 30 in the previous submission for HCB and now only 13 categories where the notation key NE (not estimated) is used (previously there were 50 categories where NE was reported) for PCB.. The reasons why there are still categories reported as NE are either that the entire category is not reported in the Danish inventory, e.g. due to a lack of activity data or that it has not been possible to find an emission factor for HCB and/or PCBs but there are indications that emissions of HCB and/or PCBs could occur. Such indication is for example reported and well-documented emission factors for dioxins and furans.

The results of the improved emission inventory for HCB can be seen in Table S.1 below.

Table S.1 Danish emission inventory for HCB for 1990-2011, kg.

	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Energy industries	4.4	3.3	1.5	1.2	1.7	1.5	1.4	1.4	1.4	1.2
Manufacturing industries and construction	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Transport	0.4	0.4	0.5	0.6	0.6	0.7	0.7	0.6	0.7	0.7
Other sectors (e.g. households)	1.5	1.0	0.4	0.4	0.4	0.5	0.5	0.4	0.5	0.5
Industrial processes	2.0	2.3	2.7	1.4	0.7	0.7	0.6	0.01	0.01	0.01
Solvents and other product use	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.001	0.001	0.001
Agriculture	18.4	0.6	0.4	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Waste	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Total	26.8	7.8	5.6	3.8	3.7	3.6	3.3	2.7	2.8	2.6
Total emission reported in 2013	3.2	1.9	0.6	0.5	0.5	0.5	0.6	0.5	0.6	0.6

The significant decrease in the HCB emission is caused by a decrease in the emissions from agriculture. The decrease is partly caused by a decrease in the use of pesticides and fungicides known to contain impurities of HCB and partly by the fact that the level of HCB impurities in certain pesticides was substantially reduced in the beginning of the 1990's.

The emission from energy industries has also decreased due to flue gas abatement targeting dioxin and furans installed at the waste incineration plants. The emission from industrial processes has decreased due to the closure of steel production and secondary aluminium production in Denmark. For the remaining sectors the emissions are quite low and stable over time.

The results of the improved emission inventory for PCBs can be seen in Table S.2 below.

Table S.2 Danish emission inventory for PCBs for 1990-2011, kg.

	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Energy industries	0.9	0.6	0.3	0.2	0.3	0.3	0.2	0.3	0.3	0.3
Manufacturing industries and construction	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.1
Transport	90.8	23.1	23.6	26.9	27.7	28.5	27.1	24.5	25.3	25.9
Other sectors	17.2	14.3	13.3	14.5	15.0	16.1	16.6	14.8	16.3	16.3
Industrial processes	1.6	1.9	1.8	0.8	0.2	0.2	0.2	0.1	0.1	0.1
Solvents and other product use	0.001	0.001	0.002	0.002	0.003	0.002	0.001	0.002	0.001	0.001
Agriculture	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002
Waste	0.02	0.02	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.02
Total	110.7	40.2	39.1	42.5	43.4	45.2	44.2	39.8	42.0	42.7

The significant decrease in the PCBs emission is caused by a decrease in the emissions from road transport. Leaded gasoline has high emission factors for PCBs and when this fuel was phased out in the beginning of the 1990's, the emissions also decreased significantly.

The other dominant emission sources are both road and non-road diesel vehicles and the emission from these sources has increased due to an increase in the diesel consumption. The emission from energy industries has decreased due to flue gas abatement targeting dioxin and furans installed at the waste incineration plants. The emission from industrial processes has decreased due to the closure of steel production and secondary aluminium production in Denmark. For the remaining sectors the emissions are quite low and stable over time.

The screening for emission sources for pentachlorobenzene (PeCB) showed that the available data are extremely limited and in many cases the reported data are based on highly uncertain assumptions. While it can be assumed that emissions of PeCB can occur from the same sources as HCB, very few measurements have been reported in the literature.

Based on the available data, it appears that waste incineration will be the largest source of PeCB emissions to air. However, the data foundation is at the moment not strong enough to facilitate the establishment of an emission inventory.

Sammenfatning

De danske luftemissionsopgørelser for hexachlorbenzen (HCB) og polychlorerede bifenyl (PCBs) har hidtidigt ikke været komplette. I dette projekt er emissionsopgørelserne for de to forureningskomponenter blevet forbedret ved inddragelsen af de nyeste tekniske retningslinjer angivet i EMEP/EEA guidebogen. I tillæg til informationerne i Guidebogen er der gennemført et litteraturstudie for at komplementere og verificere de emissionsfaktorer, der er oplyst i Guidebogen.

Baseret på de tilgængelige data i Guidebogen og den øvrige videnskabelige litteratur har det været muligt at forbedre kompletheden af rapporteringen betragteligt. Der er for HCB kun ni kategorier, hvor emissionerne rapporteres som ikke estimeret (NE - not estimated) sammenlignet med 30 ved sidste emissionsopgørelse og 13 kategorier sammenlignet med 50 kategorier for PCB, hvor emissionerne rapporteres som ikke estimeret (NE - not estimated). Årsagerne til at der fortsat rapporteres NE for nogle kategorier er enten, at hele kategorien ikke er rapporteret i den danske emissionsopgørelse, f.eks. på grund af manglende aktivitetsdata eller at det ikke har været muligt at finde en emissionsfaktor for HCB og/eller PCBs, men at der er indikationer på at emissioner af HCB og/eller PCBs kan forekomme. Sådanne indikationer kan for eksempel være, at der er rapporterede og veldokumenterede emissionsfaktorer for dioxiner og furaner (PCDD/F).

Resultatet af den forbedrede emissionsopgørelse for HCB er vist i Tabel S.1 nedenfor.

Tabel S.1 Dansk emissionsopgørelse for HCB for 1990-2011, kg.

	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Energi- og konverteringssektoren	4,4	3,3	1,5	1,2	1,7	1,5	1,4	1,4	1,4	1,2
Fremstillingsvirksomhed og bygge/-anlæg	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,1
Transport	0,4	0,4	0,5	0,6	0,6	0,7	0,7	0,6	0,7	0,7
Andet energiforbrug	1,5	1,0	0,4	0,4	0,4	0,5	0,5	0,4	0,5	0,5
Industrielle processer	2,0	2,3	2,7	1,4	0,7	0,7	0,6	0,01	0,01	0,01
Anvendelse af opløsningsmidler og produkter	0,001	0,001	0,001	0,001	0,002	0,001	0,001	0,001	0,001	0,001
Landbrug	18,4	0,6	0,4	0,1	0,1	0,1	0,1	0,1	0,1	0,1
Affald	0,01	0,01	0,01	0,01	0,01	0,01	0,01	0,01	0,01	0,01
Total	26,8	7,8	5,6	3,8	3,7	3,6	3,3	2,7	2,8	2,6
Total rapporteret i 2013	3,2	1,9	0,6	0,5	0,5	0,5	0,6	0,5	0,6	0,6

Det markante fald i HCB-emissionen skyldes et fald i emissionen fra landbrugssektoren. Faldet skyldes dels et fald i anvendelsen af pesticider og fungicider, der indeholder HCB urenheder og dels at koncentrationen af HCB-urenheder i pesticider blev reduceret betragteligt i begyndelsen af 1990'erne.

Emissionen fra energi- og konverteringssektoren er også faldet på grund af installationen af røggasrensingsudstyr rettet mod reduktioner af PCDD/F fra affaldsforbrændingsanlæg. Emissionen fra industrielle processer er faldet på grund af nedlukningen af produktionen af stål og sekundær (omsmeltet) aluminium i Danmark. For de resterende sektorer er emissionerne ret lave og har været forholdsvis konstante gennem tidsserien.

Resultatet af den forbedrede emissionsopgørelse for PCBs er vist i Tabel S.2 nedenfor.

Tabel S.2 Dansk emissionsopgørelse for PCBs for 1990-2011, kg.

	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Energi- og konverteringssektoren	0,9	0,6	0,3	0,2	0,3	0,3	0,2	0,3	0,3	0,3
Fremstillingsvirksomhed og bygge/-anlæg	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,0	0,1	0,1
Transport	90,8	23,1	23,6	26,9	27,7	28,5	27,1	24,5	25,3	25,9
Andet energiforbrug	17,2	14,3	13,3	14,5	15,0	16,1	16,6	14,8	16,3	16,3
Industrielle processer	1,6	1,9	1,8	0,8	0,2	0,2	0,2	0,1	0,1	0,1
Anvendelse af opløsningsmidler og produkter	0,001	0,001	0,002	0,002	0,003	0,002	0,001	0,002	0,001	0,001
Landbrug	0,00002	0,00002	0,00002	0,00002	0,00002	0,00002	0,00002	0,00002	0,00002	0,00002
Affald	0,02	0,02	0,02	0,02	0,02	0,03	0,03	0,03	0,03	0,02
Total	110,7	40,2	39,1	42,5	43,4	45,2	44,2	39,8	42,0	42,7

Det betydelige fald i PCBs-emissionen skyldes et fald i emissionen fra vejtransport. Blyholdig benzin har høje emissionsfaktorer for PCBs, så da blyholdig benzin blev forbudt og dermed udfaset i begyndelsen af 1990'erne faldt emissionen også kraftigt.

De andre dominerende emissionskilder til PCBs er både vejgående og ikke-vejgående dieselmotorer. Emissionen fra disse kilder er steget i takt med det stigende dieselforbrug. Emissionen fra energi- og konverteringssektoren er faldet på grund af installationen af røggasrensingsudstyr rettet mod reduktioner af PCDD/F fra affaldsforbrændingsanlæg. Emissionen fra industrielle processer er faldet på grund af ophøret af produktionen af stål og sekundær aluminium i Danmark. For de resterende sektorer er emissionerne ret lave og har været forholdsvis konstante gennem tidsserien.

Den gennemførte screening for emissionskilder til pentachlorbenzen (PeCB) viser, at de tilgængelige data er meget sparsomme og i mange tilfælde er de rapporterede data baseret på meget usikre antagelser. Det kan antages, at emissioner af PeCB kan forekomme fra samme kilder som HCB-emission, men der er udført meget få målinger, som er rapporteret i litteraturen.

Baseret på de tilgængelige data lader det til, at affaldsforbrænding vil være den vigtigste kilde til PeCB-emissioner til luften. Datagrundlaget er dog på nuværende tidspunkt så spinkelt, at der ikke er grundlag for at lave en egentlig national emissionsopgørelse.

1 Introduction

The reporting of emissions of persistent organic pollutants (POPs) is regulated by international conventions. As part of the international regulation, Denmark is obligated to report emission inventories.

To improve the Danish emission inventories for POPs, it is necessary to complete the hexachlorobenzene (HCB) emission inventory and implement an emission inventory for PCBs.

The purpose of this report is to document the methodologies for estimating emissions of HCB and PCBs and including the results in the 2014 reporting to the Convention on Long-Range Transboundary Air Pollution (CLRTAP).

When the work on improving the HCB and PCBs emission inventory begun, the latest historic year was 2011. Hence the data showed in this report are only for 1990-2011. In some cases recalculation of activity data means that the emission number contained in this report will be slightly different from the emission values that will be reported to the CLRTAP in the spring of 2014.

1.1 International obligations

The international regulation of POPs is covered by two major conventions: The Convention on Long-Range Transboundary Air Pollution (CLRTAP) under the United Nations Economic Commission for Europe (UNECE) and the Stockholm Convention on Persistent Organic Pollutants.

The CLRTAP contains a number of protocols including the Protocol on Persistent Organic Pollutants (the POP Protocol), which Denmark has ratified. The POP Protocol requires that Parties report emission inventories for dioxins and furans (PCDD/F), polycyclic aromatic hydrocarbons (PAHs), hexachlorobenzene (HCB) and polychlorinated biphenyls (PCBs). Emissions are to be reported from 1990 onwards and covers all source sectors as defined in the UNECE CLRTAP reporting guidelines (UNECE, 2009) and included in the EMEP/EEA Guidebook (EEA, 2009).

In addition to the two UN conventions, the EU E-PRTR regulation (Council Regulation (EC) No 166/2006) includes POPs. However, in the E-PRTR reporting there are threshold values that determine when facilities are obligated to report emissions. The threshold values are included in Table 1.

Table 1 Threshold values for HCB, PCBs and PeCB in the EU E-PRTR regulation.

PRTR pollutant	Threshold value, kg per year
HCB	10
PCBs	0.1
PeCB	1

1.2 Danish emission inventories

Denmark annually reports emission inventories of air pollution to the CLRTAP Convention. The reporting follows the agreed reporting format, NFR (Nomenclature for Reporting). The Danish emission inventories for PCDD/F and PAHs are considered complete in terms of coverage of sectors. Until

now there has not been an emission inventory for PCBs and the reporting for HCB has only covered some source categories within stationary combustion and waste.

In general many of the source categories where emissions of PCDD/F occur will also be relevant to consider as possible source categories for HCB and PCBs.

The list of NFR categories is included in Chapter 1.3.3 including an indication of whether each category is considered relevant for emissions of HCB and PCBs.

1.3 Relevant source categories

1.3.1 Stockholm Convention

Existing sources that may result in unintentional formation and emissions of HCB and PCBs, as listed in the Annex C of the Stockholm Convention, are as follows:

Industrial source categories according to Annex C of the Stockholm Convention, Part II, “having the potential for comparatively high formation and release of these chemicals to the environment”:

- Waste incinerators, including co-incinerators of municipal, hazardous or medical waste or of sewage sludge
- Cement kilns firing hazardous waste
- Production of pulp using elemental chlorine or chemicals generating elemental chlorine for bleaching
- The following thermal processes in the metallurgical industry:
 - Secondary copper production
 - Sinter plants in the iron and steel industry
 - Secondary aluminium production
 - Secondary zinc production

Source categories according to Annex C of the Stockholm Convention, Part III, “that may also unintentionally form and release”:

- Open burning of waste, including burning of landfill sites
- Thermal processes in the metallurgical industry not mentioned in Part II
- Residential combustion sources
- Fossil fuel-fired utility and industrial boilers
- Firing installations for wood and other biomass fuels
- Specific chemical production processes releasing unintentionally formed persistent organic pollutants, especially production of chlorophenols and chloranil
- Crematoria
- Motor vehicles, particularly those burning leaded gasoline
- Destruction of animal carcasses
- Textile and leather dyeing (with chloranil) and finishing (with alkaline extraction)
- Shredder plants for the treatment of end of life vehicles
- Smouldering of copper cables
- Waste oil refineries

The majority of sources identified in Annex C of the Stockholm Convention are already source categories that are considered within the Danish air emission inventories for other air pollutants. Exceptions are activities that do not occur in Denmark, e.g. pulp production and sinter plants.

However, one exception is possible emissions from shredder plants. This activity is not covered by the Danish emission inventories. There is also no guidance available in the EMEP/EEA Guidebook (EEA, 2009) on estimating emissions from this activity.

1.3.2 E-PRTR

Council Regulation (EC) No 166/2006 requires reporting for a large number of activities if the emission exceeds specific thresholds. The thresholds for HCB, PCBs and PeCB are shown in Table 1. The activities covered by the regulation with relevance for HCB and PCBs include: energy industry (e.g. power plants and refineries), metal industry, mineral industry, chemical industry, waste and wastewater management and paper and wood production and processing.

In 2012, no Danish companies reported emissions of HCB or PeCB to the Danish Environmental Protection Agency. One company reported an emission of PCBs of 0.64 kg. This company is involved with recycling (shredding) of waste. The reporting to the Danish EPA does not contain any information on the calculation methodology for the emission.

1.3.3 The POP Protocol

The POP Protocol refers to the general guidelines for reporting of national air emission inventories. The list of source categories is included in Table 2. Not all source categories are relevant for Denmark even though HCB and/or PCBs are indicated as relevant. This is due to the fact that the specific activity is not occurring in Denmark.

Table 2 List of reporting categories under the Convention on Long-Range Transboundary Air Pollution.

NFR category	HCb relevance	PCBs relevance
1 A 1 a Public electricity and heat production	X	X
1 A 1 b Petroleum refining	X	X
1 A 1 c Manufacture of solid fuels and other energy industries	X	X
1 A 2 a Manufacturing industries and construction: Iron and steel	X	X
1 A 2 b Manufacturing industries and construction: Non-ferrous metals	X	X
1 A 2 c Manufacturing industries and construction: Chemicals	X	X
1 A 2 d Manufacturing industries and construction: Pulp, Paper and Print	X	X
1 A 2 e Manufacturing industries and construction: Food processing, beverages and tobacco	X	X
1 A 2 f i Manufacturing industries and construction: Other	X	X
1 A 2 f ii Mobile Combustion in manufacturing industries and construction	(X)	(X)
1 A 3 a ii (i) Civil aviation (Domestic, LTO)	-	-
1 A 3 a i (i) International aviation (LTO)	-	-
1 A 3 b i Passenger cars	(X)	(X)
1 A 3 b ii Light duty vehicles	(X)	(X)
1 A 3 b iii Heavy duty vehicles	(X)	(X)
1 A 3 b iv Mopeds & motorcycles	(X)	(X)
1 A 3 b v Gasoline evaporation	NR	NR
1 A 3 b vi Automobile tyre and brake wear	NR	NR
1 A 3 b vii Automobile road abrasion	NR	NR
1 A 3 c Railways	(X)	(X)
1 A 3 d i (ii) International inland waterways	(X)	(X)
1 A 3 d ii National navigation (Shipping)	(X)	(X)
1 A 3 e Pipeline compressors	-	-
1 A 4 a i Commercial/institutional: Stationary	X	X
1 A 4 a ii Commercial/institutional: Mobile	(X)	(X)
1 A 4 b i Residential: Stationary plants	X	X
1 A 4 b ii Residential: Mobile	(X)	(X)
1 A 4 c i Agriculture/Forestry/Fishing: Stationary	X	X
1 A 4 c ii Agriculture/Forestry/Fishing: Off-road vehicles and other machinery	(X)	(X)
1 A 4 c iii Agriculture/Forestry/Fishing: National fishing	(X)	(X)
1 A 5 a Other stationary (including military)	X	X
1 A 5 b Other, Mobile (including military)	(X)	(X)
1 B 1 a Coal mining and handling	NR	NR
1 B 1 b Solid fuel transformation	NR	NR
1 B 1 c Other fugitive emissions from solid fuels	NR	NR
1 B 2 a i Exploration, production, transport	NR	NR
1 B 2 a iv Refining / storage	NR	NR
1 B 2 a v Distribution of oil products	NR	NR
1 B 2 b Natural gas	NR	NR
1 B 2 c Venting and flaring	-	-
1 B 3 Other fugitive emissions	NR	NR
2 A 1 Cement production	X	X
2 A 2 Lime production	X	X
2 A 3 Limestone and dolomite use	NR	NR
2 A 4 Soda ash production and use	NR	NR
2 A 5 Asphalt roofing	-	-
2 A 6 Road paving with asphalt	-	-
2 A 7 a Quarrying and mining of minerals other than coal	-	-
2 A 7 b Construction and demolition	NR	-
2 A 7 c Storage, handling and transport of mineral products	NR	NR
2 A 7 d Other Mineral products	NR	NR
2 B 1 Ammonia production	NR	NR
2 B 2 Nitric acid production	NR	NR
2 B 3 Adipic acid production	NR	NR
2 B 4 Carbide production	NR	NR
2 B 5 a Other chemical industry	-	-
2 B 5 b Storage, handling and transport of chemical products	NR	NR
2 C 1 Iron and steel production	X	X
2 C 2 Ferroalloys production ¹	-	-
2 C 3 Aluminium production	(X)	(X)
2 C 5 a Copper production ¹	(X)	X
2 C 5 b Lead production	(X)	X
2 C 5 c Nickel production ¹	(X)	(X)
2 C 5 d Zinc production ¹	(X)	X
2 C 5 e Other metal production	-	-
2 C 5 f Storage, handling and transport of metal products	NR	NR
2 D 1 Pulp and paper	-	-
2 D 2 Food and drink	NR	NR
2 D 3 Wood processing	NR	NR
2 E Production of POPs	-	-
2 F Consumption of POPs and heavy metals (e.g. electrical and scientific equipment)	-	X

NFR category	HCB relevance	PCBs relevance
<i>Continued</i>		
2 G Other production, consumption, storage, transportation or handling of bulk products	-	-
3 A 1 Decorative coating application	NR	(X)
3 A 2 Industrial coating application	NR	(X)
3 A 3 Other coating application	NR	(X)
3 B 1 Degreasing	NR	-
3 B 2 Dry cleaning	NR	NR
3 C Chemical products	(X)	(X)
3 D 1 Printing	NR	NR
3 D 2 Domestic solvent use including fungicides	(X)	(X)
3 D 3 Other product use	(X)	(X)
4 B 1 a Cattle dairy	NR	NR
4 B 1 b Cattle non-dairy	NR	NR
4 B 2 Buffalo	NR	NR
4 B 3 Sheep	NR	NR
4 B 4 Goats	NR	NR
4 B 6 Horses	NR	NR
4 B 7 Mules and asses	NR	NR
4 B 8 Swine	NR	NR
4 B 9 a Laying hens	NR	NR
4 B 9 b Broilers	NR	NR
4 B 9 c Turkeys	NR	NR
4 B 9 d Other poultry	NR	NR
4 B 13 Other	NR	NR
4 D 1 a Synthetic N-fertilizers	NR	NR
4 D 2 a Farm-level agricultural operations	NR	NR
4 D 2 b Off-farm storage, handling and transport of bulk agricultural products	NR	NR
4 D 2 c N-excretion on pasture range and paddock	NR	NR
4 F Field burning of agricultural wastes	(X)	(X)
4 G Agriculture other	(X)	-
6 A Solid waste disposal on land	NR	NR
6 B Waste-water handling	NR	NR
6 C a Clinical waste incineration	(X)	(X)
6 C b Industrial waste incineration	(X)	(X)
6 C c Municipal waste incineration	(X)	(X)
6 C d Cremation	(X)	(X)
6 C e Small scale waste burning	(X)	(X)
6 D Other waste	-	-

¹ This activity is not occurring in Denmark.

'X' means that the EMEP/EEA Guidebook (EEA, 2013) contains methodological guidance for estimating emissions.

'(X)' means that the EMEP/EEA Guidebook (EEA, 2013) does not contain guidance but emission factors have been identified from the scientific literature.

'-' means that no guidance has been found.

'NR' means that the source is not considered relevant for emissions of HCB and/or PCBs.

2 Emission inventory methodology for hexachlorobenzene

2.1 Stationary combustion

In the 2013 reporting, HCB emissions was included for combustion of wood, straw, waste, coal, gas oil fuelled engines and biogas fuelled engines. The improved emission inventory includes emission factors for all fuels and all years.

2.1.1 Methodology

All fuel combustion data have been included in the inventory. HCB emission factors have been added for all fuels and some existing emission factors have been revised.

Table 3 shows the references for HCB emission.

Table 3 HCB emission factors for stationary combustion, all references.

Reference	Fuel	Emission factor, ng/GJ	Technology and comment
EEA (2013)	Coal	6700	Public electricity and heat production. Refers to Grochowalski & Konieczynski (2008)
Grochowalski & Konieczynski (2008)	Coal	6200-26 700	Power plants, 4 fluid bed boilers. Including combustion of coal silt and waste coal for two plants.
Grochowalski & Konieczynski (2008)	Coal	6700 (4900-8900)	Power plants, 2 fluid bed boilers.
Pacyna et al. (2003)	Coal	656	Refers to Pacyna et al. (1999)
Bailey (2001)	Coal	533	Power plants. Literature study.
Andrijewski (2004)	Coal	533	Power plants.
EEA (2013)	Coal	620 000	Residential combustion. Refers to earlier updates of the Guidebook.
Syc et al. (2011)	Coal	23 000-2 450 000	Residential boiler, bituminous coal
Andrijewski (2004)	Coal	5100	Residential plants.
EEA (2013)	Gas oil	220	Engines. Refers to Nielsen et al. (2010)
Pacyna et al. (2003)	Diesel	636	Refers to Pacyna et al. (1999). Recalculated from 21 ng/veh.km based on 33 MJ/km (DCE assumption).
Nielsen et al. (2010)	Gas oil	<220	Engines, emission measurement from 1 plant and the emission was below the detection limit.
Bailey (2001)	Waste	73 300- 48 600 000	Literature study, geometric mean estimated for each of the references.
Nielsen et al. (2010)	Waste	4 300 (200-10 500)	CHP plant, waste incineration, emission measurements from 3 plants.
Pacyna et al. (2003)	Waste	190 000	Refers to Pacyna et al. (1999)
Pacyna et al. (2003)	Solid waste	950 000	Refers to Pacyna et al. (1999)
Pacyna et al. (2003)	Sewage sludge	48 000 000	Refers to Pacyna et al. (1999)
Pacyna et al. (2003)	Medical waste	1 800 000	Refers to Pacyna et al. (1999)
Wegiel et al. (2011)	Waste	81 189 (1681-812 000)	Emission measurements from industrial waste incinerators in Poland. Recalculated from 155 ng/Nm ⁻³ . The PCDD/F emission from the plants is 15 times the emission factor for Denmark and the HCB is 18 times the emission factor in Nielsen et al. (2010).
EEA (2013)	Biomass/Wood	5000	Public electricity and heat production. Refers to Bailey (2001)
EEA (2013)	Biomass	5000	Small combustion, refers to Syc et al. (2011)
Bailey (2001)	Biomass	1300-13 000	Literature study.
Nielsen et al. (2010)	Straw	113 (100-150)	CHP plants, emission measurements from 2 plants.
Hedman et al. (2006)	Wood pellets	27	Residential wood, pellet boiler
Hedman et al. (2006)	Wood	42 (39-44)	Old boilers
Gullet et al. (2003)	Wood	684	Woodstove, steel, lined. Calculation based on 19 MJ/kg.
Gullet et al. (2003)	Wood	18 000	Fireplace. Calculation based on 19 MJ/kg.
Kakareka & Kukharchyk (2005a)	Wood	3300	Residential wood combustion
Joas A. (2006)	Wood	27 800	Residential wood combustion
Syc et al. (2011)	Wood	575-10 200	Residential boilers and stoves, beech logs
Syc et al. (2011)	Wood	524-1 040 000	Residential boilers, spruce logs
Pacyna et al. (2003)	Wood	4100	Refers to Pacyna et al. (1999)
Andrijewski (2004)	Wood	4100	Residential
Syc et al. (2011)	Maize straw pellets	3300	Residential boiler, automatic modern boiler.
Nielsen et al. (2010)	Biogas	190	Engines, emission measurements from 1 plant.
Nielsen et al. (2010)	Producer gas	800	Engines, emission measurements from 1 plant.
CEMENT			
EEA (2013)	(Cement)	4.6 µg/te clinker	Refers to SINTEF (2006)
Bailey (2001)	(Cement)	1.7*10 ⁻⁷ kg HCB/tonne clinker	Cement production

For coal, the emission factor from Grochowalski & Koniecznyński (2008) will be applied for energy industries and for industrial plants. This emission factor is also applied in the EEA Guidebook (EEA, 2013).

For residential plants, the emission factor 1,200,000 ng per GJ will be applied referring to Syc et al. (2011). For commercial/institutional plants and for plants in agriculture / forestry the lower end of the value in Syc et al. (2011) (23 000 ng per GJ) will be applied. The lower value is chosen since the plants in these sectors are usually larger than residential plants and therefore the combustion conditions will be better resulting in lower emissions.

The emission factor for gas oil fuelled CHP engines (220 ng per GJ) referring to Nielsen et al. (2010) will be applied for all liquid fuels except for LPG and refinery gas.

For gaseous fuels, LPG and refinery gas no data are available and the emission is negligible.

For waste combustion, emission data from Danish plants are available and these data will be applied (Nielsen et al., 2010). The emission factor 4300 ng per GJ will be applied for 2005 onwards. The HCB emission factor for 1990 refers to Pacyna et al. (2003). The emission of HCB is related to emission of PCDD/F and the decline rate between 1990 and 2005 is based on the decline rate for PCDD/F.

Recent emission measurements from Polish industrial waste incineration plants confirms the emission factor level for waste incineration considering that the PCDD/F emission level is 15 times the PCDD/F emission level for Danish plants.

For wood combustion, the emission factors from EEA (2013) will be applied for both energy industries, industrial plants and for non-industrial plants. For residential wood combustion, it would be relevant to estimate a time series. However, the currently available data are considered insufficient for this estimate.

The chlorine content in straw is higher than in wood (Villeneuve et al., 2012) and thus the emission from straw combustion might potentially be higher. However, the emission factor for CHP plants combusting straw reported in Nielsen et al. (2010) is lower than the emission factor applied for wood. The measured data for Danish plants therefore do not support the assumption. Differences can also be caused by different contents of metals that can function as catalyst for the HCB formation.

The emission factor for energy industries and industrial combustion refer to Nielsen et al. (2010). For non-industrial plants, the EEA (2013) emission factor will be applied.

The emission factors for biogas and producer gas both refer to Nielsen et al (2010).

Table 4 shows the emission factors and references for the Danish emission factors.

Table 4 Emission factors for HCB, stationary combustion.

Fuel	NFR (SNAP)	Emission factor, ng/GJ	Reference
Coal	1A1, 1A2	6700	Grochowalski & Koniecznyński (2008); EEA (2013)
Coal	1A4b	1 200 000	Syc et al. (2011)
Coal	1A4a and 1A4c	23 000	Syc et al. (2011)
Other solid fuels	1A1, 1A2	6700	Assumed equal to coal.
Other solid fuels	1A4	1 200 000	Assumed equal to coal.
Liquid fuels ¹⁾	1A1, 1A2, 1A4	220	Nielsen et al. (2010)
Gaseous fuels	1A1, 1A2, 1A4	-	Negligible
Waste	1A1, 1A2, 1A4	4300	Nielsen et al. (2010). A time series have been estimated. The emission factor for 1990 (190,000 ng/GJ) refer to Pacyna et al. (2003).
Wood	1A1, 1A2	5000	EEA (2013)
Wood	1A4	5000	EEA (2013)
Straw	1A1, 1A2	113	Nielsen et al. (2010)
Straw	1A4	5000	EEA (2013)
Biogas	1A1, 1A2, 1A4	190	Nielsen et al. (2010)
Producer gas	1A1, 1A2, 1A4	800	Nielsen et al. (2010)

1. Except LPG and refinery gas.

2.1.2 Emission results

HCB emissions in 2011 are shown in Table 5. The largest emission source is public electricity and heat production.

Table 5 HCB emission from stationary combustion plants, 2011.

Sector	HCB, kg
1A1a Public electricity and heat production	1.214
1A1b Petroleum refining	0.000
1A1c Other energy industries	0.000
1A2 Industry	0.082
1A4a Commercial / Institutional	0.006
1A4b Residential	0.216
1A4c Agriculture / Forestry / Fisheries	0.037
Total	1.556

The HCB emission time series is shown in Figure 1. The emission has decreased 74 % since 1990. The decrease is mainly a result of the flue gas cleaning devices that have been installed in waste incineration plants for dioxin reduction.

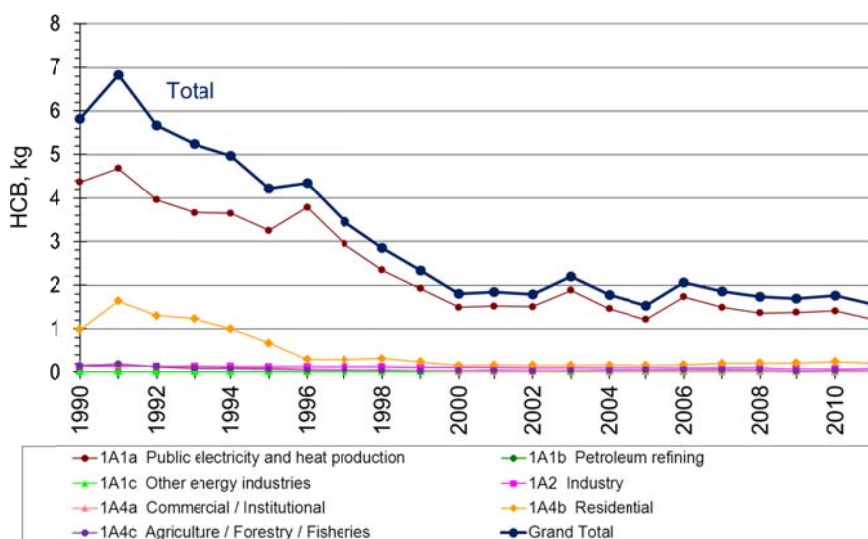


Figure 1 HCB emission time series for stationary combustion.

2.1.3 Recalculations

Emission factors

The emission factor for coal has been changed. The former emission factor for coal was 620 ng per GJ whereas the revised emission factor for the energy and industrial sectors is 6700 ng per GJ. For residential plants, the emission factor has been changed to 1 200 000 ng per GJ and for commercial/institutional plants and for plants in agriculture and forestry to 23 000 ng per GJ.

The emission factor for coal is now applied for other solid fuels as well.

The emission factor for gas oil fuelled engines (220 ng per GJ) is now applied for all technologies and for all liquid fuels except LPG and refinery gas.

The emission factor for wood combustion has been changed from 4000 ng per GJ to 5000 ng per GJ.

For straw combustion the emission factor for small combustion have been changed from 113 ng per GJ to 5000 ng per GJ.

The emission factor for producer gas fuelled engines has been added.

Emission

The estimated HCB emissions for 1990-2011 have been compared to the HCB emissions reported in the last inventory. The improved inventory results in an emission estimate there is 1.8-4.0 times larger than the latest reported inventory.

2.2 Mobile combustion

2.2.1 Methodology

For road transport, HCB emission factors are taken from EMEP/MSC East (2000) based on work made by Pacyna (1999). Based on average fuel consumption factors derived from the road transport part of the Danish emission inventories (Nielsen et al., 2013) and lower heating values for diesel and gasoline, the emission factors are recalculated from ng/km to g/TJ¹. These fuel related emission factors are also used for other mobile sources except navigation and fisheries.

For mobile sources emission factors measured by Cooper (2005) are used for marine diesel oil (MDO) and residual oil (heavy fuel oil: HFO). For aviation no emission factor could be found in the available literature.

The HCB emission factors used are shown in Table 6.

¹ Average fuel consumption factors (g/km): 80 (all diesel vehicles); 55 (gasoline cars and vans); 55 (diesel cars and vans). Lower heating values (MJ/kg): 42.7 (diesel); 43.8 (gasoline)

Table 6 HCB emission factors for mobile sources. Reference values and fuel related factors (g/TJ).

Pollutant	Category	Reference	Fuel type	Unit	Value	g/TJ
HCB	Road/Other land based	EMEP/MSC (2000)	Motor gasoline	ng/km	0.87	3.61E-04
HCB	Road/Other land based	EMEP/MSC (2000)	Unleaded motor gasoline	ng/km	0.024	9.96E-06
HCB	Road/Other land based	EMEP/MSC (2000)	Diesel	ng/km	21	6.15E-03
HCB	Navigation	Cooper (2005)	MDO	g/TJ		1.95E-03
HCB	Navigation	Cooper (2005)	HFO	g/TJ		3.50E-03

The fuel activity data taken from the latest published Danish emission inventories (Nielsen et al., 2013) are listed in Table 7.

Table 7 Fuel activity data (TJ) for mobile sources in Denmark.

NFR Category	Fuel type	1990	1995	2000	2005	2008	2009	2010	2011
Industry - Other (1A2f)	Diesel	10 158	10 324	10 773	11 753	13 970	10 284	13 015	12 678
Industry - Other (1A2f)	Gasoline	175	169	167	165	159	118	157	156
Road (1A3b) - exhaust	Diesel	59 947	64 013	69 196	88 264	103 491	97 560	101 342	105 150
Road (1A3b) - exhaust	Gasoline	66 279	80 101	83 312	77 835	72 533	67 940	63 899	60 084
Railways (1A3c)	Diesel	4010	4093	3079	3137	3199	3111	3273	3370
Navigation (1A3d)	Diesel	5628	7565	6035	5561	5395	5319	5071	4757
Navigation (1A3d)	Gasoline	309	358	396	393	361	353	346	340
Navigation (1A3d)	Residual oil	4571	3382	1444	1859	2148	2287	2456	2375
Comm./Inst. (1A4a)	Gasoline	1010	1068	1186	2214	2409	2389	2368	2348
Residential (1A4b)	Gasoline	535	544	583	806	865	862	860	858
Agriculture/forestry (1A4c)	Diesel	16 642	15 474	13 856	14 608	16 598	16 768	16 982	17 430
Agriculture/forestry (1A4c)	Gasoline	1050	740	521	419	542	574	572	571
Fisheries (1A4c)	Diesel	7920	7134	7422	8725	8106	7514	7770	7788
Military (1A5)	Diesel	146	1794	369	1271	630	1099	596	1045
Military (1A5)	Gasoline	1	0	1	7	12	9	6	0
Navigation int. (1A3d)	Diesel	11 289	26 370	22 129	11 330	10 928	10 164	11 356	10 282
Navigation int. (1A3d)	Residual oil	27 815	38 780	32 437	19 411	25 650	9416	15 682	17 120

Additional information regarding the share of leaded and unleaded gasoline sold in Denmark needed to carry out the subsequent emission calculations (Table 8) are derived from the Danish inventories (Nielsen et al., 2013).

Table 8 Share of leaded and unleaded gasoline sold in Denmark.

Fuel quality	Unit	1990	1995-
Leaded gasoline	%	42.5	0
Unleaded gasoline	%	57.5	100

Additional information regarding the amount of diesel used by cars/vans and heavy duty vehicles needed to carry out the subsequent emission calculations (Table 9) are derived from the Danish inventories (Nielsen et al., 2013).

Table 9 Diesel consumption by cars/vans and heavy duty vehicles in Danish road transport.

Unit	1990	1995	2000	2005	2008	2009	2010	2011
Diesel Cars/Vans	23 192	25 966	29 511	44 272	59 041	58 308	60 860	64 967
Diesel Heavy duty vehicles	36 755	38 047	39 685	43 992	44 450	39 252	40 482	40 183

2.2.2 Emission results

The emissions are calculated as the product of the emission factor and the fuel consumption for each NFR category/fuel type layer using the following equation:

$$E = \sum_{C,f} FC_{C,f} \cdot EF_{C,f}$$

Where: E = Emission of HCB
FC = Fuel consumption
EF = HCB Emission factor

C = NFR category

f = fuel type

The HCB emissions from mobile sources are shown in Table 10.

Table 10 HCB emissions (g) for mobile sources in Denmark.

NFR Category	Fuel type	1990	1995	2000	2005	2008	2009	2010	2011
Industry - Other (1A2f)	Diesel	62.4	63.5	66.2	72.3	85.9	63.2	80.0	77.9
Industry - Other (1A2f)	Gasoline	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Road (1A3b) - exhaust	Diesel	368.5	393.5	425.4	542.6	636.2	599.8	623.0	646.4
Road (1A3b) - exhaust	Gasoline	10.6	0.8	0.8	0.8	0.7	0.7	0.6	0.6
Railways (1A3c)	Diesel	24.7	25.2	18.9	19.3	19.7	19.1	20.1	20.7
Navigation (1A3d)	Diesel	11.0	14.8	11.8	10.8	10.5	10.4	9.9	9.3
Navigation (1A3d)	Gasoline	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Navigation (1A3d)	Residual oil	16.0	11.8	5.1	6.5	7.5	8.0	8.6	8.3
Comm./Inst. (1A4a)	Gasoline	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Residential (1A4b)	Gasoline	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Agriculture/forestry (1A4c)	Diesel	102.3	95.1	85.2	89.8	102.0	103.1	104.4	107.2
Agriculture/forestry (1A4c)	Gasoline	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Fisheries (1A4c)	Diesel	15.4	13.9	14.5	17.0	15.8	14.7	15.2	15.2
Military (1A5)	Diesel	0.9	11.0	2.3	7.8	3.9	6.8	3.7	6.4
Military (1A5)	Gasoline	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total national		612.3	629.6	630.1	766.9	882.3	825.7	865.5	892.1
Navigation int. (1A3d)	Diesel	22.0	51.4	43.2	22.1	21.3	19.8	22.1	20.0
Navigation int. (1A3d)	Residual oil	97.4	135.7	113.5	67.9	89.8	33.0	54.9	59.9

2.3 Fugitive emissions from fuels

No sources of emissions of HCB have been identified for the sector “Fugitive emissions from fuels”. This finding is in correspondence with the EMEP/EEA guidebook (EEA, 2009), that does not include methodology or emission factors for HCB for any sources in the fugitive sector.

Fugitive HCB formation could occur either due to evaporation of fuels containing HCB or due to combustion processes, e.g. flaring. Neither raw oil, natural gas nor gasoline are assumed to contain HCB. Flaring of oil and gas are the only combustion processes in the fugitive emission sector. According to European Commission (2013) it is highly unlikely that the formation conditions for PCBs occur for flaring in refineries and this is assumed to be the case for offshore flaring as well. Formation of both HCB and PCBs require presence of chlorine, and the chlorine content is assumed to be very limited

in oil and natural gas being flared offshore and in refineries. Therefore emissions of both HCB and PCBs are considered not occurring or negligible.

2.4 Industrial processes

A number of industrial processes may contribute to national emission of HCB. Based on the submitted national emission data EMEP has calculated the key sources (Gusev et al., 2011); see Table 11. Metal industry (2C1 and 2C5e), Chemical industry (2B5a), and Production of POPs (2E) contribute significantly to the emission of HCB. However, the HCB emissions are uncertain and the analysis is based on incomplete data sets from the contributing countries and therefore the analysis has to be used as an indicator of which processes to focus at in the search for emission factors.

Table 11 Key source analysis for HCB emission based on submitted nation emission inventories. Based on Gusev et al. (2011).

NFR Code	NFR Category	Contribution to total emission %	Cumulative total %
2 C 1	<i>Iron and steel production</i>	59.4	59.4
1 A 4 b i	Residential: Stationary plants	9.4	68.8
4 G	Other Agricultural processes	4.7	73.5
1 A 1 a	Public electricity and heat production	4.4	77.9
6 C b	Industrial waste incineration	3.9	81.7
2 B 5 a	<i>Other chemical industry</i>	3.8	85.6
2 E	<i>Production of POPs</i>	1.8	87.4
1 A 3 b i	Road transport: Passenger cars	1.8	89.1
2 C 5 e	<i>Other metal production</i>	1.7	90.8

Wang et al. (2010) reports HCB in environmental samples from China, however, no measured emissions were available. They identify certain parts of chemical industry and metal industry as industrial sources.

2.4.1 Methodology

The emissions of HCB are calculated as the product of the emission factor and the activity for each NFR category using the following equation:

$$E = \sum_{C,f} Act_C \cdot EF_C$$

Where: E = Emission of HCB
 Act_C = Activity
 EF_C = HCB Emission factor
 C = NFR category

Table 12 presents emission factors for HCB from industrial processes.

Table 12 Emission factors for HCB from industrial processes.

Air pollution control		Country	Emission factor mg HCB per tonne	Reference	Comment
Cement production			0.011	EEA, 2006	
Cement production			0.017-1.7	Bailey, 2001	
Cement production			0.021	Andrijewski, 2004	
Cement production			0.010	Bipro, 2006	
Lime production		Japan	0.008	Bipro, 2006	
Steelwork, Electric Arc Furnace (EAF), secondary raw materials	Sedimentation/pos-combustion chamber, cyclone and bag filter	Portugal	3.2	Antunes et al., 2012	Measured
Steelwork, Electric Arc Furnace (EAF), with secondary fusion on ladle furnace (refining)	Sedimentation/pos-combustion chamber, cyclone and bag filter	Portugal	0.094	Antunes et al., 2012	Measured
Steelwork, EAF		Japan	2.0	Bipro, 2006	
Cast iron on cubilot furnace, secondary raw materials	Sedimentation/pos-combustion chamber, cyclone and bag filter	Portugal	0.012	Antunes et al., 2012	Measured
Cast iron on cubilot furnace/induction furnace, secondary raw materials	Cyclone, electrostatic air cleaner and bag filter	Portugal	0.015	Antunes et al., 2012	Measured
Iron foundries		Poland	0.040	Bipro, 2006	
Aluminium, primary/casting			0.295	Saarinén et al., 2007	
Aluminium smelting on reverberatory furnace/ladle furnace, secondary	None	Portugal	0.0012	Antunes et al., 2012	Measured
Aluminium smelting on induction furnace, secondary	None	Portugal	0.012	Antunes et al., 2012	Measured
Aluminium smelting on reverberatory furnace, secondary	None	Portugal	0.026	Antunes et al., 2012	Measured
Aluminium, secondary			5 000	EEA, 2006	
Aluminium, secondary			2 200	Bailey, 2001	
Aluminium, secondary		Poland, Japan	20.0	Bipro, 2006	
Aluminium, secondary			0.660	Saarinén et al., 2007	
Lead smelting on crucible, secondary	None	Portugal	0.0018	Antunes et al., 2012	Measured
Lead production		Italy	0.300	Bipro, 2006	
Copper/zinc/lead smelting on induction furnace, secondary	Cyclone and bag filter	Portugal	0.010	Antunes et al., 2012	Measured
Lead/zinc/tin smelting on induction furnace/crucible, secondary	Cyclone and bag filter	Portugal	0.022	Antunes et al., 2012	Measured
Zinc, primary – smelting in hot dip galvanising process			1.456	Toda, 2005	
Zinc, secondary		Poland, Japan	10.0	Bipro, 2006	
Copper, secondary			3.90-390	Bailey, 2001	
Copper, secondary			39.0	Pacyna, 2003	
Copper, secondary		Poland, Japan	20.0	Bipro, 2006	
Pulp and paper		Japan	0.009	Bipro, 2006	

Table 13 presents the applied EFs for HCB.

Table 13 Applied EFs for HCB.

NFR Code	NFR Category	Applied EF mg HCB per tonne
2A1	Cement production	Included in 1A2
2A2	Lime production	0.008
2B5a	Other chemical industry	- ¹
2C1	Steelwork, EAF	3.2
2C1	Iron foundry	0.04
2C3	Secondary aluminium production	20
2C5b	Secondary lead production	0.3
2E	Production of POPs	- ¹
2F	Consumption of POPs and heavy metals (e.g. electrical and scientific equipment)	- ^{2,3}

¹ There is no indication of production of HCB in Denmark – either as intermediate or as fungicide.

² There is no indication of use of HCB or products containing HCB as fungicide in Denmark. A more general investigation of this issue may be necessary.

³ There is no indication that HCB has been imported for use in chemical industry.

2.4.2 Emission results

Based on the applied emission factors the emissions of HCB from industrial processes have been estimated as shown in Table 14.

Table 14 Emissions of HCB from industrial processes, kg.

NFR category	1990	1995	2000	2005	2010	2011
2A2 Lime production	0.0012	0.0009	0.0008	0.0007	0.0005	0.0004
2C1 Steelwork, EAF	1.96	2.29	2.02	0.80	0	0
2C1 Iron foundry	0.0041	0.0036	0.0041	0.0040	0.0029	0.0033
2C3 Secondary aluminium production	0	0	0.64	0.56	0	0
2C5b Secondary lead production	0.0015	0.0015	0.0015	0.0015	0.0015	0.0015
Total	1.97	2.30	2.67	1.36	0.0049	0.0052

2.5 Solvent and other product use

The reporting for solvent and other product use covers the following categories:

- 3A1 Decorative coating application
- 3A2 Industrial coating application
- 3A3 Other coating application
- 3B1 Degreasing
- 3B2 Dry cleaning
- 3C Chemical products
- 3D1 Printing
- 3D2 Domestic solvent use including fungicides
- 3D3 Other product use

2.5.1 Methodology

HCB occurs as a by-product from the production of certain chlorinated solvents (Pacyna et al., 2003). There may therefore be unintentional emissions of HCB from industrial chemical production of chlorine and the industrial synthesis of chlorinated hydrocarbon/solvents, e.g. trichloroethene, tetra-

chloromethane, tetrachloroethene, carbontetrachloride and vinylchloride. Furthermore, HCB emissions occur from application of chlorine containing solvents where HCB can be present in trace amounts.

HCB has been used in wood protection agents as fungicide in PCP where HCB emissions, especially in buildings, can be calculated from population density as surrogate parameter. HCB has also been used as fire retardant in wood and plastics and as softener especially for PVC (EMEP/CORINAIR, 2006b).

Sources to emissions are many and diverse, some emissions are direct through industrial activities or use of products, and some are indirect, e.g. via disposal or incineration of pollutant containing products and materials. Table 15 below provides information on the activities and products within solvent and other product use that lead to atmospheric emissions of HCB.

Table 15 Activities and products with HCB use and emissions to air related to NFR 3 Solvent and Other Product Use. Blank cells indicate NFR3 categories that are not relevant with respect to HCB use and emissions.

NFR 3	Product group/activity	HCB sources	HCB emission factors
NFR 3A1 Decorative coating application	Domestic use		
	Construction and buildings		
	Agriculture, forestry, fishing and fish farms		
NFR 3A2 Industrial coating application	Manufacture of automobiles ¹⁾		
	Coil coating ¹⁾		
	Car repair ¹⁾		
	Wood ¹⁾		
	Other industrial paint application ¹⁾		
	Boat building ¹⁾		
NFR 3A3 Other coating application	Other non-industrial paint application		
NFR 3B1 Degreasing	Metal degreasing ¹⁾		
	Other industrial (dry) cleaning ¹⁾		
	Electronic components manufacturing ¹⁾		
NFR 3B2 Dry cleaning	Dry cleaning ¹⁾		
NFR 3C Solvents in chemical products manufacture and processing	Polyester processing ¹⁾		
	Polyvinylchloride processing ¹⁾	Softener in PVC	No data (polyvinyl chloride production) ²⁾
	Polyurethane foam processing ¹⁾		
	Polystyrene foam processing ¹⁾		
	Rubber processing ¹⁾		
	Pharmaceutical products manufacturing ¹⁾		
	Paints manufacturing ¹⁾		
	Inks manufacturing ¹⁾		
	Vehicle manufacturing ¹⁾		
	Glues manufacturing ¹⁾		
	Adhesive, magnetic tapes, films & photographs manufacturing ¹⁾		
	Textile finishing ¹⁾		
	Leather tanning ¹⁾		

	Other ¹⁾	By-product from production of chlorinated solvents (trichloroethene, tetrachloromethane, tetrachloroethene, carbontetrachloride and vinylchloride)	1-20 g/t tetrachloromethane 1-6 g/t trichloroethylene 3-10 tetrachloroethylene (by-product from production of chlorinated solvents) ²⁾
	Asphalt blowing ¹⁾		
	Creosote-treated materials		
NFR 3D1 Printing	Printing industry ¹⁾		
NFR 3D2 Domestic solvent use including fungicides	Domestic solvent use (other than paint application)	Application of chlorine containing solvents where HCB can be present in trace amounts	2 mg/t (application of chlorine containing solvents where HCB can be present in trace amounts) ³⁾
	Personal care products		
	Pharmaceuticals (see below)		
	Household cleaning agents		
	Motor & vehicle cleaning agents		
	Use of adhesives and sealants		
	Use of textiles		
	Fungicide	Agricultural application of fungicides containing impurities of HCB	EF only available for use in agriculture ³⁾
	Pesticides	Agricultural application of pesticides containing impurities of HCB	EF only available for use in agriculture ³⁾
NFR 3D3 Other product use	Glass wool enduction ¹⁾		
	Mineral wool enduction ¹⁾		
	Fat, edible and not edible oil extraction		
	Application of glues and adhesives		
	Preservation of wood	PCP use	10-30 mg/inhabitant a (PCP use in wood protection) ⁴⁾
	Underseal treatment and conservation of vehicles		
	Vehicles dewaxing		
	Domestic use of pharmaceutical products		
	Other (preservation of seeds,...): use of pesticides in cultivations and in construction	Fire retardant in wood and plastics, softener in PVC, application of chlorine containing solvents where HCB can be present in trace amounts	No data (use of PVC) 2 mg/t (application of chlorine containing solvents where HCB can be present in trace amounts) ³⁾
	Tobacco smoking		
	Use of fireworks		

	Charcoal use for barbeques	HCB formation during the burning of charcoal	EF for residential wood combustion 95 µg/t
	Concrete, wall and floor coverings		
	Transformers and capacitors		
	Surface coating, sealants and adhesives		
	Enclosures and monitors		
	Construction and building products use		

¹⁾ Mainly industrial sources.

²⁾ Production process (EMEP/CORINAIR, 2006b).

³⁾ Open application (EMEP/CORINAIR, 2006b).

⁴⁾ Open application (Pacyna et al., 2003).

2.5.2 Emission results

In this report all possible sources to HCB emissions within this sector have been mapped and available emission factors are compiled. Omission of emission factors for certain possible sources does not mean they are insignificant. Experimental measurements and/or theoretical assessments are needed in order to get reliable estimates on missing emission factors for manufacture and use of the wide range of products. However, it is likely that the stated emission factors represent the most dominant sources. The scientific literature and other countries' emission inventories will continuously be checked for updated and new emission factors.

At present HCB emissions are only calculated for charcoal use while emissions are not estimated for any other product uses. In future reporting focus will be on investigating the availability of activity data (use) for the products and activities where emission factors are available.

The emission from charcoal use is shown in Table 16.

Table 16 Emission of HCB, 1990-2011, g.

	1990	1995	2000	2005	2008	2009	2010	2011
Charcoal use	0.68	0.75	1.27	1.42	0.99	1.11	0.74	0.64

2.6 Agriculture

Two sources for HCB emission from the agricultural sector have been identified; emissions from the use of pesticides and from field burning of agricultural waste.

2.6.1 Methodology

Pesticides

A range of pesticides are used in Denmark. Pure HCB used as pesticide is banded, but some of the pesticides used contain trace amounts of HCB.

In the period from 1990 to 2011 six types of pesticides containing HCB have been identified as used in Denmark. These are atrazine, chlorothalonil, clopyralid, lindane, pichloram and simazine. Data of amounts of active substances used in Denmark are collected from the Danish Environmental Protection Agency (DEPA, 2011a), see Table 17. The use of atrazine and lindane stopped in 1994 and the use of chlorothalonil and simazine stopped in 2000 and 2004, respectively.

Table 17 Amounts of active HCB containing substances used in Denmark, 1990-2011, kg.

	1990	1995	2000	2005	2008	2009	2010	2011
Atrazine	91 294	-	-	-	-	-	-	-
Chlorothalonil	10 512	10 980	7 340	-	-	-	-	-
Clopyralid	16 461	22 587	7 446	5 874	5 137	20 846	9 126	11 841
Lindane	8 356	-	-	-	-	-	-	-
Pichloram	-	-	-	-	-	-	723.6	1 350
Simazine	30 234	19 865	23 620	-	-	-	-	-

The emission is calculated using following equation:

$$E_{pes} = \sum a_i \cdot EF_i$$

Where: E_{pes} = emission of HCB from pesticides
 a_i = amount of active substance in the pesticide i
 EF_i = emission factor for the pesticide i

No default emission factors are given in the EMEP/EEA Guidebook (EEA, 2009). Emission factors given in Yang (2006) are used in the calculation of the emissions, see Table 18.

Table 18 Emission factors for HCB from pesticides, 1990-2011, g per tonne.

	1990	1995	2000	2005	2008	2009	2010	2011
Atrazine	100	1	1	1	1	1	1	1
Chlorothalonil	500	40	40	10	10	10	10	10
Clopyralid	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Lindane	100	50	50	1	1	1	1	1
Pichloram	100	50	50	8	8	8	8	8
Simazine	100	1	1	1	1	1	1	1

Field burning

Field burning of agricultural waste is also a source for HCB. The same activity data are used as for estimating the other emissions from field burning already included in the emission inventory. An emission factor is given by Hübner (2001) as 10 000 µg per ha. This factor has been converted to the unit g per tonne by following equation:

$$EF_{Used} = \frac{EF_{Hubner} \cdot Y}{1000000}$$

Where: EF_{Used} = emission factor in g per tonne
 EF_{Hubner} = emission factor given by Hübner (2001), 10 000 µg per ha
 Y = yield in tonne per ha

Table 19 Emission factor for HCB from field burning of agricultural waste.

	Yield, tonne per ha	EF, g per tonne
Straw from cereals	3.4	0.003
Straw from seed production	5	0.002

The emission is calculated by following equation:

$$E = \sum b_i \cdot C_f \cdot EF_i$$

Where: E = emission
 b_i = amount of burned agricultural waste, type i
 C_f = combustion factor 0.9 (EEA, 2009)
 EF_i = emission factor for waste type i

Field burning of agricultural waste are divided into two types, straw from seed production and burning of wet or broken bales of straw from cereals.

2.6.2 Emission results

Table 20 shows the emission of HCB from the agricultural sector for the years 1990-2011. The emission has decreased significantly from 1990 to 2011 due to decrease in use of pesticides containing HCB.

Table 20 Emission of HCB, 1990-2011, kg.

	1990	1995	2000	2005	2008	2009	2010	2011
Pesticides	18.28	0.50	0.33	0.01	0.01	0.04	0.02	0.03
Field burning of agricultural waste	0.08	0.10	0.12	0.13	0.11	0.12	0.09	0.09
Total	18.36	0.60	0.45	0.14	0.12	0.16	0.11	0.12

2.7 Waste

Emissions of HCB could occur from NFR waste categories 6C Waste Incineration and 6D Waste Other but not from 6A Solid Waste Disposal on Land or 6B Wastewater Handling.

In Denmark, waste incineration of municipal, industrial and hazardous waste is carried out with energy recovery. Therefore, it is included under the energy sector (stationary combustion) in the international reporting to the LRTAP convention under UNECE and the same categorisation is used in this report. For details on the emissions of HCB from waste incineration with energy recovery please refer to Chapter 2.1.

2.7.1 Methodology

In NFR waste category 6C Waste Incineration, the sources Human Cremation and Animal Cremation contribute to the emissions of HCB; however, literature on the subject is scarce. The estimated emissions are shown in Table 21.

In 6D Waste Other, the emission source Accidental fires are likely to contribute to the emissions of HCB but no data is available on this subject.

2.7.2 Emission results

Table 21 Emissions of HCB from the waste category.

	Unit	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Human cremation	g	6.2	6.2	6.3	6.5	6.5	6.6	6.6	6.5	6.3	6.4
Animal cremation	g	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.5	0.5	0.9
<i>Continued</i>		2000	2001	2002	2003	2004	2005	2006	2007	2008	2009
Human cremation	g	6.3	6.3	6.4	6.4	6.3	6.2	6.2	6.3	6.3	6.4
Animal cremation	g	1.0	1.1	1.1	1.1	1.3	1.8	2.6	3.0	3.1	3.1
<i>Continued</i>		2010	2011								
Human cremation	g	6.4	6.3								
Animal cremation	g	3.4	2.8								

3 Emission inventory methodology for polychlorinated biphenyls

Polychlorinated biphenyls (PCBs) cover a group of 209 different PCB congeners which can be divided into two groups according to their toxicological properties. One group, consisting of 12 congeners, shows toxicological properties similar to dioxins and is therefore termed 'dioxin-like PCBs' (dl-PCB) or 'co-planar PCB', with reference to their conformation. The other PCBs, referred to as 'non dioxin-like PCBs' do not primarily interact by means of a similar mode of action as the dioxins.

Data on the occurrence of PCBs in the environment have been reported in many different ways: e.g. as PCB congener 153 only; as the sum of three PCB congeners (PCB 138, 153 and 180); as the sum of six PCB congeners (PCB 28, 52, 101, 138, 153, 180), often referred to as indicator PCB; or as the sum of seven (sum of six indicator PCB plus PCB congener 118). This lack of consistency hampers a comparison of occurrence data. Other references report data for the 12 dioxin-like PCBs (PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189), either as a sum or as toxic equivalent (teq) values.

Thus the congeners included in indicator PCBs differ from the congeners included in dioxin-like PCBs. Also, there is different toxic equivalent factors being available (WHO 1998 and WHO 2005) and it is not always clear from published data, which set of toxic equivalent factors has been used.

For the sake of simplicity, in the present investigation the term PCBs has been used for all possible combinations. This of course introduces a certain amount of uncertainty to the analysis, but this uncertainty is regarded as considerably smaller than a number of other uncertainties present in this compilation.

3.1 Stationary combustion

The previous Danish emission inventories did not include PCB emissions.

PCBs can be emitted in any chemical process involving chloride and organic carbon or emitted due to incomplete combustion of PCBs in fuel (waste incineration). Waste with high levels of PCBs is only incinerated in plants with permission to incinerate this waste fraction as it requires a high combustion temperature.

3.1.1 Methodology

Emission data for stationary combustion have been collected in a literature review. Table 22 shows references for emission factors for PCBs.

Emission of PCBs is strongly related to the chlorine content of the fuel (Syc et al., 2011) and to the emission level for PCDD/F (Hedman et al., 2006; Syc et al., 2011; Pandelova et al., 2009).

The chlorine content of straw, bark and manure is higher than for wood (Villeneuve et al., 2012). Villeneuve et al. (2012) states the chlorine contents 50-60 mg per kg wood, 100-370 mg per kg bark, 1000-7000 mg per kg straw.

Different references for PCB emissions are not directly comparable because some PCB emission data are reported for individual PCB congeners, some as a sum of a specified list of PCB congeners and some PCB emission data are reported as toxic equivalence (teq) based on toxicity equivalence factors (TEF) for 12 dioxin-like PCB congeners. The emission measurements reported by Thistlethwaite (2001a and 2001b) show that the emission of non-dioxin-like PCBs is high compared to the emission of dioxin-like PCBs.

Furthermore, teq values based on TEF are reported as WHO₂₀₀₅-teq or WHO₁₉₉₈-teq. This difference is however typically less than 50 %².

² Data have been compared for a few datasets in which each dioxin-like PCB congener was specified.

Table 22 PCB emission factors for stationary combustion.

Reference	Fuel	Emission factor, ng/GJ	Technology and comment
EEA (2013)	Coal	3.3 (1.1-9.9) ng WHO ₁₉₉₈ - teq/GJ	Public electricity and heat production. Refers to Grochowalski & Konieczynski (2008)
Grochowalski & Konieczynski (2008)	Coal	3.85 ng WHO ₁₉₉₈ - teq/GJ or 904 ng \sum dl-PCB ¹ /GJ	Power plants, 4 fluid bed boilers. Including combustion of coal silt and waste coal for two plants.
Grochowalski & Konieczynski (2008)	Coal	3.16 ng WHO ₁₉₉₈ - teq/GJ or 839 ng \sum dl-PCB ¹ /GJ	Power plants, 2 fluid bed boilers.
Thistlethwaite 2001a	Coal	53 ng WHO ₁₉₉₈ - teq/GJ Or 50 800 ng \sum PCB ⁹ /GJ Or 5700 ng \sum dl-PCB/GJ	Coal fired boiler, 500 kW.
Fontelle et al. (2014)	Coal	55 000 ng/GJ	Refers to AEAT (2001) (Thistlethwaite, 2001)
EEA (2013)	Coal	170 000 ng/GJ	Non-industrial combustion. Refers to Kakareka et al. (2004)
Syc et al. (2011)	Coal	7403 (2120-16 100) ng \sum PCB ³ /GJ or 66 (4-184) ng teq/GJ	3 residential boilers, bituminous coal. TEQ according to EN 1948.
Lee et al. (2005)	Coal	264 000 (66 000-462 000) ng \sum PCB ⁶ /GJ or 6 (1-10) ng WHO ₁₉₉₈ teq/GJ	Residential plant. TEQ according to WHO 1998.
Thanner & Moche (2002)	Coal	510 ng WHO ₁₉₉₈ teq/GJ	Residential plant.
Andrijewski (2004)	Coal	13 000 ng/GJ	Energy industries and industrial plants. Hard coal. Recalculation based on 24.38 GJ/Mg.
Andrijewski (2004)	Coal	17 000 ng/GJ	Commercial/institutional plants and plants in agriculture/forestry. Recalculation based on 24.38 GJ/Mg.
Andrijewski (2004)	Coal	1 296 000 ng/GJ	Residential plants. Recalculation based on 24.38 GJ/Mg.
Kakareka & Kukharchyk (2005b)	Coal	147 000 ng \sum PCB/GJ	Refers to TNO report from 1995. Recalculated based on 24.5 MJ/kg.
Kakareka & Kukharchyk (2005b)	Coal	359 000 ng \sum PCB/GJ	Refers to Lee et al. (2005)
Kakareka & Kukharchyk (2005b)	Coal	24 000-51 000 ng \sum PCB/GJ	Residential/domestic combustion. Refers to Kakareka et al. (2004)
Andrijewski (2004)	Brown coal	98 000 ng/GJ	Non-residential plants. Brown coal. Recalculation based on 18.3 GJ/Mg.
Andrijewski (2004)	Brown coal	10 011 000 ng/GJ	Residential plants. Brown coal. Recalculation based on 18.3 GJ/Mg.
Andrijewski (2004)	Coke	123,000 ng/GJ	Non-residential plants. Recalculation based on 29.3 MJ/kg.
Dyke et al. (2003)	Recycled fuel oil	0-3.2 ng WHO ₁₉₉₈ - teq/GJ	Emission measurement from coal fired power station during start-up using recycled fuel oil. All congeners below detection limit.
EEA (2013)	Gas oil	0.13 ng WHO ₂₀₀₅ - teq/GJ	Engines. Refers to Nielsen et al. (2010)
Andrijewski (2004)	Residual oil	15,000 ng/GJ	Fuel oil, non-residential plants.
Nielsen et al. (2010)	Gas oil	<0.13 ng WHO ₂₀₀₅ - teq/GJ or < 0.11 ng WHO ₁₉₉₈ - teq/GJ or <93 ng \sum dl-PCB/GJ	Emission measurements from one engine. The emission was below the detection limit for all congeners.

Continued

Nielsen et al. (2010) ⁸⁾	Waste	<0.33 ng WHO ₂₀₀₅ - teq/GJ or < 0.28 ng WHO ₁₉₉₈ - teq/GJ or < 109 ng Σ dl-PCB/GJ	CHP plant, waste incineration, emission measurements from three plants. Some PCB congeners above detection limit.
Dyke et al. (2003)	Waste	0 - 8 ng WHO ₁₉₉₈ - teq/GJ	Emission measurement from two MSW incineration plants with injection of activated coal. Many congeners below the detection limits.
Dyke et al. (2003)	Waste – sewage sludge	0 - 6 ng WHO ₁₉₉₈ - teq/GJ	Emission measurement from one sludge incineration plant with injection of activated lignite. Many congeners below the detection limits.
Dyke et al. (2003)	Waste – sewage sludge	0 - 12 ng WHO ₁₉₉₈ - teq/GJ	Emission measurement from one medical waste incineration plants with injection of activated coal. Many congeners below the detection limits.
Dyke et al. (2003)	Waste	0.027-25 600 ng WHO ₁₉₉₈ -teq/GJ	Literature study.
Kakareka & Kukharchyk (2005b)	Municipal waste	78 000-1 760 000 ng Σ PCB/GJ	Several references included.
Andrijewski (2004)	Waste	19 000 ng/GJ	Municipal waste. Emission factors for industrial waste and hospital waste much higher unless the plants comply with the EU directive (recent years). Recalculation based on 10.5 MJ/kg.
EEA (2013)	Biomass	3500 (350-35 000)	Public electricity and heat production. Refers to US EPA (2003), chapter 1.6.
Thistlethwaite 2001a	Wood	21 ng WHO ₁₉₉₈ - teq/GJ Or 32 000 ng Σ PCB ⁹⁾ /GJ Or 2800 ng Σ dl-PCB/GJ	Wood –fired air-heater at a furniture factory.
Andrijewski (2004)	Wood	50 000 ng/GJ	Non-residential plants. Recalculation based on 18 MJ/kg.
EEA (2013)	Biomass/Wood	60 ng/GJ	Non-industrial combustion, refers to Hedman et al. (2006)
EEA (2013)	Wood	30 ng/GJ	Energy efficient stoves. Refers to Hedman et al. (2006)
EEA (2013)	Wood	7 ng/GJ	Advanced / ecolabelled stoves and boilers and automatic boilers. Refers to Hedman et al. (2006)
Syc et al. (2011)	Wood	2008 (433-4350) ng Σ dl-PCB ³⁾ /GJ or 0.96 ng WHO-teq/GJ	Five residential boilers and one stove. TEQ according to EN 1948.
Gullet et al. (2003)	Wood	441 000 ng Σ PCB ⁴⁾ /GJ 0.074 ng teq/GJ	Oak wood, stove. EPA certified. Lined steel stove. Recalculation based on 19 MJ/kg.
Hedman et al. (2006)	Wood pellets	13 (5-21) ng WHO- teq/GJ	Residential wood pellet boiler. Recalculation based on 19 MJ/kg.
Hedman et al. (2006)	Wood	53 (37-68) ng WHO- teq/GJ	Old boilers. Recalculation based on 19 MJ/kg. Data for tests including paper and plastic not included.
Hedman et al. (2006)	Wood	7 (5-11) ng WHO- teq/GJ	Residential modern wood boiler. Recalculation based on 19 MJ/kg.
Hedman et al. (2006)	Wood	26 (21-32) ng WHO- teq/GJ	Residential stove. Recalculation based on 19 MJ/kg.
Karareka & Kukharchyk (2005)	Wood	17 000-33 000 ng/GJ	Residential/domestic combustion. Recalculation based on 18 MJ/kg. Refers to Karareka et al. (2004)

<i>Continued</i>			
Lee et al. (2005)	Wood	34 000 (10 000-58 000) ng Σ PCB ⁷⁾ / GJ or 1.1 (0.3-1.8) ng WHO teq ⁵⁾ /GJ	Hardwood, residential. TEQ according to WHO 1998.
Pandelova et al. (2009)	Wood	362 ng WHO teq/GJ.	Pilot scale unit. Stove. 100 % wood.
Andrijewski (2004)	Wood	500 000 ng/GJ	Residential plants. Recalculation based on 18 MJ/kg.
Hedman et al. (2006)	Straw pellets	21 ng WHO-teq/GJ	Residential pellet boiler, straw pellets. Recalculation based on 19 MJ/kg.
Syc et al. (2011)	Mais straw pellets	3110 ng Σ PCB ³⁾ /GJ or 31.2 ng teq./GJ	Residential boiler. TEQ according to EN 1948.
Nielsen et al. (2010) ⁸⁾	Biogas (landfill gas)	<0.19 ng WHO ₂₀₀₅ -teq/GJ or < 0.13 ng WHO ₁₉₉₈ -teq/GJ or < 90 ng Σ dl-PCB/GJ	Emission measurements from 1 landfill gas fuelled engine. The emission was below the detection limit for all congeners.
Nielsen et al. (2010) ⁸⁾	Biomass producer gas	<0.24 ng WHO ₂₀₀₅ -teq/GJ or < 0.17 ng WHO ₁₉₉₈ -teq/GJ or < 144 ng Σ dl-PCB/GJ	Emission measurements from 1 engine. The emission was below the detection limit for all congeners.
CEMENT			
EEA (2013)	(Cement)	103 µg/ te clinker	
Andrijewski (2004)	(Cement)	0.007 g/Gg clinker	

1. Σ of 12 dioxin-like PCBs (77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189).
2. WHO 1997.
3. Σ of 12 dl-PCBs (77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189), PCB 170 and PCB 180.
4. 209 congeners included.
5. WHO teq. 1998.
6. 48 different congeners included. If only the 12 dioxin-like PCBs are included, the emission is 190 ng/GJ.
7. 48 different congeners included. If only the 12 dioxin-like PCBs are included, the emission is 20 ng/GJ.
8. And additional data aggregation based on the original data.
9. A total 23 congeners have been measured. All 12 dioxin-like congeners included.

Table 23 shows the emission factors that have been selected for the Danish PCB emission inventory and reference for each emission factor. All emission factors are dioxin-like PCBs (but not teq values). PCB emission factors have been added for all fuels except LPG, refinery gas and natural gas. The emission from these three fuels is considered negligible.

Recent field emission measurements from up to date technologies have been prioritised in the selection of the applied reference. In addition, the emission factor level should preferably be confirmed by other references.

Data with specification of which PCB congeners were included and with both a sum of all dioxin-like congeners and a WHO-teq value have been prioritised.

The emission factor for waste incineration refers to recent Danish field measurements. Historical data are not available, but a time series have been estimated based on the assumption that the dl-PCB emission factor follows the PCDD/-F emission factor. The estimated emission factor for 1990 is 45671 ng per GJ or 117 ng WHO-teq per GJ. This emission level is confirmed by other references (Kakareka & Kukharchyk, 2005b; Andrijewski, 2004). The emission factor time series is shown in Table 24.

For residential wood combustion, technology specific emission factors in toxicological equivalence are available from Hedman et al. (2006), see Table 25. However, sums of dioxin-like PCBs are not included in the reference. The emission factors for dioxin-like PCBs have been estimated based on the data for toxicological equivalence and the sum of dioxin-like PCBs in Thistlethwaite (2001a). Thus, the teq factors referring to Hedman (2006) have been multiplied by 2800/21. This assumption is highly uncertain, but the resulting emission factors seem to be in agreement with other references for residential wood combustion. A technology distribution time series for residential wood combustion in Denmark is available and have been applied for estimating the time series for the aggregated emission factor shown in Table 24.

Table 23 Emission factors for Σ dl-PCB, stationary combustion, 2011.

Fuel	NFR (SNAP)	Emission factor, Σ dl-PCB, ng/GJ	Emission factor, PCB, ng WHO ₁₉₉₈ - teq/GJ	Reference
Coal	1A1	839	3.16	Grochowalski & Koniecznyński (2008)
Coal	1A2	5700	53	Thistlethwaite (2001a)
Coal	1A4	7403	66	Syc et al. (2011)
Other solid fuels	1A1	839	3.16	Assumed equal to coal.
Other solid fuels	1A2	5700	53	Assumed equal to coal.
Other solid fuels	1A4	7403	66	Assumed equal to coal.
Residual oil	1A1, 1A2, 1A4	839	3.2	The teq value refers to Dyke et al. (2003).
The TEQ value is equal to the emission factor for coal combustion in power plants and the sum of dioxin-like PCB congeners has been assumed equal to the corresponding factor for coal.				
Gas oil	1A1, 1A2, 1A4	93	0.11	Nielsen et al. (2010)
Other liquid fuels ¹⁾	1A1, 1A2, 1A4	93	0.11	Assumed equal to gas oil.
Gaseous fuels	1A1, 1A2, 1A4	-	-	Negligible
Waste	1A1, 1A2, 1A4	109 (time series)	0.28 (time series)	Nielsen et al. (2010). A time series have been estimated. The emission factor for 1990 (46,000 ng/GJ / 117 ng WHO ₁₉₉₈ teq/GJ) have been estimated based on the assumption that the PCB emission factor time series follow the PCDD/F time series.
Wood	1A1, 1A2, 1A4a/c	2800	21	Thistlethwaite (2001a)
Wood	1A4b	3179 (time series)	23.9 (time series)	Hedman et al. (2006). A time series have been estimated based on time series for technologies applied in Denmark.
Straw	1A1, 1A2	3110	31.2	Assumed equal to residential plants.
Straw	1A4	3110	31.2	Syc et al. (2011)
Biogas	1A1, 1A2, 1A4	90	0.13	Nielsen et al. (2010)
Producer gas	1A1, 1A2, 1A4	144	0.17	Nielsen et al. (2010)

1. Except LPG and refinery gas.

Emission factor time series for waste incineration and for residential wood combustion are shown in Table 24.

Table 24 Emission factor time series for waste incineration and for residential wood combustion.

Year	Waste incineration		Residential wood combustion	
	Σ dl-PCB,	dl-PCB,	Σ dl-PCB,	dl-PCB,
	ng/GJ	ng WHO-teq/ GJ	ng/GJ	ng WHO-teq/ GJ
1990	45 671	117	5468	41.1
1991	38 063	98	5468	41.1
1992	30 433	78	5468	41.1
1993	22 825	59	5468	41.1
1994	19 773	51	5468	41.1
1995	16 721	43	5468	41.1
1996	13 690	35	5468	41.1
1997	10 638	27	5468	41.1
1998	7586	19	5468	41.1
1999	5515	14	5468	41.1
2000	3423	9	5468	41.1
2001	3423	9	4894	36.8
2002	3423	9	4657	35.0
2003	3423	9	4589	34.5
2004	1766	4.5	4513	33.9
2005	109	0.28	4298	32.3
2006	109	0.28	4055	30.5
2007	109	0.28	4082	30.7
2008	109	0.28	3836	28.8
2009	109	0.28	3532	26.6
2010	109	0.28	3348	25.2
2011	109	0.28	3179	23.9
2012	109	0.28	3025	22.7

Table 25 Technology specific emission factors for residential wood combustion.

Technology	dl-PCB emission factor, ng WHO- teq/GJ	Σ dl-PCB emission factor, ng/GJ	Reference and assumptions
Old stove	53	7049	Hedman (2006), old boiler
New stove	53	7049	Hedman (2006), old boiler
Modern stove	7	931	Hedman (2006), modern boiler
Eco labelled stove	3.5	466	Hedman (2006), assumed ½ modern boiler
Other stove	53	7049	Hedman (2006), old boiler
Old boiler with acc. tank	53	7049	Hedman (2006), old boiler
Old boiler without acc. tank	53	7049	Hedman (2006), old boiler
New boiler with acc. tank	7	931	Hedman (2006), modern boiler
New boiler without acc. tank	7	931	Hedman (2006), modern boiler
Pellet boilers/stoves	3.5	466	Hedman (2006), assumed ½ modern boiler

3.1.2 Emission results

Emissions of dioxin-like PCBs in 2011 are shown in Table 26. The largest emission sources are public electricity and heat production and residential plants.

Table 26 dl-PCB emission from stationary combustion plants, 2011.

Sector	dl-PCB, kg
1A1a Public electricity and heat production	0.264
1A1b Petroleum refining	0.000
1A1c Other energy industries	0.000
1A2 Industry	0.058
1A4a Commercial / Institutional	0.003
1A4b Residential	0.115
1A4c Agriculture / Forestry / Fisheries	0.015
Total	0.456

The PCBs emission time series is shown in Figure 2. The emission has decreased 62 % since 1990. The decrease is mainly a result of the flue gas cleaning devices that have been installed in waste incineration plants for dioxin reduction.

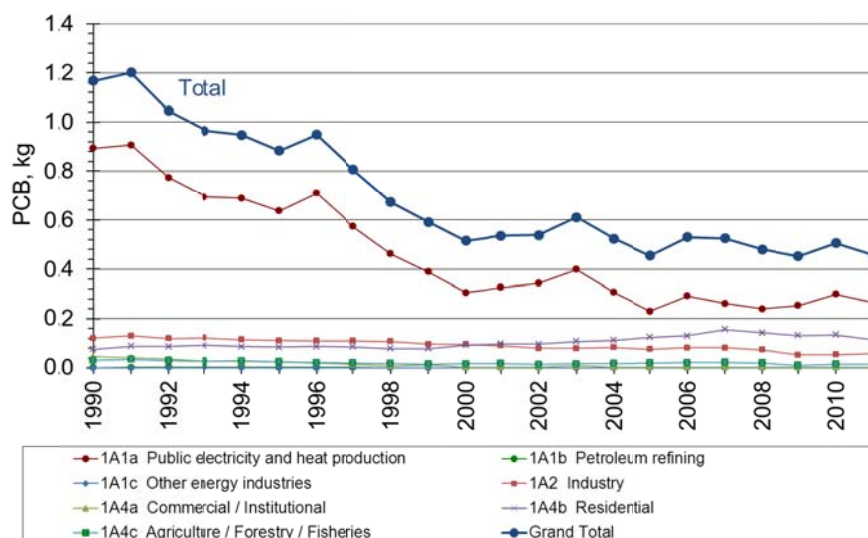


Figure 2 dl-PCB emission time series for stationary combustion.

3.2 Mobile combustion

3.2.1 Methodology

For road transport, PCB emission factors are taken from Andrijewski (2004). Based on average fuel consumption factors derived from the road transport part of the Danish emission inventories (Nielsen et al., 2013) and lower heating values for diesel and gasoline, the emission factors are recalculated from g per km to g per TJ³.

The gasoline fuel related emission factors for road transport are used for other gasoline fuelled mobile sources as well. The fuel related emission factors derived for heavy duty vehicles are used for railways and diesel machinery in agriculture, forestry, industry and military.

For navigation and fisheries emission factors measured by Cooper (2005) are used for marine diesel oil (MDO) and residual oil (heavy fuel oil: HFO). For aviation no emission factor could be found in the available literature.

The PCBs emission factors used are shown in Table 27.

³ Average fuel consumption factors (g/km): 240 (heavy duty vehicles); 55 (gasoline cars/vans); 55 (diesel cars/vans). Lower heating values (MJ/kg): 42.7 (diesel); 43.8 (gasoline).

Table 27 PCBs emission factors for mobile sources. Reference values and fuel related factors (g per TJ).

Pollutant	Category	Reference	Fuel type	Unit	Value	g/TJ
PCBs	Road/Other land based	Andrijewski (2004)	Motor gasoline	g/Gg	106	2.42E+00
PCBs	Road/Other land based	Andrijewski (2004)	Unleaded motor gasoline	g/Gg	0.02	4.57E-04
PCBs	Road	Andrijewski (2004)	Diesel PC and LDV	g/km	5.00E-08	2.13E-02
PCBs	Road/Other land based	Andrijewski (2004)	Diesel HDV	g/km	5.39E-06	5.26E-01
PCBs	Navigation	Cooper (2005)	MDO	g/TJ		8.76E-03
PCBs	Navigation	Cooper (2005)	HFO	g/TJ		1.40E-02

The fuel activity data taken from the latest published Danish emission inventories (Nielsen et al., 2013) are listed in Table 28.

Table 28 Fuel activity data (TJ) for mobile sources in Denmark.

NFR Category	Fuel type	1990	1995	2000	2005	2008	2009	2010	2011
Industry - Other (1A2f)	Diesel	10 158	10 324	10 773	11 753	13 970	10 284	13 015	12 678
Industry - Other (1A2f)	Gasoline	175	169	167	165	159	118	157	156
Road (1A3b) - exhaust	Diesel	59 947	64 013	69 196	88 264	103 491	97 560	101 342	105 150
Road (1A3b) - exhaust	Gasoline	66 279	80 101	83 312	77 835	72 533	67 940	63 899	60 084
Railways (1A3c)	Diesel	4010	4093	3079	3137	3199	3111	3273	3370
Navigation (1A3d)	Diesel	5628	7565	6035	5561	5395	5319	5071	4757
Navigation (1A3d)	Gasoline	309	358	396	393	361	353	346	340
Navigation (1A3d)	Residual oil	4571	3382	1444	1859	2148	2287	2456	2375
Comm./Inst. (1A4a)	Gasoline	1010	1068	1186	2214	2409	2389	2368	2348
Residential (1A4b)	Gasoline	535	544	583	806	865	862	860	858
Agriculture/forestry (1A4c)	Diesel	16 642	15 474	13 856	14 608	16 598	16 768	16 982	17 430
Agriculture/forestry (1A4c)	Gasoline	1050	740	521	419	542	574	572	571
Fisheries (1A4c)	Diesel	7920	7134	7422	8725	8106	7514	7770	7788
Military (1A5)	Diesel	146	1794	369	1271	630	1099	596	1045
Military (1A5)	Gasoline	1	0	1	7	12	9	6	0
Navigation int. (1A3d)	Diesel	11 289	26 370	22 129	11 330	10 928	10 164	11 356	10 282
Navigation int. (1A3d)	Residual oil	27 815	38 780	32 437	19 411	25 650	9416	15 682	17 120

Due to the split in emission factors, additional information regarding the share of leaded and unleaded gasoline sold in Denmark and the share of diesel fuel used by cars/vans and heavy duty vehicles is needed to carry out the subsequent emission calculations (Table 29). The fuel data are derived from the Danish inventories (Nielsen et al., 2013).

Table 29 The percentage share of leaded and unleaded gasoline sold in Denmark and the share of diesel fuel used by cars/vans and heavy duty vehicles used in Danish road transport.

	Unit	1990	1995	2000	2005	2008	2009	2010	2011
Leaded gasoline	%	42.5	0	0	0	0	0	0	0
Unleaded gasoline	%	57.5	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Diesel cars/vans	%	39	41	43	50	57	60	60	62
Diesel heavy duty vehicles	%	61	59	57	50	43	40	40	38

3.2.2 Emission results

The emissions are calculated as the product of the emission factor and the fuel consumption for each NFR category/fuel type layer using the following equation:

$$E = \sum_{C,f} FC_{C,f} \cdot EF_{C,f}$$

Where: E = Emission of PCB
FC = Fuel consumption
EF = HCB Emission factor
C = NFR category
f = fuel type

The PCB emissions from mobile sources are shown in Table 30.

Table 30 PCB emissions (g) for mobile sources in Denmark.

NFR2 category	Fuel type	1990	1995	2000	2005	2008	2009	2010	2011
Industry - Other (1A2f)	Diesel	5343	5430	5666	6182	7348	5409	6846	6668
Industry - Other (1A2f)	Gasoline	180	0	0	0	0	0	0	0
Road (1A3b) - exhaust	Diesel	19 825	20 564	21 501	24 080	24 636	21 886	22 588	22 518
Road (1A3b) - exhaust	Gasoline	68 252	37	38	36	33	31	29	27
Railways (1A3c)	Diesel	2109	2153	1619	1650	1682	1636	1721	1772
Navigation (1A3d)	Diesel	49	66	53	49	47	47	44	42
Navigation (1A3d)	Gasoline	318	0	0	0	0	0	0	0
Navigation (1A3d)	Residual oil	64	47	20	26	30	32	34	33
Comm./Inst. (1A4a)	Gasoline	1040	0	1	1	1	1	1	1
Residential (1A4b)	Gasoline	551	0	0	0	0	0	0	0
Agriculture/forestry (1A4c)	Diesel	8753	8139	7288	7683	8730	8819	8932	9167
Agriculture/forestry (1A4c)	Gasoline	1082	0	0	0	0	0	0	0
Fisheries (1A4c)	Diesel	69	62	65	76	71	66	68	68
Military (1A5)	Diesel	77	944	194	668	332	578	314	550
Military (1A5)	Gasoline	1	0	0	0	0	0	0	0
Total national		107 714	37 443	36 446	40 452	42 910	38 506	40 578	40 848
Navigation int. (1A3d)	Diesel	99	231	194	99	96	89	99	90
Navigation int. (1A3d)	Residual oil	389	543	454	272	359	132	220	240

3.3 Fugitive emissions from fuels

No sources of emissions of PCBs have been identified for the sector “Fugitive emissions from fuels”. This finding is in correspondence with the EMEP/EEA guidebook (EEA, 2009), that does not include methodology or emission factors for PCBs for any sources in the fugitive sector.

Fugitive formation of PCBs could occur either due to evaporation of fuels containing HCB/PCBs or due to combustion processes, e.g. flaring. Neither raw oil, natural gas nor gasoline are assumed to contain HCB or PCBs. Flaring of oil and gas are the only combustion processes in the fugitive emission sector. According to European Commission (2013) it is highly unlikely that the formation conditions for PCBs occur for flaring in refineries and this is assumed to be the case for offshore flaring as well. Formation of both HCB and PCBs require presence of chlorine, and the chlorine content is assumed to be very limited in oil and natural gas being flared offshore and in refineries. Therefore emissions of both HCB and PCBs are considered not occurring or negligible.

3.4 Industrial processes

3.4.1 Methodology

The emissions of PCB are calculated as the product of the emission factor and the activity for each NFR category using the following equation:

$$E = \sum_{C,f} Act_C \cdot EF_C$$

Where: E = Emission of PCB
 Act_C = Activity
 EF_C = PCB Emission factor
 C = NFR category

Table 31 on next page presents emission factors for PCB from industrial processes. The emission factors may be given as \sum PCB or as I-TEQ (International toxicity equivalents).

Table 31 Emission factors for PCB from industrial processes.

	Air pollution control	Country	Emission factor ng I-TEQ per tonne	Reference	Comment
Cement production			2 000 000 ¹	Bipro, 2006	
Cement production			7 000 ¹	Andrijewski, 2004	
Lime production			150 000 ¹	Bipro, 2006	
Steelwork, Electric Arc Furnace (EAF), secondary raw materials	Sedimentation/post-combustion chamber, cyclone and bag filter	Portugal	659	Antunes et al., 2012	Measured
Steelwork, Electric Arc Furnace (EAF), secondary raw materials	Sedimentation/post-combustion chamber, cyclone and bag filter	Portugal	597	Antunes et al., 2012	Measured
Steelwork, EAF			3 600 000 ¹	EEA, 2006	
Steelwork, EAF			2 500 ¹	EEA, 2013	
Steelwork, EAF			1 500 000- 4 500 000 ¹	IPPC, BREF	
Steelwork, EAF			5 000 000 ¹	Bipro, 2006	
Cast iron on cubilot furnace, secondary raw materials	Sedimentation/post-combustion chamber, cyclone and bag filter	Portugal	24	Antunes et al., 2012	Measured, raw material cleaner than raw material used in EAF
Cast iron on cubilot furnace/induction furnace, secondary raw materials	Cyclone, electrostatic air cleaner and bag filter	Portugal	121	Antunes et al., 2012	Measured, raw material cleaner than raw material used in EAF
Foundry, pig iron	Bag filter, cyclone, inertial force dust collector	S. Korea	33.4	Yu et al., 2006	Measured
Foundry, steel	Bag filter, cyclone, inertial force dust collector	S. Korea	10.9	Yu et al., 2006	Measured
Foundry, iron			500 000 ¹	Bipro, 2006	
Aluminium smelting on reverberatory furnace/ladle furnace, secondary	None	Portugal	113	Antunes et al., 2012	Measured
Aluminium smelting on induction furnace, secondary	None	Portugal	87	Antunes et al., 2012	Measured
Aluminium smelting on reverberatory furnace, secondary	None	Portugal	259	Antunes et al., 2012	Measured
Aluminium production	Bag filter, cyclone, wet scrubber, absorption tower	S. Korea	292	Yu et al., 2006	Measured
Aluminium, secondary			3 400 000 ¹	Bipro, 2006	
Lead smelting on crucible, secondary	None	Portugal	2.7	Antunes et al., 2012	Measured
Lead production	Bag filter, wet scrubber	S. Korea	310	Yu et al., 2006	Measured
Lead production			7 250 000 ¹	Bipro, 2006	
Lead/zinc/tin smelting on induction furnace/crucible, secondary	Cyclone and bag filter	Portugal	1.0	Antunes et al., 2012	Measured
Copper, primary	Cyclone, wet scrubber, electrostatic precipitator	S. Korea	11.9	Yu et al., 2006	Measured
Copper, secondary	Cyclone, wet scrubber, electrostatic precipitator	S. Korea	9 770	Yu et al., 2006	Measured
Copper, primary			100 000 ¹	Bipro, 2006	
Copper, secondary – smelting and refining			2 600 000 ¹	Bipro, 2006	
Copper/zinc/lead smelting on induction furnace, secondary	Cyclone and bag filter	Portugal	0.49	Antunes et al., 2012	Measured
Zinc production	Bag filter, cyclone, wet scrubber, inertial force dust collector	S. Korea	24.8	Yu et al., 2006	Measured
Zinc, secondary			1 000 000 ¹	Bipro, 2006	
Pulp and paper			700 000 ¹	Bipro, 2006	
Consumption of POPs and heavy metals (e.g. electrical and scientific equipment)			0.1 g Σ PCB per capita	EEA, 2009	

¹ Unit: ng PCB per tonne (Σ PCB).

Table 32 presents the applied EF for PCB.

Table 32 Applied EFs for PCBs.

NFR Code	NFR Category	Applied EF mg PCBs/tonne
2A1	Cement production	Included in 1A2
2A2	Lime production	0.15
2A7b	Construction and demolition	- ¹
2C1	Steelwork, EAF	2.5
2C1	Iron foundry	0.5
2C3	Secondary aluminium production	3.4
2C5b	Secondary lead production	7.25
2E	Production of POPs	- ²
2F	Consumption of POPs and heavy metals (e.g. electrical and scientific equipment)	- ³

¹ Demolition of buildings may result in emission of PCBs, however, PCB containing building materials are treated as hazardous waste according to special legislation. A more general investigation of emission to the environment may be necessary as focus has been on indoor environment.

² There has not been production of PCBs in Denmark.

³ The proposed default emission factor by EEA (2009) seems very high as the EF will result in a total PCB emission in Denmark at 558 kg PCB per year – more than 10 times the emission reported from other sources. Therefore, this EF has not been used in the inventory. The EF is probably based on the use of PCBs in capacitors in electrical equipment, however, this use of PCBs have not occurred in Denmark through the whole time-series.

3.4.2 Emission results

Based on the applied emission factors the emissions of PCBs from industrial processes have been estimated as shown in Table 33.

Table 33 PCBs emissions from industrial processes in Denmark, kg.

	NFR category	1990	1995	2000	2005	2010	2011
2A2	Lime production	0.023	0.017	0.015	0.013	0.0092	0.0074
2C1	Steelwork, EAF	1.54	1.79	1.58	0.63	0	0
2C1	Iron foundry	0.052	0.045	0.051	0.050	0.036	0.042
2C3	Secondary aluminium production	0	0	0.11	0.094	0	0
2C5b	Secondary lead production	0.036	0.036	0.036	0.036	0.036	0.036
	Total	1.65	1.89	1.79	0.82	0.082	0.085

No emissions of PCBs from demolition have been included in the inventory as no EF exists for emission of PCBs from building materials to the environment. PCB containing building materials are treated as hazardous waste according to special legislation due to the health effects of PCB.

No emissions of PCBs from electrical equipment have been included in the inventory as the default EF proposed by EEA (2009) seems very high, and as the EF will result in a total PCBs emission in Denmark of 558 kg PCBs per year – more than 10 times the emission reported from other sources.

A more general investigation of emission of PCBs from building materials and electrical equipment to the environment may be necessary in order to clarify the magnitude of the problem.

3.5 Solvent and other product use

3.5.1 Methodology

The following list of known and suspected sources of PCB sources is based on PCB Fact Sheet (2003), ATSDR (2006), EIP Associates (1997), UNEP (1999), EPA (2011), EEA (2007), de Voogt & Brinkman (1989).

Primary Applications:

- Dielectric fluids and transformers.
- Capacitors.
- Fluorescent light ballasts.
- Electromagnets.
- Miscellaneous electrical equipment.
- Heat transfer systems.
- Hydraulic fluids.
- Plasticizers.
- Lubricants.
-

Other applications of PCBs:

- Dust control (dedusting agents).
- Pesticides.
- Fire retardants.
- Paints, coatings.
- Carbonless copy paper.
- Printing inks.
- Investment casting waxes.
- Wood treatment.
- Laminating and impregnating agents.
- Adhesives.
- Waxes.
- Additives to cement and plaster.
- Casting agents.
- Sealing liquids.

Due to the long service life of many PCB-containing items and the use of PCBs in some durable, relatively inert products, PCB-containing materials will continue to be disposed of and processed in waste and recycling operations. Waste products and recycling operations that may process significant quantities of PCB-containing materials are:

PCBs sources in Waste Materials and Recycling Operations:

- Material or Operation.
- Scrap metal recycling.
- Auto salvage yards, auto crushing.
- Repair activities.
- Used oil.
- Recycled paper.
- Effluent.
- Asphalt roofing materials, tar paper, and roofing felt.
- Building demolition.
- Dredge spoils.
- Landfills.
- Wastewater treatment plant sludge.

Care must be taken when a product containing the pollutant is disposed of and recycled. If an emission considers the total amount of the pollutant containing product, then the recycled amount must not be added again to avoid double counting.

Breivik et al. (2002) distinguishes between use of PCB-containing products in open systems, nominally closed systems, small capacitors and closed systems. Open systems are used in applications where the PCBs are unrecoverable or consumed during use; i.e. plasticizers, carbonless copy paper, lubricating oils, inks, paints, adhesives and more. Nominally closed systems are neither open nor closed and comprise hydraulic and heat transfer fluids. Small capacitors are used in a wide range of products, such as cars and electrical household appliances, and may contain only a few grams of PCBs per unit. Closed systems have been the dominant PCB use category since the mid-1970s and most importantly comprise transformers and large capacitors, containing on average between 0.9 and 1.3 t PCBs per unit and up to approximately 10 kg of PCBs per unit, respectively.

For countries where no information on distribution of use amounts of PCBs into use categories are available Breivik et al. (2002) assumed a historical global usage pattern of; open systems (26 %), nominally closed systems (13 %), small capacitors (20 %) and closed systems (41 %). Furthermore it was assumed that use in open systems was gradually reduced to 0 % in 1980, and use in nominally closed systems was reduced to 0 % in 1985.

The main uses of PCBs are thus in industry as heat exchange fluids, in electric transformers and capacitors, and as additives in paint, carbonless copy paper, sealants and plastics.

Approximate usage of PCBs in the US is summarized as follows (EIP Associates, 1997):

- Closed system and heat transfer fluids (transformers, capacitors, fluorescent light ballasts, etc.): 60 %.
- Plasticizers: 25 %.
- Hydraulic fluids and lubricants: 10 %.
- Miscellaneous uses: 5 %.

No substance flow analysis has been made for PCBs in Denmark. Surveys have primarily dealt with specific activities, such as energy production, mobile sources and waste disposal. No PCB air emission inventories for the use of product categories are available for Danish conditions. Even though PCB in consumer products is banned, the SPIN database and Statistics Denmark holds use data for some relevant product categories, which may serve as activity data in an emission inventory. Emission factors for use of products are not available at present. Thomsen et al. (2009) provides activity data from Statistics Denmark for use of chlorine containing solvents in organic chemicals production in the chemical industry (187 tonnes in 2005). Emission factors for PCBs emissions from the chemical industry are not known. In the EMEP/EEA guidebook (EMEP/CORINAIR, 2009) there is a Tier 1 default emission factor for category NFR 2F Consumption of Persistent Organic Pollutants and Heavy Metals of 0.1 g PCB/capita, which constitutes an average value of estimated emission factors for leaks from transformers and capacitors for European countries in 1990 (EEA, 2009; Berdowski et al., 1997). The Tier 1 emission factor thus comprise some of the sources relevant to the Sol-

vent and Other Product Use sector but also includes recycling of ferrous scrap, which falls outside this sector.

In the Nordic countries the regulations and trends in PCB use are comparable (Fauser et al., 2011). In a recent project at the Finnish Environment Institute SYKE for the OECD PRTR Task Force and funded by the Nordic Council of Ministers (SYKE, 2010), information related to release estimation techniques for releases from the use of products was compiled from the literature, and through surveys to national research institutes and authorities. The results of the study are used to prioritize product groups to be included in national PRTR registers, using the following criteria: (1) releases of a chemical from the use of the product is likely, (2) the product has widespread and high volume use, and (3) the releases from use of the product reach the environment in relevant volumes. Conclusions from the SYKE project are that at the moment the knowledge of emissions from the use of products is restricted and there is not much knowledge of the actual contribution of emissions from the use phase of end-products to the total emissions of most pollutants. However, there is a clear indication that a large part of the national total emissions of certain pollutants may originate from the use of products.

In Norway there have been studies on estimating emissions of PCBs to air primarily from energy production, mobile sources and waste disposal. No PCBs air emission inventories for the use of product categories are available for Norwegian conditions. Emission factors for use of products are not available at present. It has been forbidden to produce, sell and use substances or preparations that contain PCBs in Norway since 1980. There are PCBs in materials in buildings, particularly in plaster, jointing paste and paint, used in the period 1940-1980.

In Sweden the EPA Report "Omhändertagande av PCB i byggnader" (2002, Dnr 643-2492-02) provides estimates of the amount of PCBs in buildings presented. The amounts in sealants used were estimated to be between 100 to 500 tonnes. The corresponding figure for PCBs in insulating glass units were estimated to originally be about 115 tonnes, but the estimated remaining amount are 35 tonnes of PCBs. PCBs in acrylic floor mass was estimated to be 20 - 30 tonnes while the amounts in capacitors in fluorescent light was estimated to be 20 tonnes. Lilliehorn and Bernevi-Rex (2010) indicate that the total initial amount of PCBs in buildings was around 260 tonnes of which about 100 tonnes remains to be cleaned.

In conclusion the available data on PCBs amounts in products, use amounts of products and emission factors are limited in Denmark and the Nordic countries, due to the diversity of product groups, their uses, disposal, and emission patterns. Available PCBs emission factors are compiled in Table 34 below.

Table 34 Activities and products with PCBs use and emissions to air related to NFR 3 Solvent and Other Product Use. Blank cells indicate NFR3 categories that are not relevant with respect to PCBs use and emissions.

NFR 3	Product group/activity	PCB sources	PCBs emission factors
NFR 3A1 Decorative coating application	Domestic use	Paints, coatings	80 kg/t (surface coating) ³⁾
	Construction and buildings	Paints, coatings, repair activities	80 kg/t (surface coating) ³⁾
	Agriculture, forestry, fishing and fish farms		
NFR 3A2 Industrial coating application	Manufacture of automobiles ¹⁾		
	Coil coating ¹⁾		
	Car repair ¹⁾	Repair activities	
	Wood ¹⁾		
	Other industrial paint application ¹⁾	Paints, coatings, repair activities	80 kg/t (surface coating) ³⁾
	Boat building ¹⁾		80 kg/t (surface coating) ³⁾
NFR 3A3 Other coating application	Other non-industrial paint application	Paints, coatings, repair activities	80 kg/t (surface coating) ³⁾
NFR 3B1 Degreasing	Metal degreasing ¹⁾		
	Other industrial (dry) cleaning ¹⁾		
	Electronic components manufacturing ¹⁾	Lubricants, heat transfer systems, miscellaneous electrical equipment, capacitors	
NFR 3B2 Dry cleaning	Dry cleaning ¹⁾		
NFR 3C Solvents in chemical products manufacture and processing	Polyester processing ¹⁾		
	Polyvinylchloride processing ¹⁾	Plasticizers	
	Polyurethane foam processing ¹⁾		
	Polystyrene foam processing ¹⁾		
	Rubber processing ¹⁾	Plasticizers	
	Pharmaceutical products manufacturing ¹⁾		
	Paints manufacturing ¹⁾	Paints, heat transfer systems	2-10 kg/t PCB used ⁴⁾
	Inks manufacturing ¹⁾	Printing inks	
	Vehicle manufacturing ¹⁾		
	Glues manufacturing ¹⁾	Heat transfer systems	
	Adhesive, magnetic tapes, films & photographs manufacturing ¹⁾	Adhesives, plasticizers, heat transfer systems	
	Textile finishing ¹⁾	Fire retardants, heat transfer systems	
	Leather tanning ¹⁾		
	Other ¹⁾	Fire retardants in various products, pesticides, dust control (dedusting agents), recycled paper, lubricants, plasticizers, hydraulic fluids, heat transfer systems, capacitors, dielectric fluids and transformers, sealing liquids, waxes, casting agents, laminating and impregnating agents	0.1 kg/kg PCB used (production of capacitors and transformers) ⁵⁾
	Asphalt blowing ¹⁾	Asphalt roofing materials, tar paper, and roofing felt (IE CRF2A5) Heat transfer systems	
	Creosote-treated materials		

NFR 3D1 Printing	Printing industry ¹⁾	Printing inks	
NFR 3D2 Domestic solvent use including fungicides	Domestic solvent use (other than paint application)		
	Personal care products		
	Pharmaceuticals (see below)		
	Household cleaning agents	Dust control (dedusting agents)	
	Motor & vehicle cleaning agents		
	Use of adhesives and sealants	Adhesives, plasticizers	80 kg/t (adhesives) ³⁾
	Use of textiles	Fire retardants in textiles	
	Fungicide		
	Pesticides	Pesticides	
NFR 3D3 Other product use	Glass wool enduction ¹⁾		
	Mineral wool enduction ¹⁾		
	Fat, edible and not edible oil extraction		
	Application of glues and adhesives	Adhesives, Plasticizers	80 kg/t (adhesives) ³⁾
	Preservation of wood	Wood treatment	
	Underseal treatment and conservation of vehicles	Used oil, repair activities, lubricants, hydraulic fluids, auto salvage yards, auto crushing	80 kg/t (surface coating) ³⁾
	Vehicles dewaxing		
	Domestic use of pharmaceutical products		
	Other (preservation of seeds,...): use of pesticides in cultivations and in construction	Investment casting waxes, carbonless copy paper, fire retardants in various products, pesticides, dust control (dedusting agents), recycled paper, lubricants, plasticizers, hydraulic fluids, heat transfer systems, miscellaneous electrical equipment, electromagnets, fluorescent light ballasts, dielectric fluids and transformers, sealing liquids, waxes, casting agents, laminating and impregnating agents	83.6 kg/t (sealants) ²⁾
	Tobacco smoking		
	Tobacco manufacturing ¹⁾		
	Use of fireworks		
	Charcoal use for barbeques	PCBs formation during burning of charcoal	EF for residential wood combustion 134 µg/t
	Concrete, wall and floor coverings		
	Transformers and capacitors	Capacitors	0; 0.06 kg/t; 0.13 g/capita/year (leaks from transformers) ⁶⁾ 0; 0.8 kg/t (leaks from capacitors) ⁶⁾
	Surface coating, sealants and adhesives	Coatings, fire retardants, plasticizers	83.6 kg/t (sealants) ²⁾ 80 kg/t (surface coating, adhesives) ³⁾
	Enclosures and monitors		

	Construction and building products use	Fire retardants in building materials, repair activities, plasticizers	83.6 kg/t (sealants) ²⁾
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¹⁾ Mainly industrial sources.

²⁾ Open application (Breivik et al., 2002).

³⁾ Open application (SYKE, 2010).

⁴⁾ Paint production (EMEP/CORINAIR, 2006a).

⁵⁾ Production process (EMEP/CORINAIR, 2006a).

⁶⁾ SYKE (2010).

3.5.2 Summary on PCBs for solvent and other product use

Directive 96/59/EC on the disposal of PCBs and PCTs aims at disposing completely of PCBs and equipment containing PCBs as soon as possible. This Directive sets the requirements for an environmentally sound disposal of PCBs. EU Member States have to make an inventory of big equipment containing PCBs, have to adopt a plan for disposal of inventoried equipment, and outlines for collection and disposal of non-inventoried equipment (small electrical equipment very often present in household appliances manufactured before the ban on marketing of PCBs). The PCB Directive further mandates that Member States had to dispose of big equipment (equipment with PCB volumes of more than 5 litres) by the end of 2010 at the latest. The Commission will verify the implementation of this provision.

Since October 1986 production, import, use and sale of PCBs and apparatus containing PCB has been banned in Denmark. Furthermore, unintentional emissions, primarily from industrial production, energy production and incineration, must not exceed 0.0001 mg PCBs/(normal m³ emitted air). Regulations were applied in 1986 to ensure environmentally safe disposal of PCBs containing waste. A PCBs fact sheet issued by the Danish Environmental Protection Agency describes how PCBs in building joints and other building materials, in indoor air, in the working environment and in waste must be handled (DEPA, 2011). The regulations are not directed towards the activities in NFR 3 Solvent and Other Product Use sector but give directions on how to handle waste and disposed products, which are associated with this sector.

It has been forbidden to produce, sell and use substances or preparations that contain polychlorinated biphenyls in Norway since 1980. Although PCBs are banned, there are still PCBs in some products and materials produced before 1980. Use and handling of PCB containing products and materials are now largely regulated by:

- Regulations on restrictions on the use of hazardous chemicals and other products (Product Regulations).
- Regulations relating to the recycling of waste (Waste Regulations).

By the end of 2008 it was expected that approximately 90 % of PCB products in Norway were taken out of use. It was nevertheless estimated that nearly 150 tonnes of PCBs was still in use in products and materials. The most important known national sources for the remaining PCBs are buildings from the period 1940-1980, contaminated soil and sediments and oil in transformers and capacitors. There are large uncertainties associated with estimating the amount of PCBs used in Norway. (SFT, 2009).

Finland does not have national legislation restricting emissions of PCB emissions over those regulations by the UN, the UNECE or the EU.

In Sweden all use of PCBs was banned in 1978 and PCBs have been phased out gradually since then. PCBs were primarily used as insulating and lubricating oil in capacitors and transformers in sealants, paints, carbonless paper, etc. (KemI, www.kemi.se). Since 2007 it is regulated that buildings and structures erected between 1956 and 1973 should be investigated and remediated for PCBs in the sealants and flooring materials. The total amount of PCBs detected by the inventory is very difficult to assess but a rough calculation indicates that the initial amount of PCBs was around 260 tonnes of which about 100 tonnes remains to be cleaned. It is estimated that the company SAKAB, who has the only facility for the destruction of PCBs in Sweden, have destroyed around 40 tonnes of pure PCBs in sealants during the period 1998-2009 (Lilliehorn & Bernevi-Rex, 2010). The PCB regulation (SFS 2007:19) requires the removal of sealants containing PCBs to be completed in 2011 and 2013, respectively, depending on the type of building. The Swedish EPA suggested in 2010 that the regulation time limits should be moved. The two dates for the remediation to be completed is thus suggested to be 30 June 2014 and 30 June 2016, respectively.

3.5.3 Emission results

In this report all possible sources to PCBs emissions within this sector have been mapped and available emission factors are compiled. Omission of emission factors for certain possible sources does not mean they are insignificant. Experimental measurements and/or theoretical assessments are needed in order to get reliable estimates on missing emission factors for manufacture and use of the wide range of products. However, it is likely that the stated emission factors represent the most dominant sources. The scientific literature and other countries' emission inventories will continuously be checked for updated and new emission factors.

At present PCBs emissions are only calculated for charcoal use while emissions are not estimated for any other product uses. In future reporting focus will be on investigating the availability of activity data (use) for the products and activities where emission factors are available. Although surface coating is a PCBs source it is not all coating products that contain PCBs, therefore an assessment of PCBs content of specific products and activities, within the group of coating products, must be made.

The emission from charcoal use is shown in Table 35.

Table 35 Emission of PCBs, 1990-2011, g.

	1990	1995	2000	2005	2008	2009	2010	2011
Charcoal use	0.96	1.06	1.79	2.00	1.39	1.56	1.05	0.90

3.6 Agriculture

One source of PCBs emission from the agricultural sector have been identified; emissions from field burning of agricultural waste.

3.6.1 Methodology

The emission of PCBs from field burning of agricultural waste is calculated as shown for HCB in Chapter 2.5.1. No default emission factor is given in the

EMEP/EEA Guidebook (EEA, 2009). Emission factors given in Black et al. (2012) are used, see Table 36.

Table 36 Emission factor for PCBs.

	EF, µg TEQ per tonne
Straw from cereals	3
Straw from seed production	0.05

3.6.2 Emission results

Table 37 shows the emission of PCBs from the agricultural sector 1990- 2011.

Table 37 Emission of PCBs, 1990-2011, g.

	1990	1995	2000	2005	2008	2009	2010	2011
Field burning of agricultural waste	0.021	0.018	0.019	0.018	0.018	0.020	0.017	0.017

3.7 Waste

Emissions of PCBs could occur from NFR waste categories 6C Waste Incineration and 6D Waste Other but not from 6A Solid Waste Disposal on Land or 6B Wastewater Handling.

In Denmark, waste incineration of municipal, industrial and hazardous waste is carried out with energy recovery. Therefore, it is included under the energy sector (stationary combustion) in the international reporting to the LRTAP convention under UNECE and the same categorisation is used in this report. For details on the emissions of PCBs from waste incineration with energy recovery please refer to Chapter 3.1.

3.7.1 Methodology

In NFR waste category 6C Waste Incineration, the sources Human Cremation and Animal Cremation contribute to the emissions of PCBs; however, literature on the subject is scarce. The estimated emissions are shown in Table 38.

In 6D Waste Other, the emission source, Accidental fires, is likely to contribute to the emissions of PCBs but no data are available on this subject.

3.7.2 Emission results

Table 38 Emissions of PCBs from the waste category.

	Unit	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Human cremation	g	17.0	18.1	17.2	16.9	17.1	17.3	17.3	17.5	17.4	17.1
Animal cremation	g	1.0	1.3	2.8	4.9	7.1	8.2	8.5	8.5	9.2	7.8

4 Screening for pentachlorobenzene

Pentachlorobenzene (PeCB) is a semi-volatile chemical with an overall half-life in the environment measured in years and capable of long range transport. There are no large scale uses of PeCB at present.

PeCB is a chlorinated aromatic hydrocarbon with the molecular formula C_6HCl_5 . Its CAS registry number is 608-93-5.

Global emissions of PeCB around the year 2000 have been estimated at 85000 kg per year. However, it is noted that there is considerable uncertainty about the size of each of the estimated PeCB emissions, potentially an order of magnitude. (Bailey, 2007)

PeCB has also been used in a chlorobenzenes mixture with PCBs in electrical equipment. However, PCBs have not been used in Denmark in the time series considered, and hence this potential source is not relevant for Denmark.

4.1 Stationary combustion

This is considered the main source of emissions of PeCB. Especially, combustion of waste and biomass has been reported as the most significant sources of PeCB emissions (Bailey, 2007).

The observation of PeCB as a trace product of incomplete combustion has been widely reported. Nearly all fuels contain some chlorine, especially biomass and waste. The yield of PeCB from combustion of different fuels under different conditions has been reported to vary widely (Bailey, 2007; Hogendoorn et al., 2009).

4.1.1 Waste incineration

Bailey (2007) reviewed available data for emission factors for waste incineration. The emission factors reported varied greatly from 7 mg per tonne to 273 mg per tonne. Even the lowest of the emission factors reported are higher than the assumed emission factors for biomass and coal, see Chapters 4.1.2 and 4.1.3.

As expected waste incineration would be the largest source of PeCB from stationary combustion. However, it can be assumed that the same measures that reduce emissions of PCDD/F from waste incinerators will also have a reducing effect on the emission of PeCB. Therefore, the emission from waste incineration plants in Denmark will have decreased considerably since 1990 due to abatement measures implemented to reduce emissions of PCDD/F.

4.1.2 Biomass combustion

The only emission factor for PeCB from burning biomass found was from a German study where waste wood was burned in a high temperature incinerator (Zimmerman et al., 2001). No information on PeCB emissions from smouldering combustion or other combustion conditions was found. The emission concentration reported by Zimmerman et al. (2001) can be converted to an emission factor of approximately 0.6 mg per tonne.

Since the only emission factor available is for waste wood, it is not considered applicable to apply this emission factor in general to cover all wood combustion.

Some studies have tried to establish correlations between emission factors for PeCB with emission factors for other pollutants such as PCDD/F (dioxins and furans). However, this approach is very uncertain and the data material would have to be examined thoroughly before it could form the basis of establishing emission factors for PeCB.

4.1.3 Coal combustion

There are typically trace elements of chlorine in coal, amounting to about 0.1% chlorine (Bailey, 2007). No information on PeCB emissions from coal combustion has been found in the literature. However, since there are data available in the literature for HCB emission from coal combustion both in small scale boilers and stoves as well as in large utility or industrial boilers, emissions of PeCB must be expected from coal combustion.

Based on Oberg and Bergstrom (1985), Bailey (2007) calculates PeCB emission from small scale coal combustion by multiplying the HCB emission factor with five. Bailey (2007) does not consider emission of PeCB from large scale combustion of coal. The PeCB emission factor is calculated to 2.45 mg per tonne.

Small scale combustion of coal in Denmark is very limited and almost the entire consumption of coal is combusted at large power plants or large industrial plants. However, it cannot be ruled out that PeCB emissions occur also from large plants.

4.2 Mobile combustion

In Chapter 2 the available emission data for HCB from mobile sources are presented. The occurrence of HCB emissions from mobile sources could suggest that PeCB could also be emitted. However, it should be noted that little information on HCB emissions from mobile sources is available and the emission levels reported are very low. No information has been found suggesting that PeCB should be formed during combustion of liquid fuels and hence, it is considered that emissions of PeCB from mobile combustion sources are not applicable.

4.3 Fugitive emissions from fuels

No information is available regarding potential emissions of PeCB from any process related to fugitive emissions from fuels. Therefore, it is considered that emissions of PeCB from fugitive emission sources are not applicable.

4.4 Industrial processes and solvent and other product use

Emissions of PeCB from several processes within metal industry have been reported. This includes aluminium production (Aittola et al., 1996; Westberg et al., 1997), magnesium production (Knutzen and Oehme, 1989) and copper production (Doering et al., 1992). However, none of these processes are occurring in Denmark.

Another possible source of emissions is industrial chlorination reactions, where it is possible to produce PeCB as a by-product. There are other processes which produce a variety of chlorinated aromatics that may contribute

PeCB even if PeCB has not been explicitly detected and reported yet (Bailey, 2007).

No other industrial sources that are considered to take place in Denmark have been identified where PeCB emissions could occur.

4.5 Agriculture

Emissions of PeCB from the use of pesticides have been reported (Bailey, 2007). However, it has not been possible to establish particular pesticides that would contain impurities of PeCB. The global emission of PeCB from pesticide use as reported by Bailey (2007) is much lower than the emissions reported for combustion of especially waste and biomass.

4.6 Waste

In Denmark, waste incineration of municipal, industrial and hazardous waste is carried out with energy recovery. Therefore, it is included under the energy sector (stationary combustion) in the international reporting to the LRTAP convention under UNECE and the same categorisation is used in this report. For details on the emissions of PeCB from waste incineration please refer to Chapter 4.1.1.

Other emission sources in the waste sector include cremations, incineration of carcasses and accidental fires. It has not been possible to find any information in the literature on PeCB emissions from these categories, nor has it been possible to find any documented ratios between other POP emissions and PeCB. As a consequence it is not possible to provide estimates of PeCB from the waste category. Since emissions of PCDD/F, HCB and PCBs do occur from these activities it cannot be concluded that emissions from the sector do not occur.

4.7 Conclusions

The available data for PeCB emissions to air are extremely limited and in many cases the reported data are based on highly uncertain assumptions. The most detailed analysis is provided by Bailey (2007), where an attempt of making a global inventory for PeCB has been made.

From this study, the largest sources are waste incineration and biomass combustion, with significant contributions coming from coal combustion and aluminium casting. Of these sources aluminium casting is not occurring in Denmark, while the combustion of coal and biomass as well as the incineration of waste are prevalent in Denmark.

The remaining quantified sources in Bailey (2007) are: chlorinated solvents, pesticide use and chemical manufacturing and waste disposal. None of these sources are thought to be large sources in Denmark.

The emission factors available for the combustion related releases of PeCB to the atmosphere are only well documented in the case of waste incineration.

For coal the emission factor used by Bailey (2007) is based on a correlation with a HCB emission factor reported for small scale coal combustion. It is not considered that this emission factor can be representative for all coal combustion.

For biomass combustion the emission factor use is referenced to a single German study where waste wood was combusted in a boiler with a thermal capacity of 1 MW. According to Zimmerman et al. (2001) the wood used during the experiment was contaminated with plastics and paint. The precise degree of contamination is unknown. Therefore, the emission factor might be more representative for waste incineration than for combustion of wood or other biomass without any contamination of chlorinated compounds.

As mentioned, more data are available for waste incineration than for the other types of fuel combustion. However, the reported emission factors display large variations and it is not possible to identify an emission factor that in general could represent the Danish conditions.

Based on the available data, it appears that waste incineration will be the largest source of PeCB emissions to air. However, the data foundation is at the moment not strong enough to facilitate the establishment of an emission inventory with even a modicum of certainty.

5 Conclusions

The work on improving the Danish air emission inventories for HCB and PCBs was carried out for all sectors in the Danish inventory system. For some sectors few datasets are available and even for the sectors with more available emission measurements the uncertainties are still considered to be significant.

Until this project the Danish inventory for HCB was incomplete as it only included few stationary combustion sources and some sources within the waste sector. In this project the completeness has been greatly improved, so that all reporting sectors have been considered. As a result there is now only 9 categories where the notation key NE (not estimated) is used compared to 30 in the previous submission. These categories are related to the industrial processes sector, the solvent and other product use sector and the waste sector. For these categories there are no methodologies and default emission factors available in the EMEP/EEA Guidebook nor has it been possible to find emission factors in the literature. For some categories, it is the case that the entire category is not considered in the Danish inventory, e.g. small scale waste burning. When it has been decided to use the notation key NE for these categories rather than NA (not applicable), it is because of indications that emissions of HCB could occur. Such indication is for example reported and well-documented emission factors for PCDD/F.

The results of the improved emission inventory for HCB can be seen in Table 39 and Figure 3 below.

Table 39 Danish emission inventory for HCB for 1990-2011, kg.

	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Energy industries	4.4	3.3	1.5	1.2	1.7	1.5	1.4	1.4	1.4	1.2
Manufacturing industries and construction	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Transport	0.4	0.4	0.5	0.6	0.6	0.7	0.7	0.6	0.7	0.7
Other sectors	1.5	1.0	0.4	0.4	0.4	0.5	0.5	0.4	0.5	0.5
Industrial processes	2.0	2.3	2.7	1.4	0.7	0.7	0.6	0.01	0.01	0.01
Solvents and other product use	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.001	0.001	0.001
Agriculture	18.4	0.6	0.4	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Waste	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Total	26.8	7.8	5.6	3.8	3.7	3.6	3.3	2.7	2.8	2.6
Total emission reported in 2013	3.2	1.9	0.6	0.5	0.5	0.5	0.6	0.5	0.6	0.6

The significant decrease in the HCB emission is caused by a decrease in the emissions from agriculture. The decrease is partly caused by a decrease in the use of pesticides and fungicides known to contain impurities of HCB and partly by the fact that the level of HCB impurities in certain pesticides was greatly reduced in the beginning of the 1990'ties.

The emission from energy industries has also decreased due to flue gas abatement targeting dioxin and furans installed at the waste incineration plants. The emission from industrial processes has decreased due to the closure of steel production and secondary aluminium production in Denmark. For the remaining sectors the emissions are quite low and stable over time.

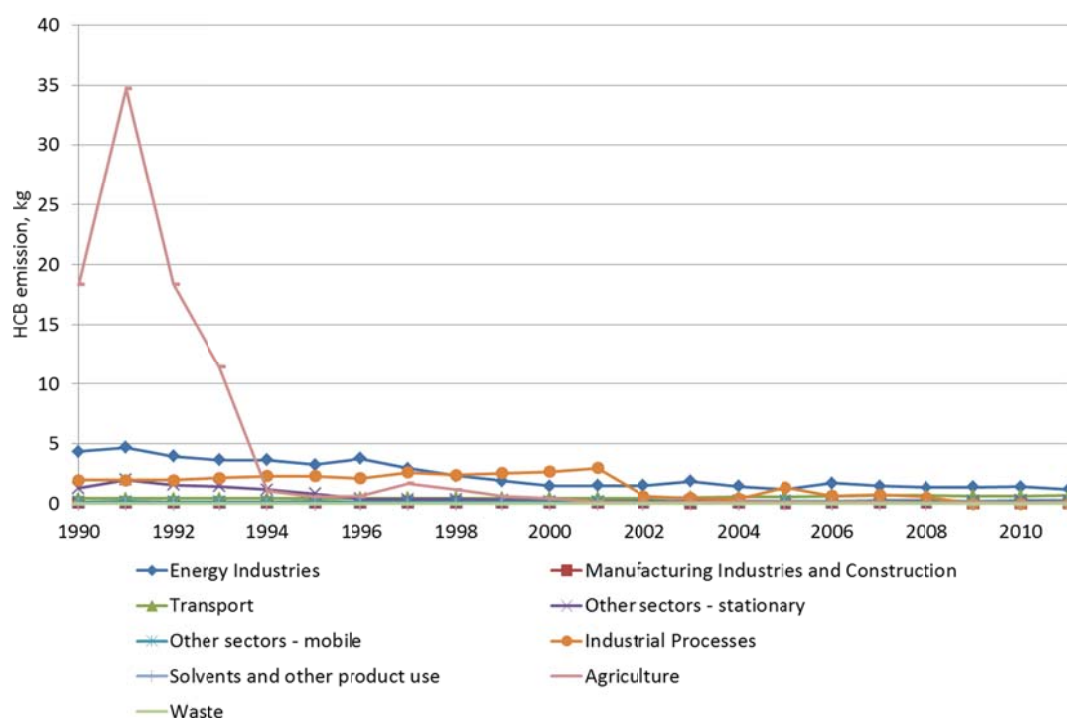


Figure 3 Danish emissions of HCB 1990-2011.

Previously the Danish inventories did not include PCBs and therefore the completeness has been greatly improved by the work undertaken in this project. As a result there is now only 13 categories where the notation key NE (not estimated) is used (previously there were 50 categories where NE was reported). These categories are related to the industrial processes sector, the solvent and other product use sector and the waste sector. For these categories there are no methodologies and default emission factors available in the EMEP/EEA Guidebook nor has it been possible to find emission factors in the literature. For some categories, it is the case that the entire category is not considered in the Danish inventory, e.g. small scale waste burning. When it has been decided to use the notation key NE for these categories rather than NA (not applicable), it is because of indications that emissions of PCBs could occur. Such indication is for example reported and well-documented emission factors for PCDD/F.

The results of the improved emission inventory for PCBs can be seen in Table 40 and Figure 4 below.

Table 40 Danish emission inventory for PCBs for 1990-2011, kg.

	1990	1995	2000	2005	2006	2007	2008	2009	2010	2011
Energy industries	0.9	0.6	0.3	0.2	0.3	0.3	0.2	0.3	0.3	0.3
Manufacturing industries and construction	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.1
Transport	90.8	23.1	23.6	26.9	27.7	28.5	27.1	24.5	25.3	25.9
Other sectors	17.2	14.3	13.3	14.5	15.0	16.1	16.6	14.8	16.3	16.3
Industrial processes	1.6	1.9	1.8	0.8	0.2	0.2	0.2	0.1	0.1	0.1
Solvents and other product use	0.001	0.001	0.002	0.002	0.003	0.002	0.001	0.002	0.001	0.001
Agriculture	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002	0.00002
Waste	0.02	0.02	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.02
Total	110.7	40.2	39.1	42.5	43.4	45.2	44.2	39.8	42.0	42.7

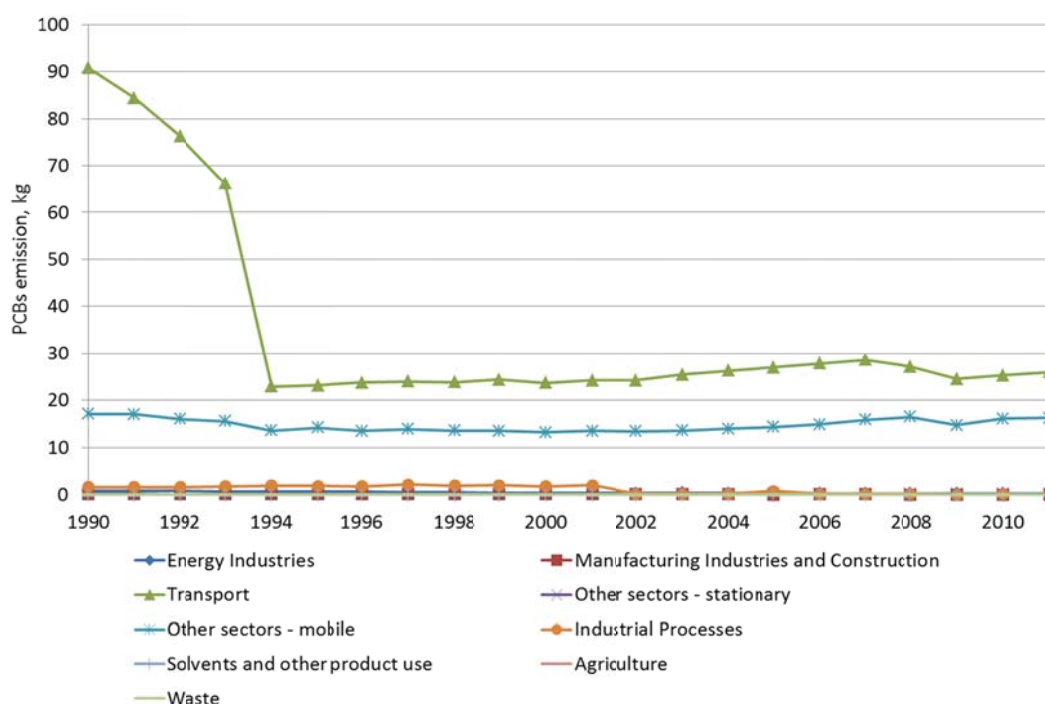


Figure 4 Danish emissions of PCBs 1990-2011.

The significant decrease in the PCBs emission is caused by a decrease in the emissions from road transport. Leaded gasoline had high emission factors for PCBs and when this fuel was phased out in the beginning of the 1990'ties, the emissions also decreased significantly.

The other dominant emission sources are both road and non-road diesel vehicles and the emission from these sources has increased due to an increase in the diesel consumption. The emission from energy industries has decreased due to flue gas abatement targeting dioxin and furans installed at the waste incineration plants. The emission from industrial processes has decreased due to the production stop for steel and secondary aluminium. For the remaining sectors the emissions are quite low and stable over time.

The screening for emission sources for PeCB showed that the available data are extremely limited and in many cases the reported data are based on highly uncertain assumptions. While it can be assumed that emissions of PeCB can occur from the same sources as HCB, very few measurements have been reported in the literature.

Based on the available data, it appears that waste incineration will be the largest source of PeCB emissions to air. However, the data foundation is at the moment not strong enough to facilitate the establishment of an emission inventory.

6 References

- Aittola, J.-P., Paasivirta, J., Vattulainen, A., Sinkkonen, S., Koistinen, J., Tarnanen, J., 1996: Formation of chloroaromatics at a metal reclamation plant and efficiency of stack filter in their removal from emission. *Chemosphere* 32: 99-108.
- Andrijewski, M. (ed.), 2004: Poland, National implementation plan for the Stockholm Convention. Enabling activities to facilitate early action on the implementation of the Stockholm Convention on Persistent Organic Pollutants (POPs), 2004. Available at: <http://pdc.ceu.hu/archive/00002817/01/NIP-Poland-eng.pdf> (25-05-2014)
- Antunes, P., Viana, P., Vinhas, T., Rivera, J. & Gaspar, E.M.S.M., 2012: Emission profiles of polychlorinated dibenzodioxins, polychlorinated dibenzofurans (PCDD/Fs), dioxin-like PCBs and hexachlorobenzene (HCB) from secondary metallurgy industries in Portugal. *Chemosphere* 88 (2012) 1332–1339.
- ATSDR, 2006: Case Studies in Environmental Medicine – Polychlorinated Biphenyls (PCB) Toxicity. Environmental Alert. U.S. Department of Health and Human Services - Agency for Toxic Substances and Disease Registry - Division of Toxicology and Environmental Medicine. ATSDR Publication No.: ATSDR-HE-CS-2003-0001.
- Bailey, R.E., 2001: Global hexachlorobenzene emissions, *Chemosphere*, 43, 2001.
- Bailey, R.E., 2007: Pentachlorobenzene –Sources, environmental fate and risk characterization, Science dossier, Euro Chlor, 41 pages.
- Berdowski, J.J.M., Baas, J., Bloos, J.P.J., Visschedijk, A.J.H., Zandveld, P.Y.J., 1997: The European Atmospheric Emission Inventory for Heavy Metals and Persistent Organic Pollutants. Umweltforschungsplan des Bundesministers für Umwelt, Naturschutz und Reaktorsicherheit. Luftreinhaltung. Forschungsbericht 104 02 672/03. TNO, Apeldoorn, The Netherlands.
- Bipro 2006. Identification, assessment and prioritisation of EU measures to reduce releases of unintentionally produced/released Persistent Organic Pollutants. REFERENCE:O7.010401/2005/419391/MAR/D4. European Commission, Brussels.
- Black, R.R., Meyer, C.P., Touati, A., Gullett, B.K., Fiedler, H. & Mueller, J.F., 2012: Emission factors for PCDD/PCDF and dl-PCB from open burning of biomass, *Environment International*, Volume 38, Issue 1, January 2012, Pages 62-66
- Breivik, K., Sweetman, A., Pacyna, J.M. & Jones, K.C., 2002: Towards a global historical inventory for selected PCB congeners – a mass balance approach, 2. Emissions. *The Science of the Total Environment* 290 (199-224).
- Cooper, D.A., 2005: HCB, PCB, PCDD and PCDF emissions from ships, *Atmospheric Environment* 39 (2005) 4901–4912.

Council Regulation (EC) No 166/2006 of 18 January 2006 concerning the establishment of a European Pollutant Release and Transfer Register and amending Council Directives 91/689/EEC and 96/61/EC. Official Journal of the European Communities L 33/1, 04.02.2006.

de Voogt, P. & Brinkman, U.A.Th., 1989: Production, properties and usage of polychlorinated biphenyls. In: Kimbrough, R.D., Jensen, A.A. (Eds.), Halogenated Biphenyls, Terphenyls, Naphtalenes, Dibenzodioxins and Related Products. Topics in Environmental Health. Elsevier Science Publishers, Amsterdam, pp. 3–45.

Denier van der Gon, H., van het Bolscher, M., Visschedijk, A., & Zandveld, P., 2007: Emissions of persistent organic pollutants and eight candidate POPs from UNECE-Europe in 2000, 2010 and 2020 and the emission reduction resulting from the implementation of the UNECE POP protocol. Atmospheric Environment 41 (2007) 9245–9261.

DEPA, 2011a: Statistics for use of pesticides in agriculture. Available at (in Danish).

http://www.mst.dk/Virksomhed_og_myndighed/Bekaempelsesmidler/Pesticider/pesticidstatistik/Landbrug/ (25-05-2014)

DEPA, 2011b: PCB fact sheet – update 21 January 2011 (in Danish). Available at:

<http://mst.dk/media/mst/70131/PCB%20faktaark%20opdateret%2025%20%20januar%202011.pdf> (25-05-2014)

Doering, J., Damberg, M., Gamradt, A. & Oehme, M., 1992: Screening method based on the determination of perchlorinated aromatics for surface soil contaminated by copper slag containing high levels of polychlorinated dibenzofurans and dibenzo-p-dioxins. Chemosphere 25(6): 755-762.

Dyke, P.H., Foan, C. & Fiedler, H., 2003: PCB and PAH releases from power stations and waste incineration processes in the UK, Chemosphere, 50, 2003.

EMEP/MSC, 2000: Hexachlorobenzene: Properties, Emissions and Content in the Environment, EMEP Meteorological Synthesizing Centre – East, Technical note 6/2000, 86 pages.

EEA, 2007: Emission inventory guidebook - 2007 (Internet version). Group 6. A joint EMEP/CORINAIR production. Available at:

<http://reports.eea.europa.eu/EMEPCORINAIR5/en/page002.html>

EEA, 2009: EMEP/EEA air pollutant emission inventory guidebook – 2009. Available at:

<http://www.eea.europa.eu/publications/emep-eea-emission-inventory-guidebook-2009>

EEA, 2013: EMEP/EEA air pollutant emission inventory guidebook – 2013. Available at:

<http://www.eea.europa.eu/publications/emep-eea-guidebook-2013>

EIP Associates, 1997: Polychlorinated Biphenyls (PCBs) Source Identification. Prepared for the Palo Alto Regional Water Quality Control Plant. Palo Alto, CA 1997.

EMEP/CORINAIR, 2006a: Sources of PCB emissions. EMEP/CORINAIR Guidebook – September 2006.

EMEP/CORINAIR, 2006b: Sources of HCB emissions. EMEP/CORINAIR Guidebook – September 2006.

EPA, 2011: U.S. Environmental Protection Agency. Polychlorinated Biphenyls (PCBs) Homepage. Available at:
[http://yosemite.epa.gov/R10/OWCM.NSF/webpage/Polychlorinated+Biphenyls+\(PCBs\)+Homepage/](http://yosemite.epa.gov/R10/OWCM.NSF/webpage/Polychlorinated+Biphenyls+(PCBs)+Homepage/)

European Commission, 2013: Best Available Techniques (BAT) Reference Document for the Refining of Mineral Oil and Gas. Industrial Emissions Directive 2010/75/EU (Integrated Pollution Prevention and Control). Final draft (July 2013).

Fausser, P., Saarinen, K., Aasestad, K. & Danielsson, H. 2011: Emissions of mercury, PAHs, dioxins and PCBs related to NFR 3 "Solvent and Other Product Use" in Nordic countries. TemaNord, 98 p.

Fontelle, J.-P., Chang, J.-P., Vincent, J., Andre, J.-M., Taieb, N., Bort, R., De-florenne, E., Druart, A., Dulhoste, S., Gavel, A., Gueguen, C., Jabot, J., Jacquier, G., Jeannot, C., Kessouar, S., Martin, E., Mathias, E., Nicco, L., Serveau, L. & Vilmain, J.-B., 2014: Organisation et méthodes des inventaires nationaux des émissions atmosphériques en France, 11ème édition.: National Inventories of Air Emissions in France, OMINEA. 2014.

Grochowalski, A. & Konieczynski, J., 2008: PCDDs/PCDFs, dl-PCBs and HCB in the flue gas from coal fired CFB boilers, *Chemosphere*, 73, 2008.

Gullet, B.K., Touati, A. & Hays, M.D., 2003: PCDD/F, PCB, HxCBz, PAH and PM Emission Factors for Fireplace and Woodstove Combustion in the San Francisco Bay Region, *Environmental Science & Technology*, vol. 37, 2003.

Gusev, A., Dutchak, S., Rozovskaya, O., Shatalov, V., Sokovykh, V., Vulykh, N., Aas, W. & Breivik, K. 2011: Persistent Organic Pollutants in the Environment. EMEP Status Report 3/2011 June 2011.

Hedman, B., Näslund, M. & Marklund, S. 2006: Emission of PCDD/F, PCB, and HCB from Combustion of Firewood and Pellets in Residential Stoves and Boilers, *Environmental Science & Technology*, vol. 40, 2006.

Hogendoorn, E.A., Bruinen de Bruin, Y. & Janssen, M.P.M. 2009: Inventory emission factors for pentachlorobenzene. RIVM Letter report 601773002.

Hübner, C., 2001: Österreichische Emissionsinventur für POPs 1985–1999. Studie im Auftrag des Umweltbundesamt. Interner Bericht, Bd. IB-650. FTU – Forschungsgesellschaft Technischer Umweltschutz GmbH, Wien.

Kakareka, S. & Kukharchyk, 2005a: Sources of hexachlorobenzene emissions.

Kakareka, S. & Kukharchyk, 2005b: Sources of polychlorinated biphenyls emission.

Knutzen, J. & Oehme, M. 1989: Polychlorinated dibenzofuran (PCDF) and dibenzo-p-dioxin (PCDD) levels in organisms and sediments from the Frierfjord, Southern Norway. *Chemosphere* 19(12): 1897-1909.

Lee, R.G.M., Coleman, P., Jones, J.L., Jones, K.C. & Lohmann, R., 2005: Emission Factors and Importance of PCDD/Fs, PCBs, PCNs, PAHs and PM10 from the Domestic Burning of Coal and Wood in the U.K., *Environmental Science & Technology*, vol. 39, 2005.

Lilliehorn, P. & Bernevi-Rex, G., 2010: Uppföljning av inventering och sanering av PCB i fog- och golvmassor. Available at (in Swedish): http://www.fogspecialisten.se/uploads_files/uppfoljning_pcb.pdf (25-05-2014)

Nielsen, M., Nielsen, O.-K. & Thomsen, M. 2010: Emissions from decentralised CHP plants 2007 - Energinet.dk Environmental project no. 07/1882. Project report 5 - Emission factors and emission inventory for decentralised CHP production. National Environmental Research Institute, Aarhus University. 113 pp. - NERI Technical report No. 786. Available at: <http://www.dmu.dk/Pub/FR786.pdf>

Nielsen, O.-K., Winther, M., Mikkelsen, M.H., Hoffmann, L., Nielsen, M., Gyldenkerne, S., Fauser, P., Plejdrup, M.S., Albrechtsen, R., Hjelgaard, K. & Bruun, H.G. 2013. Annual Danish Informative Inventory Report to UNECE. Emission inventories from the base year of the protocols to year 2011. Aarhus University, DCE - Danish Centre for Environment and Energy, 699 pp. Scientific Report from DCE - Danish Centre for Environment and Energy No. 53. Available at: <http://www.dmu.dk/Pub/SR53.pdf>

Oberg, T. & Bergstrom, J.G.T., 1985: Hexachlorobenzene as an indicator of dioxin production from combustion. *Chemosphere* 14(8): 1081-1086.

Pacyna, J.M., Breivik, K., Münch, J. & Fudala, J., 2003: European atmospheric emissions of selected persistent organic pollutants, 1970-1995, *Atmospheric Environment*, 37, 2003.

Pandelova, M., Stanev, O., Hendelmann, B., Lenoir, D. & Schramm, K.-W., 2009: Correlation of PCDD/F and PCB at combustion experiments using wood and hospital waste. Influence of (NH₄)₂SO₄ as additive on PCDD/F and PCB emissions. *Chemosphere*, 75, 2009.

PCB Fact Sheet, 2003: Fact Sheet: Sources of Polychlorinated Biphenyls. Available at: <http://www.deq.state.or.us/lq/cu/nwr/PortlandHarbor/docs/SourcePCBs.pdf>

Saarinen, K., Lindh, P., Kokko, M., Mikkola-Pusa, J., Suomi, O., Nurmi, E. & Mattila, S. 2007: Finnish HCB air emissions 1990-2005 to the Secretariat of the UNECE Convention on Long-Range Transboundary Air Pollution. Finnish Environmental Institute, Expert Services Department, Environmental Management Division, Air Emission team.

SFT, 2009: Handlingsplan for reduserte utslipp av PCB i 2009-2012. (Action plan for reducing emissions of PCBs in 2009-2012). Available at: http://www.miljodirektoratet.no/Global/dokumenter/tema/kjemikaler/pcb_handlingsplan090709.pdf (25-05-2014)

Swedish EPA Report, Dnr 643-2492-02, 2002. Omhändertagande av PCB i byggnader. Available at (in Swedish):
http://www.naturvardsverket.se/upload/stod-i-miljoarbetet/vagledning/pcb/pcb_i_byggnader.pdf (25-05-2014)

Šyc, M. Horák, J., Hopan, F., Krpec, K., Tomšej, T., Ocelka, T. & Pekárek, V., 2011: Effect of Fuels and Domestic Heating Appliance Types on Emission Factors of Selected Organic Pollutants, *Environmental Science & Technology*, vol. 45, 2011.

SYKE, 2011: Releases from the use of products. Finnish Environment Institute, Centre for Sustainable Consumption and Production. Environmental Performance Unit, Helsinki 5th October 2010.

Thanner, G. & Moche, W., 2002: Emission von dioxinen, PCBs und PAHs aus kleinfeuerungen. Umweltbundesamt, Federal Environmen Agency - Austria.

Thistlethwaite, G. 2001a: Determination of Atmospheric Pollutant Emission Factors at a Small Coal-fired heating boiler, 2001. A report produced for the Department of the Environment, Transport and the Regions, the National Assembly for Wales, the Scottish Executive and the Department of the Environment for Northern Ireland by AEA Technology Environment.

Thistlethwaite, G. 2001b: Determination of Atmospheric Pollutant Emission Factors at a Small Industrial Wood-Burning Furnace, 2001. A report produced for the Department of the Environment, Transport and the Regions, the National Assembly for Wales, the Scottish Executive and the Department of the Environment for Northern Ireland by AEA Technology Environment.

Thomsen, M., Nielsen, O.-K. & Illerup, J., 2009: Unintentional formation and emission of the persistent organic pollutants HCB and PCBs in the Nordic countries. *TemaNord* 2009:518.

Toda, 2005: POPs and heavy metals emission inventory of Japan. TFEIP & ESPREME Workshop on "Heavy Metals and POPs Emissions, Inventories and Projections", 18-19 October 2005, Rovaniemi, Finland.

UNECE, 2009: Guidelines for estimating and reporting emission data under the Convention on Long-range Transboundary Air Pollution (ECE/EB.AIR-/97).

UNEP, 1999: United Nations Environment Programme. Guidelines for the Identification of PCBs and Materials Containing PCBs. First Issue August 1999. Available at:
<http://www.chem.unep.ch/Publications/pdf/GuidIdPCB.pdf>

Villeneuve, J., Palacios, H. P., Savoie, P. & Bodbout, S., 2012: A critical review of emission standards and regulations regarding biomass combustion in small scale units (<3 MW), *Bioresource Technology*, 111, 2012.

Wang, G., Lu, Y., Han, J., Luo, W., Shi, Y., Wang, T. & Sun Y. 2010: Hexachlorobenzene sources, levels and human exposure in the environment of China. *Environment International* 36 (2010) 122-130.

- Wegiel, M., Chrzaszcz, R., Maslanka, A. & Grochowalski, A. 2011: Study on the determination of PCDD/Fs and HCB in exhaust gas. *Chemosphere*, 85, 2011.
- Wenzel K.-D., Hubert A., Weissflog L., Kühne, R., Popp P., Kindler A. & Schüürmann G., 2006: Influence of different emission sources on atmospheric organochlorine patterns in Germany, *Atmospheric Environment* 40, 943-957.
- Westberg, H.B., Selden, A.I. & Bellander, T. 1997: Emissions of some organochlorine compounds in experimental aluminum degassing with hexachloroethane. *Appl. Occup. Environ. Hyg.* 12: 178-183.
- Zimmerman, R., Blumenstock, M., Heger, H.J., Schramm, K.-W. & Kettrup, A., 2001: Emission of non-chlorinated and chlorinated aromatics in the flue gas of incineration plants during and after transient disturbances of combustion conditions: Delayed emission effects. *Environ. Sci. Technol.* 35: 1019-1030.
- Yang, C., 2006: Estimating HCB Releases from Pesticide Applications, HCB Releases from Pesticide Applications in USA and Canada – All Years. Environment Canada.
- Yu, B.-W., Jin, G.-Z., Moon, Y.-H., Kim, M.-K., Kyoung, J.-D. & Chang, Y.-S. 2006: Emission of PCDD/Fs and dioxin-like PCBs from metallurgy industries in S. Korea. *Chemosphere* 62 (2006) 494–501.

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This report documents the improved air emission inventories for hexachlorobenzene (HCB) and polychlorinated biphenyls (PCBs). The emission inventories now include all sources covered by the EMEP/EEA Guidebook. The completeness and accuracy of the inventories for these pollutants have been significantly improved through the results of this project. The project also included a screening for pentachlorobenzene (PeCB); the result of this screening was that the current data available do not allow for the elaboration of an emission inventory.

ISBN: 978-87-7156-074-9
ISSN: 2245-0203



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Scientific Report from DCE – Danish Centre for Environment and Energy

No. 103

2014



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