Identification and Quantification of Sources to PCDD, PCDF, PCB and HCB in Sweden

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Introduction

During 1988 - 1992 an extensive national survey of dioxin sources was carried out in Sweden[1]. A second, less comprehensive, survey was performed during 2003 – 2005[2]. In the current survey we have focussed not only on dioxins but also on sources to PCB and hexachlorobenzene, HCB. The aim of this study was to update the state of art in Sweden as a basis for the national implementation of the Stockholm Convention. Thus, the substances covered in this survey are identical to those listed in annex C of the Stockholm Convention. The survey included literature studies, questionnaires to regional authorities and industry, as well as analyses of samples taken in connection to a few prioritised primary and secondary sources.

Environmental monitoring of dioxins and PCB in Sweden since the late 1960s indicates a profound decrease of these substances in biota. Since 1967 the general decrease in some species studied in the national monitoring programme (herring, pike and guillemot) has been estimated to be 3-10 % per year[3]. This positive trend has however become less obvious during the last 10-15 years, and in some areas it is to day difficult to detect any temporal trend.

The Swedish national food administration has estimated that some ten percent of the population has an exposure to dioxins and dioxin-like PCBs that exceed the current TDI set by EU at 2 pg TEQ/kg b.w. It is therefore urgent to further reduce formation and emission of the current substances as well as to develop and improve techniques for this purpose.

The survey

The first phase contained a survey of relevant literature. This was followed up by a set of questionnaires sent to industries and local and regional authorities in order to get an overview of the current knowledge. The survey included both primary sources such as different industrial branches (pulp and paper industry, metal industry, cement kilns, chemical industry etc.), shipping and combustions processes as well as secondary sources such as dump sites and contaminated soil and sediments. Combustion processes studied were both large scale plants as well as small scale furnaces in individual homes. Based on the outcome of the first phase, a number of prioritized areas were identified where the need for complementary analyses were considered to be particularly motivated. In total, some 150 samples were taken for this purpose. These samples included emissions to air, water and wastes from industries, shipping, combustion processes and contaminated sites.

Results and conclusions

Compared to the situation in the early 1990s, our knowledge of the magnitude and character of the emissions from some industrial sources has improved. In most cases, however, it is obvious that we still need more up to date information from industrial sources and different combustion processes to get an adequate picture of the magnitude and proportions of the major sources of unintentionally produced contaminants. Based both on emission factors and measurements the following results are given (Table 1).

Table 1. Estimated current emissions from a number of primary dioxin sources to air, water and products/waste compared to the emissions as they were estimated in the early 1990s. The quantification of dioxins (often applying different systems of TEQ calculation) is given in grams/year. The purpose of the table is mainly to give an overview of the magnitude between the different sources. For some of the sources, the estimation of current emissions can be interpreted as being higher than previous estimations. The reason for this is, in most cases, lack of reliable current data from representative sampling and analysis. This has led to great uncertainty and the interval around the “true”

value has sometimes increased. Lack of reliable data are indicated as "-".

	Emissions to air g TEQ/yr		Emissions to water g TEQ/yr		Products and waste g TEQ/yr	
	1993	2004	1993	2004	1993	2004
Iron- and steel industry	2-20	5,9-8,6	-	-	28	-
Foundries and non-ferrous metal industry	5	5,6-10,3	-	-	3-17	<2
Cement industry	3-6	0,2-0,3	-	-	-	-
Pulp- and paper industry	1	1,2	1,5-5	<0,1	3-9	<5
Chloralkali industry	-	-	0,28-0,6	0,001-0,02	0,25	0,008-0,26
Fossil fuel burning	0,7-3	<4	-	-	-	-
Small- and large scale bio fuel burning	3,5-18	<14	-	-	-	<11
Municipal waste incineration	3	1,1	-	-	0,6-2,4	C:a 160
Accidental fires at dump sites	2,8-30	0,4-65	-	-	-	-
Road traffic	0,2-1,4		-	-	-	-
Shipping, to and from Swedish ports		0,2-0,5	-	-	-	-

The results indicate that most industries have decreased their emissions since the 80's (data not shown) and probably also since the 90's. This later reduction is however not that pronounced and the data are scarce and uncertain for many source categories. Reductions could be the result of optimisation of the process variables specifically to accomplish a reduction of formation and emission of dioxins. Other changes to reduce the emissions more in general, as dust filtering, has probably also had a positive effect on the emissions of dioxins.

The metal industry is an important industry in Sweden and one of those that contribute to a great extent to the production and emission of dioxins[4]. The main factors regulating the emissions are the process/furnace used, the amount of scrap-metal added and the quality of the scrap-metal. The main contributor is the secondary non-ferrous works. The waste products from the metal industry contains a non neglectable amount of dioxins. However, due to the recirculation of the waste the amount of unintentionally produced contaminants emitted to the environment is unclear. In an international perspective the emissions from the Swedish metal industry are still relatively low, with regards to the large amounts produced.

From the available results of the ships, there is no firm rationale for differentiating the emission factors for engine types regarding engine speed, but it is relevant to differentiate between fuel types[5]. The present results suggest that a lower emission factor for PCDD/PCDF WHO-TEQ and a higher emission factor for HCB than previous marine emission factors, (table 2). In general, the measured emissions can be considered as relatively low.

Table 2. HCB, PCB, PCDD and PCDF emissions factors for different marine fuels[5].

Unit	Marine Distillate		Residual Oil	
	ng kWh^{-1}	g TJ supplied $^{-1}$ [a]	ng kWh^{-1}	g TJ supplied $^{-1}$ [a]
HCB	20	1.95×10^{-3}	30	3.50×10^{-3}
Total PCB	90	8.76×10^{-3}	120	1.40×10^{-2}
PCDD/PCDF WHO-TEQ	0.03	2.92×10^{-6}	0.1	1.17×10^{-5}

a: The conversion from ng kWh^{-1} to g TJ $supplied^{-1}$ takes in to account the different diesel engine types and their distribution (corresponds to ca. 45% efficiency for the diesel engine).

Combustion of bio fuels as wood and woodchips generally give rise to minor emissions of dioxins and related compounds. The few data from large scale bio fuel burning indicates low emissions, but due to the large number of combustion plants in Sweden their total environmental impact could be considerable[2, 4]. The emissions from small scale bio fuel burning are to a large extent dependent on the quality, in generally strongly correlated to the age, of the furnaces/stoves. This pattern is significant for dioxins and PCB as well as other hazardous substances such as PAH, VOC and particles. Contamination of the fuel with plastic, paint or wood impregnated with chlorophenols could also severely affect the outcome for both large- and small scale bio fuel burning.

Combustion tests simulating domestic small scale combustion of waste, “backyard burning”, were performed within this survey (discussed in abstract from Hedman, et al, 2005 at this conference). Different fuel combinations were used but no obvious relation between the content of the waste fuel and the emissions of dioxin and PCB could be shown except for the tests including PVC and computer scrap. The higher emissions in those later tests were probably due to the very high content of chlorine. For waste with lower chlorine content other factors, more difficult to determine, seems to have larger influence of the emissions.

The emissions from accidental fires at dump sites can be a very important source. It is of course highly variable depending on frequency, composition of the material at the site, depth and the extent of the fire as well as the method used to put out the fire.[7]

As the emissions from various point sources have decreased during the last decades, the relative contribution from secondary sources as diffuse emissions from contaminated sites and long range transport has increased.

There are several contaminated areas due to the former treatment of timber with PCP and other chlorophenols contaminated with dioxins. An unknown portion of the thus treated material might still be in use in the society today and there is a need to investigate the possibilities to identify and quantify this material, developing possible ways of remediation etc. Preliminary calculations show that the total amount of dioxins that could have been present in this process could be some 200 kg TEQ and that up to 30 kg TEQ might still be present in buildings and other constructions in Sweden[2].

Actions

The Swedish EPA realizes the need for more and more representative data on the current emissions of dioxins, PCB and hexachlorobenzene from primary industrial sources. Therefore industry will be encouraged to increase the number of measurements within the Intrinsic Control as stated in the Environmental Code and report to the responsible authorities. Further contributions are needed in order to get reliable data of the formation, emission and distribution of dioxin and other organic pollutants. Although present knowledge is scarce we can still propose the following actions:

· Technical developments are needed to decrease the emissions of dioxins and other organic pollutants and should always be considered in connection to the issue of operating permits and licensing review of different activities.

· There is also a great, but not acute need for better techniques to remediate contaminated sites.

· There is a big variance in the age and thereby quality of furnaces/stoves in Sweden today and hence the emission factor is very variable. It is therefore proposed to stimulate and thereby speed up the replacement of the out of date furnaces with modern, environmentally certified equipment.

· There is a need to give more and better information about what should and should not be burnt in individual furnaces and how the combustion should be carried out in order to achieve the highest efficiency.

· The supervision of landfill sites as well as the contributions in order to prevent fires in landfills should increase.

References

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