INVENTORY OF PCDD/F RELEASES TO AIR, LAND AND WATER IN TARRAGONA PROVINCE, SPAIN

Gerard Fuster[1,2], Marta Schuhmacher[1,2] and José Luis Domingo[1]

[1]Laboratory of Toxicology and Environmental Health, "Rovira i Virgili" University, San Lorenzo 21, 43201 Reus, Spain
[2]Environmental Engineering Laboratory, Department of Chemical Engineering, "Rovira i Virgili" University, Ctra. Salou s/n, 43005 Tarragona, Spain

Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are well known environmental pollutants. They may be found in all environmental media usually at low concentrations. Because PCDD/Fs are persistent compounds that can be moved around, the estimation of the extent of PCDD/F releases is rather difficult. Although a number of recent studies have reported inventories of emissions of these pollutants to air, little work has been done on PCDD/F releases to land or water[1,2].

The specific objectives of this study were the following: a) to prepare a global inventory of PCDD/F releases to air, land, and water from industrial and non-industrial sources in Tarragona Province (Catalonia, NE Spain) for 1999, and b) to compare these results with previous inventories undertaken in other countries.

Error! Bookmark not defined.**Methods and Materials**

Tarragona Province with a surface of approximately 6,259 km^2 and a population of 580,245 inhabitants, has an important industrial activity. A PVC facility, a perchloroethylene facility, a petrochemical complex, a municipal solid waste incinerator and a hazardous waste incinerator, among other important industrial facilities, are located in the Province.

The current study aimed on the industrial and non-industrial processes that have the potential to form PCDD/Fs. Some processes that may lead to releases of PCDD/Fs (although these pollutants are not directly formed in the process: the disposal of waste oils, for example) were also included. It should be taken into account that the potential adverse impact of PCDD/F releases depends on the treatment and final destination of the materials in which they are contained, as well as on the behavior of these contaminants in the environment.

In the first stage of this study, processes were screened to identify those which should be included in the area studied by employing criteria previously developed[3,4]. To estimate emissions to air primary information was used. Primary information is based on original engineering test reports utilizing adequate quality control procedures. When measurement data was not available, an extrapolation was necessary. Such extrapolation involves deriving emission factors, which are representative of the industrial class studied. An emission factor (EF) relates the mass of PCDD/F released per unit of activity level. Annual releases from a single source or combined sources can be estimated by multiplying the emission factor by the annual activity level as showed in this equation:

$E_{total} = E_{tested\ i} + (EF_i * A_i)_{untested}$

E_{total} is the annual PCDD/F emission from all sources in g I-TEQ/y
$E_{tested\ i}$ is the annual PCDD/F emission from all tested facilities in source class i in g I-TEQ/y
EF_i is the mean emission factor for the tested facilities in source class i in g I-TEQ/y
A_i is the activity level for untested facilities in source class i in kg/y

With respect to PCDD/F releases to land and water, data on the amounts of material discharged, as well as their contamination with PCDD/Fs, was obtained from the literature[4] and from discussions with experts from the Spanish industry. Spanish data were used whenever possible. However, when they were absent or incomplete, information was supplemented by relevant data from other countries. Anyhow, at present, data on PCDD/F releases to media other than air are very scant either in Spain or in other countries. The present study has therefore shown that for a number of countries with old and limited data sets, more contemporary data are needed in order to complete this global inventory.

Releases of PCDD/Fs are given in toxic equivalent quantities (I-TEQ) using the International Toxic Equivalent System (NATO-CCMS). Because PCDD/Fs are generated and are present at very small amounts, there may be processes other than those considered here which also lead to the formation of these compounds. Consequently, the current study could only to identify the major processes.

Error! Bookmark not defined.**Results and Discussion**
The inventory of PCDD/F releases for industrial and non-industrial processes in Tarragona Province is summarized in Table 1. Because few data were available to estimate these releases, values are uncertain and the ranges of possible releases are also given. Numerical values are presented when the data were sufficient to calculate a release. Due to the extreme scarcity of data on PCDD/F in releases to water, the potential for PCDD/F releases to this medium was classified as high (H), medium (M), low (L) and negligible. Those denoted as negligible corresponded to processes which probably had not any liquid discharge. In turn, the distribution of probability for global PCDD/F releases estimated by the Monte Carlo method is depicted in Figure 1.

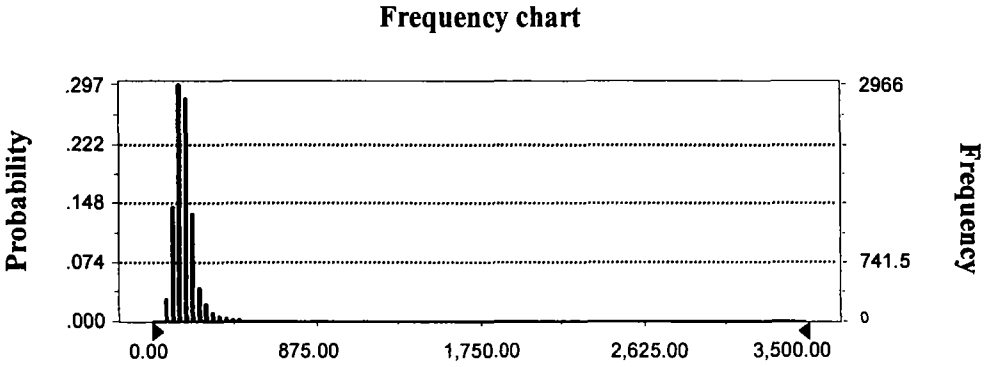

Fig. 1. Total PCDD/F release distribution for Tarragona Province in 1999

The global inventory releases in Tarragona Province for 1999 ranged from 131.9 to 259.4 g I-TEQ/y. The inventory shows a release to land in an rank between 130.0 and 254.5 g I-TEQ/y (98.6% of global releases), which is significantly higher than that to air estimated between 1.61 and 4.06 g I-TEQ/y (1.1% of global releases), or that for water estimated to be between 0.27 and 0.87 g I-TEQ/y (0.3% of global releases). It was not unexpected taking into account the nature of the processes that form PCDD/Fs, as well as the propensity of PCDD/Fs to bind tightly to solid materials. Emissions from cement kilns, diesel motor vehicles and MVC/PVC facilities are the most important contributors to PCDD/Fs emissions to air. Respect to releases to land, perchloroethylene production, accidental fires and pesticide production are the main contributors.

Table 1. Global Inventory of PCDD/F releases (g I-TEQ/y) in Tarragona Province (1999)

		Activity Level	Emission to air	Release to land	Potential release to water[d]
Municipal Solid Waste Incinerator		1.4E8 kg/y	0.0007-0.0013	2.69	M
Hazardous Waste Incinerator		3.0E7 kg/y	0.0018-0.0023	0.0006-0.204	0.00187-0.112
Oil Combustion Facilities [a]		3.5E8 kg/y	0.0013-0.013	NQ, but low	Negligible
		2.1E7 L/y	0.022-0.23	NQ, but low	Negligible
		1.4E8 kg/y[b]	0.0092-0.092	NQ, but low	Negligible
Motor	Leaded	1.0E9 km/y	0.015-0.15	NQ, but low	Negligible
	Unleaded	0.8E9 km/y	0.0004-0.0042	NQ, but low	Negligible
	Diesel	2.3E9 km/y	0.13-1.24	NQ, but low	Negligible
Cement kilns		1.5E6 tn clink/y	0.40-0.72	0.0000445 –1.33	Low
		1.7E6 tn clink/y	0.42-0.84	0.000051-1.55	Low
Accidental fires[c]		-	0.0027-0.027	1.73-35.81	0.0173-0.358
MVC/PVC facility		2.7E8 kg/y	0.0049-0.366	0.00-0.63	0.00050-0.00376
Crematoria		578 body/y	0.00009-0.0009	NQ	Negligible
Perchloroethylene production		4.0E7 kg/y	NQ	106-193	0.0215-0.123
Paper/Pulp Processes		7.7E8 kg/y	Negligible	0.0424-0.171	Medium
Pesticide production		4.4E4 kg waste/y	Negligible	0,130-29,7	0.000130-0.0297
Chlorine production		1.5E8 kg/y	NQ	0.993	Low
		2.1E8 kg/y	NQ	1.34	Low
Waste oil disposal		7.7E6 kg waste/y	Negligible	Not quantified	0.124-0.466
PCBs		252.7-379.1 kg PCB/y	NQ	0.0211-0.0317	Medium
Open use of chemical	PCP	3.1E7 kg wood/y	NQ	0.0712	High
	Other pesticide	-	NQ	0.01-0.06	H
Sewage sludge disposal		8.1E6 kg sludge/y	NQ	0.162-0.649	0.00474-0.0189
TOTAL			1.61-4.06	130.0-254.5	0.27-0.87[e]

[a] Fuel and natural gas were not included [1]. [b]This plant is 5 km out of Tarragona Province. [c] Accidental fires include those in buildings, outdoor fires and vehicle fires. [d] Releases denoted as high are thought to be greater than 0.01 g I-TEQ/y, medium between 0.01 and 0.001, and low below 0.001. [e]Summing up only numerical data. NQ= Not quantified

Table 2 shows the densities of PCDD/Fs in a number of countries. While the density of emissions to air in Tarragona Province (0.35 mg I-TEQ/km^2.y) is very similar to those of Germany (0.81) and USA (0.28), the density of releases to land (10.82) is relatively similar to that reported for Belgium (15.88) and the United Kingdom (17.38). With respect to the densities of potential releases to water, that found for Tarragona (0.09) is rather similar to that of Belgium (0.12).

Table 2. Densities of PCDD/F (mg I-TEQ/km^2.y) for a number of countries[1]

	Surface (km^2)	Year	emissions to air	releases to land	potential releases to water
France	543,965	1997	1.60	-	-
Belgium	30,528	1995	21.6	15.88	0.12
Germany	356,854	1994	0.81	-	-
UK	244,110	1997	1.80	17.38	-
USA	9,529,063	1995	0.28	0.021	0.002
Canada	9,970,610	1997	0.03	0.01	5.0E-7
Japan	377,835	1994	16.31	-	-
Australia	7,682,300	1998	0.07	-	-
Tarragona	6,259	1999	0.35	10.82	0.09

In summary, among the evaluated sources of PCDD/Fs in Tarragona Province, cement kiln factories followed by diesel motor vehicles, oil combustion facilities, and MVC/PVC factories were the main responsible for PCDD/F emissions in this area. On the other hand, the comparison of the density of these pollutants in this region is, in general terms, of the same order of magnitude than that reported for other industrialized countries.

References

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