

THE SPANISH DIOXIN INVENTORY: PROPOSAL AND PRELIMINARY RESULTS FROM MUNICIPAL WASTE INCINERATOR EMISSIONS

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I. Introduction

In 1.998, the Spanish Ministries of Environment and Industry supported CIEMAT (*Energetic, Environmental and Technological Research Center of the Ministry of Industry*) and CSIC (*Spanish Council for Scientific Research*) to elaborate a Spanish Dioxin and Furans Inventory from Industrial Sources.

The aim of this project is to evaluate the emissions into the atmosphere and water and to characterize the residues from industrial activities (fly ash, slag, etc.), to compare their relative contribution and the total release in the environment. The methodology of this National Programme has been established by this way :

- a) The emitting sources were classified according to the international well accepted criteria on formation of these types of compounds.
- b) The installations are being studied in terms of their thermal processes, gases filter systems , products, residues and waste water production to estimate a balance in the formation, of these compounds. The releases to land and water will be considered the main objective to reach.
- c) A review of data previously reported as described in reference 1.
- d) The activity rate and operational conditions and are considered in order to confirm the actual emission factors.
- e) In the next future major types of sources and industrial sectors such as sintering plants should be measured in order to complete the inventory.

In this paper we present a preliminary data of dioxin contents in samples from stack gas emissions reported by the MWI operators since 1997. Data about dioxins and furans levels presents in fly ashes and slags samples is also included.

II. Materials and Methods : The Municipal Waste Incineration Sector.

In Spain, the waste production was estimated in 15,200,000 Mg/yr (1997). The urban solid waste management combines different strategies such as recycling, landfill disposal, composting and combustion processes (Table 1). Therefore, the total material incinerated was around 1,140,000 Mg/yr, which represented 7.45% of the total Spanish waste.

Today, there are operating eight different municipal waste incinerators (MWI) : IRSUSA (Girona), REMESA (Melilla), SIRUSA (Tarragona), TERSA (Sant Adrià de Besós and Montcada) , TIRMADRID (Madrid), TIRME (Palma de Mallorca) y TVRM (Mataró). In a near future two new installations will be operating in the Northeast Region : SOGAMA in La Coruña

and ZABALGARBI in Bilbao. Most of the incineration facilities are mass burn travelling grate combustors with the exception of TIRMADRID AND SOGAMA which burn refuse derived fuel (RDF) in a fluidized bed. The system cleaning gas vary depending on the type of MWI even though semi-dry system combined with the used of carbon for dioxin removal is the most extended.

Table 1 : Urban solid waste management

<i>Disposal System</i>	<i>% Total</i>
Recycling	2.94
Uncontrolled Landfill	11.76
Controlled Landfill	62.18
Composting	15.68
Incineration	7.45

The incineration facilities are periodically controlled : monthly or every third or fourth months, principally from stack emissions of dioxins and furans by the Autonomic Environmental Authorities, so there are a large number of analysis of stack emissions, overall in SIRUSA, TIRMADRID, TIRME and MELILLA installations because of the great interest of their managers to improve their system cleaning gas and get down the levels of PCDD/Fs emitted. The majority of the analysis have been made by the CSIC-CID group, directed by Prof. Rivera, that has developed the methodology to analyze these type of contaminants in different matrices [1].

III. Results and Discussion

Air emissions : In this report we considered 78 stack gas analysis in the period January 1997-April 1999. Up to now all the plants fulfil the operations requirements in relation to temperature and residence time, and most of them observe the limit values for PCDD/Fs emissions (0.1 ng/Nm³). Table 3 shows the characteristics of the MWI revised and their ranges of emissions

Table 2: Characteristics of the MWI and their ranges of emission. Mean values Jan 97-April 99

MWI	Annual Capacity (Mg/yr)	Flue gas emission (Nm ³ /yr)	Range Emission and Mean value* (ng I-TEQ/ Nm ³)	Stack gas (mg I-TEQ/yr)
MWI1	273,902	1,280,000,000	0.003-0.007	3.84-8.96
MWI2	217,722	1,300,000,000	0.05- 0.1	65-130
MWI3	121,294	652,500,000	0.002-0.004	1.3-2.6
MWI4	31,000	200,000,000	0.03-0.7	6-140
MWI5	137,378	720,000,000	1.08*	777
MWI6	50,053	280,000,000	0.1*	28
MWI7	273,311	1,040,000,000	0.1*	104
MWI8	35,339	200,000,000	0.1*	20
TOTAL	1,140,000	4,502,500,000		1,005-1,210

In most cases, the levels of PCDDs/PCDFs were less than 0.1 ng I-TEQ/Nm³, only two plants which are performing the system cleaning gas exceed the limit value (0.7 ng/Nm³ and 1.08 ng/Nm³). Since 1996 a great effort has been made by all the incineration facilities to modernise their flue gas cleaning systems. Actually, all of them have flue gas cleaning systems composed of spray absorber (semi-dry) and fabric filter except Mataró's plant that works with spary absorber/electrostatic precipitator. In addition, some of them have activated coke addition systems. The modernisation could result in some cases in both, reducing production of PCDD/Fs in air emissions by increasing capture of PCDD/Fs formed, which may increase the burden in discharges in fly ashes and slags.

Solid residues : Fly Ashes and Slags. Table 3 shows their annual production. There are relatively few reported data about these types of residues because the installations are not obligated to control it. The fly ashes management follow two differnt ways: landfills or inmovilized in cement. In some cases the slags are recycled to construction materials. 42 analysis have been compiled from 5 MWI. Their mean values are : 3.5, 1, 0.37, 0.25 and 0.07 ng i-TEQ/g

Applying a conservative criteria in the estimations of the rest of incineration facilities, the total amount of PCDD/Fs in fly ashes are in the range of **111.6-46.6 g i-TEQ/y**

With regard to the slags, only three installations have made analysis. The mean values are 0.006, 0.013 and 0.098 ng i-TEQ/g. Estimating a maximum level of contents in the rest of the incinerator plants, a maximum total amount of **19 g** is expected by this way.

Table 3 : Production of fly ashes and slags

MWI	Annual production of fly ashes (Mg/y)	Annual production of slags (Mg/y)
MWI1	24000	60000
MWI2	26000	11000
MWI3	3500	29500
MWI4	750	8680
MWI5	6000	40000
MWI6	900	50053
MWI7	5000	69000
MWI8	714	7500
TOTAL	66864	275733

PCDD/Fs amounts present in the initial USWs have been recently analysed in order to examine the total dioxin output or mass balance in one of the incinerators. The levels are around 60-80 ng i-TEQ/kg USW [2] It appears according the levels found by other scientific groups. [3, 4, 5]. A mean of 70 ng i-TEQ/kg is considered to evaluate this preliminary balance. This quantity exceeds

the results of previous studies in other installation [6]. Considering this conservative result the Table 4 shows the preliminary dioxin mass balance in MWIs.

Table 4 : Preliminary PCDD/Fs mass balance in spanish incinerators (Mean values period Jan 1997-April 1999)

Mgs of USW incinerated (Mg/y)	PCDD/Fs in USW (ng i-TEQ/kg)	Total PCDD/Fs in USW incinerated (gr i-TEQ/y) INPUT	Total PCDD/Fs in Stack gases (g i-TEQ/y) OUTPUT	Total PCDD/Fs in Fly ashes (g i-TEQ/y) OUTPUT	Total PCDD/Fs in Slags (g i-TEQ/y) OUTPUT
1,140,000	70	79.8	1-1.2 (78 samples)	46.6-111.6 (42 samples)	2-19 (10 samples)

Air emissions have been drastically decreased with respect to the data reported by Rivera et al. [1], that established the emission into the atmosphere in around 20 g I-TEQ/y only in Catalonia. New data are considered representative of the emission levels with a tendency to be diminished when the gas cleaning systems will be optimised in the plants that are now in a test period. With regard to the ash flies and slags a measurement programme has been established to analyse more samples and evaluate the real contribution of these matrices to the balance. Also it is essential to improve the knowledge of PCDD/Fs in USWs in each MWI. The USW composition depends on the geografic situation, local industrial activities and the existence of a previous selective separation strategy.

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