DEVELOPMENT OF DIOXIN AND FURAN AIR EMISSION FACTORS FROM MUNICIPAL SOLID WASTE INCINERATORS IN REGIONE LOMBARDIA (ITALY)

Cristina Martines*, Roberto Fanelli**, Giuseppe Pastorelli***, Luigi Pistone****

* ASL Città di Milano, U.O. Chimica PMIP, Via F. Juvara 22, I-20129 Milano, Italy
** Istituto Ricerche Farmacologiche "Mario Negri", Via Eritrea 62, I-20157 Milano, Italy
*** Fondazione Lombardia per l'Ambiente, Foro Bonaparte 12, I-20121 Milano, Italy
**** AIDIC - Associazione Italiana di Ingegneria Chimica, P.le Morandi 2, I-20121 Milano, Italy

Introduction

Incineration of municipal solid waste (MSW), hospital waste and hazardous waste has for about 20 years been considered one of the most important sources of dioxin emissions and pollution. This is the reason why regulators have set in the past and are currently setting stricter and stricter air emission limit values (e.g., 0.1 ngI-TEQ Nm^{-3} in the European Union).
Polychlorinated dibenzo-*p*-dioxin (PCDD) and polychlorinated dibenzofuran (PCDF) air emission factors from MSW incinerators are commonly found in the technical-scientific literature from handful of sources. The U.S. EPA (1995, 1997) reports the following average values (expressed as sum of all PCDD and PCDF congeners, both toxic and non-toxic) for mass burn waterwall combustors (the most common type of MSW incineration device):

- 835 μg(PCDD+PCDF) $(tMSW)^{-1}$ before any control device;
- 585 μg(PCDD+PCDF) $(tMSW)^{-1}$ for plant equipped with electrostatic precipitators only;
- 311 μg(PCDD+PCDF) $(tMSW)^{-1}$ for plant equipped with spray dryers and electrostatic precipitators;
- 80 μg(PCDD+PCDF) $(tMSW)^{-1}$ for plant equipped with dry sorbent injection systems and fabric filters;
- 33 μg(PCDD+PCDF) $(tMSW)^{-1}$ for plant equipped with spray dryers and fabric filters.

The Danish EPA (1997) has obtained an average emission factor of 1300 μg(PCDD+PCDF) $(tMSW)^{-1}$ from an analysis of more than 200 flue gas samples collected after particulate matter control devices. Eduljee & Dyke (1996) used air emission factors of 184-231 μgI-TEQ $(tMSW)^{-1}$ for old incinerators and 0.6 μgI-TEQ $(tMSW)^{-1}$ for new plants for the development of the last PCDD/F inventory in the UK.
A widely accepted value of PCDD/F air emission factor still lacks for Italian MSW incinerators. Seven facilities located in Regione Lombardia (detailed in Table 1) were chosen in order to produce a first evaluation of field-based values useful for an Italian emission inventory not merely based on literature emission factors.

Table 1 - Characteristics of the seven MSW incinerators used to evaluate PCDD/F air emission factors (Beone *et al.*, 1998; De Stefanis *et al.*, 1998; Motawi, 1998).

Facility	Construction (last upgrading) year	Combustor	Energy recovery	Air pollution control devices	Nominal capacity [tMSW d^{-1}]	Nominal flue gas flowrate [Nm^3 h^{-1}]	MSW incinerated in 1997 [tMSW $year^{-1}$]	Operating days
1. Abbiategrasso (MI)	1970 (1985)	RK	no	ESP+WS	1×48	15,000	12,240	255
2. Bergamo	1965 (1987)	MG	yes	ESP+WS	2×75	49,800	46,500	300
3. Como	1967 (1988)	MG	yes	ESP+WS	1×100	42,000	39,560	n.a.
4. Desio (MI)	1976 (1990)	MG	yes	ESP+WS	2×120	70,000	64,678	302
5. Milano - Via Silla	1975 (1986)	MG	yes	ESP+WS	2×225	160,000	77,952	300
6. Milano - Via Zama	1968 (1990)	MG	yes	ESP+WS	2×240	220,000	86,261	300
7. Valmadrera (LC)	1981 (1988)	MG	yes	ESP+WS	2×120	90,000	77,650	293

Legend: RK = rotary kiln; MG = moving grate; ESP = electrostatic precipitator; WS = wet scrubber; n.a. = not available.

Materials and Methods

According to U.S. EPA (1995), an air emission factor is a representative value that attempts to relate the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant. PCDD/F emission factors from MSW incinerators are usually expressed as the weight of PCDD/F (expressed as μgI-TEQ) divided by the weight of MSW incinerated (expressed as tMSW). The data used for the evaluation are the PCDD/F concentrations in the flue gases sampled from seven MSW incinerators located in Regione Lombardia (Northern Italy) during the period 1991-1997. The data (obtained according to the Italian standard method, UNICHIM no. 825) were retrieved from the archive of the regional air pollution control laboratory (PMIP Milano). The original data were expressed as ng(PCDD+PCDF) Nm^{-3} (dry flue gas @ normal conditions - 273 K, 101.3 kPa - and 11%O_2), as prescribed by the Italian air emission control regulation in force until February 1998. Therefore a preliminary elaboration was needed in order to convert the original data into ngI-TEQ Nm^{-3} (dry flue gas @ normal conditions and 11%O_2) as prescribed by the new Italian MSW incinerators regulation and the common scientific use. The original database has been thoroughly analysed in order to apply the correct I-TEF. This preliminary elaboration has allowed to obtain a practical evaluation of the average value of the ratio of the emission concentrations expressed as total PCDD+PCDF and I-TEQ.
Since it was impossible to define (1) the actual weight of MSW incinerated during each sampling episode and (2) the actual, corrected flue gas flowrate (dry flue gas @ normal conditions and 11%O_2), it was decided to use a conventional, normalised flue gas factor (FGF) of 5.85 Nm^3 $(kgMSW)^{-1}$, corresponding to a stoichiometric air requirement (SAR) of 2.81 Nm^3 $(kgMSW)^{-1}$. These figures have been calculated considering a typical Northern Italian MSW having a lower heating value (LHV) of 9,450 kJ $(kgMSW)^{-1}$, that is 2,260 kcal $(kgMSW)^{-1}$, with the following composition (Cernuschi *et al.*, 1995):

- 53,15% combustible matter (obtained as sum of 28,09% C, 3,72% H, 21,18% O, 0,16% S);
- 23,16% inert matter;
- 23,69% moisture content (W).

Mathematical expressions used to calculate the LHV, the SAR and the normalised FGF are the following enthalpy and stoichiometric balance equations (1), (2) and (3):

$$LHV = 330.55 \cdot C + 1482.42 \cdot (H - O/8) + 93.16 \cdot S - 25.12 \cdot (W + 9 \cdot H) \quad (1)$$

$$SAR = 0.0897 \cdot C + 0.2692 \cdot (H - O/8) + 0.0337 \cdot S \quad (2)$$

$$FGF = (0.0187 \cdot C + 0.0070 \cdot S + 0.792 \cdot SAR)[(20.8 - 0.0)/(20.8 - 11.0)] \quad (3)$$

where the parameters C, H, O, S and W represent the composition of MSW as a w/w percentage.
Emission factors (EFs, expressed as μgI-TEQ $(tMSW)^{-1}$) have been then calculated using equation (4) as a function of the PCDD/F effluent concentration (CONC):

$$EF = CONC \cdot FGF \quad (4)$$

Results and Discussion

Both PCDD+PCDF and I-TEQ concentrations for each facility do not show any specific trend around the average value during the whole period 1991-1997; the deviation is generally lower using I-TEQ (CV 0,17-1,20 vs 0,08-1,47; see Table 2). Apart from the facility B (that is a rotary kiln incinerator), average I-TEQ concentrations are in very little range (0,76-1,71 ngI-TEQ Nm^{-3}). Therefore air emission factors can be evaluated in the range 4,4-10,0 μgI-TEQ $(tMSW)^{-1}$, with an average value of 6,8 μgI-TEQ $(tMSW)^{-1}$ (calculated excluding data from facility B that has an emission factor of 19,9 μgI-TEQ $(tMSW)^{-1}$). This average value can be reasonably used for Italian MSW incinerators built as moving grate combustors equipped with wet scrubbers and electrostatic precipitators (a very common technical solution).
Apart from facility A (characterised by an anomalous sample very rich of PCDF and poor of PCDD), the (PCDD+PCDF)/I-TEQ ratio shows slight deviation (CV lower than 0,71) around the average values of 36-53. These values are slight lower than the ones reported in the literature: 50 (U.S. EPA, 1997), 55 (Cernuschi *et al.*, 1995) and 67 (Danish EPA, 1997).
By using a (PCDD+PCDF)/I-TEQ ratio of 50 (as assumed by the U.S. EPA), it is possible to find that the emission factors here calculated are in the same range of those calculated by the U.S. EPA for MSW incinerators equipped with electrostatic precipitators (with or without spray dryers).
All the data analysed show that the current level of technology is able to comply with the old standard (4000 ng(PCDD+PCDF) Nm^{-3}) but not with the new one (0,1 ngI-TEQ Nm^{-3}). This will imply the use of combustion/pollution control technologies able to reduce the emission factor to values lower than 0,6 μgI-TEQ $(tMSW)^{-1}$ (an order of magnitude lower than the current level).

Table 2 - PCDD/F emissions from the seven MSW incinerators detailed in Table 1 (one of them has a sampling point in each of the two stacks) and (PCDD+PCDF)/I-TEQ ratio.

Facility	# of data	PCDD+PCDF [ng Nm^{-3}]					I-TEQ [ng Nm^{-3}]					(PCDD+PCDF)/I-TEQ [-]				
		Min	Max	Avg	SD	CV	Min	Max	Avg	SD	CV	Min	Max	Avg	SD	CV
A	9	14,5	413,8	89,3	124,7	1,40	0,33	3,36	1,05	0,93	0,88	22,5	866,8	146,4	273,2	1,87
B	7	26,6	865,8	203,3	298,3	1,47	0,67	12,15	3,41	4,09	1,20	30,6	71,3	52,7	16,6	0,31
C	6	15,4	48,8	34,3	11,3	0,33	0,47	3,48	1,27	1,10	0,86	12,1	70,9	36,7	20,7	0,56
D	5	5,6	166,2	72,0	61,2	0,85	0,44	4,84	1,61	1,83	1,14	12,7	111,4	51,9	36,8	0,71
E	4	8,2	85,2	43,5	37,7	0,87	0,32	1,87	1,12	0,76	0,68	13,1	47,6	36,4	15,9	0,44
F	4	5,4	76,8	44,1	29,4	0,67	0,36	1,62	0,82	0,56	0,67	14,8	84,4	52,3	29,3	0,56
G	3	35,7	41,7	38,7	3,0	0,08	0,66	0,91	0,76	0,13	0,17	46,0	58,8	51,5	6,6	0,13
H	3	40,7	64,4	52,4	11,9	0,23	1,03	1,86	1,44	0,41	0,29	34,6	39,5	36,9	2,4	0,07
-	41	5,4	865,8	83,3	143,0	1,72	0,32	12,15	1,54	2,01	1,30	12,1	866,7	68,0	130,7	1,92

Legend: Avg = average value; SD = standard deviation; CV = coefficient of variation (SD/Avg).

Acknowledgments

The research was carried out as one of the activities of the AIDIC Working Group on Dioxins. The authors wish to acknowledge Dr. Roberta Miglio for reviewing the paper.

References

1. Beone, G., Luciani, G., Luciani, M. (1998). *Banca dati termodistruttori italiani. 2° edizione su web*, http://wwwamb.casaccia.enea.it/Tein/moditerm/hometerm.htm, ENEA - Dipartimento Ambiente, Aprile 1998.
2. Cernuschi, S., Giugliano, M., Lonati, G., Ragazzi, M. (1995). Flussi di materiali e di energia per il bilancio ambientale. In: *Incenerimento di RSU e recupero di energia*, a cura di de Fraja Frangipane, E. & Giugliano, M.. Collana Ambiente 11, C.I.P.A. Editore, Milano, pp. 145-174.
3. Danish EPA (1997). *Dioxins - Sources, Levels and Exposures in Denmark*. Danish Environmental Protection Agency, Copenhagen, October 1997.
4. De Stefanis, P. *et al.* (1998). *La valorizzazione energetica in impianti dedicati. Documento di indirizzo.* Conferenza Nazionale Energia e Ambiente, Roma, 25-28 novembre 1998.
5. Eduljie, G.H., Dyke, P. (1996). An updated inventory of potential PCDD and PCDF emission sources in the UK. *The Science of the Total Environment*, **177**, 303-321.
6. Motawi, A. (1998). *Impianti di smaltimento: Analisi sui termocombustori RSU*. Federambiente, Roma, novembre 1998.
7. U.S. EPA (1995). *Compilation of Air Pollutant Emission Factors, Volume 1: Stationary Point and Area Sources, Fifth Edition, AP-42*. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards. Research Triangle Park, North Carolina.
8. U.S. EPA (1997). *Locating and estimating air emissions from sources of dioxins and furans*. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards. Research Triangle Park, North Carolina, EPA-454/R-97-003, May 1997.