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Inventory of U.S. 2012 dioxin emissions to atmosphere

Henri Dwyer, Nickolas J. Themelis *

Earth Engineering Center, Columbia University in the City of New York, United States

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ABSTRACT

In 2006, the U.S. EPA published an inventory of dioxin emissions for the U.S. covering the period from 1987–2000. This paper is an updated inventory of all U.S. dioxin emissions to the atmosphere in the year 2012. The sources of emissions of polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), collectively referred to in this paper as "dioxins", were separated into two classes: controlled industrial and open burning sources. Controlled source emissions decreased 95.5% from 14.0 kg TEQ in 1987 to 0.6 kg in 2012. Open burning source emissions increased from 2.3 kg TEQ in 1987 to 2.9 kg in 2012. The 2012 dioxin emissions from 53 U.S. waste-to-energy (WTE) power plants were compiled on the basis of detailed data obtained from the two major U.S. WTE companies, representing 84% of the total MSW combusted (27.4 million metric tons). The dioxin emissions of all U.S. WTE plants in 2012 were 3.4 g TEQ and represented 0.54% of the controlled industrial dioxin emissions, and 0.09% of all dioxin emissions from controlled and open burning sources.

1. Introduction

After the 1970 Clean Air Act, the U.S. started regulating emissions from all industrial plants. Following the Clean Air Act Amendments of 1990, the U.S. Environmental Protection Agency (EPA) promulgated the Maximum Achievable Control Technology (MACT) regulations for air pollutants, including dioxins. The MACT standards resulted in large reductions in toxic air emissions across all industries, by over 90% for most pollutants (US EPA, 2000). In particular, these standards resulted in significant emission reductions from municipal waste combustors (MWCs), the regulatory definition that EPA applies to both early municipal solid waste (MSW) incinerators and modern waste-to-energy facilities (WTE).

The first U.S. waste incinerator plant was built in 1885, in New York City, and hundreds more were operating by the middle of the 20th century. Those early plants were quite different from today's modern municipal solid waste (MSW) thermal treatment facilities, which produce steam and electricity, recover metals, and are generally referred to as Waste-to-Energy (WTE) power plants. Most of the current WTE capacity in the U.S. was built between 1980–1996. In the late nineties, the new EPA MACT requirements resulted in the closing of nearly forty of the older incinerators and the retrofitting of the remaining WTE plants with emission controls needed to meet the new MACT standards. As of 2014, there were 84 waste-to-energy facilities in 23 states, processing 27.4 million metric tons annually and generating over 14.5 billion kilowatt hours of electricity, corresponding to 0.36% of the 4 TWh U.S. total (Michaels, 2014).

This study examined the current level of dioxin emissions to the atmosphere from various U.S. sources and the relative contribution of the WTE industry to the total dioxin emissions. The generic term "dioxins" refers to 17 structurally related halogenated tricyclic aromatic hydrocarbons, 53 polychlorinated dibenzo-p-dioxins, and 54 polychlorinated dibenzofurans. Dioxins are persistent organic pollutants, released into the environment from several sources. The health effects due to exposure to dioxins, in particular one of the most toxic dioxin compound – 2,3,7,8-Tetrachlorodibenzo-p-diox in (TCDD) – have been studied extensively (US EPA, 2012). Dioxin toxicity equivalency factors (TEFs), are used to compare the toxicities of 16 other toxic dioxins to that of 2,3,7,8 TCDD and thus provide a total Toxic Equivalent (TEQ) amount for all 17 dioxin compounds. The TEFs used in this study are those provided by the World Health Organization (WHO, 1998) and are known as the WHO I-TEF. The total TEQ thus provides a universal basis to assess dioxin emissions from various sources.

In 2006, the U.S. EPA published an inventory of dioxin emissions for the U.S. covering the period from 1987 to 2000 (US EPA, 2006). This paper is a 2012 inventory of all dioxin emissions in the U.S.

* Corresponding author.
E-mail address: njt1@columbia.edu (N.J. Themelis).

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2. Methodology used

Emissions from most sources were calculated using the protocol provided by the Environmental Protection Agency in their reports on dioxin emissions. The approach used is based on an emissions factor that relates the "*mass of [P]CDDs/[P]CDFs released into the environment with some measure of activity (e.g., kilograms of material processed per year, vehicle miles traveled per year, etc.). It is developed by averaging the emission factors for the tested facilities or activities within the particular classification of sources*" (US EPA, 2013). Emissions are calculated by multiplying this "average emissions factor" by the activity level for that source.

Unless specified, the emission factors used in this report are the same as those published in the EPA report (US EPA, 2006) while dioxin quantities emitted are expressed in toxic equivalent grams (grams TEQ), as described in the previous section. Emissions are separated into two source categories: For controlled industrial sources, accurate emissions tests and activity levels were used, while for open burning sources, best emissions tests and estimates were used; of course, emissions from this second category are more uncertain.

2.1. Sources of information

The controlled sources of dioxins are grouped into five classes:

- **Waste to energy**.
- **Waste incineration**.
- **Electricity and heat generation**: fuel combustion for electricity generation, heating, and vehicles.
- **Metallurgical processes**: metal smelting, refining, and processing.
- **Cement and asphalt production**.

Other sources are grouped into a sixth class:

- **Open burning processes**: refer to minimally or non controlled combustion including, backyard barrel burning, agricultural burning, construction debris, yard waste and fires (forest, vehicle, landfill, building).

For each emissions class, the EPA 2000 activity levels were first recalculated in order to test the validity of the calculations. Classes with annual emissions of less than 2 g TEQ, for 1995 and 2000 (US EPA, 2013), were not included in the inventory.

The following sections describe the calculations for emissions directly linked to the management of municipal solid waste: waste to energy plants, landfill gas combustion, and landfill fires. For more information on other dioxin sources mentioned in this paper, the reader is referred to the Columbia University thesis by Dwyer (2014).

2.2. WTE plants

Dioxin emissions from WTE plants were calculated using the results of the 2012 stack tests, as reported to state environmental agencies, of all facilities operated by the two major U.S. WTE companies, Covanta Energy and Wheelabrator Technologies. All together, these plants represent 84% of the U.S. WTE capacity (Van Brunt, 2014; Porter, 2014; Shin, 2014). The other U.S. plants belong to various municipalities and were assumed to have the same average emissions per short ton of waste processed.

The U.S. EPA standard, or MACT limit, for *total* dioxin concentration is 30 ng per dry standard cubic meter (dscm) corrected to 7% oxygen (O_2) [ref. to 40 CFR 60 Subpart Cb], for units constructed prior to 2006. WTE facilities are required annually to measure total dioxin concentrations in the stack gas exhaust to demonstrate compliance with the MACT limit.

For this study, measured stack exhaust dioxin concentrations were converted to annual mass emissions based on the EPA estimate of volume of combustion components per unit of heat content (*F*-Factor) of 0.257 dry standard cubic meters (dscm) at 0% O_2 per MJ [ref. to 40 CFR 60 Appendix A, Method 19, Table 19-2]

Table 1
2012 dioxin emissions of 57 U.S. WTE plants. *Sources:* Van Brunt (2014) and Porter (2014).

Facility #	Total dioxin concentration (ng/dscm)	TEQ dioxins (ng/dscm)	Annual dioxin emissions (g TEQ)	Ratio of total dioxins to TEQ dioxins
1	1.7	0.0153	0.0474	111.1
2	1.1	0.0197	0.0635	56.8
3	4.0	0.0322	0.0221	124.2
4	0.9	0.0114	0.0280	75.5
5	10.9	0.1133	0.0640	96.2
6	2.7	0.0311	0.1288	88.0
7	0.6	0.0077	0.0099	79.3
8	6.3	0.0503	0.1175	125.6
9	5.1	0.0396	0.0226	128.0
10	2.7	0.0321	0.1573	83.4
11	2.8	0.0480	0.0315	57.5
12	1.2	0.0166	0.0732	72.8
13	0.9	0.0126	0.0282	67.4
14	7.7	0.0385	0.0289	200.8
15	2.4	0.0214	0.0279	114.2
16	2.5	0.0262	0.0182	94.8
17	1.6	0.0178	0.0320	91.4
18	0.7	0.0107	0.0147	69.8
19	1.8	0.0195	0.0159	92.3
20	0.4	0.0064	0.0066	60.3
21	1.8	0.0092	0.0178	199.0
22	8.2	0.0939	0.1552	87.7
23	0.8	0.0135	0.0299	60.5
24	2.3	0.0284	0.0969	82.1
25	1.3	0.0214	0.0152	60.7
26	0.9	0.0134	0.0130	64.4
27	0.9	0.0122	0.0159	74.3
28	2.4	0.0481	0.1933	49.7
29	0.7	0.0130	0.0179	55.7
30	10.0	0.0986	0.1722	101.1
31	11.6	0.2204	0.0793	52.6
32	1.3	0.0212	0.0224	60.6
33	0.7	0.0164	0.0126	42.9
34	0.6	0.0091	0.0069	68.8
35	0.7	0.0077	0.0079	90.9
36	0.0	0.0005	0.0007	43.5
37	1.5	0.0126	0.0200	117.9
38	2.0	0.0169	0.0246	119.1
39	1.4	0.0159	0.0586	84.7
40	2.3	0.0323	0.0752	70.2
41	1.6	0.021	0.0617	76.6
42	1.2	0.011	0.0316	114.3
43	1.3	0.017	0.0044	80.3
44	0.4	0.003	0.0020	116.7
45	1.0	0.017	0.0336	61.2
46	1.8	0.016	0.0123	112.5
47	6.0	0.045	0.0284	132.3
48	0.3	0.002	0.0013	125.0
49	3.1	0.022	0.0291	143.2
50	0.2	0.001	0.0021	143.5
51	1.4	0.014	0.0288	95.3
52	3.1	0.025	0.0743	126.1
53	12.0	0.115	0.3091	103.9
54	2.5	0.028	0.0557	90.1
55	8.8	0.062	0.1857	143.0
56	0.5	0.007	0.0075	71.4
57	0.8	0.007	0.0196	115.4
Total			2.86	
Average	2.73	0.030		92

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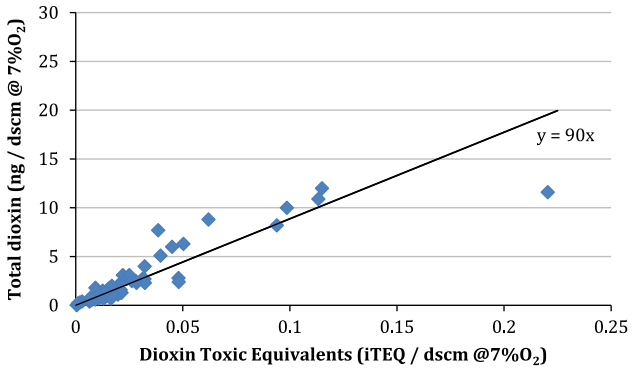

Fig. 1. Relationship of total dioxins to TEQ dioxins.

and the MSW higher heating value (HHV) of 11.63 MJ/kg MSW, using the following equation:

$$M_{\mathrm{iTEQ}} = C_{\mathrm{iTEQ}} F_{\mathrm{d}} \frac{20.9}{(20.9-7)} HHV_{\mathrm{MSW}} W$$

where:

M_{iTEQ} = Mass dioxin equivalent (iTEQ) emissions per year
C_{iTEQ} = Stack concentration of dioxin equivalent
F_{d} = Volume of combustion components per unit of heat content factor
HHV_{MSW} = Higher heating value of MSW
W = Mass of waste combusted at facility in 2012

and 20.9% is the concentration of oxygen in air.

The compilation of data in Table 1 shows that, in total, 2.86 g TEQ were emitted by 57 WTE plants that combusted 23.0 million tonnes of municipal solid waste (84% of the total MSW combusted in U.S. WTE facilities) in 2012. To estimate the emissions of all U.S. WTE plants, this number was scaled up to the total of 27.4 million tonnes combusted in all U.S. WTE plants (Shin, 2014), i.e. $2.86 \frac{27.4}{23.0} = 3.4$ g TEQ.

The ratio of *total dioxins* to *TEQ dioxins*, for each plant, is also shown in Table 1 and is plotted in Fig. 1. It can be seen that most of the data can be represented by the approximate relationship, 90 ng of total dioxins = 1 ng TEQ dioxins.

Table 2 shows the emissions for the entire WTE industry from 1987 to 2012.

Table 2
Dioxin emissions from WTE facilities, 1987–2012.

Emissions factor (calculated)
- 1987: 694 ng TEQ/kg of waste feed
- 1995: 40.3 ng TEQ/kg of waste feed
- 2000: 2.6 ng TEQ/kg of waste feed
- 2012: 0.1 ng TEQ/kg of waste feed

Activity levels
- 1987: 13.7 million metric tons
- 1995: 29.8 million metric tons
- 2000: 29.4 million metric tons
- 2012: 27.4 million metric tons

Releases
- 1987: 9510 g TEQ
- 1995: 1200 g TEQ
- 2000: 77 g TEQ
- 2012: 3.4 g TEQ

2.3. Landfill fires

According to a 2002 report by the U.S. Fire Administration (USFA) (TriData Corporation, 2002), landfill fires are primarily caused by spontaneous combustion of methane emitted from landfills and are a complex problem that has existed for decades. USFA reports that these fires can be difficult to extinguish and can last weeks before even being discovered. The number of landfill fires was estimated using the National Fire Incident Reporting System (NFIRS), a national system where fire departments send their statistics. This system is not mandatory, so not all fire departments use it, but. NFIRS considers it to be reasonably representative of the nation. This report estimates that only 30% of the landfill fires are being reported annually. In 2011, the number of fires in landfills reported to NFIRS was 3324, so the total fires were estimated at about 11,000. The NFIRS data (TriData Corporation, 2002) is available at the Federal Emergency Management Agency website (https://www.fema.gov/resource-document-library).

Using the EPA emission factor of 700 ng TEQ/kg burned and the average value of 225,000 kg burned per fire (Table 3), the estimated dioxin emissions from landfill fires in 2012 were: $11,000 \text{ fires} * 700 \frac{\text{ng TEQ}}{\text{kg}} * 225,000 \frac{\text{kg}}{\text{fire}} = 1733$ g TEQ. However, because of the uncertainty in the number of non-reported landfill fires, in the statistics following Table 3, the authors assumed that dioxin emissions from landfill fires in 2012 remained at the same level as in earlier years, i.e. 1300 g TEQ.

2.4. Landfill gas flaring

The EPA report "Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2012" (US EPA, 2014) reports methane emissions

Table 3
Dioxin emissions from landfill fires.

Emissions factor
- 1987: 700 ng TEQ_{DF}/kg burned
- 1995: 700 ng TEQ_{DF}/kg burned
- 2000: 700 ng TEQ_{DF}/kg burned
- 2012: 700 ng TEQ_{DF}/kg burned

Activity levels
- 1987: 1.9 million metric tons
- 1995: 1.9 million metric tons
- 2000: 1.9 million metric tons
- 2012: 2.5 million metric tons

Releases
- 1987: 1300 g TEQ
- 1995: 1300 g TEQ
- 2000: 1300 g TEQ
- 2012: 1733 g TEQ

Table 4
Dioxin emissions from flaring of landfill gas.

Emissions factor
- 1987: 1.4 ng TEQ_{DF}/m^3
- 1995: 1.4 ng TEQ_{DF}/m^3
- 2000: 1.4 ng TEQ_{DF}/m^3
- 2012: 1.4 ng TEQ_{DF}/m^3

Activity levels
- 1987: 1.35 Gm^3
- 1995: 4.7 Gm^3
- 2000: 16 Gm^3
- 2012: 11.4 Gm^3

Releases
- 1987: 2 g TEQ
- 1995: 7 g TEQ
- 2000: 22 g TEQ
- 2012: 16 g TEQ

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Table 5
Dioxin emissions from controlled sources (g TEQ/year).

Classes	Subclasses	1987	1995	2000	2012
WTE industry	Waste-to-energy (WTE) industry	9500	1200	77	3.4
	Category subtotal	9500	1200	77	3
Waste Incineration	Hazardous waste incineration	5	6	3	2
	Medical waste incineration	2700	510	400	7
	Sewage sludge incineration	6	14	10	0
	Category subtotal	2711	530	410	9
Electricity and heat generation	Gasoline fuel	55	6	7	8
	Diesel fuel	70	75	87	118
	Other oils fuel	25	20	22	11
	Wood fuel	114	97	106	92
	Coal combustion	109	112	114	85
	Category subtotal	373	310	336	314
Metallurgical processes	Primary magnesium smelting and refining	13	13	8	8
	Secondary aluminum smelting	11	20	8	8
	Secondary zinc production	4	7	7	8
	Secondary copper smelting	990	270	1	0
	Sinter plants	33	28	24	16
	Secondary ferrous metal smelting smelting/ refining	37	46	59	64
	Ferrous foundries	13	19	16	6
	Coke production	10	9	8	4
	Category subtotal	1111	412	131	114
Cement and asphalt production	Cement kilns burning hazardous wastes	120	160	19	19
	Cement kilns burning alternative fuels	13	17	17	18
	Asphalt mixing plants	5	5	5	3
	Category subtotal	138	182	41	40
Total of controlled sources		14,024	2789	1173	634

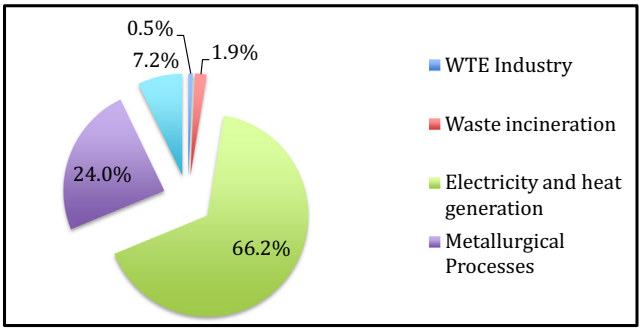

Fig. 2. Distribution of controlled-source U.S. dioxin emissions to air in 2012.

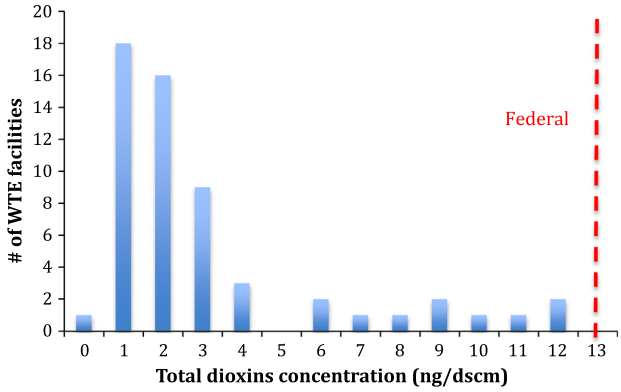

Fig. 3. Histogram of 2012 measured stack concentrations of total dioxins in WTE plants.

from landfills: 13,331 Gg (metric tons) of methane were produced in 2012, 4040 Gg of which were flared. The molar mass of methane is 16, so the volume of methane flared, at standard temperature and pressure, is:

$$4040\ \text{Gg} * 22.414 \frac{\text{L}}{\text{mol}} * \frac{1}{16 \frac{\text{g}}{\text{mol}}} = 5.7\ \text{billion m}^3$$

Landfill gas consists of about 50% methane and 50% CO_2, according to the U.S. EPA Landfill Methane Outreach Program. Thus, the total landfill gas volume flared in 2012 was estimated at 11.4 billion m^3. Using the EPA emissions factor of 1.4 ng TEQ/m^3, the dioxin emissions from landfill gas flaring in the U.S. were estimated at 16 g TEQ (Table 4).

3. Results and discussion

Table 5 summarizes the controlled industrial sources of dioxin emissions, divided into five classes, and the U.S. emissions for each source, at four reference years: 1987, 1995, 2000, and 2012. Table 6 does the same for the sixth class of all open burning sources of dioxin emissions. Since 1987, dioxin emissions from controlled sources have decreased over 95%. In contrast, the dioxin emissions from open burning processes have increased 43%. This increase is primarily due to a larger number of landfill and forest fires that, together, account for 77% of the open burning emissions and 93% of the increase. Part of this increase may be due to better reporting

Table 6
Dioxin emissions from non-controlled sources (g TEQ/year).

Classes	Subclasses	1987	1995	2000	2012	% of 2012
Fires and open burning processes	Combustion of landfill gas	2	7	22	16	0.55
	Building fires	24	18	16	15	0.52
	Vehicle fires (cars, etc.)	72	72	78	86	2.96
	Landfill fires	1300	1300	1300	1300	44.8
	Forest and brush fires	180	170	730	837	28.9
	Backyard burning	610	630	600	385	13.3
	Residential yard burning	4	5	5	6	0.21
	Land clearing debris burning	83	79	66	72	2.48
	Open burning of construction debris	22	22	22	53	1.83
	Agricultural burning (sugarcane, etc.)	28	31	162	131	4.52
Total open burning processes		2325	2334	3001	2901	100

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Table 7
WTE as a % of total dioxin emissions.

	1987	1995	2000	2012
Total controlled sources	14,024	2789	1173	634
WTE as % of controlled emissions	67.7	43.0	6.6	0.54
Total of all sources	16349	5123	4174	2901
WTE emissions as % of all sources	58.1	23.4	1.8	0.09

of landfill and forest fires in recent years and part to an actual increase of such fires due to higher temperatures or other factors.

The data of Table 5 are plotted in the form of a pie chart in Fig. 2.

Fig. 3 shows the municipal waste combustor total dioxin stack emissions, all of the plants were below the federal emissions limit of 13 ng/dscm for large new WTE plants.

Table 7 shows that over the period 1987–2012, the WTE dioxin emissions constantly decreased, from 67.7% of all the controlled sources to 0.54% in 2012. When both controlled and non-controlled sources are considered, the WTE contribution to the U.S. toxic dioxin emissions in 2012 was 0.09%.

4. Conclusions

The U.S. annual toxic dioxin emissions were examined over the period 1987 to 2012. Dioxin emission data were compiled for 57 waste-to-energy plants, located in eighteen states of the Union and representing 84% of the total U.S. WTE capacity. The average dioxin concentration of these plants was 0.029 ng TEQ/dscm equivalent (TEQ) per standard dry cubic meter of stack gas, i.e., only one third of the E.U. standard (0.1 ng TEQ/dscm) for WTE plants. The total amount of dioxins emitted by all U.S. WTE plants in 2012 was estimated at 3.3 g TEQ. The inventory of all U.S dioxin emissions was divided into controlled sources, which in the period of 1987–2012 were reduced from 14.0 kg TEQ to 0.6 kg; and open burning sources which have increased from 2.3 kg TEQ, in 1987, to 2.9 kg TEQ in 2012.

The study showed that, by 2012, the dioxin emissions of the U.S. WTE industry have been reduced to 0.54% of all controlled sources and 0.09% of both controlled and non-controlled sources. An estimated 89% of the U.S. total dioxin emissions is due to three major non-controlled sources: landfill fires, forest and brush fires, and backyard burning.

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