Manuscript prepared for Atmos. Chem. Phys.
with version 3.0 of the LaTeX class copernicus.cls.
Date: 28 January 2010

Supplementary Information for the article, entitled "The Municipal Solid Waste Landfill as a Source of Ozone-Depleting Substances in the United States and United Kingdom "

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Table 2 summarizes ratios of ozone-depleting substances (ODSs) and methane (CH_4) from three sources. The average ODS/CH_4 ratios from EPA (1995) for both total emissions and gas sampled from the gas collection system are listed. CFC-12 and CH_3CCl_3 both had relatively similar emission and gas collection system ratios, while the ratio of CFC-11 to CH_4 was slightly more than double in the gas collection system. The US Environmental Protection Agency model, LandGEM (version 3.02 from www.epa.gov/ttn/catc/products.html#software), predicted landfill emission ratios which were very similar to those detected in the EPA (1995) study and to our study. The largest difference between ratios detected in the gas collection system (EPA (1995) and this study) and emitted ratios (EPA (1995) and LandGEM) was for CFC-11. Table 2 provides evidence that the ratios measured in the gas collection system are similar to what we would expect in the emitted ratios for CFC-12, CFC-113, and CH_3CCl_3. We would expect our study to provide an upper estimate of CFC-11 landfill emissions.

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3 ODS Emissions Estimated with National Waste Statistics

To provide a comparison to the estimates in the main text of this paper, which were made using recovered ODS/CH_4 ratios, we regressed recovered ODS against national landfilled waste statistics. The waste statistics used for the United Kingdom (UK) and United States (US) correlate with recovered CH_4. Thus, we would expect the estimates made using recovered ODS/CH_4 ratios to be proportional. Unlike the national CH_4 statistics, the waste statistics do not take into account recovered landfill gas. Therefore, we would expect the estimates made using the waste statistics to be roughly equivalent to the maximum emission estimates made using recovered ODS/CH_4 ratios in the main text of this paper.

To calculate ODS emissions we used Equation 1, which is based on the same principle as Equation 3 in the main text,

$$ODS_(emitted) = \frac{ODS_{(recovered)}}{waste_{(on-site)}} \times waste_{(national)} \qquad (1)$$

where on-site waste is the waste landfilled at each individual sample landfill site and national waste is the total waste landfilled in the country. Both on-site and national waste statistics are summed over the years 1994-2005 in the US and 1997-2005 in the UK. These are the years for which both on-site and national landfilled waste statistics were available.

National US landfilled waste for 1994-2000, 2002, and 2004 was calculated using reported total annual US municipal solid waste (MSW) generation and percent MSW landfilled in Table 1 of Simmonds et al. (2006). Annual landfilled MSW in 2001 and 2003 were calculated as linear interpolations of the year before and after. For 2005, US population data was used to scale the Simmonds et al. (2006) 2004 estimate (U.S. Census Bureau, 2009). Reported MSW

in Simmonds et al. (2006) included total solid waste which is more than just MSW depending on how the states recorded their waste statistics. US on-site waste statistics are from the Massachusetts (MA) Department of Environmental Protection. The division of accepted waste for the MA landfills is shown in Table 1. National annual UK waste is the sum of reported landfilled municipal waste statistics from England, Scotland, Wales, and Northern Ireland (annual waste statistics are reported by the Department for Environment, Food, and Rural Affairs; Environment and Heritage Service, Northern Ireland; Scottish Environment Protection Agency, and the Welsh Assembly Government; see www.defra.gov.uk/environment/statistics/waste/kf/wrkf20.htm for a list of links). UK on-site waste statistics were provided from Viridor Waste Management, the owner and/or operator of all of the UK study sites, and include mainly commercial, industrial, and domestic waste.

The available waste statistics were often a mixture not only of MSW, but also of other solid waste streams, such as construction and demolition, industrial, etc. Thus, the resulting emission estimates shown in Table 3, should be considered as rough benchmarks only. Two of the US data sets, CFC-12 and CFC-113, had linear relationships with landfilled waste. If we compare Table 3 with the estimates in Tables 4 and 6 in the main text, we see that the ODS emissions estimated using the waste statistics are between the US best and maximum ODS emissions estimated using recovered ODS/CH_4 ratios and equal to the UK best estimates. This suggests that if there is any underestimation with the recovered ODS/CH_4 ratios, it is due to sample bias and not due to the regression methodology. This is also further evidence that the maximum estimates calculated from recovered ODS/CH_4 ratios are real upper limits and that the true emissions of CFC-12, CFC-11, CFC-113, and CH_3CCl_3 from US and UK landfills are below these values.

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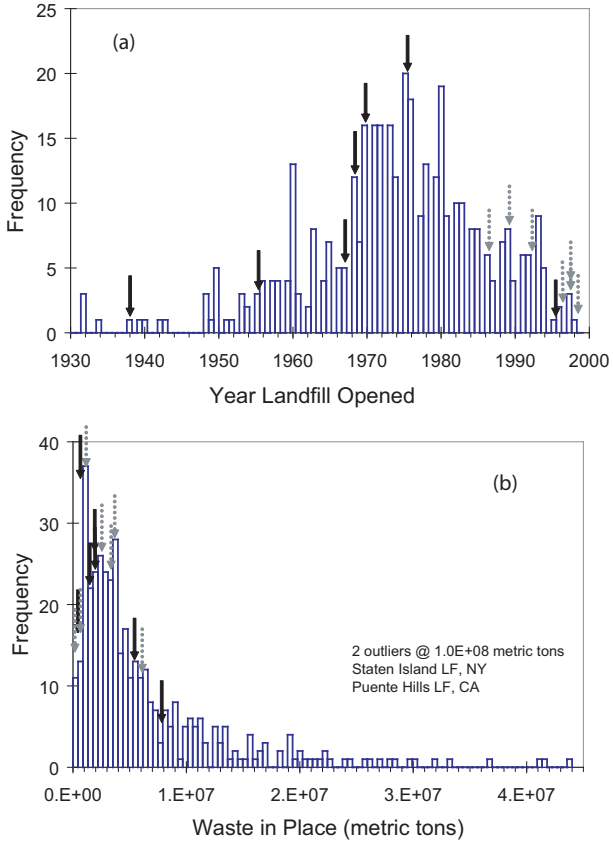

Fig. 1. Relative age (a) and total landfilled waste (b) of sampled landfills in the US (black solid arrows) and UK (gray dashed arrows) compared to an October, 2007, distribution of landfills in the US with operational gas-to-energy projects (LMOP, 2007).

Table 1. Demographic and waste statistics for the US and UK study sites

Landfill No.	Landfill Type	Year Closed	% Waste Type in Landfill: Dom[a]	I/C[a] MSW[b]	C&D[b]	Total Waste as of 1/1/06	Year opened	Megawatt capacity	Main Type of Gas Collection
			%	%	%	metric tons		MW	
US sites[c]									
1	municipal	open		80	17	5.1E+06	1967	5.7	reciprocating engine
2	municipal	open		94	6	1.1E+06	1995	3.2	reciprocating engine
3	corporate	open		71	28	1.9E+06	1968	1.0	reciprocating engine
4	municipal	open		82	8	7.5E+05	1969	0	flare
5	corporate	open		66	28	1.8E+06	1938	1.9	reciprocating engine
6	corporate	1998		75	4	7.4E+06	1975	5.6	reciprocating engine
7	municipal	1997		76	0	1.5E+06	1955	0.2	reciprocating engine
UK sites[d]									
1	corporate	open	52	44		3.0E+06	1973	2.1	reciprocating engine
2	corporate	open	67	32		1.3E+06	1998	2.4	reciprocating engine
3	corporate	open	76	18		2.2E+06	1997	2.8	reciprocating engine
4	corporate	open	60	24		1.3E+06	1982		reciprocating engine
5	corporate	open	63	35		5.5E+06	1982	5.1	reciprocating engine
6	corporate	open	52	27		2.9E+06	1992	2.2	reciprocating engine
7	corporate	open	78	14		5.7E+05	1996	1	reciprocating engine
8	corporate	2005	75	20		6.5E+05	1997	1	reciprocating engine
9	corporate	2001	65	33		3.3E+06	1986	1	reciprocating engine

[a]The two major waste types for the UK landfills are domestic, which includes household waste, and industrial and commercial waste (I/C). % waste type in landfill was calculated by averaging over the years between when the landfill opened and 2005.

[b]The two major waste types for the US landfills are municipal solid waste (MSW), which includes domestic and commercial waste, and construction and demolition waste (C&D). % waste type in landfill was calculated by averaging over the portion of the years 1994-2005 for which the landfill was open. Disaggregated waste data for the US landfills was not reliable previous to 1994 (DEP, 2007).

[c]US data on landfill type, operating status, waste type, total waste, and year opened was provided by the Massachusetts Department of Environmental Protection (MA DEP). The MA DEP maintains a large publicly available database on waste statistics which is updated annually through mandatory reporting by the individual landfills. US data on megawatt capacity was taken from the Landfill Methane Outreach Program (LMOP, 2007).

[d]All of the UK data was provided by Viridor Waste Management.

Table 2. Comparison of ODS vs. CH_4 ratios[a]

	EPA (1995)[b]			This study
ODS/CH_4	surface	gcs	LandGEM[c]	
CFC-11/CH_4	3.8E-06	8.8E-06	3.7E-06	5.9E-06
CFC-12/CH_4	1.4E-05	1.7E-05	1.7E-05	1.4E-05
CFC-113/CH_4	nd	nd	8.7E-07	9.3E-07
CH_3CCl_3/CH_4	2.0E-06	2.4E-06	2.0E-06	1.9E-06

[a] All ratios are unitless. nd = no data, surface = sum of surface flux emissions and passive vents, gcs = gas collection system

[b] Table 5-17 in EPA (1995)

[c] We provided the US Environmental Protection Agency emission model, LandGEM version 3.02, with waste data (Table 1 in the SI) and average CH_4 and ODS mole fractions (Table 1 in the main text) for all 5 open US landfills and corrected for air infiltration. The emissions were linearly regressed. The listed value is the resulting slope.

Table 3. 2006 US and UK MSW landfill emission estimates using national waste statistics for the extrapolation

Country/ Compound	2006 Estimate (Waste)[a]	% Uncertainty of Estimate[c]	% of Total Emissions (Mean)[b]
US/CFC-12	0.14	±55%	1.1%
US/CFC-113	0.0085	±95%	1.2%
UK/CFC-12	0.032	±70%	6.2%
UK/CFC-11	0.0084	±86%	1.2%

[a]Units are Gg y^{-1}

[b]Calculated like in Table 5 in the main text

[c]The uncertainty is the lower and upper 95% confidence interval. Only the regression error is included, because the waste statistics do not have error estimates.