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**MSW landfill ODS
emissions**

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The municipal solid waste landfill as a source of ozone-depleting substances in the United States and United Kingdom

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Abstract

This study provides observation-based national estimates of CFC-11, CFC-12, CFC-113, and 1,1,1-trichloroethane emissions for the United States (US) and United Kingdom (UK) from municipal solid waste (MSW) landfills. The scarcity of national estimates has led to the assumption that a significant fraction of the lingering ozone-depleting substance (ODS) emissions, which have been detected in industrialized countries, could be emitted from landfills. Spatial coverage was achieved through sampling at seven landfills in Massachusetts and through data provided by nine UK landfills. Linear least square regressions of recovered ODS vs. CH₄ were used in combination with national estimates of landfill CH₄ emissions to estimate 2006 national US and UK ODS landfill emissions. The ODS landfill emission estimates were then compared to recent estimates of total US and UK ODS emissions. US ODS landfill emissions were 0.4%–0.9% (0.006–0.09 Gg/year) of total US emissions. UK ODS landfill emission estimates were 1% (0.008 Gg/year) and 6% (0.03 Gg/year) of total UK CFC-11 and CFC-12 emissions, respectively. This indicates that landfills are only a minor source of lingering ODS emissions in the US, but may be more significant for CFC-12 emissions in the UK. The implications are that the majority of current ODS emissions in industrialized countries are likely coming from equipment still in use.

1 Introduction

The Montreal Protocol has had great success in reducing global emissions of ozone-depleting gases since it came into effect in industrialized countries in the mid-1990s. The rate of accumulation of ozone-depleting substances (ODSs) in the troposphere has declined to the point where tropospheric concentrations are now stable or decreasing (AGAGE, 2009). However, ODSs continue to be important to the study of stratospheric ozone recovery and to climate change. In particular, accurate predictions of future ODS emissions are needed to devise strategies which could minimize stratospheric ozone

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loss and greenhouse gas (GHG) emissions over the coming decades.

The four ODSs in this study are CFC-11 (trichlorofluoromethane), CFC-12 (dichlorodifluoromethane), CFC-113 (trichlorotrifluoroethane), and CH_3CCl_3 (1,1,1-trichloroethane). It is the combination of ozone depleting potential (ODP), the sheer volume historically released to the atmosphere, and their long lifetimes which make chlorofluorocarbons (CFCs) still highly important to the study of ozone recovery. CFC-11 and CFC-12 continue to have the highest global background concentration of any Montreal Protocol gas, and only HCFC-22 has surpassed CFC-113 (AGAGE, 2009). CH_3CCl_3 concentrations have decreased sharply in recent years so that it is no longer as important a factor in ozone recovery, but it remains important as a method of estimating OH mole fractions (Prinn et al., 2005; Bousquet et al., 2005; Wang et al., 2008). With regard to climate, CFCs have very high global warming potentials (GWPs), possessing ~5000–15 000 times the effect which the same amount of added CO_2 would have on warming the atmosphere over 100 years (WMO, 2007).

As the Montreal Protocol has come into effect and production of ODSs has decreased sharply, “banks” have become a significant source of future projected CFC emissions. Banks comprise products still in use, stockpiled products in case of equipment replacement, and discarded products in landfills. An increase in estimated CFC banks (i.e. by ~1100 Gg for CFC-11 and 700 Gg for CFC-12 for the year 2002) is considered to be a significant cause for the recent increase in projected ozone recovery time (5 years) compared to previous ozone recovery estimates (WMO, 2007; Daniel et al., 2007).

Landfills have the potential to be important emitters of banked ODSs. CFC-11, CFC-12, CFC-113, and CH_3CCl_3 have all been detected in landfill gas significantly above ambient concentrations (Rettenberger and Stegmann, 1996; Allen et al., 1997; Giess et al., 1999; Scheutz et al., 2008; and references therein). Major landfilled products include aerosol cans (propellant residue), refrigerant liquid, and foam blowing (mainly polystyrene) for CFC-12; aerosol cans, closed cell polyurethane foam (e.g. refrigeration insulation), soft foam plastics (e.g. furniture and mattresses) for CFC-11; sol-

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vent residues (e.g. dry-cleaning), foams, refrigerant, and propellant for CFC-113; and solvent residues (e.g. stain removers, detergents, adhesives, lacquers) for CH_3CCl_3 (Deipser et al., 1996; McCulloch and Midgley, 1998; McCulloch et al., 2001, 2003; IPCC/TEAP, 2005).

5 There have been significant advances in the understanding of processes controlling the magnitude and timing of CFC and CH_3CCl_3 landfill emissions, but more in-situ measurements are still needed. Several laboratory studies have investigated parameters important to the study of CFC landfill emissions. The size of shredded foam has been shown to have a large influence on the release time of CFC-11, which could delay landfill emissions by at least several decades (McCulloch et al., 2001; Kjeldsen and Jensen, 2001; Kjeldsen and Scheutz, 2003). All of the compounds in this study have been shown to degrade under simulated anaerobic landfill conditions (Deipser and Stegmann, 1997; Scheutz and Kjeldsen, 2003; Scheutz et al., 2007), but the effects in-situ are just beginning to be investigated (Scheutz et al., 2008). ODS concentrations have been measured in-situ and several countries have default national landfill concentration values for individual ODSs (Parker et al., 2002; EPA, 2008). There are fewer site-specific in-situ measurements of CFC and CH_3CCl_3 emissions (EPA, 1995; Allen et al., 1997; EPA, 2009b) and the United States Environmental Protection Agency maintains an online model which can estimate single landfill emissions for a range of ODSs (EPA, 2009c). Yet, as of the latest assessment report from the Intergovernmental Panel on Climate Change, there were no existing national inventory methods to quantify national ODS emissions from landfills (Bogner et al., 2007).

15 In this study, we estimate national emission rates from municipal solid waste (MSW) landfills for two industrialized countries with significant landfilling of waste, the United States (US) and United Kingdom (UK). National MSW landfill emissions are based on whole landfill mole fractions and flow rates, which were collected from US and UK landfills during the fall of 2006. The data was used to create linear regressions of ODS and methane (CH_4) recovery rates. These regressions, combined with national estimates of landfill CH_4 emissions, yielded national estimates of MSW landfill emissions

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of ODSs for the US and UK.

2 Methods

2.1 Design of study

5 Estimating emissions of CFC-11, CFC-12, CFC-113, and CH_3CCl_3 from landfills in industrialized countries required a targeted approach. Because any field sampling would only cover a fraction of the more than 1800 open landfills in 2006 in the US (Arsova et al., 2008), it was important to design the field study carefully to achieve the most unbiased estimates possible.

10 The study focused on estimating landfill ODS emissions for the US, with one-time field sampling at several US MSW landfills as well as monthly sampling for over one year at one US MSW landfill. Data was made available for several UK MSW landfills to provide a comparison with another industrialized country.

15 To narrow the focus of the study, we decided to sample or collect information from non-hazardous waste landfills which accept more than 50% of their waste from domestic and commercial sources (MSW landfills). In the US, MSW landfills account for the majority of total landfills and total generated landfill gas (EPA, 1994, 2009a,d). Within MSW landfills, sampling was constrained to landfills which pumped their landfill gas to the surface (active gas collection) to increase the number of measurements for statistical analysis.

20 In order to sample a representative cross section of US and UK landfills, we controlled for two important variables, namely landfill age and size, as shown in Fig. 1 in the Supplementary Information (SI) (see <http://www.atmos-chem-phys-discuss.net/9/22803/2009/acpd-9-22803-2009-supplement.pdf>). The intention was to sample a cross-section of small, medium, and large landfills covering a range of ages. On average, the US study sites in the state of Massachusetts were older and smaller than the
25 national average. The UK study sites were better at covering the range of sizes, but

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were younger than the US landfills.

2.2 Study sites

The sample sites consisted of seven US and nine UK MSW landfills with active gas management. The detailed demographic information for each landfill is shown in Table 1 in the SI. The UK sites are all owned by one landfill company, while the US sites are partly municipal and partly privately owned landfills. Two US (landfills 5 and 6) and two UK (landfills 8 and 9) landfills had reached capacity and no longer accepted waste at the time of sampling. The accounting of the waste type which is landfilled is slightly different for the US and UK. For the UK landfills, domestic and I/C (industrial and commercial) are the two main waste categories, while for the US landfills, MSW, which includes domestic and commercial waste, is the dominant category. For this study, US landfills with more than 50% MSW and UK landfills with more than 50% domestic waste were considered MSW landfills.

2.3 Measurement data

Table 1 presents data for each sample site. All of the mole fractions and flow rates are whole landfill averages, which were obtained by sampling at ports located on the main gas line directly before either the flare or the gas-to-energy plant. For the US sites, all of the data shown in Table 1 was used for the emission estimates. Multiple sample dates per site were averaged to yield one data point. For the UK sites, only the data from 2006 and 2007 was used for the emission estimates because of high intra-landfill variability in the CFC-11 mole fractions. UK data from 2004 is added as a comparison. CFC-12 mole fractions within each UK sample sites showed almost no intra-annual variability within landfill sites. Flow rates in Table 1 are yearly averages for 2006 and are the ones used in the subsequent analyses to calculate emissions.

A 1.5 year-long study (not shown) at US landfill 2 was undertaken during 2005–2006 to monitor the intra-annual variability of ODS, CH₄, and flow rates and to lend credibility

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to the following emission estimates, which used data from one-time field sampling. The standard deviation on the monthly-sampled landfill mole fractions was 19% (CFC-12), 29% (CFC-11), 23% (CFC-113), 12% (CH_3CCl_3), and 2.8% (CH_4), while the standard deviation for the flow rate was 12.5%.

2.4 US gas collection and analysis

Data collection for the US and UK landfill sites was considerably different. For each of the seven US sample sites, we collected and analyzed triplicate canister samples during the fall of 2006. The canister samples were collected in 0.8 L electropolished stainless steel cylinders filled in succession to 30 psig using a metal-bellows pump on a sampling line already flushed with the sample landfill gas. Three-fold evacuations to 100 mTorr of each sample canister as close as possible to the time of sampling ensured canister cleanliness. Selected canisters were analyzed for lingering traces of CH_4 to further prohibit cross-contamination between sampling.

The US gas samples were analyzed on an AGILENT 6890 gas chromatograph with both a flame ionization detector (FID) and a micro electron capture detector (μ -ECD). Samples analyzed for CFC-11, CFC-12, CFC-113 and CH_3CCl_3 were introduced by static fixed loop injection (50 μl , inject time=30 s), with separation on a 25M \times 0.32 mm ID CP Sil 5 CB capillary column followed by detection by μ ECD. The GC oven program was 45°C for 1 min, 10°C/min to 100°C, hold at 100°C for 0.5 min, 30°C/min to 200°C, and hold at 200°C for 1 min. CH_4 was analyzed by static fixed loop injection (10 μl , inject time=4.8 min) followed by separation on a 50M \times 0.32 mm i.d. Alumina (Al_2O_3) PLOT column (Na_2SO_4 modified) at an isothermal oven temperature of 45°C.

Samples were analyzed in triplicate and bracketed by calibration standards. The ODS standard (Scott specialty gases) contained CFC-11, CFC-12, CFC-113 and CH_3CCl_3 all at certified accuracy of less than 5%. CH_4 was calibrated using pure standard (research grade, 99.999% pure, BOC gases).

Calibration curves were constructed over the experimental range for the ODS mea-

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measurements and these were applied to analytical samples. The limits of quantitation (signal/noise=10) were 0.57 ppb for the ODSs and 1330 ppb for CH₄. All samples measured exceeded these values. Average measurement precision for the GC-μECD/FID was 0.95% (CFC-12), 0.90% (CFC-11), 2.5% (CFC-113), 2.9% (CH₃CCl₃) and 0.47% (CH₄). For more detail regarding the measurement methodologies, refer to Hodson (2008).

2.5 UK gas collection and analysis

CFC-11, CFC-12, and CH₄ mole fractions for the nine UK landfills were obtained from Viridor Waste Management. All of the data from Viridor had well documented sampling and analysis procedures, but had only one analysis per sample date. For the gas samples collected before May of 2007, Viridor collected the sample and CERAM (Stoke-on-Trent, Staffordshire, UK) performed the analysis. CERAM is a UKAS (United Kingdom Accreditation Service) ISO 17025:2005 accredited testing laboratory specializing in landfill gas analysis. Viridor filled one or more Tedlar bags using a GA2000 Infrared Gas Analyzer (Geotechnical Instruments, Chelmsford, England) fitted with a water filter. The bags were filled to ~3 cm thick and shipped to CERAM. At CERAM, the gas samples were analyzed using their UKAS in-house method BCRL-C51 and BCRL-C72 for the CFCs and CH₄, respectively. Both methods were accredited in 1999. All of the gas samples were analyzed on a GC-MS at most two weeks after sampling.

For the two UK samples analyzed in May of 2007, C&P Environmental (Bedford, Bedfordshire, UK) collected the samples and performed the analysis. C&P used a GA2000+ analyzer to fill and evacuate their Tedlar bags two or more times before filling the final sample. Two Tedlar bags were filled in case one leaked before the gas analysis. Bags were kept at ambient temperature in a dark, cool box during transportation after sampling and before analysis. Analysis was done one week after sampling on a GC-MS at C&P Environmental which is an UKAS accredited testing laboratory.

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2.6 Flow rates

For US landfills 1–6, instantaneous flow rates were provided by the landfill operators and extrapolated to yearly averages. A comparison at US landfill 2 between the built-in, continuous monitoring orifice plate (accuracy $\pm 0.6\%$ of flow, Oripac Model, Lambda Square Inc., Bay Shore, NY, USA) and a traverse with a Dwyer 166 T Telescoping Pitot Tube (Dwyer Instruments Inc, Michigan City, IN, USA) combined with a Dwyer 477A-1 Handheld Digital Manometer (scale 0"–20" water column, accuracy is 0.1% full scale) yielded flow rates within 4%. Landfills 1–6 all had similar built-in flow monitoring technologies. US landfill 7 had only a flare and no built-in flow monitoring technology. Thus, the flow rate for landfill 7 was measured with the Dwyer pitot tube and manometer.

For the UK, Viridor provided yearly averaged flow rates for each site. The flow rates were given in units of normal meters cubed per year (1 atm and 0°C). Viridor did not measure daily flow rates, but rather used a model to calculate annual emissions based on input parameter including landfill size, depth, age of waste, and type of landfilled waste. Estimated error on the annual flow projections is 10–20%.

It was necessary to standardize the US and UK data sets by calculating 2006 annual averages of flow rates for each landfill. The US instantaneous flow rates were converted into average annual flow rates using seasonal correction factors derived from the long term study at US landfill 2. Flow rates were averaged in the long term study for 30 days before and after the sample date of the flow rate to be corrected. The average ratio between the 60 days and running 12-month averages (calculated from 18 months of gas-to-energy plant data for US landfills 1–6 and 3 years of flare data for US landfill 7) was then applied to the data set to be corrected. No corrections were necessary for the annual averaged flow rates obtained for the UK landfills.

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2.7 Emission estimation methodology

The fate of landfill CH₄ and ODSs can be expressed by the following simplified equations

$$\text{CH}_{4(\text{emitted})} = \text{CH}_{4(\text{produced})} - R - \text{Ox} \quad (1)$$

$$\text{ODS}_{(\text{emitted})} = \text{ODS}_{(\text{volatilized})} - R - \text{Md} \quad (2)$$

where R=recovered CH₄ or ODS from flaring or gas-to-energy plants, Ox=CH₄ oxidized in the landfill soil cover, and Md=microbial degradation of the respective ODSs (Bogner et al., 2007; Scheutz et al., 2007, and references therein). Two further pathways important on longer time scales are lateral migration underground and internal changes in landfill gas storage (Bogner et al., 2007).

In this study, we measured recovered ODS and CH₄ gas fluxes and made use of available national inventory statistics for landfill CH₄ to scale the individual measurements to national landfill ODS emissions. This bootstrap method can be expressed as

$$\text{ODS}_{(\text{emitted})} = \frac{\text{ODS}_{(\text{recovered})}}{\text{CH}_{4(\text{recovered})}} \times \text{CH}_{4(\text{emitted})} \quad (3)$$

where all units are in mass/time. While this method allowed us make use of available inventory data and landfill gas technology, we assumed that each ODS/CH₄ ratio was the same in the recovered and emitted landfill gas. To check this assumption, we calculated ODS/CH₄ ratios for our five US open landfills using the US Environmental Protection Agency (EPA) landfill gas emissions model LandGEM (EPA, 2009c). We then compared the LandGEM emission ratios and the ratios from this study to surface emission ratios and gas collection system ratios, which were measured at Freshkills landfill in New York (EPA, 1995). Table 2 in the SI presents the results. The ratio of ODS/CH₄ was within 25% from all data sources for CFC-12, CFC-113, and CH₃CCl₃. CFC-11/CH₄ ratios were 130% higher in the gas collection system compared to the emitted gas (EPA, 1995). Likewise, the CFC-11/CH₄ ratio in this study was 60% higher

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than the predicted emissions from the LandGEM model. One reason for this difference in ratio variability may come from recent studies of anaerobic degradation. In simulated anaerobic landfill conditions, CFC-11 degrades relatively quickly compared to the other ODSs in this study (Scheutz and Kjeldsen, 2003; Scheutz et al., 2007). Moreover, a recent field study observed very constant soil concentration profiles to a depth of 80 cm for CFC-113 and CH_3CCl_3 (Scheutz et al., 2008). Taking the more conservative ratio, we would expect our CFC-11 emission estimates to overestimate emissions by approximately 130% and that our CFC-12, CFC-113, and CH_3CCl_3 estimates would be within 25% of their true values.

The national inventory statistics for emitted landfill CH_4 , which were used for this study, along with their confidence intervals are shown in Table 2. Both the US and UK estimates calculate landfill CH_4 emissions using a more complex version of Eq. (1) (Choudrie et al., 2008; EPA, 2009a). The best estimates in Table 2 include all of the parameters in Eq. (1). The maximum estimates do not subtract recovered CH_4 (R) or oxidized CH_4 (Ox). A second US CH_4 estimate from EIA (2008) was not included in Table 2 because it was only 5.7% higher than the EPA (2009a) estimate and did not provide either a maximum estimate or annually updated confidence intervals.

Several other bootstrap parameters besides total emitted landfilled CH_4 were considered such as total and annual landfilled waste, landfill age, megawatt capacity of gas-to-energy plants, and total landfill surface area. Estimates made using landfilled waste are presented in the SI. Their major disadvantage was that they did not allow subtraction of the recovered fraction of landfill gas. Landfill age did not have a linear relationship with ODS recovery rates and it was reported in a partially audited database (LMOP, 2009). It is possible that landfill age might be useful as a secondary parameter in a multiple linear regression, but the small sample size for this study did not support anything more complicated than using a single linear relationship for scaling to emission estimates. Megawatt capacity is also reported in the same partially audited database as landfill age. There is no national annual estimate of total landfill surface area.

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Scatter plots of recovered ODSs vs. CH_4 for all of the sample sites are shown in Fig. 1. The data is separated into four categories, including US, UK, open and closed landfills. The sampled US landfills emitted statistically significantly less CFC-12 per unit mass of CH_4 than the UK landfills. This could indicate either 1) UK landfills release CFC-12 faster than US landfills due to age, climate, engineering design, etc.; or 2) that the US and UK waste have more total CFC-12 landfilled mass at least in the sample sites. For CFC-11, the comparison between countries was less clear because of one highly concentrated US site. If we compare open and closed landfills, not all of the compounds had the same ODS/ CH_4 ratio between the two landfill types. In particular, CFC-113 and CH_3CCl_3 closed and open landfills could not be included in the same linear regression. Thus, for the following analysis, the US and UK data sets were analyzed separately. Each data set was used to provide quantitative estimates for its respective country only. Furthermore, except for the UK CFC-11 data set, only open landfills were used to develop the linear regressions used in the methane bootstrap method (Eq. 3).

The six linear regressions for each ODS and country combination are presented in Fig. 2. The regression method used in this study was weighted linear least squares, which is appropriate for predicting a dependent variable (recovered ODS) given an independent variable (recovered CH_4) (Isobe et al., 1990). The regression for UK CFC-11 was the only one which included closed landfills. Adding the closed landfills did not change the mean regression fit or the final mean emission estimates, but it did significantly reduce the regression uncertainty (p -value was reduced from 0.07 to 0.007). The linear regression for the US CFC-11 data set was fit by removing the outlier in Fig. 1, which was considerably outside the 99% confidence interval. A discussion of how this outlier may have affected the final results is included in the following section.

3 Results and discussion

The 2006 US and UK estimates of CFC-11, CFC-12, CFC-113, and CH_3CCl_3 landfill emissions are presented in Table 3. The best estimates used national CH_4 emission statistics which subtract recovered and oxidized CH_4 . The maximum estimates represent the total possible ODS landfill emissions, using the methodology in this study, if all landfill gas produced or volatilized in landfills were emitted to the atmosphere, and are considered to be an upper threshold to the estimates. The US CFC-11 best estimate (0.037 Ggy^{-1}) is significantly lower than the emissions expected from instantaneous release of decommissioned appliances (Kjeldsen and Scheutz, 2003). Moreover, the UK best estimates (0.008 and 0.03 Ggy^{-1}) are significantly lower than a previous order of magnitude estimate of 1 Ggy^{-1} for UK CFC landfill emissions in 1995 (Allen et al., 1997). For the US estimates, CFC-12 had the largest landfill mole fractions and emissions, followed by: $\text{CFC-11} > \text{CH}_3\text{CCl}_3 > \text{CFC-113}$. This ranking is consistent with default US concentrations (EPA, 2008) and some landfill studies (e.g. EPA, 1995; Allen et al., 1997). However, other landfill studies have detected larger relative concentrations of CH_3CCl_3 or CFC-11 (e.g. Deipser et al., 1996; Parker et al., 2002; Scheutz et al., 2008). The US and UK per capita emissions, calculated by dividing the Table 3 estimates by Census data (ONS, 2007; US Census Bureau, 2009), were statistically indistinguishable for CFC-11 (US= 0.12 , UK= $0.14 \text{ g person}^{-1} \text{ y}^{-1}$) and CFC-12 (US= 0.30 , UK= $0.53 \text{ g person}^{-1} \text{ y}^{-1}$) due to the estimate uncertainties. The UK CFC-12 emission estimates had the narrowest 95% confidence intervals (+44% and -36%) resulting from the comparatively constant ODS/ CH_4 ratios amongst all of the open UK landfill sites.

With our landfill emission estimates, it was possible to quantify the importance of landfills as a source of ODS emissions in the US and UK by comparing our estimates to total ODS emissions. Table 4 summarizes recent observation-based estimates of total US and UK ODS emissions. The UK CFC-12 and CFC-11 total emissions are averages of two estimation methods. The first estimation method uses mole fractions

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measurements taken at Mace Head, Ireland in combination with inverse modeling as described in Manning et al. (2003). The second estimation method calculates ODS/CO ratios using Mace Head ODS mole fractions and a model-derived CO time series, and then uses CO maps from the European Monitoring and Evaluation Programme to extrapolate to the UK scale. The estimates from both methods are within 10% (A. Manning, personal communication, 2007).

Landfill emission estimates from this study are plotted against the recent US and UK total ODS emission estimates in Fig. 3. Except for UK CFC-12, the landfill emission best estimates are ~ 2 orders of magnitude below the best estimates of total country emissions. This provides clear evidence that the US ODS and UK CFC-11 MSW landfill emissions are a small fraction of total ODS emissions. Moreover, we projected that the methodology used in this study could significantly overestimate CFC-11 landfill emissions.

Using the total country estimates (Table 4), we calculated source strengths for our landfill emission estimates in Table 5. The best estimates of US landfill emissions were below 1% of total US ODS emissions for each compound, with an upper limit (upper 95% confidence interval of the maximum estimate) between 1.6%–4.2%. UK CFC-11 landfill emissions (1.2%) were within the same range as the US landfill emissions. Only CFC-12 landfill emissions in the UK were estimated to be a potentially significant fraction of total UK CFC-12 emissions (6.3% with an upper limit of 30%). The highly linear correlation between recovered CFC-12 and CH_4 for the UK samples (Fig. 2) lends further credibility to the UK CFC-12 estimates. The difference between the best and maximum percentages was larger for the UK estimates because a larger fraction of landfill gas is recovered (Table 2).

ODS landfill emission estimates calculated using waste statistics as an alternative to landfill methane emissions are presented in the SI. The results were either equal to the best estimates in Table 3 (UK estimates) or between the best and maximum estimates (US estimates). Because the waste statistics do not account for gas recovery, this provides further evidence that the maximum estimates calculated using landfill

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methane statistics (Tables 3 and 5) are upper limits and that the true emissions of CFC-11, CFC-12, CFC-113, and CH_3CCl_3 from US and UK landfills are lower than our maximum estimates.

With a small sample size, one of the main concerns was that the data sets were not representative of landfill emissions in the US or UK. The UK regressions for CFC-11 and CFC-12 provided a useful way to check how a much higher ODS/ CH_4 ratio would affect US ODS landfill emission estimates (see Figs. 1 and 2). Using the higher UK ODS/ CH_4 ratios increased the best estimates of US CFC-11 and CFC-12 landfill emissions to 0.65% and 1.6% of total US emissions, respectively. The US maximum landfill source strength increased to 1.3% (CFC-11) and 3.3% (CFC-12). Likewise, if we used the US outlier ODS/ CH_4 ratio from Fig. 1, US CFC-11 emissions increased to 2% of total US CFC-11 emissions. Even these comparisons, which use $\sim 2\times$ higher CH_4 statistics and much different ODS/ CH_4 ratios, were still only a small fraction of total US emissions. This indicates that the US estimates are relatively robust to large changes in both the generated ODS/ CH_4 ratio and to the national CH_4 value used with the regressions. Large biases within the data would not change the overall conclusion that landfills are not a significant source of lingering ODS emissions in the US.

4 Conclusions

The results support the hypothesis that US and UK MSW landfills are not significant sources of CFC-11, CFC-12, CFC-113, and CH_3CCl_3 in the US and CFC-11 in the UK. US MSW landfills were estimated to emit all four MPGs at a rate of less than 1% of total US emissions. Even the upper limits, which take into account estimate uncertainty and assume emission of all generated landfill gas, still are small percentages of total US ODS emissions (1.6%–4.2%). CFC-11 emission estimates for the UK were approximately the same fraction of total UK CFC-11 and only UK CFC-12 emissions were estimated to be a higher fraction of total country emissions (6.3%). This should be further investigated in the future. The results were very robust to both the selec-

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tion of sampled actively-managed landfills and to the national statistics used for the extrapolation.

The conclusions from this study support the idea that continued emissions in industrialized countries of CFC-11, CFC-12, CFC-113, and CH_3CCl_3 are still coming from faster emitting sources such as installation, leakage, and destruction of ODS-containing material as described for the global scale in IPCC/TEAP (2005). However, the potential remains for increased banking of ODSs in landfills for industrialized countries where landfilling is still commonly practiced, such as the US or UK. Especially in the case of CFC-11, where a significant fraction is used in blown foams, landfills could become important as repositories of CFC-11-containing products in the future. If what was measured in this study is small because the lifetimes of ODSs within landfills are quite long, there could be a long tail of emissions from landfills which would last over many decades. It is unclear if landfill emissions have peaked or if the peak is yet to come. If landfill emissions do increase or even maintain their current emission levels in the coming decades, this could be important for stratospheric ozone recovery in polar regions, which is expected to recover to 1980s levels in the year 2065, ~16 years later than in the mid-latitudes (WMO, 2007).

Acknowledgements. The research for this study was funded by NASA grants NNX07AE89G and NAG512669 and NSF Grant ATM-0120468 with additional resources from the MIT Joint Program on the Science and Policy on Global Change. In addition, we are thankful to many individuals who assisted with our research: to the US landfill and gas plant operators and managers for their permission to sample and for providing the instantaneous flow rate data for each US landfill; to Brian Holdridge at the Massachusetts Department of Environmental Protection for the Massachusetts waste data; to Stuart James at Viridor Waste Management for providing the UK gas, flow rate, and waste data; and to Alistair Manning at the UK Met Office, whose research was funded by DECC UK government contract GA0201, for providing the total UK ODS emission estimates. Lastly, we would like to thank Stefan Reimann for providing helpful comments on earlier paper drafts.

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Table 1. US and UK landfill gas mole fractions and flow rates^a.

Site	Sample Date	N ^b	CFC-11 (ppb)	CFC-12 (ppb)	CFC-113 (ppb)	CH ₃ CCl ₃ (ppb)	CH ₄ (%)	Flow Rate ^c (Nm ³ y ⁻¹)
US sites								
1	20.11.06	9	1500	850	26	70.	47.3	2.88E7
2	2006 ^d	108	140	730	9.3	44	55.0	1.46E7
3	19.12.06	9	130	490	5.0	19	48.6	1.87E7
4	28.11.06	9	36	580	6.0	11	40.7	1.27E7
5	19.10.06	9	160	590	15	74	50.4	2.34E7
	18.12.06	9	130	600	20.	40.	45.5	2.34E7
6	31.10.06	9	25	410	3.7	4.9	45.4	2.59E7
7	20.11.06	9	24	200	4.7	7.2	46.0	1.26E6
UK sites ^e								
1	21.03.06	1	360	2300			52.8	1.68E7
2	2004	1		1800				1.48E7
	03.05.07	1		1800			47.8	1.48E7
3	10.01.07	1	300	2800			56.2	1.98E7
4	07.04.06	1		2800			40.4	6.61E7
5	23.04.04	1	320	2500				3.82E7
	03.05.07	1	530	2200			46.6	3.82E7
6	28.06.04	1	940	2300				1.12E7
	07.04.06	1	780	2200			49.6	1.12E7
7	07.04.06	1	590	2300			39.5	5.69E6
8	07.04.06	1	200	1200			51.3	5.95E6
9	21.03.06	1	250	1600			55.6	4.71E6

^a Units are parts per billion (ppb), % total gas stream by volume (%), and normal cubic meters per year (Nm³y⁻¹).

^b Number of gas analyses which were averaged to yield the mole fractions on the corresponding rows.

^c Represents yearly averaged flow rates for 2006 corrected for intra-annual variability.

^d US site 2 sampled monthly in 2006.

^e Blank spaces indicate that concentrations were below the UK analysis detection limit of 1 μg m⁻³.

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Table 2. 2006 US and UK national estimates of CH₄ emissions from landfills^a.

Country	Source	2006 Estimates		Confidence Intervals	
		Best	Max	Upper	Lower
US	EPA (2009a) ^b	6211	12 543	+ 33%	–39%
UK	Jackson et al. (2008) ^c	926.5	3309	+ 20%	–20%

^a Estimates are in Ggy^{–1}.

^b The maximum estimate adds the CH₄ which is oxidized and recovered (EPA, 2009a).

^c The maximum estimate is calculated by assuming the Jackson et al. (2008) CH₄ best estimate is 30% of potential UK CH₄ emissions (Choudrie et al., 2008). The upper and lower confidence intervals are from all CH₄ sources and are equal to $\pm 2 \times (\text{standard deviation}) / \text{mean} \%$ (~95% confidence interval) (Jackson et al., 2008).

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Table 3. 2006 US and UK MSW landfill emission estimates for CFC-11, CFC-12, CFC-113, and CH₃CCl₃.

Country	Compound	Estimate Mean ^a		
		Best (Ggy ⁻¹)	Max (Ggy ⁻¹)	95% CIs ^b Upper, Lower
US	CFC-11	0.037	0.074	+80%, -61%
US	CFC-12	0.089	0.18	+148%, -93%
US	CFC-113	0.0058	0.012	+148%, -93%
US	CH ₃ CCl ₃	0.012	0.024	+123%, -81%
UK	CFC-11	0.0082	0.027	+91%, -68%
UK	CFC-12	0.032	0.11	+44%, -36%

^a Calculated by using the corresponding Best and Max estimates in in Table 2 along with the regressions in Fig. 2.

^b CI=Confidence Interval. Includes the uncertainty in the regression estimates and the uncertainty in the landfill CH₄ emission estimates.

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Table 4. Recent observation-based best estimates of total US and UK emissions of CFC-11, CFC-12, CFC-113, and CH₃CCl₃^a.

Country/ Compound	Li et al. (2005) ^b	Hurst et al. (2006) ^c	Millet et al. (2009) ^d	Other
US/CFC-11	7.3	7.0	11	
US/CFC-12	16.4	14.2	8.8	
US/CFC-113	0.6	1.5	ND	
US/CH ₃ CCl ₃	2.2	3.8	2.8	3.7 ^e
UK/CFC-11				0.68 ^f
UK/CFC-12				0.51 ^f

^a All estimates are in Ggy⁻¹. ND=not detectable.

^b Combines measurements at Trinidad Head, California and Harvard Forest, Massachusetts as explained in Li et al. (2005). Data is averaged over the years 1999–2002 for CFC-12, CFC-11, and CFC-113 and 2001–2002 for CH₃CCl₃.

^c Calculated by multiplying 2003 emission rates derived from the COBRA-NA airplane campaign (Hurst et al., 2006) with a 2003 US population of 290, 796, 023 (US Census Bureau, 2009).

^d Combines recent airplane campaigns with a chemical transport model and represents an average of the years 2004–2006 (Millet et al., 2009).

^e US CH₃CCl₃ emissions estimated for 2002 from three urban field campaigns as described in Millet and Goldstein (2004).

^f UK emission are estimated for the year 2005/2006 (updated results from A. Manning (personal communication, 2007) are based on Manning et al., 2003).

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Table 5. US and UK landfill emissions as a percentage of total country emissions

Country/ Compound	% of Total	
	Best ^a mean (95% range)	Maximum ^b mean (95% range)
US/CFC-11	0.44 (0.17–0.79)%	0.88 (0.34–1.6)%
US/CFC-12	0.68 (0.05–1.7)%	1.4 (0.10–3.4)%
US/CFC-113 ^c	0.85 (0.06–2.1)%	1.7 (0.13–4.2)%
US/CH ₃ CCl ₃	0.37 (0.07–0.83)%	0.75 (0.14–1.7)%
UK/CFC-11	1.2 (0.39–2.3)%	4.0 (1.3–7.7)%
UK/CFC-12	6.3 (4.0–9.1)%	21 (13–30.)%

^a Mean estimates and 95% confidence intervals calculated by multiplying (best estimate of landfill ODS emissions from Table 3) $\times 100 \div$ (best estimate of US or UK total ODS emissions as shown in Table 4).

^b Mean estimates and 95% confidence intervals calculated by multiplying (maximum estimate of landfill ODS emissions from Table 3) $\times 100 \div$ (best estimate of US or UK total ODS emissions as shown in Table 4).

^c ND in Table 4 was assumed to be equal to zero.

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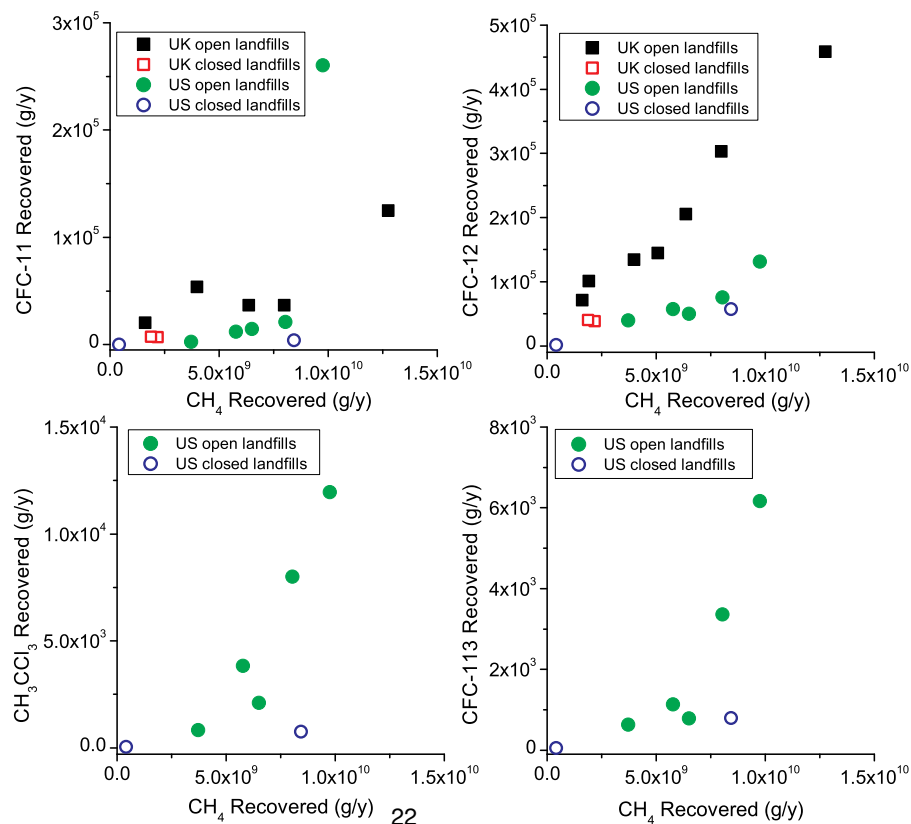


Fig. 1. Recovered CFC-11, CFC-12, CFC-113, and CH₃CCl₃ plotted against recovered CH₄ for all US and UK landfill sites.

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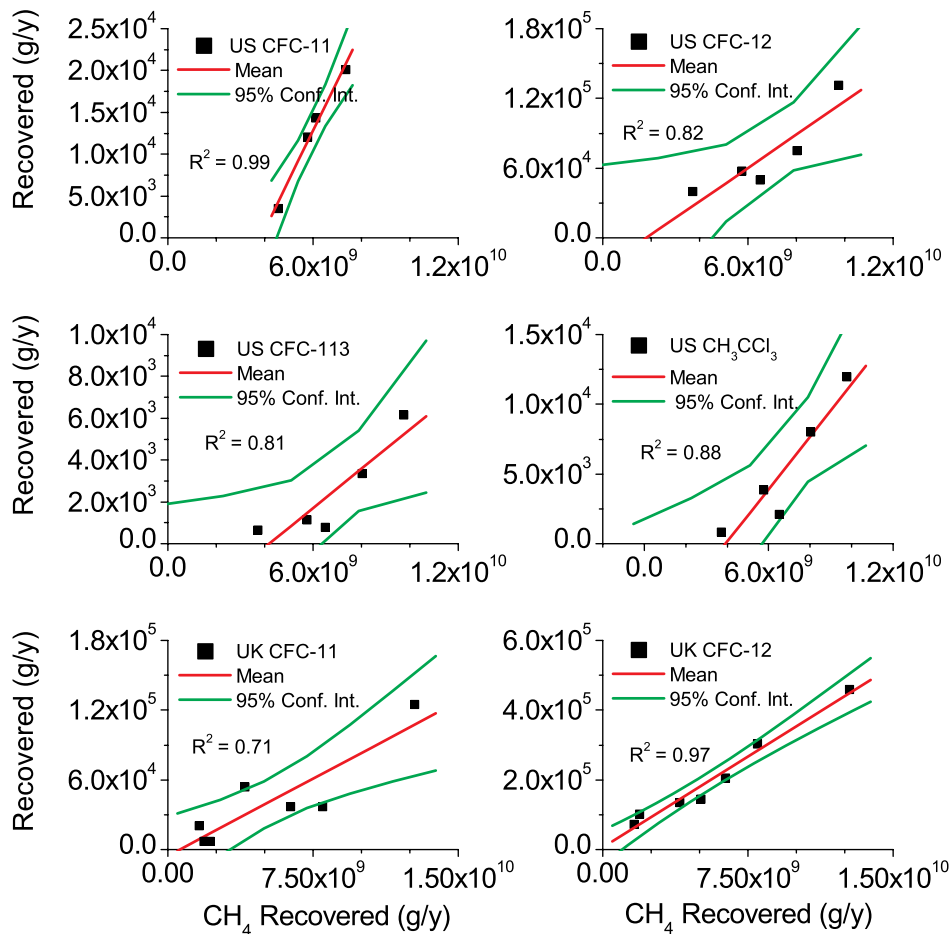


Fig. 2. Mean linear regression (red line), regression 95% confidence intervals (green lines), and regression fit (R^2) for each labeled country and ODS combination.

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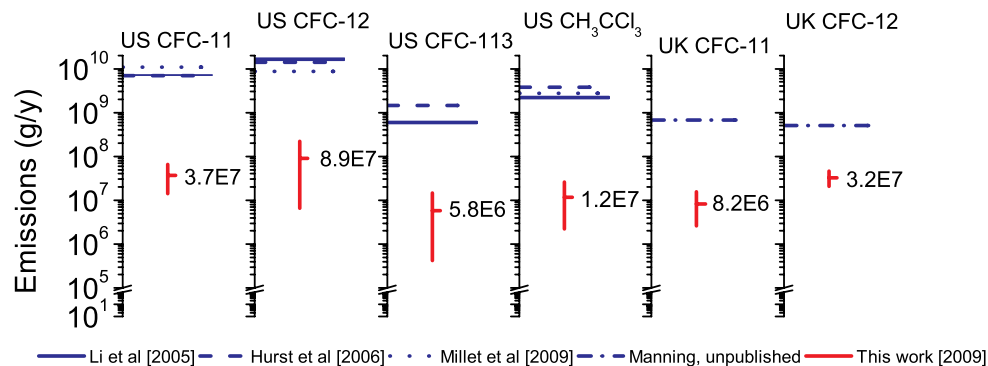


Fig. 3. The 2006 landfill emission estimates (vertical red lines) plotted against select total emission estimates from Table 4 (horizontal blue lines). The vertical red lines represent the mean and 95% confidence interval of the landfill ODS best estimates with the mean value explicitly labeled (Table 3).

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