

Airborne Emissions of Mercury from Municipal Landfill Operations: A Short-Term Measurement Study in Florida

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ABSTRACT

Large quantities of mercury (Hg) have been placed in municipal landfills from a wide array of sources, including fluorescent lights, batteries, electrical switches, thermometers, and general waste. Despite its known volatility, persistence, and toxicity in the environment, the fate of this Hg has not been widely studied. Using automated flux chambers and atmospheric sampling, we quantified the primary pathways of Hg vapor releases to the atmosphere at two municipal landfill operations in south Florida for eight days in April 1997. These pathways included landfill gas (LFG) releases from passive and active vent systems, passive emissions from landfill surface covers of different ages (including CH₄ "hot spots"), and emissions from daily activities at a working face (WF). Hg vapor was released to the atmosphere at readily detectable rates from all sources measured. Emission rates ranged from ~1 to 20 ng m⁻² hr⁻¹ over aged surface covers (generally comparable to background soils), from ~6 to 2400 ng/hr from LFG vents and flares, and from ~5 to 60 mg/hr at

the WF. In general the fluxes increased from older to newer landfills, from fresh to aged cover, and from passive to active venting systems. Limited data suggest that methyl- and other organo-mercury compounds may also be emitted from these sites, suggesting an important area for future research. We estimate that atmospheric Hg releases from municipal landfill operations in the state of Florida are on the order of 10 kg/yr, or <1% of the estimated total anthropogenic Hg releases to air in this region.

INTRODUCTION

Mercury (Hg) is a potentially toxic environmental pollutant that is among the most highly bioconcentrated trace metals in the human food chain, and several international committees have targeted Hg for special attention with regard to its emissions, cycling, and health effects.¹ As a result of health and environmental concerns associated with elevated levels of Hg observed in the edible tissues of certain species of fish in more than half of Florida's fresh water bodies, the Florida Department of Environmental Protection (DEP) has been striving to understand the inputs and mechanisms driving Hg cycling between the atmosphere and the surface as well as the bioaccumulation of Hg in various food chains. Recently, researchers have observed high to possibly fatal levels of Hg in predators at the top of the wetland food chains such as raccoons and the endangered Florida panther. A multi-year coordinated research program under way since the late 1980s has had some success in unraveling these facets of Florida's Hg problem.²

Municipal solid waste combustors (MWCs) and medical waste incinerators (MWIs) are significant anthropogenic sources of Hg in Florida.³ Hg emissions from the former are documented via annual stack testing, and are on the decline due to the reduction of Hg content of

IMPLICATIONS

Quantitative estimates of Hg emissions from municipal solid waste landfills based upon field measurements can enable solid waste managers, environmental regulators, and legislators to better evaluate existing waste management practices for common Hg-bearing wastes. Besides increasing our understanding of the Hg cycle, these measurements help agencies to evaluate the effectiveness of the following in reducing Hg releases: industry source reduction efforts, state recycling programs for Hg-bearing wastes, and landfill design requirements. Quantifying landfill emissions for comparison with Hg emissions from point sources such as waste combustors and electricity generating plants will assist in prioritizing resources needed to address the problem of anthropogenic Hg emissions.

Table 1a. Summary characteristics of landfill sites sampled in Florida during April 1997 (LFG = landfill gas).

Site Code	Name	Location	Class ^a	Status	Dates of Operation	Waste Mass (MT)	Surface Area (ha) ^b	LFG Collection	Dates Sampled
PB (83-90)	Dyer	Palm Beach Cty	I	Closed	1983-90	2,128,182	35.0	Active/FLARE ^d	4/16, 6/25
PB (90-)	Site 7	Palm Beach Cty	I	Operating	1990-	2,516,364	20.6	Active/FLARE ^d	4/16
MC (85-90)	Phase I, Cell 1-3	Martin County	I	Closed	1985-90	752,465	4.2	Passive/VENT ^d	4/18-23
MC (90-95)	Phase II, Cell 1	Martin County	I	Closing	1990-95 ^c	562,669	6.7	Passive/VENT ^d	4/18-23
MCWF (96-)	Phase II, Cell 3	Martin County	I	Operating	1996-	122,560	2.1	None	4/18-23
MCIII (90-)	Phase II Cell 4	Martin County	III	Operating	1990-	381,171	6.6	None	4/18-23

^aFlorida defines Class I waste as general, nonhazardous household, commercial, industrial, and agricultural waste. Class III waste is yard trash, construction and demolition debris, waste tires, asbestos, carpet, cardboard, paper, glass, plastic, furniture, and other wastes which are not putrescible and are not expected to produce leachate.

^bPB(90-) includes an active WF area of 836 m²; the proportions of the MC landfill areas characterized as top cover are as follows: MC(85-90) = 60%, MC(90-95) = 20%; MCWF(96) = 100%, MCIII(90-) = 3%. MCWF includes an active WF area of 418 m².

^cIntermediate cover in place, February–March 1997.

^dEstimated annual LFG collection rates (in m³) are PB(83-90) = 7×10^6 , PB(90-) = 11×10^6 , MC(85-90) = 6×10^3 , MC(90-95) = 18×10^3 .

manufactured products, front end removal, recycling of Hg-bearing wastes, and back-end Hg emission controls. Emissions from MWIs have also been documented and will soon be limited by federal regulations.⁴ However, Hg occurs in important quantities in both municipal and industrial waste, and these wastes are still being placed in municipal landfills. Prior to 1994, Hg levels in waste were estimated to be ~4 mg/kg, with >80% originating from alkaline batteries, but other important sources include fluorescent lights, switches, and paints.⁵ Because of its unique chemistry, Hg is subject to volatilization even at ambient temperatures, and its release from waste disposal processes such as agricultural application of municipal sludge have been documented.⁶ In addition, studies of Hg accumulation in mosses growing near municipal waste incinerators have suggested that somewhat increased levels exist near these sources.⁷

The goal of this study was to develop a preliminary assessment of Hg emissions from Florida's solid waste landfills, one of the least studied sources of Hg in the solid waste management system. Besides increasing our understanding of the Hg cycle, our measurements will assist agencies in evaluating the effectiveness of industry source reduction efforts, Florida's recycling programs for Hg-bearing wastes, and landfill design requirements in reducing the amount of Hg released to the environment. Because many active industrial atmospheric Hg sources have now been reduced, processes such as volatilization of Hg in waste previously and currently landfilled or otherwise placed in near-surface deposits increase in relative importance.⁸ However, until recently there have been no methods by which to quantify airborne emissions of Hg from large-scale surfaces, especially passive, gaseous releases. New methods are now

available that allow such measurements to be applied to the question of landfill Hg emissions.⁹

In April 1997, we performed an eight-day study of Hg emissions from several municipal landfills in two counties in south Florida. We report here the first known direct measurements of all potentially important emission pathways for release of Hg vapor to the air during various phases of routine landfill operations. Our methods allowed us to quantify total airborne Hg vapor (primarily the elemental form, designated Hg⁰) in surface soil emissions over several landfills of different age and cover material, from active and passive landfill gas (LFG) vents and wells, and from working face (WF) operations.

APPROACH

Sites

From April 16 to 23, 1997, we measured Hg concentrations in air and Hg fluxes from LFG vents and over cover material at the municipal landfill operations located in Martin County (MC) and Palm Beach County (PB), FL. All potential Hg sources were sampled at the MC site, while only those associated with active LFG collection and flaring (gas vents and LFG condensate) were sampled at the much larger PB site (because of scatter in the April data, LFG vents at PB were resampled in June 1997; LFG condensate was only sampled in June). Each landfill consisted of multiple sites of varying characteristics as described in Table 1. Background soils, such as those normally used to provide cover on the WF, were sampled for Hg content and Hg fluxes at a site ~200 m from the MC WF.

Methods

Hg fluxes over landfill surfaces were measured using a Teflon dynamic flux chamber (FC) system developed

Table 1b. Further characteristics of landfill cover.

Site Code	Cover Type	Top Detail	Side Detail
PB (83-90)	Permanent	30 cm sand; 20-mil PVC liner; 46-cm sand; 15-cm top soil amended with sewage sludge, grass	46-cm sand, 15-cm top soil amended with sewage sludge, grass
PB (90-)	Intermediate and Daily	15-cm MSW incinerator ash (appl. daily on WF), 30-cm top soil (intermediate on inactive areas)	15-cm sand, 15-cm top soil, grass
MC (85-90)	Permanent	30-cm soil, 46-cm compacted clay ($K_{max} = 1 \times 10^{-5}$ cm/sec); 61-cm erosion/vegetative support (soil); grass	30-cm soil, 61-cm erosion/vegetative support (dirt); grass
MC (90-95)	Intermediate	30-cm soil	30-cm soil
MCWF (96-)	Daily	15-cm soil	15-cm soil
MCIII (90-)	Daily	15-cm soil	15-cm soil

in our laboratory (see Reference 10 and illustration therein). An FEP-Teflon chamber was constructed at Oak Ridge National Laboratory (ORNL) with an open bottom and deep stretching skirts (dimensions of 60 ¥ 20 ¥ 20 cm) and was supported by an external aluminum frame. For each experiment, the airstream at the inlet and outlet port is sampled simultaneously for Hg⁰ using gold-coated sand absorbers (gold traps) at a constant flow rate of about 400 mL/min⁻¹. This chamber was built to facilitate simultaneous collection of three replicate air samples each from the inlet and outlet ports to better characterize the true mean concentrations of Hg entering and exiting the flux chamber. Air samples are drawn for periods of approximately 1–2 hr using a multiple replicate sampling system equipped with six separate mass flow controllers (MFCs).¹¹ Flow into and out of the chamber is also maintained at a constant flow rate of 5 L/min using a high-capacity MFC (corresponding to about 5-min turnover time for the internal volume of the FC). The flux of Hg⁰ from the surface covered by the FC is computed as follows:

$$F = \frac{(C_o - C_i) \times Q}{A} \quad (1)$$

where F is Hg⁰ flux in ng m⁻² hr⁻¹; C_i and C_o are the mean Hg⁰ concentrations in ng/m³ at the inlet and outlet ports; A is the bottom surface area of chamber in m²; and Q is the flushing flow rate through the chamber in m³/hr.

The blank levels of our FC are routinely measured by sealing the chamber over a large, clean sheet of FEP Teflon place on a table in the field. Measurements of field chamber blanks and intercomparisons between different sampling approaches have been published.^{6,10,12,15} We have found that consistent storage of the chamber in background air reduces the blank levels, and tests during this study showed very small blanks (see below).

Hg vapor in air is collected in the field on reusable gold traps that are blanked by heating to 450 °C prior to use, then sealed with Teflon plugs. Immediately following field operations, the traps are analyzed in our laboratory by cold vapor atomic fluorescence spectrometry (CVAFS). The CVAFS system is calibrated using gas-tight syringe samples of a Hg⁰-saturated atmosphere maintained in a constant temperature bath. This procedure yields very high precision.⁹ The detection limit for Hg⁰ in air using a 1.5-hr sampling time is ~0.05 ng/m³ based on typical gold trap blanks (absolute DL = 2pg). All handling and analysis of gold traps is done in class 100 laminar flow clean air benches.

Given the excellent capabilities of CVAFS, we found that the limitation on precise Hg⁰ determination lies largely with sample collection and handling. With our sampling and modified CVAFS system we can consistently sample Hg⁰ in air at background levels (sub pptv) using 1–2-hr sampling period with an overall precision of ≤3%. Another requirement is that adjacent samplers result in comparable concentration data. We extensively tested our MFC samplers for biases between adjacent sampling systems and found none which were significant.¹¹

In addition to our manual gold trap sampling procedure, the FC was operated in an automated mode, utilizing a commercially available Tekran mercury analyzer to quantify air concentrations at the inlet and outlet of the FC. This approach is a substantial improvement over our manual method as it provides continuous, near-real-time (20-min) fluxes. A Tekran 2537A was operated in normal 5-min sampling mode to sample two lines via a dual-solenoid switching unit controlled by the analyzer to provide alternating paired samples from the FC inlet and outlet (e.g., two inlet samples followed by two outlet samples.). The Tekran Automated Dual Sampling System (TADS) reduced possible sampling biases resulting from the two alternating gold

amalgamation traps used in the Tekran for collection of Hg°. The Tekran was also employed to monitor air concentrations 50-m downwind of the WF at the MC site over a 24-hr period, during which time over 400 metric tons (MT) of waste were delivered, spread, compacted and buried.

Hg concentrations in LFG samples were measured using both activated, iodated charcoal traps and gold traps, each type operated in tandem mode using a primary and backup trap to assess possible breakthrough due to elevated levels of reduced S and organic gases present in LFG. Both trap designs were employed in replicate and for different sampling durations to assure detection of Hg in short-term samples (~2–15 min) because levels of Hg in LFG had not been previously published. Samples of LFG were collected at flow rates of ~400 mL/min from two types of systems: active gas collection systems with flares (pumped systems, PB only) and passive gas collection wells (open flow systems, MC only). Flares were sampled between the flame arrestor and the open flame under a slight positive pressure via stainless steel ports. All traps were heated slightly above LFG temperatures (~45–55 °C) to eliminate condensation. Total LFG flow rates from the sampled flares were provided by continuous recording flowmeters. The LFG wells at MC were sampled similarly, but traps were situated directly in the open mouth of each well, extending 20 cm into the “goose-neck” well pipe opening to prevent dilution with ambient air. Total LFG flow rates from MC wells were estimated from measurements of face velocity through a known area using a directed flow pipe. Total and methyl Hg were also measured in LFG condensates at the PB site (collected from wells by local landfill personnel), while total Hg was measured in nearby background soils and soils used for landfill cover at the MC site. These samples were analyzed by CVAFS after digestion/extraction procedures performed in a commercial laboratory.¹³

Table 2. Mercury in soils at the Martin County (MC) landfill site.

Site Code	Description	Soil T	Soil Total Hg (ng/g)	Comments
MCWF (96-)	aged working face	25	39.6	6 weeks old
MC (85-90)	grass, top	20	178	hot spot ^a
MC (85-90)	grass, side slope	25	61.9	grass covered
MC	control soil 1	23	28.9	grass covered
MC	control soil 2	26	62.7	grass covered
MC (96-)	side slope 1	36	31.2	hot spot ^a
MC (96-)	side slope 2	36	369	hot spot ^a
MC (96-)	top	20	35.1	bare soil
MCIII (90-)	top	26	146	bare soil

^a Hot spots of CH₄ releases were identified by dead vegetation (see text).

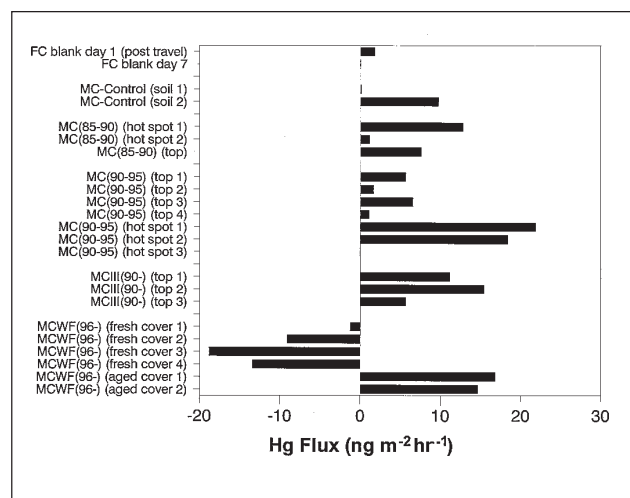


Figure 1. Measured Hg fluxes over landfill surfaces at the Martin County site using the ORNL Teflon flux chamber (FC). By convention, negative fluxes indicate deposition to the surface, positive indicates emission. WF = working face. Sites are defined in the text and in Table 1.

RESULTS AND DISCUSSION

Surface Emissions from Landfill Covers

Hg fluxes were measured over landfill covers of different composition, age, and permeability at four separate landfills at the MC site (Table 1). Where possible, fluxes were measured for several hours over multiple locations on landfill top cover and side slopes, including so-called LFG “hot spots.” Hot spots were identified from patches of vegetation damaged or killed by exposure to elevated levels of CH₄. We also sampled Hg emissions through a ~0.15-m-thick cover of fresh (<1 hr post application) and aged (~40 day) bare soil at the MC Class I landfill WF. Concentrations of total Hg in these cover soils were generally comparable to background soils (~30–60 ng/g), and indicated no significant surface accumulation of mercury from landfill operations with the possible exception of one hot spot (Table 2).

The mean surface emission fluxes of Hg measured at the MC site were generally comparable to those measured elsewhere over background soils, and suggest that this pathway was not a significant source of Hg loss at this landfill. Many of the fluxes measured over landfill surfaces were in the same range as those measured over the MC control soils, and some were not significantly above our mean FC blank values (Figure 1). Landfill surface fluxes were also highly variable across the range of sites sampled, and several interesting trends can be discerned from this limited data set (Figure 1). Areas of dead vegetation on the side slopes associated with elevated CH₄ emissions¹⁴ are apparently also conduits for Hg release, and were characterized by the highest surface fluxes measured (~18–22 ng m⁻² hr⁻¹). Fluxes over most of the top landfill surfaces were low, suggesting

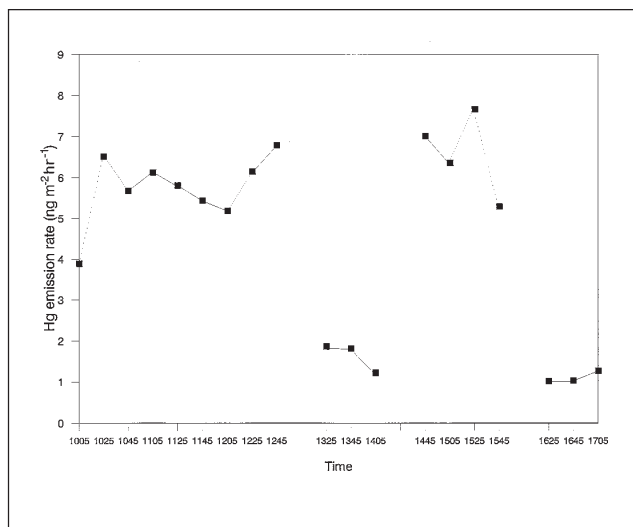


Figure 2. Temporal trend in Hg emission over the intermediate bare soil cover on the Martin County closing landfill site [MC(90-95); site closed 1996]. Data shown for several adjacent measurement plots.

that the cap designs described in Table 1 are effective barriers. For example, the fluxes measured over the dirt cover of the recently closed Class I site (MC 90-95) were small and relatively stable over a 4-hr period (Figure 2). Emissions over the older and newer landfills were comparable, and those over the Class III waste ($\sim 11 \text{ ng m}^{-2} \text{ hr}^{-1}$) were moderately higher than those over the Class I sites ($\sim 4\text{--}7 \text{ ng m}^{-2} \text{ hr}^{-1}$). However, it must be emphasized that none significantly exceeded the fluxes over MC control soils ($\sim 5 \text{ ng m}^{-2} \text{ hr}^{-1}$) or background soils in Tennessee (mean $\sim 7 \text{ ng m}^{-2} \text{ hr}^{-1}$ for forest soils, $\sim 10\text{--}40 \text{ ng m}^{-2} \text{ hr}^{-1}$ for field soils).^{12,15} There was no apparent relationship between total Hg in surface cover and Hg emission rates ($r^2 = 0.08$, $n = 9$). Although we measured fluxes only during daytime, our earlier studies over soils indicated strong relationships between Hg flux, solar radiation and temperature, with peak fluxes occurring at midday and decreasing to near zero at night.^{6,9} Because the surface fluxes reported here were measured during midday, and under moderately elevated temperatures, they may represent daily maxima.

Fresh undisturbed dirt cover (referred to as daily cover) effectively eliminates the surface emission of Hg from the newly buried underlying wastes, and fluxes over fresh cover consistently revealed uptake of Hg from air to soil (-1 to $-19 \text{ ng m}^{-2} \text{ hr}^{-1}$). The fresh cover was applied to the WF as a 15-cm layer ~ 1 hr prior to sampling, and was characterized by a much higher moisture content than aged cover. Airborne Hg is readily absorbed by wet soils, as we have noted elsewhere,¹⁵ and any volatile Hg in the buried waste must first re-equilibrate with the soil gas in this fresh cover before emission can occur. Fluxes measured over aged

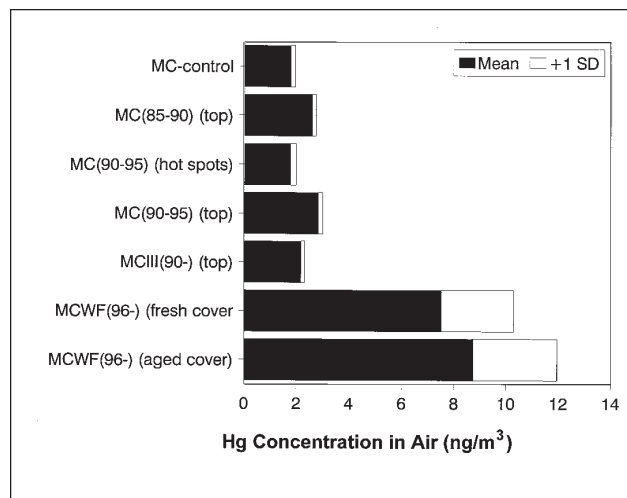


Figure 3. Mean Hg concentrations in air measured over landfill surfaces in Martin County (site codes are in Table 1).

(~ 40 day) and dried cover on an adjacent WF plot revealed significant Hg emission ($\sim 15\text{--}17 \text{ ng m}^{-2} \text{ hr}^{-1}$), suggesting that the effectiveness of the daily cover as a barrier against Hg release is temporary. However, in this limited study, we were unable to assess extended temporal trends in surface fluxes or to determine controlling factors. We were also unable to assess the influence of cover type (e.g., PVC liner vs. clay cap) on Hg emissions with these limited measurements. We observed that vehicular activity on fresh cover disrupted the continuity of the surface and exposed underlying wastes. Such disturbed cover would be a less effective barrier.

The Hg levels measured in the air over these surfaces during our FC studies were generally near continental background (e.g., $\sim 1.5\text{--}2.5 \text{ ng/m}^3$),¹⁵ with the

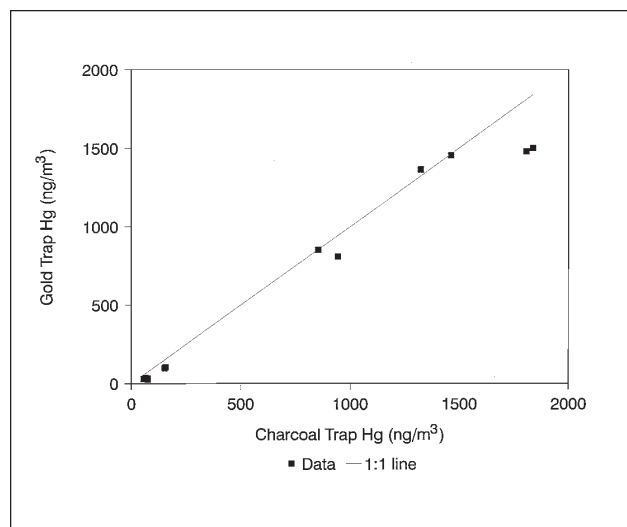


Figure 4. Relationship between Hg concentrations measured in LFG flares and wells using gold and charcoal traps. Data are from all sites (the points farthest from the 1:1 line were collected at the PB-7 site).

interesting exception of one location (Figure 3). The concentrations of Hg measured in air at the landfill WF (~7 ng/m³) were significantly elevated over the other sites, confirming this area of operations to be a potentially important source of Hg emissions, as discussed below.

Emissions from LFG Vents

Concentrations of total gaseous Hg in LFG ranged over nearly three orders of magnitude, from ~30 to ~1800 ng/

m³, well above ambient air (Table 3). All concentrations are expressed in m³ at STP as measured using MFCs. Since the exact composition of the LFG was not available, the data are not normalized to a 100% CH₄/CO₂ mixture). Samples collected sequentially over 60 min showed small temporal variations ($\pm 20\%$), as did samples from the same well collected four days apart (replicate samples showed a precision of ~5%). Despite the presence of complex organics and reduced sulfur gases in the LFG (as an indicator, the Cu screens in

Table 3. Summary of LFG analyses for Hg at Palm Beach and Martin County landfill sites, April 1997. All concentrations are expressed at STP on an as-measured basis (see text). We received limited data on LFG composition from Palm Beach following our study (CH₄, CO₂, N₂, O₂, in % by volume for the PB 90- flare were as follows on November 4, 1997: 54, 24, 14, 4.5; on December 6, 1996: 59, 27, 12, 1.4; during April 1997 the monthly average CH₄ concentration was 58%. Data source: Palm Beach County Solid Waste Authority, North County, Solid Waste Disposal Facility Landfill Gas Management System: Annual Operational Report, 1997 and 1996). Similar data were not available from the MC site.

Site	Date Sampled (day)	Time	Gas flow (m ³ /hr) ^a	Hg Conc. Charcoal (ng/m ³)	Hg Conc. Gold trap (ng/m ³)	Hg Conc. Difference (ng/m ³)	Charcoal Breakthrough (%)	Gold Trap Breakthrough (%)
Palm Beach County								
North flare (PB 90-)	4/16/97	1300	1309	1810	1480	330	0%	nd
North flare (PB 90-)	4/16/97	1330	1309	1840	1500	340	0%	nd
Dyer flare ^a (PB 83-90)	4/16/97	1430	833	90.7 ^b	4.72 ^b	- ^b	425% ^b	nd
Dyer flare ^a (PB 83-90)	4/16/97	1445	832	18.7 ^b	0.65 ^a	- ^b	50% ^b	nd
Dyer flare (PB 83-90)	6/25/97	900	800	48.3	32.9	15	0%	0%
Dyer flare (PB 83-90)	6/25/97	910	800	40.6	36.0	5	0%	0%
Dyer flare (PB 83-90)	6/25/97	930	800	27.2			0%	
Dyer flare (PB 83-90)	6/25/97	950	800	36.5				0%
Martin County								
Closed landfill (MC 85-90):	4/17/97							
Well 1	4/17/97	1330	0.27	74.2	31.5	43	0%	nd
Well 1	4/17/97	1400	0.27	74.5	25.8	49	0%	nd
Well 2	4/17/97	1500	0.04	151	97.8	53	0%	nd
Well 2	4/17/97	1515	0.04	156	104	52	17%	nd
Well 9	4/17/97	1615	0.11	55.1	29.1	26	204% ^c	nd
Well 9	4/17/97	1630	0.11	64.2	34.9	29	0%	nd
Closing landfill: (MC 90-95)	4/18/97							
Well 11	4/18/97	1515	0.06	945	809	136	0%	nd
Well 11	4/18/97	1530	0.06	905			6%	
Well 12	4/18/97	1430	0.77	1404			- ^e	
Well 12	4/18/97	1415	0.77	1325	1364	-39	0%	nd
Well 12	4/18/97	1745	0.77	1464			3%	
Well 12 ^c	4/22/97	913	0.77		1407			0.4%
Well 12 ^c	4/22/97	913	0.77		1501			0%
Well 14	4/22/97	954	0.24		343			0.3%
Well 15	4/18/97	1615	0.15	855	851	4	5%	nd
Well 16	4/18/97	1700	0.44	180			0%	
Well 16	4/18/97	1715	0.44	166			0%	

^a All gas volumes are expressed at STP as measured with MFCs. The exact composition of LFG samples and proportions of constituents (CH₄/CO₂/O₂/N₂) are unknown, and concentrations are expressed on an as-measured basis.

^aData from April 16 were compromised by excessive condensation due to heater failure, which is why the site was resampled on June 25, 1997.

^bWe suspect this apparent breakthrough was actually a contaminated backup trap.

^cWell 12 data from April 22 were side-by-side replicates.

^dCharcoal trap was loaded backwards, and both portions were summed to get concentration.

Table 4. Concentrations of total Hg and MeHg in LFG condensates at the Palm Beach County, FL, landfill sites.

Sample ^a	Total Hg (ng/L)	MeHg (ng/L)	MeHg/Total Hg %
PB (83-90) DDW	0.3	<0.013	
PB (83-90) bailer rinse	1.5	<0.013	
PB (83-90) cond 1	18.0	0.34	1.9
PB (83-90) cond 1-r	18.8		
PB (83-90) cond 1-r	18.1		
PB (83-90) cond 2	16.9	0.37	2.1
PB (83-90) cond 2-r	17.0		
PB (83-90) cond 2-r	17.5		
Mean (±SD) ^b	16.2±0.7	0.35±0.02	2.0
PB (90-) DDW	0.4	<0.013	
PB (90-) bailer rinse	0.9	<0.013	
PB (90-) cond 1	5100	23.1	0.45
PB (90-) cond 1-r	5090		
PB (90-) cond 2	4330	21.2	0.49
PB (90-) cond 2-r	4430		
Mean (±SD) ^b	4740±16	22.2	0.47

^aDDW = distilled deionized rinse water; bailer rinse = DDW flush of sampling bailer; cond = condensate water (1 or 2 indicates a replicate sample, r indicates a replicate analysis).

^bThese statistics are for blank corrected (using the bailer rinse) concentrations.

our charcoal traps turned a brilliant red upon exposure), we believe these Hg concentrations are accurate. Despite earlier reports of breakthrough during LFG sampling,¹⁶ we found little evidence of Hg breakthrough using either tandem gold or charcoal Hg traps (median breakthrough 0%, ≤6% in 24 of 26 tests). An earlier unpublished study reported Hg levels in LFG from sites in Minnesota ranging from ~250 to ~1800 ng/m³, but the investigators stated that these charcoal trap samples were compromised by breakthrough.¹⁶ A study at the Fresh Kills, NY, landfill reported Hg levels in LFG from ~6 to 20 µg/m³.¹⁷

Concentrations measured by both charcoal and gold traps agreed well, within 10% in most cases (Figure 4). Since charcoal traps collect both inorganic and organic forms of gaseous Hg, while gold traps collect primarily Hg^o, higher concentrations in charcoal compared to gold traps could be interpreted to represent gaseous organic Hg, most likely mono and dimethyl Hg. Although concentrations determined by difference are subject to large uncertainties, they were quite consistent at two of the sites: ~300 ng/m³ in the operating landfill flare at PB (PB 90-), and ~30–50 ng/m³ at the closed Class I landfill at MC (MC 85-90). Further research would be necessary to confirm the identity and concentration of these “other” gaseous Hg species (as discussed below, analyses of the LFG condensates indicated that MeHg was present). We are not aware of

other reports of MeHg in LFG. We have reported gaseous MeHg emissions from soils treated with municipal sludge,¹⁸ and the MeHg levels we measured in the ambient air near our sludge-amended soils (~1–5 pg/m³) were far lower than the “difference method” suggests may occur in LFG. These data suggest that gas-phase Hg speciation is an important area for future research since organic Hg may be present in LFG at levels well above these in ambient air.

Two interesting trends are apparent in our limited data on LFG: Hg concentrations were not generally diluted by elevated LFG flow rates (the highest levels were often associated with the largest flows), and Hg concentrations in LFG decrease significantly with increasing landfill age (Table 3). The lack of a dilution effect suggests that CH₄ and Hg^o releases may be influenced by a similar parameter, most likely the age and to a lesser extent the composition of waste in the radius of influence of a given well or vent. The sharp decrease in Hg concentrations (and fluxes) with landfill age was unexpected, especially over the relatively short time period these facilities had been closed. Considering that the estimated Hg content of municipal solid waste has decreased by a factor of 3 to 4 between 1985 and 1995⁵ (suggesting that less Hg is contained in the newer landfills) makes these trends even more intriguing. Mean LFG Hg concentrations decreased from ~1,800 ng/m³ in the flare of the PB active landfill (PB 90-) to ~40 ng/m³ in the flare of the PB closed landfill (PB 83-90). Overall gas flow rates were ~40% lower at the closed site. At the MC site, LFG Hg levels at the landfill closed in 1995 (MC 85-90) averaged ~800 ng/m³, while those at the landfill closed in 1990 (MC 85-90) averaged ~90 ng/m³. Gas flow rates at the older site were about half those at the newer site.

These trends in LFG Hg concentrations were independently confirmed at the two PB sites by our analyses of Hg in the gas condensate wells. Replicate LFG condensate samples drawn from the closed PB landfill wells (PB 83-90) exhibited a mean total Hg concentration of 16 ng/L, while those from the operating PB landfill condensate wells (PB 90-) exhibited a mean total Hg concentration of 4,700 ng/L (Table 4). Without data on condensate generation rates it is difficult to quantify the Hg removed, but it is clear that higher airborne Hg emissions would occur with LFG in the absence of condensate collection.

The gas condensate also provided some evidence of the speciation of the total Hg measured in the LFG itself, as we found readily detectable levels of MeHg in all samples (~0.3–20 ng/L). In line with other trends, the concentrations of MeHg were also highest in the condensate from the PB (90-) landfill, exceeding those

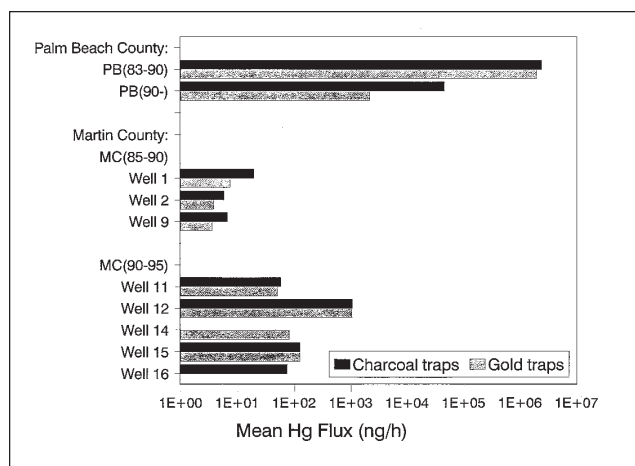


Figure 5. Mean fluxes of Hg estimated from LFG flow rates and measured Hg concentrations in flares and wells at PB and MC sites (n = 2–4 reps each; see Table 3).

in the closed PB (83-90) landfill by about an order of magnitude (Table 4). MeHg represented ~0.5 (PB 90-) to 2.2% (PB 83-90) of total Hg in these condensates. Measurements of Hg in LFG condensates collected in Minnesota yielded somewhat higher MeHg levels (14–69 ng/L¹⁶), and Gilmour and Bloom¹⁹ reported MeHg concentrations in this same range in effluents from a municipal wastewater treatment plant. Since our samples were collected from wells, it is not possible to determine if the MeHg was condensed (or scrubbed) from the gas phase in the LFG, or was formed directly in the collection wells.

The fate of the Hg in these condensates is also not known. The operational PB landfill (PB 90-) is a Class I landfill accepting primarily ash and processed residuals from the on-site, refuse-derived-fuel waste-to-energy (WTE) facility and occasional quantities of Class I MSW during peak deliveries and scheduled outages at the WTE facility. The PB (83-90) site is a closed (operational from 1983–1990) Class I landfill which accepted Class I MSW before the PB Solid Waste Authority began processing its MSW via WTE. Both PB landfills employ an active LFG collection system with multiple gas collection wells connected to a single manifold under negative pressure. The collected LFG is flared with a series of condensate knockouts removing moisture prior to flaring. Estimates of LFG condensate generation rates as a function of LFG generation could not be estimated because an unknown quantity of the condensate from most knockouts discharges back into the landfill. The condensate from the landfills is ultimately pumped through the leachate collection system and then to the deep well injection system.²⁰

As a result of the wide range in LFG flows and Hg concentrations, total Hg emission rates measured in LFG varied significantly across the range of wells and

flares sampled (Figure 5). The highest fluxes were measured from the pressurized-flow (i.e., active) LFG collection system flare at the operating PB landfill (PB 90-) (~2400 µg/hr), while the lowest occurred from the passive wells over the closed landfill at the MC (85-90) site (0.006 µg/hr). The decreases in both Hg concentrations and LFG flow rates from newer to older landfills result in drastically lower Hg emission rates from the closed landfills at both sites compared to the operating landfills. The average Hg fluxes at the two actively pumped flares at the PB sites decreased from ~2 mg/hr from the operating landfill (PB 90-) to ~0.02 mg/hr at the closed landfill site (PB 83-90). Fluxes overall were much lower at the smaller MC landfill which employed a passive LFG emission system, but also decreased dramatically from ~0.3 µg/hr at the closing landfill (MC 90-95) to ~0.01 µg/hr at the closed landfill (MC 85-90).

Recent research at the Fresh Kills Landfill serving New York City attempted to quantify Hg emissions in LFG from this site. As one of the largest U.S. landfills, Fresh Kills covers over 1,200 ha in mounds up to 46-m high, and is loaded with ~12,000 T/day of MSW (compared to ~400 T/day at MC). The landfill is vented through a series of passive vents, and includes an active gas collection system as well. This system produces ~4 × 10⁵ m³/day of LFG, which is cleaned, sold, and burned. Hg emissions were initially assessed at the Fresh Kills Landfill in 1995.²¹ Hg levels were determined in the LFG collection system and from a subset of the passive vents by a portable Jerome analyzer (detection limit in the µg/m³ range). The reported Hg concentrations in LFG were unexpectedly high (in the mg/m³ range), and the authors estimated that the landfill produced Hg vapor at a rate of ~30 mg/sec, comparable to the fluxes of Hg from large coal-fired power plants.²² However, at the request of the U.S. Environmental Protection Agency (EPA) and in response to criticism of the sampling methods, McGaughey and Burklin¹⁷ repeated parts of the Fresh Kills study using EPA Standard Methods (SM29 and 7470) with lower detection limits. They resampled LFG from the headers of the collection system, and found concentrations nearly 3 orders of magnitude lower than previously reported (~6–20 µg/m³). The wide variation in the data collected in these two different studies of the same landfill was one justification of the study reported here.

Hg Emissions During Routine Waste Handling and Disposal Operations

Another potential source of Hg emissions from landfill operations is routine waste handling including transport, dumping, spreading, compacting, and

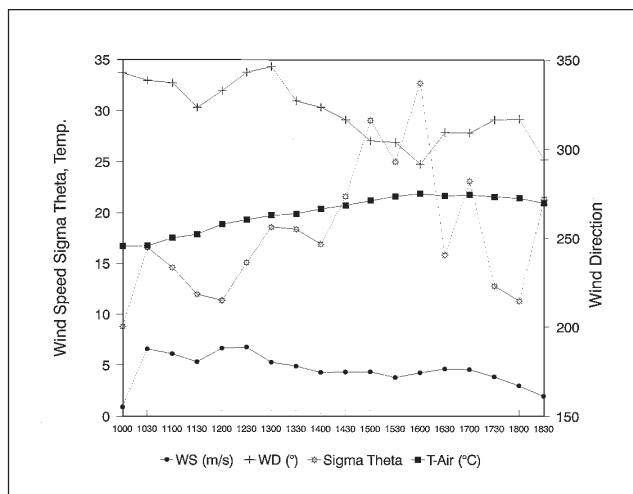


Figure 6. Meteorological conditions over the Martin County WF on April 18, 1997 during Hg monitoring. Data were used to estimate overall Hg losses during waste disposal activities (see text).

burial. If delivered waste includes significant quantities of Hg⁰-containing materials such as fluorescent bulbs and electrical switches, the release of this volatile Hg during dumping and compacting should be detectable. We designed a simple short-term experiment to evaluate the potential for Hg losses during all but the collection and transport phases of these operations (which occur largely outside of the landfill itself). Over a 24-hr period we monitored Hg concentrations in air downwind of the WF at the MC landfill (MCWF 96-), including the typically most active day of waste delivery (Friday). We measured Hg levels continuously with a Tekran mercury vapor analyzer whose intake line was located 2 m above the ground ~50 m from the edge of WF operations (an area of ~200 m²). Sampling took place one day after a cold front, and winds remained steady from the NW (mean WD = 320 ± 15°), passing directly over the landfill to our sampling point (Figure 6). The levels of Hg in the background incoming air were estimated by monitoring for 8 hr on the following day at an upwind site unaffected by landfill operations. During this entire period we also collected routine meteorological data onsite from a 3-m tower equipped with a standard measurement package and data logger.¹⁵

The concentration of Hg⁰ in air downwind of the WF clearly appears to be influenced by delivery, dumping, and crushing operations (Figure 7). By 1100 hr when our monitoring commenced, ~110 MT of waste had been delivered and the atmospheric concentration of Hg downwind of the site (~5 ng/m³) was already well above background (mean upwind Hg concentration = 2.37 ± 0.14 ng/m³, n = 101). Concentrations remained between 5 and 10 ng/m³ until 1230 hr, when airborne Hg began to exhibit a series of spikes that

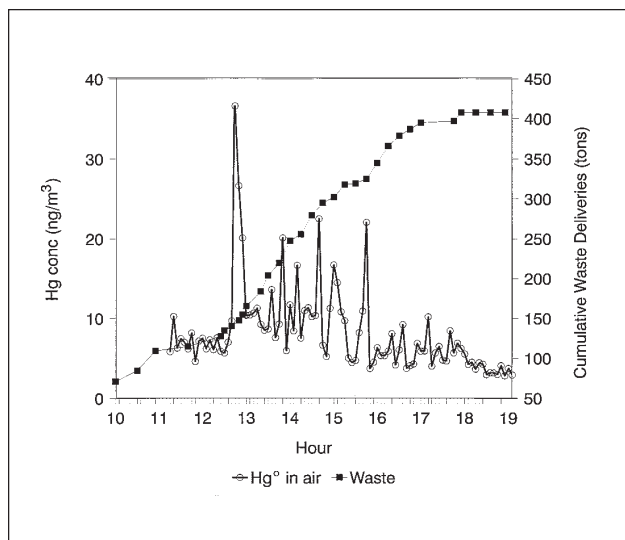


Figure 7. Temporal trends in atmospheric Hg[∞] concentrations downwind of the WF of the Martin County landfill measured with a Tekran analyzer on April 18, 1997. Also shown is the cumulative waste mass delivered to the site over this time period in MT.

persisted until ~1530 hr (one > 35 ng/m³, several > 20 ng/m³). Nearly half of the day's 400 MT of waste was delivered and spread during this 3-hr period.

We can only speculate on the exact origin of these spikes because the waste composition is unknown. They were unrelated to major shifts in wind direction which remained relatively constant (Figure 6); however, they do correspond remarkably well with activity on the WF (Figure 7). The parameters waste delivery rate and wind speed alone explain 44% of the variance in the air concentrations of Hg⁰ ($r = 0.66$, $P < 0.01$). The largest spike was associated with resumed deliveries following the lunch break when as many as five trucks were observed to be waiting in line at the WF. Despite continued deliveries, the large spikes ended abruptly at 1545 hr when application of the daily soil cover began, and Hg remained in the range of 5–10 ng/m³ until the landfill closed at 1800 hr. As indicated by the surface flux data discussed above, newly applied soil cover is an effective short-term barrier against gaseous Hg emissions. By 1900, the air concentrations of Hg began to approach upwind levels, and at night Hg remained near continental background (1.9 ± 0.04 ng/m³, from 0300 to 0600 hr). With the resumed but lower activity the next day (38 MT delivered from 0830 to 1130 hr), Hg levels again increased to around 4–5 ng/m³. We also collected one 24-hr aerosol sample using the Tekran pre-filter, which yielded a total particulate Hg concentration of 0.43 ng/m³. Assuming that most of the particle load was collected during the 8 hr of activity at the WF, particulate Hg represents ~10% of total atmospheric Hg under the

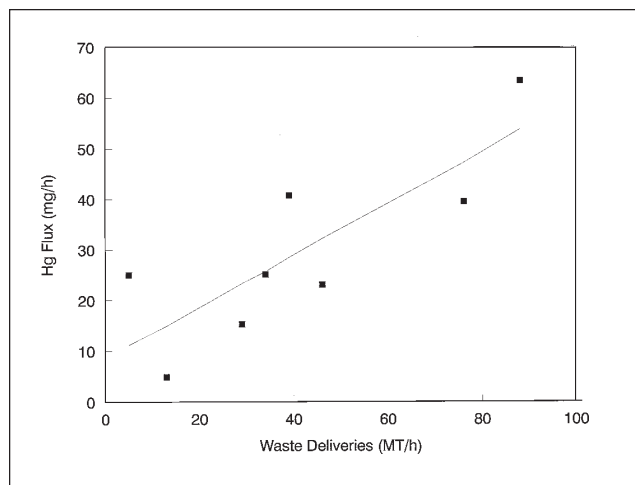


Figure 8. Relationship between the modeled hourly fugitive atmospheric emission of Hg^0 during activities at the Martin County landfill WF and the recorded hourly delivery of waste to the landfill on April 18, 1997.

dusty conditions experienced here. Aerosol Hg typically averages $0.01\text{--}0.03\text{ ng/m}^3$ at rural sites, or $<1\%$ of airborne Hg.²³

It is possible to estimate the quantity of Hg released during daytime WF activities using a simple box model. The WF was treated as a ground-level area source, the emissions from which were uniformly dispersed by turbulent mixing throughout an atmospheric “box” upwind of our sampler. The box dimensions were estimated from dispersion parameters computed from our on-site meteorological data using the methods of Turner.²⁴ We assumed that the Tekran inlet was sampling the plume center line, that Hg concentrations were horizontally uniform within the area of the box, that concentrations exhibited a decreasing logarithmic profile with height above the ground, and that the “box” was defined by $3\sigma_z$ and $3\sigma_y$ (where y and z are vertical and horizontal turbulent dispersion parameters). Hg fluxes were then estimated for each hour of monitoring data, and corrected for Hg in the incoming air mass measured upwind. Dispersion conditions during this period were neutral to slightly unstable (Pasquill-Gifford stabilities C and D), and the box dimensions ranged from 7–16-m high (by a nearly constant ~50-m wide, which approximated the actual dimensions of the WF). Because of the local terrain, it is possible that vertical dispersion was much greater during this time, and our flux estimates may well be biased low. Our estimates of the hourly fluxes of Hg during WF activities ranged from ~5–60 mg/hr, and were significantly correlated with the rate of waste delivery (Figure 8; $r = 0.82$, $P < 0.01$). Given our simple approach, this relationship is reassuring since the fluxes were estimated independently of the waste delivery data. Assuming a uniform emission rate over the active area of the

WF, this translates to a mean surface flux of $\sim 70,000\text{ ng m}^{-2}\text{ hr}^{-1}$, far greater than the emission rates measured over the landfill surfaces with final or intermediate cover in place. For the 8-hr period of heaviest WF activities we estimate a total airborne Hg emission of $\sim 240\text{ mg}$ (given our various assumptions, we place an uncertainty on this value of $\pm 50\%$ with the understanding that our approach was more likely to under- than over-estimate the actual flux).

The slope of the regression of Hg emission on waste mass provides an estimate of the average volatilized Hg fraction of the waste delivered, and a further check on our overall emission estimates. For the zero-intercept regression, this value is $0.7 \pm 0.2\text{ mg/T}$ or roughly $1\text{ }\mu\text{g Hg/kg}$ waste, a small fraction of the estimated $\sim 4\text{ mg/kg}$ of Hg in overall waste.⁵ This suggests that $<0.1\%$ of the Hg estimated to be in the waste when it is initially discarded is lost during these WF activities. The $>99\%$ of the Hg not accounted for in the WF emissions is either lost during waste collection and transport activities (prior to delivery to the landfill), is sequestered in the landfill in a nonvolatile form, or is volatilized over time after cover is applied. Our estimates of Hg losses from the older (closed) MC landfill (MC 85-90) suggest that the Hg which is unaccounted for is most likely sequestered in the landfill more or less permanently, or is actually lost prior to landfilling. An evaluation of a landfill “mining” demonstration from 1976–1979²⁵ found Hg concentrations in the mined MSW residues of 0.36 mg/kg or $<10\%$ of the estimated 4 mg/kg Hg in the MSW originally placed in the landfill. This could suggest that the bulk of the Hg in that study (as much as $\sim 90\%$) was lost prior to landfilling. However, it is not clear whether these “mined residues” included dry cell batteries which were at that time the largest source of Hg in MSW. Clearly, further research into emissions during storage/transport prior to landfilling would be necessary for a complete mass balance for the total MSW handling and storage process.

Estimates of Annual Hg Emissions from Landfills in Florida

The data we have summarized here are clearly very limited. However, they are also unique, representing the only data set of their kind of which we are aware. Because of the intense interest in the Hg issue in Florida, and because more extensive data are not available, we performed a simple scaling exercise. To emphasize the uncertainty associated with our data, we estimated ranges of annualized fluxes from the sites studied, and then scaled our fluxes to Florida’s landfills using state-wide MSW regulatory inventories.

Several assumptions were made to estimate annual Hg fluxes from the PB and MC landfills. Annual Hg

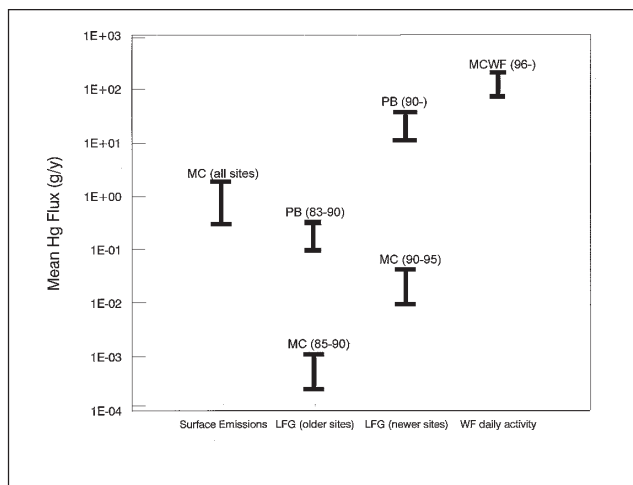


Figure 9. Estimates of annual Hg emissions from various sources at the Martin County, Florida municipal landfill (MC). Ranges of fluxes were derived from different scaling assumptions as explained in the text. For comparison, also shown are the estimated Hg fluxes from the active LFG collection system at the Palm Beach County, FL, Landfill (PB) (other sources were not measured at PB).

fluxes through the landfill surface-covers at the MC sites were estimated from the mean surface emission rates measured over each landfill (i.e., from Figure 1, corrected for the mean control soil flux), and the total surface areas (sides plus tops) of each site. Ranges of fluxes were derived by assuming that measured fluxes were constant over the diel cycle, or that measured fluxes represented midday maxima (operative for 6 hr, then reduced by 50% for 4 hr, and going to 0 at night, following patterns we have observed over background soils¹⁵). For the special case of surface fluxes through the MC-WF aged cover, we assumed that the MSW in place was emitting Hg at the estimated rates for only

six months to account for the fact that the actual residence time of waste in the WF during a year could be anywhere from 1 to 365 days. Hg fluxes in LFG were computed from the flow-weighted mean Hg concentrations at each landfill (i.e., from Table 3) and annual LFG production estimates (derived from measured gas flows at all MC wells and recorded flows at both PB flares). The LFG fluxes shown in Figure 9 represent the ranges derived individually for each site (the lowest ranges are for the passive well-type LFG systems at MC, the highest are for the actively pumped systems at PB). Finally, annual emissions from daily waste handling activities at the MC WF were estimated from the relationship we derived for Hg release per ton of waste and the annual mass of waste delivered to the MC site.¹⁴ The range of fluxes from the WF shown in Figure 9 was derived using two assumptions of the spatial variability in the concentration of Hg in the area source “plume” we measured at this site (lowest value assumes concentrations decrease logarithmically with height, the highest value assumes a uniform concentration with height).

Overall, we estimate that the MC landfill could emit on the order of ~80–100 g/yr of Hg from all sources (Table 5). Annual emissions from LFG alone at the actively pumped PB site could be on the order of 20 g/yr (other pathways were not measured at PB). Several observations are apparent from the annual estimates shown in Figure 9: Hg fluxes are dominated by the releases that occur during routine waste handling operations at the WF, emissions via surface cover can probably be ignored (these fluxes do not significantly exceed those over background soils), and emissions coincident with LFG losses may be important but are highly variable be-

Table 5. Annualized Hg emissions from Florida Class I MSW landfills based upon direct measurements at three Martin County, FL (MC), landfill study sites.

MC Site Description	MC MSW Age	MC MSW Mass (T)	MC Annual Hg Emissions (g/yr)	MC Hg Emission Factor (g/T of MSW)	FL MSW Age	FL MSW ^a Mass (T)	FL Annual Hg Emissions (kg/yr) ^b
Closed, Class I	1985-1990	750,000	0.3-1.0 ^c	4E-07 to 1E-06	1988-1990	23,000,000	0.009-0.031
Closing, Class I	1990-1996	560,000	0.6-1.7 ^c	1E-06 to 3E-06	1990-1995	50,700,000	0.054-0.15
	Subtotal	1,310,000	0.9-2.7			73,700,000	0.063-0.18
Open, Class I	1996-Present:						
	WF	76,000	75-110	1E-03 to 2E-03	1996 est	9,300,000	9.2-14
	Aged Cover	110,000	0.3-1.0	3E-06 to 9E-06	1996 est	9,300,000	0.028-0.082
Total			76-110				9.3-14

^aThe estimated uncertainties on these waste masses are ± 5 –10% (for period 1996–present, and 1990–1996) and ± 50 % (for 1985–1990).

^bAssumption 1: All MSW placed in all Florida landfills is comparable in composition to the MSW in place at the MC sites.

Assumption 2: All Florida landfill site characteristics, that is, meteorological conditions, landfill construction, MSW decomposition mechanisms, and landfill surface area to MSW mass ratios, are comparable to MC site characteristics.

^cTotal estimated Hg emissions from surface cover plus LFG generation via passive wells at MC. Hg emissions from LFG at larger landfills with actively pumped LFG collection systems could be much larger based on our limited data from the Palm Beach Landfill (LFG Hg emissions ~20g/yr; see text).

tween sites. As mentioned earlier, LFG emissions of Hg decrease strongly from the actively pumped PB site to the passive wells at MC, and within each site, LFG emissions decrease significantly from newer to older landfills. Landfills using active gas collection systems such as the PB facility might be expected to generate higher Hg fluxes coincident with LFG releases, but further measurements are necessary to test this hypothesis.

We used the Hg emissions data from the MC sites to estimate Hg losses from Class I MSW landfilled statewide in Florida. The MC sites are generally representative of many Florida landfills in size, waste mass handled, and waste composition. Because of the apparent relationship between waste age and LFG emissions, this exercise was done for MSW that roughly corresponded in age with that in place at the MC sites, and we scaled primarily from relationships between emission rates and waste mass in place at each MC landfill. The official data available from the Florida Department of Environmental Protection²⁶ for waste age correspond relatively well, but not exactly, with the data available from the MC sites (see Table 5). For example, the MC (85-90) site data were matched with the MSW placed in Florida landfills from July 1988 to June 1990 (statewide data for 1985–June 1988 were not available). Similarly, the MC (90-96) site data were matched with state MSW data from July 1990 to December 1995. Finally, the WF site (WF 96-) was matched with the best available estimate of recent statewide data (we used the arithmetic mean of the MSW landfilled from 1991 to 1995, since 1996 data were not available).

For the MC (85-90) and (90-95) landfill study sites, the estimated annual Hg fluxes in LFG plus surface cover emissions were summed for each site, while only surface cover fluxes were used to represent the MC-WF (96-) site where a LFG collection system was not yet in place. We then divided these total annual Hg emissions/site by the mass of MSW in place at each site to obtain an emissions rate in g/T of MSW for each site. For routine activities at the MC-WF (96) site, we used the Hg emission rate/T of waste relationships derived above. The various per-ton emission rates were then applied to official Florida DEP data on MSW landfilled statewide during the years best corresponding to the age of the MSW in place at each of the three sites as explained above.

The input values and resulting annualized emissions from the MC sites and MSW landfilled in all Florida Class I landfills are shown in Table 5. Our calculations suggest that all Florida Class I MSW landfills could release on the order of 10 kg/yr of Hg to the atmosphere. We emphasize that this exercise is not intended to be a statistically significant model of actual Hg emissions from Florida landfills, but the most reasonable "order-of-magnitude" estimate that we can make from our data. An

estimate of annual Hg emissions from various anthropogenic source categories statewide in Florida during 1990²⁷ listed MSW combustion at ~4,200 kg, electric utility industry at ~1,400 kg, medical waste incineration at ~1,500 kg, and all anthropogenic sources at ~13 T.²⁸ That same estimate listed annual Hg emissions from landfills at <1 kg. While it appears that statewide Hg emissions from landfills have been underestimated by perhaps an order of magnitude, our estimates from the measurements at MC site still suggest that landfill emissions are significantly below the three major anthropogenic source categories listed above, accounting for <1% of all anthropogenic emissions in Florida. In fact, it appears that our estimated total annual Hg emission from Florida landfills is less than emissions from most individual MSW combustion facilities (these facilities report a range of Hg emissions of ~10–400 kg/yr, mean ~140 kg/yr²⁸). However, since we have accounted for only a very small fraction (<1%) of the Hg estimated to be contained in MSW, the potential for significant Hg emissions from landfills or MSW handling activities remains large.

Our intent in this exercise was to scale up our limited site-specific data to a larger universe of landfills to encourage discussions on the significance of this source of Hg emissions as it pertains to additional research needs and current MSW management policies. Our results suggest four questions for further research into Hg emissions from MSW landfills: (1) Are the results from our study of two landfill operations in Florida representative of MSW landfills in Florida and across the United States? (2) Are there significant Hg emissions from MSW prior to placement in a landfill, that is, from collection containers, transport vehicles, and MSW transfer stations? (3) What is the relationship between the various Hg-bearing fractions of the MSW stream (e.g., fluorescent lamps, thermometers, thermostats, Hg switches) and Hg emissions at the landfill WF and during MSW collection and transport activities? (4) Are there significant releases of methyl- or other organo-Hg compounds in LFG or its condensates?

AUTHORS' NOTE

The authors recently completed a more extensive mercury study at a second Florida landfill.²⁹ At this site we found much higher levels of Hg in air downwind of the working face, as well as in landfill gas (by factors of 2–5). We also positively identified gaseous dimethyl mercury in landfill gas. It is clear that mercury emissions from landfills will be site specific, and that organo-mercurials are being formed in landfills.

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