Climate Change Effects of Gases from Municipal Solid Waste Landfills

Derek Manheim i), Nazli Yesiller ii) and James L. Hanson iii)

i) Research Fellow, Civil and Environmental Engineering Department, California Polytechnic State University, San Luis Obispo, CA 93407, U.S.A.
ii) Director, Global Waste Research Institute, California Polytechnic State University, San Luis Obispo, CA 93407, U.S.A.
iii) Professor, Civil and Environmental Engineering Department, California Polytechnic State University, San Luis Obispo, CA 93407, U.S.A.

ABSTRACT

Direct and indirect effects of landfill gases and interrelationships between direct and indirect climate-forcing emissions and cover soil characteristics were analyzed at 5 municipal solid landfills in California. Fluxes of 80 chemicals were quantified across daily, intermediate, and final covers using static flux chambers. Whole-site emissions were scaled up using the flux data. Direct and indirect climate-forcing emissions were estimated through application of global warming (direct), atmospheric degradation (direct), tropospheric ozone forming (indirect), stratospheric ozone depletion (indirect), and secondary aerosol forming (indirect) potentials reported for each chemical. Methane emissions accounted for 46 to 99% of the total direct climate-forcing emissions. Nitrous oxide and F-gases collectively accounted for 1 to 99% of the total direct climate-forcing emissions. Overall direct emissions accounted for 93 to nearly 100% of the total combined (direct and indirect) climate-forcing emissions. Correlations were observed: a) between direct and indirect climate-forcing emissions of greenhouse gases and water content parameters as well as void ratio and b) between direct and indirect climate-forcing emissions of NMVOCs and mass of water in a cover profile, volumetric air content, temperature, and volumetric solids content.

Keywords: landfill, municipal solid waste, cover, climate change, soil property, climate forcing

1 INTRODUCTION

A significant challenge in the operation of municipal solid waste (MSW) landfills is to effectively control the release of harmful chemicals into the environment. Fugitive emissions of toxic, noxious, and persistent chemicals through various pathways affect human and ecosystem health and have direct and indirect effects on global climate change (IPCC 2013). Biodegradation of organic constituents in MSW and volatilization of organic compounds from anthropogenic sources generates landfill gas (LFG). The primary components of LFG include methane (55-60% v/v), carbon dioxide (40-45% v/v), and trace components, including carbon monoxide, nitrous oxide and NMVOCs (<1% v/v) (Tchobanoglous et al. 1993). Methane, carbon dioxide, and nitrous oxide, are well-known greenhouse gases (GHGs) that absorb outgoing radiation from Earth’s surface, exerting a strong, positive radiative forcing (RF) on climate (IPCC 2013). In addition, trace fluorinated gases, including (hydro)chlorofluorocarbons (CFCs, HCFCs, and HFCs), collectively termed F-gases (originating from insulation foams, refrigerants, solvents), are anthropogenic GHGs that contribute to climate change.

NMVOCs are a broad class of highly reactive and volatile organic compounds that originate from anthropogenic and biogenic sources in landfills (Kansal 2009, Nair et al. 2019). NMVOCs volatilize from anthropogenic sources including personal care products, cleaning solvents, household sprays, textiles, electronics, paints, furniture, and cooking fuels in the waste mass. In addition to generation from decaying wood or green wastes, biogenic NMVOCs are generated as oxidation or fermentation byproducts of aerobic and anaerobic biological conversion processes in the waste mass and/or cover soils (Komilis et al. 2004).

Once released into the atmosphere, NMVOCs participate in numerous photochemical reactions in the troposphere and, in addition to oxides of nitrogen (NOx), are regarded as critical precursors of ozone, photochemical smog, and secondary organic aerosols (Atkinson and Arey 2003). Through these complex photochemical reactions, NMVOCs indirectly affect global climate change by reducing the efficiency of potent GHG oxidation and promoting the build up of GHGs in the atmosphere (Collins et al. 2002). Atmospheric degradation of NMVOCs leads to the production of CO₂, which is a known secondary direct effect on climate change (IPCC 2013). Some NMVOCs
are also stratospheric ozone depleting substances (ODSs), which significantly alter the radiative balance of Earth’s climate (WMO 2014).

In active MSW landfills, three categories of covers are used: daily, intermediate, and final. These covers remain in place over the short, medium, and long term for daily, intermediate, and final covers, respectively. Daily covers are used to temporarily cover the waste mass at the end of a work day prior to placement of wastes over the next day and function mainly to isolate the waste mass from the surrounding environment. Intermediate covers, while also temporary, remain in place for extended durations when the waste mass reaches a relatively constant height and function to isolate the waste mass and limit fugitive emissions. Final covers are permanent systems used to limit fugitive emissions and prevent ingress of water into the waste mass. Soil is used for all cover categories. Alternative materials such as green waste, autofluff, sludge, tires, construction and demolition waste, and various spray-on and synthetic materials are also used in daily covers. Geosynthetics also are used for final covers. The covers are built with increasing engineering designs and controls from daily to intermediate to final covers.

Methane emissions from landfills contribute 627 Mt CO₂-eq. to global climate change (IPCC 2013). While effects of methane have been considered in climate change analysis (IPCC 2013), investigation of climate change effects of other GHGs including nitrous oxide and F-gases from landfills have been highly limited. Yesiller et al. (2018) indicated that due to their high global warming potentials (GWP), F-gases exert disproportional direct effects on climate change. Detailed analysis of effects of soil properties on landfill gas emissions also have been highly limited. Yesiller et al. (2018) identified strong correlations between F-gas flux and cover soil fines content, degree of saturation, void ratio, water content, and density. To the authors’ knowledge, climate effects of trace NMVOCs or correlations to soil properties have not been determined.

The objectives of this investigation were two-fold: identify climate change effects of trace landfill gases including both direct and indirect effects and identify correlations between direct and indirect emissions and landfill cover characteristics to assess larger scale impacts of landfill systems on global climate change. Data and analysis are included herein from an extensive field and laboratory testing program conducted to determine emissions at 5 landfill sites over 2 seasons for 3 cover categories and 80 chemical species.

2 EXPERIMENTAL STUDY

2.1 Field testing program

The field-testing program included determination of surface fluxes of 2 primary GHGs (methane and nitrous oxide) and 78 NMVOCs including 13 species of primary GHG F-gases. The NMVOCs were grouped under 11 chemical families: fluorinated gases (F-gases), reduced sulfur compounds (RSCs), halogenated hydrocarbons (HH), organic alkyl nitrates (ON), alkanes (Alk), alkenes (Alke), Aldehydes/Alkynes (A/A) aromatic hydrocarbons (Ar), monoterpenes (Mon), alcohols (Alc), and ketones (Ket). Tests were conducted in 3 different climatic regions over two main seasons in California: wet (winter) and dry (summer).

Details for the landfills included in the study are provided in Table 1. The landfills selected for the study included both medium (SMRL, TDL) and large (PHL, SAL, CCL) landfills. The 5 sites collectively contain 13% of waste in place in active California landfills. The landfills are located in three climate zones Csb (temperate, dry summer, hot summer), Csa (temperate, dry summer, warm summer), and Bsk (arid, steppe, cold) (Peel et al. 2007). Daily covers had clean and contaminated soils and alternative materials (auto shredder residue, green waste, and wood waste). Interim cover types included soil and green waste. Final cover systems were all conventional covers and consisted of compacted clay liners with the exception of SMRL with a composite GCL-compacted clay liner. All of the sites had operational gas collection and removal systems in place at the time of testing.

Table 1. Landfill sites.

| Site | Waste in Place (m³) | Avg. Daily Temp (°C) | Ppt. (mm) | Climate Zone | Daily Interim Final (%) |
|------|--------------------|----------------------|-----------|--------------|------------------------|
| SMRL | 1,360,577          | 14.9                 | 462       | Csb          | 0.1 69.3 30.6          |
| TDL  | 3,038,622          | 17.4                 | 278       | Bsk          | 15.5 84.5 0            |
| PHL  | 26,454,935         | 16.1                 | 630       | Csa          | 3 91 6                |
| SAL  | 44,173,397         | 15.8                 | 387       | Csb          | 0.6 89.5 9.9           |
| CCL  | 55,227,178         | 18.2                 | 462       | Csb          | 8.3 63.8 27.8          |

2.2 Determination of gas flux

Static flux chambers were used to measure surface fluxes of the gases. Custom-built large-scale chambers constructed of stainless steel with dimensions of 1x1 m (1 m² measurement area) x 0.4 m (height) were used. A fan is operated inside the chambers to circulate the accumulating gas to ensure uniform distribution prior to sampling. Gas samples were obtained using custom-built, 2-L capacity stainless steel evacuated canisters and analyzed using two integrated VOC analytical systems. At each measurement location, two chambers (60 and 120 minute tests) were used to collect data. A total of 5 gas samples over time are obtained per chamber. Flux is determined for each chamber as the product of the concentration gradient (i.e., change in concentration with time, dC/dt) and the volume:area ratio of the chamber following the procedure described in Rolston (1986). The change in concentration with time was determined using a linear regression with a target coefficient of determination value, R² ≥ 0.70. The methodology used in the test program is similar to the
approach used in Yesiller et al. (2018).

2.3 Determination of soil properties
A total of 232 field samples were collected from the 5 landfill sites during the dry and wet season testing. Analysis only for soil covers are included herein. Density of the covers were determined in place using the sand-cone method (ASTM D1556). Samples of the cover soils were collected to determine moisture content (ASTM D2216), specific gravity (ASTM D854), grain size distribution (ASTM D422), and Atterberg limits (ASTM D4318).

2.4 Direct and indirect climate change effects
Chemical-specific, whole site, annual emissions were calculated using the fluxes measured for the different cover types at a given landfill. For each landfill, the relative areas of the different cover types and the total area of the landfill (Table 1) are used to scale-up average fluxes for each cover type to calculate annual emissions for the entire landfill.

Calculations for climate change analysis are summarized in Table 2. Primary direct climate-forcing emissions (DGWR1, Mg CO2-eq./yr) were estimated using Equation 1, where DGWP is the chemical-specific direct GWP (available for methane, nitrous oxide, and F-gases) based on 100-year values (IPCC 2013). The secondary production of carbon dioxide from atmospheric degradation of all 80 chemicals (DGWR2, CO2-eq./yr) was estimated using Equation 2, where MWi and Ni,i represent the chemical specific molecular weights and total number of carbon atoms, respectively (Majumdar and Srivastava 2012). The net direct emissions (in terms of CO2-eq./yr) for each landfill site was calculated by summing the primary (DGWR1) and secondary (DGWR2) direct emissions.

Table 2. Summary of indirect and direct emission calculations.

| Eq. | Mathematical Expression | References |
|-----|-------------------------|------------|
| 1   | $DGWR_1 = \sum_{i=1}^{15} \left[ E_{LF,i} \cdot DGWP_i \right]$ | IPCC (2013) |
| 2   | $DGWR_2 = \sum_{i=1}^{80} \left[ \frac{E_{LF,i}}{MW_i} \cdot N_{i,i} + 44 \right]$ | Majumdar and Srivastava (2012) |
| 3   | $IGWR = \sum_{i=1}^{78} \left[ E_{LF,i} \cdot IGWP_i \right]$ | Daniel and Solomon (1998), Collins et al. (2002) |
| 4   | $IOFP = IGWP_{OFP} \cdot \sum_{i=1}^{78} \left[ E_{LF,i} \cdot OFP_i \right]$ | Carter (2009), Wuebles et al. (2014) |
| 5   | $ISOAP = IGWP_{SOA} \cdot \sum_{i=1}^{78} \left[ E_{LF,i} \cdot SOAP_i \right]$ | Grosjean and Seinfeld (1989), Unger (2014) |
| 6   | $IODP = \sum_{i=1}^{78} \left[ E_{LF,i} \cdot IGWP_{ODP} \right]$ | WMO (2014), IPCC (2013) |

Indirect climate change effects were evaluated using indirect GWP, tropospheric ozone formation potentials (OPFs), secondary organic aerosol formation potentials (SOAPs), and stratospheric ozone depletion potentials (ODFs). The overall indirect effect on global climate change (IGWR, Mg CO2-eq./yr) was calculated using Equation 3 (Table 2), where IGWPi and Ei,i represent chemical-specific indirect GWP and surface emissions from a given landfill, respectively. Available indirect GWP values were incorporated for 15 NMVOC species (Daniel and Solomon 1998, Collins et al. 2002). If indirect NMVOC GWP values were not reported, these were estimated using scattered, 3-D interpolation by applying the natural neighbor method (MATLAB 2018).

Since indirect GWPs are representative of atmospheric reactivity, experimental reaction rate coefficients of each NMVOC with hydroxyl, chlorine, and nitrate radicals (McGillen et al. 2020) were used as predictor variables in this analysis.

Tropospheric OPFs for each site were quantified using the maximum incremental reactivity (MIR) scale (Carter 2009). The overall indirect climate effect for OPF (IOFP, Mg CO2-eq./yr) was calculated using Equation 4 (Table 2), where OFPj (g Oeq/g NMVOC) is the chemical-specific ozone formation value based on FAC and IGWPi is the indirect GWP of tropospheric ozone (Wuebles et al. 1994).

Tropospheric SOAPs for each site were quantified using fractional aerosol coefficients (FAC) reported in Grosjean and Seinfeld (1989). The overall indirect climate effect for SOAs (ISOAP, Mg CO2-eq./yr) was calculated using Equation 5 (Table 2), where SOAPi (g SOAP/g NMVOC) is the chemical-specific SOA formation value based on FAC and IGWPi is the indirect GWP of tropospheric SOA (Unger 2014).

The overall indirect climate-forcing for ODSs (IODP, Mg CO2-eq./yr) was calculated using Equation 6 (Table 2), where IGWPi is the chemical-specific indirect GWP regarding stratospheric ODP (WMO 2014, IPCC 2013). Ultimately, the net indirect emissions (Mg CO2-eq./yr) for each landfill site were calculated by summing the IGWR, IOFP, ISOAP, and IODP values obtained from Equations 2-6.

2.5 Development of correlations
Spearman’s correlation coefficient ($\rho$) was applied to assess the relative strength and direction of correlations between site-specific geotechnical properties and direct and indirect climate-forcing emissions. Spearman’s $\rho$ range from -1 to 1 with the bounding limit values indicating perfect negative or perfect positive non-linear, monotonic association, respectively. Statistical significance of the resulting correlations was assessed using a permutation test, which estimates and compares $p$-values to a standard $\alpha$ significance level ($\alpha = 0.10$ used herein). $p$-values that are below $\alpha$ indicate statistically significant correlations. Various non-linear and linear curve fits were applied in MATLAB r2018a
to obtain mathematical expressions of the developed correlations.

## 3 RESULTS AND DISCUSSION

### 3.1 Direct and indirect climate-forcing emissions

Direct climate-forcing emissions are summarized in Table 3. The highest direct climate-forcing emissions were for methane followed by emissions of nitrous oxide and F-gases except for SMRL. The alkane family contributed to indirect emissions as high-GWP gases that directly absorb incoming shortwave and outgoing terrestrial radiation in the atmosphere. Aside from the F-gases, the alkane and monoterpenoid families were highest, on average, for the F-gas and HH families, as these families contain a significant number of ODSs. In particular, a large, negative indirect value was obtained for CCL, given the high emissions of ODP chemicals and SOA precursors, which contribute to a net negative RF. In comparison to direct emissions, the indirect emissions were significantly lower than total values of direct climate-forcing emissions (Tables 3 and 4). After combining indirect and direct emissions, direct emissions accounted for 93 to nearly 100% of the total combined climate-forcing emissions at a given landfill.

| Table 3. Summary of direct emissions (Mg/year). |
|-----------------------------------------------|
| **Ch** | **N** | **F-gas** | **RSC** | **HH** | **ON** | **Alk** | **A/A** | **Ar** | **Mon** | **Alc** | **Ket** |
|-------|-------|-----------|---------|--------|--------|--------|--------|-------|--------|--------|--------|
| Ch4   | -     | 3.75E+04  | 9.53E+00| 1.29E+00| 1.12E+00| 1.59E+00| 1.71E+00| 7.84E+00| 1.99E+00| 5.82E+00| 1.37E+00|
| N2O   | 2.31E+04| 5.99E+00| 1.26E+00| 7.18E+00| 1.55E+00| 7.36E+00| 1.84E+00| 5.71E+00| 1.33E+00| 6.65E+00| 1.37E+00|
| F-gas | 7.68E-01| 3.89E+00| 2.17E+00| 1.98E+00| 1.22E+00| 1.63E+00| 1.71E+00| 5.82E+00| 1.50E+00| 6.81E+00| 1.37E+00|
| RSC   | 1.33E-03| 7.74E-03| 1.29E-01| 7.28E-03| 8.77E-02| 5.12E-02| 1.08E-01| 1.08E-01| 1.22E+00| 3.04E+00| 5.94E+00|
| HH    | 6.67E-04| 1.62E-02| 5.88E-02| 2.39E-02| 1.08E-01| 1.08E-01| 1.08E-01| 1.08E-01| 1.22E+00| 3.04E+00| 5.94E+00|
| ON    | 3.15E-05| 2.21E-02| 1.55E-03| 1.74E-04| 1.48E-03| 1.48E-03| 1.48E-03| 1.48E-03| 1.22E+00| 3.04E+00| 5.94E+00|
| Alk   | 5.50E-03| 7.18E-01| 4.50E+00| 2.46E+00| 1.31E+02| 1.31E+02| 1.31E+02| 1.31E+02| 1.22E+00| 3.04E+00| 5.94E+00|
| A/A   | 9.53E-03| 1.93E+00| 1.60E+00| 2.11E+00| 1.22E+00| 1.22E+00| 1.22E+00| 1.22E+00| 1.22E+00| 3.04E+00| 5.94E+00|
| Ar    | 8.67E-03| 1.12E+01| 3.75E+01| 4.22E+02| 1.24E+01| 1.24E+01| 1.24E+01| 1.24E+01| 1.24E+01| 3.04E+00| 5.94E+00|
| Mon   | 2.31E-02| 2.44E+00| 8.72E+00| 1.21E+01| 1.54E+00| 1.54E+00| 1.54E+00| 1.54E+00| 1.54E+00| 3.04E+00| 5.94E+00|
| Alc   | 2.40E-02| 1.07E+00| 7.84E+00| 4.50E+01| 7.85E+01| 7.85E+01| 7.85E+01| 7.85E+01| 7.85E+01| 7.85E+01| 7.85E+01|
| Ket   | 2.12E-02| 3.02E+01| 1.26E+00| 1.88E-01| 1.49E+00| 1.49E+00| 1.49E+00| 1.49E+00| 1.49E+00| 1.49E+00| 1.49E+00|
| Total | 2.40E+01| 3.79E+04| 4.26E+04| 2.98E+04| 2.56E+04| 2.56E+04| 2.56E+04| 2.56E+04| 2.56E+04| 2.56E+04| 2.56E+04|

NEG - Net emissions were negative (atmospheric uptake)

Indirect climate-forcing emissions are summarized in Table 4. Total (positive) indirect climate-forcing emissions were highest, on average, for the alkane and methane families. The alkane family contains multiple species with high indirect GWP values and low SOAP/ODPs, thereby contributing to high, net positive indirect emissions. As methane has a reported MIR value, it indirectly produces tropospheric ozone and contributes to indirect emissions as compared to nitrous oxide. Total (negative) indirect climate forcing emissions were highest, on average, for the F-gas and HH families, as these families contain a significant number of ODSs. In particular, a large, negative indirect value was obtained for CCL, given the high emissions of ODP chemicals and SOA precursors, which contribute to a net negative RF. In comparison to direct emissions, the indirect emissions were significantly lower than total values of direct climate-forcing emissions (Tables 3 and 4). After combining indirect and direct emissions, direct emissions accounted for 93 to nearly 100% of the total combined climate-forcing emissions at a given landfill.

### 3.2 Correlations to geotechnical properties

Selected correlations between site-specific geotechnical properties and direct climate-forcing emissions are presented in Figure 1 for both GHGs and NMVOCs. For GHGs, correlations were observed between direct and indirect climate-forcing emissions and water content parameters (w, θw, and S) as well as void ratio. For NMVOCs, correlations were observed between direct and indirect climate-forcing emissions of NMVOCs and mass of water (Mw), volumetric air content (θa), temperature, and volumetric solids content (θs). The correlation results were developed using power series functions (y = axb + c), where R² values ranging from 0.87 to 0.99 indicated that the model fit to the experimental data was acceptable.

For a majority of the correlations developed between soil properties and GHG direct emissions, the mathematical relationship appeared hyperbolic, where there was potentially a limiting value for each soil property (e.g., S < 30% or s < 1.4) where direct climate-forcing emissions leveled off. In addition, soil moisture was an important factor related to direct emissions of GHGs, where water content (gravimetric and volumetric) and degree of saturation all exhibited significant correlations. The NMVOCs demonstrated different trends, where the total mass of water in a 1 m²
column through the depth of the cover profile decreased exponentially with direct emissions (Figure 1). In general, fluctuating soil water contents in the field (e.g., short term during precipitation events and longer term during dry and wet seasons) will either enhance or retard GHG and NMVOC transport due to variations in chemical solubility, molecular diffusion coefficients between the aqueous and gaseous phases, sorption, and biological oxidation. For example, methane and F-gas fluxes were observed to increase with increasing soil water content due to a reduction in sorptive and biological oxidation capacities of the soils under saturated conditions (Yesiller et al. 2018). The results of this study support the trends reported by Yesiller et al. (2018), in which direct GHG climate-forcing emissions were observed to increase as a function of water content and degree of saturation present in the soil covers.

Statistically significant correlations were observed between all depicted soil properties and indirect GHG emissions ($p < 0.10$). Similar to results presented in Figure 1, the correlation results were developed using power series functions, where $R^2$ values ranging from 0.87 to 0.99 indicated that the model fit to the experimental data was acceptable.

For NMVOCs, the correlations were of similar strength to the direct emissions correlations, but were opposite in direction (e.g., as $\theta_s$ increased, direct and indirect climate-forcing emissions increased and decreased, respectively). Accounting for stratospheric ozone depletion likely resulted in the reverse direction trends. While some of the parameters that correlate to emissions have potential for engineering control during design of cover systems (e.g., $\theta_s$ or $e$), others do not (water content parameters and temperature). The interrelationships between particle packing arrangements, void structures, and diffuse double layer effects (for soils containing clays) with transport and transformation of chemical species through covers as well as the resulting direct and indirect climate forcing emissions warrant further research.

The soil parameters correlated to direct emissions also were correlated to indirect emissions. The correlations between site-specific geotechnical properties and indirect climate-forcing emissions are presented in Figure 2 for both GHGs and NMVOCs.

![Fig. 1. Selected correlations between geotechnical properties and direct emissions for GHGs (a-d) and NMVOCs (e-h).](image1)

![Fig. 2. Selected correlations between geotechnical properties and indirect emissions for GHGs (a-d) and NMVOCs (e-h).](image2)
4 CONCLUSIONS

Direct and indirect climate effects of landfill gases including the main landfill gas methane (known GHG), trace gases nitrous oxide and F-gases (both known GHGs), and trace NMVOCs were investigated in this study. Extensive testing was conducted across 5 California landfills and two seasons (wet and dry). Interrelationships between direct and indirect climate-forcing emissions and cover soil characteristics were analyzed.

Methane emissions accounted for 46 to 99% of the total direct climate-forcing emissions. Nitrous oxide and F-gases collectively accounted for 1 to 99% of the total direct climate-forcing emissions. Overall direct emissions accounted for 93 to nearly 100% of the total combined (direct and indirect) climate-forcing emissions. While the inventories used in climate change assessments typically include only methane emissions, this investigation identified the potential significant emissions contributions from trace landfill gases including nitrous oxide and F-gases. Statistically significant correlations were observed a) between direct and indirect climate-forcing emissions of GHGs and water content parameters as well as void ratio and b) between direct and indirect climate-forcing emissions of NMVOCs and mass of water, volumetric air content, temperature, and volumetric solids content. The examples provided herein demonstrate the complex coupling between larger scale indirect and direct climate forcing and site-specific geotechnical properties. Controlling geotechnical properties during design, construction, and operation of landfills may have a considerable impact on global climate change.

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