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**RESEARCH ARTICLE**

# Alternative carbon dioxide modelling approaches accounting for high residual gases in LandGEM

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**Abstract** High Canadian waste disposal rates necessitate landfill gas monitoring and accurate forecasting. $CO_2$ estimates in LandGEM version 3.02 currently rest on the assumptions that $CO_2$ is a function of $CH_4$, where the two gases make up nearly 100% of landfill gas content, leading to overestimated $CO_2$ collection estimates. A total of 25 cases (five formulas, five approaches) compared annual $CO_2$ collection at four western Canadian landfills. Despite common use in literature, the 1:1 ratio of $CH_4$ to $CO_2$ was not recommended to forecast landfill gas collection in cold climates. The existing modelling approach significantly overestimated $CO_2$ production in three of four sites, resulting in the highest residual sum of squares. Optimization resulted in the most accurate results for all formulas and approaches, which had the greatest reduction in residual sums of squares (RSS) over the default approach (60.1 to 97.7%). The 1.4 Ratio approach for $L_o{:}L_{o\text{-}CO2}$ yielded the second most accurate results for $CO_2$ flow (mean RSS reduction of 50.2% for all sites and subsection models). The annual $k$-modified LandGEM calculated $k$'s via two empirical formulas (based on precipitation) and yielded the lowest accuracy in 12 of 20 approaches. Unlike other studies, strong relationships between optimized annual $k$'s and precipitation were not observed.

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## Introduction

### Land disposal and landfill gas

Canada's municipal solid waste (MSW) generation rate is among the highest in the world (Bruce et al. 2016; Richter et al. 2017). Per capita non-hazardous waste generation ranged between 876 and 961 kg per year between 1996 and 2010, with a peak of 1033 kg in 2006 (Bruce et al. 2015; Wang et al. 2016). Permanent land disposal is widely practiced, and thus, proper landfill management and planning remains a priority. In Canada, landfill gas (LFG) collection remains an underutilized technology due to project costs and a low population density (4 vs. 35 cap/km$^2$ in the USA) (World Bank Group 2016); only 52 landfills across the vast country (9.985M km$^2$ land) operated LFG collection systems by the mid-2000s (Thompson et al. 2009). Additionally, accurate LFG field measurement can be expensive, and accurate modelling remains a challenge.

LFG is generated under anaerobic conditions within landfills during operation, and generation continues for decades after final closure, depending on site conditions and organic content of waste. The major components of LFG are the common greenhouse gases methane ($CH_4$) and carbon dioxide ($CO_2$), along with other residual gases including nitrogen ($N_2$), oxygen ($O_2$), and non-methane organic compounds (NMOCs). Field gas data often shows that $O_2$ and $N_2$ concentrations make up the bulk of residual gas concentrations, suggesting minor to significant air intrusion (Aguilar-Virgen et al. 2014; Tolaymat et al. 2010) is a common issue in collection systems. LFG collection systems are used primarily for economic benefits, to mitigate emissions that pose health and safety risk to humans, and to reduce global warming potential in the environment.

Early work on LFG production by Barlaz et al. (1989a) used an idealized chemical equation (Table 1) for anaerobic

decay and compared the methane potential of cellulose, hemi-cellulose, protein, and sugar. The study found that cellulose and hemicellulose accounted for 91.1% of methane potential in MSW, and their early work may have contributed to the broad modelling assumption that the ratio between $CH_4$ and $CO_2$ is nearly 1:1 as per the ideal decay products, which is less suitable when forecasting field LFG collection due to $CO_2$'s higher solubility. Barlaz et al. (1989a) cautioned that the lab conditions, including shredded waste, leachate recycling, and relatively homogeneous materials, may have overestimated $CH_4$ yields.

Table 1 summarizes the results of other studies on $CH_4$, $CO_2$, and residual gas ratios in LFG. The field studies' average ratio between $CH_4$ and $CO_2$ was 1.4:1, while theoretical and lab basis yielded 1.2:1.

## $CH_4$ modelling

A common focus in LFG studies is $CH_4$, which has higher volatility than $CO_2$, more utility in heating and electricity projects, and higher global warming potential (28 $CO_2$e, 100-year horizon) (Myhre et al. 2013). This focus on $CH_4$ has resulted in multiple first-order decay (FOD) models of varying complexity which estimate only $CH_4$ generation (i.e. the Intergovernmental Panel on Climate Change (IPCC) model) or LFG components (such as $CO_2$, NMOCs) as a function of $CH_4$ generation (i.e. Afvalzorg model, LandGEM model). A

remaining issue with these generation models is that they are difficult to calibrate due to the size and complexity of sites, and the operational issues with collection systems. Assumed collection efficiencies are often used in order to approximate LFG generation from collection data (Amini et al. 2013; Maciel and Juca 2011).

LandGEM version 3.02 (Alexander et al. 2005) assumes that nearly all degradable carbon degrades equally into $CH_4$ and $CO_2$. $CH_4$ estimates are largely sensitive to site-specific or selected variables "$k$" ($CH_4$ generation rate constant, or decay constant) and "$L_o$" ($CH_4$ generation potential) (Aguilar-Virgen et al. 2014; Amini et al. 2013; Machado et al. 2009). The decay rate $k$ is dependent on waste moisture content and precipitation rates, nutrients, pH, bacterial culture, and waste temperature. $L_o$ largely depends on the amount and type of organic waste, which changes slowly over time due to shifts in disposal trends (Thompson et al. 2009). Although $L_o$ is traditionally understood in terms of $CH_4$, it can be adapted for use in $CO_2$ modelling ($L_{o-CO2}$) due to their similar substrates (Table 1); however, lab studies are required to develop and verify a range of reasonable $L_{o-CO2}$ values.

## LandGEM $CO_2$ modelling and objectives

Few LFG studies focus on $CO_2$. Several recent studies utilize LandGEM for various applications, such as cost analyses and

**Table 1** Observed methane:carbon dioxide:residual gas ratios at various locations

| Basis | Location | Notes | $CH_4$:$CO_2$:residual (% v/v) | Reference |
|---|---|---|---|---|
| Field study | Recife City, Brazil | 5 vertical wells, passive ventilation 9-m-thick cell Reported as STP | 54.3:40.7:5.0 | Maciel and Juca (2011) |
| | Ensenada, Mexico | Well sampling at well 21 days in fall, 3 readings/day Well 3 was deepest, low intrusion Wells 1, 2, 4 had severe intrusion Varied temp. | 55.1:44.4:0.5 (well 3) 42.2:33.4:24.4 (wells 1, 2, 4) | Aguilar-Virgen et al. (2014) |
| | Winnipeg, Canada | Five 10-m probes (to cell bottom) 15 samples biweekly for 1 year Varied temp. and pressure | 56.1:40.1:3.8 (average) | Thompson and Tanapat (2005) |
| | Louisville, USA | 1-m clay-capped control cell Daily cover on bioreactor cell Vertical gas collection wells Unknown temp. and pressure | 58:41:1 (cc, low change) 51: 37: 12 (bioreactor) | Tolaymat et al. (2010) |
| Empirical or lab (at STP) | $C_aH_bO_cN_d + [(4a - b - 2c + 3d)/4]$ $H_2O$ ➔ $[(4a + b - 2c - 3d)/8]$ $CH_4 + [(4a - b + 2c + 3d)/8]$ $CO_2$ | | | Barlaz et al. (1989a), Tchobanoglous and Kreith (2002) |
| | Cellulose $C_6H_{10}O_5 + H_2O$ ➔ $3CO_2 + 3CH_4$ | | 50.1:49.9:0 | |
| | Protein $C_{3.2}H_5ON_{0.86}$ ➔ $1.548CO_2 + 1.653CH_4$ | | 51.7:48.2:0 | |
| | Approx. organic fraction in MSW $C_{99}H_{149}O_{59}N$ ➔ $53CO_2 + 46CH_4 + NH_3$ | | 54.0:46.0:0 Assumes 50% biodegradable | Worrell and Vesilind (2012) |
| | Wisconsin (lab) | 2-L reactors Shredded refuse, leachate recycling | 58.1:41.9:0 (111 days old) | Barlaz et al. (1989b) |

model comparisons, and assume the 1:1 ratio in their models (Calabro et al. 2011; Chalvatzaki and Lazaridis 2010; Goswami et al. 2011; Kumar and Sharma 2014; Marroni et al. 2010; Rezaee et al. 2014). The assumption is used for simplicity, despite expected ranges of 30 to 45% $CO_2$ (Abushammala et al. 2012; Ahmed et al. 2015; Saquing et al. 2014).

Models using field $CH_4$ content can also overestimate $CO_2$ generation in curve fitting and forecast applications due to the governing formula being a function of $CH_4$ content. Lower field $CH_4$ content is observed at sites with significant air intrusion due to damaged collection lines (Guter and Nuerenberg 1987) and shallow or permeable cover, which causes partial aerobic production of $CO_2$ (Jeong et al. 2015), as well as increased $N_2$ content. Final cover systems in Canada are more susceptible to degradation due to cold climate freeze-thaw mechanisms. Both factors result in lowered $CH_4$ content and increased $N_2$, the latter of which will be erroneously reported as increased $CO_2$ under the default LandGEM assumption: $CH_4$ and $CO_2$ represent almost 100% of LFG composition. These overestimations serve as poor forecasts for collected $CO_2$, which affects flares and LFG energy projects: higher $CO_2$ content results in lower efficiencies (Lee and Hwang 2007), which may affect collection system pre-design.

The three issues are thus a lack of $CO_2$ LFG modelling studies, an unrealistic assumption of 1:1 $CH_4$ to $CO_2$ ratio in LFG studies, and the effects of residual gases on $CO_2$ estimates in LandGEM. To address these issues, the study objective is to compare the effects of residual gases on $CO_2$ collection estimates for five different input approaches in LandGEM version 3.02: (i) default LandGEM assumption (Total LFG = $CH_4$ + $CO_2$) with field $CH_4$ content; (ii) default LandGEM assumption with adjusted $CH_4$ content; (iii) $CH_4$ to $CO_2$ $L_o$ ratio of 1.2 (as observed from lab studies); (iv) $CH_4$ to $CO_2$ $L_o$ ratio of 1.4 (as observed from field studies and the four study sites); and (v) optimized $k$ and $L_o$ using site-specific data. $CO_2$ generation estimates were calculated using formulas dependent and independent of $CH_4$ content to establish a ranking by accuracy. Five variations of LandGEM's governing formula (four with different divisors, one with annual $k$'s) were used to determine whether modifications affected $CO_2$ model results. Two empirical $k$ formulas were also evaluated with an annual $k$-modified LandGEM.

## Methodology

### Site history and details

Four landfills in western Canada with active LFG collection systems were studied. Three of the four sites were active at the end of the study periods (HL, CC, and SK). Three of the sites

tended to collect significant amounts of residual gases (Fig. 1). SK had the lowest range (0.4 to 6.5% monthly), and HL had the highest (7.8 to 28.2%). CC (7 to 13%) and RE (5.8 to 19.8%) were similarly high, increasing the uncertainty when using average collected $CH_4$ content in LandGEM. To verify this, LandGEM was run using average measured $CH_4$ content for each site (denoted "Default w/ Residuals") and compared to results which assumed total collected LFG was the sum of $CH_4$ and $CO_2$ (denoted "Default w/o Residuals"). $CO_2$ data were unavailable in 2009 at CC, 2014 at RE, and 2008 at HL, while $CH_4$ data were still available.

LFG was measured at each site using continuous gas analyzer systems. All wells (SK), or a subset of control wells (CC, HL) were monitored monthly by operators to minimize air intrusion. Collection efficiencies were based on designed cover systems and the B.C. Ministry of Environment's LFG generation model. The ranges of collection efficiencies were estimated from field reports and other published values from literature. Waste data for the landfills were collected and verified from available annual reports. Degradable organic carbon (DOC) was calculated using available waste audit data and Eq. 1:

$$DOC = \frac{0.4A + 0.17B + 0.15C + 0.3D}{E} \quad (1)$$

where $A$ = mass of paper and textile waste; $B$ = mass of garden, park, and putrescible waste; $C$ = mass of food waste; $D$ = mass of wood and straw waste; and $E$ = total mass of waste audited.

Annual DOC values were averaged to calculate site-specific $L_o$. Study and site-related data are available in Table 2.

### Cache Creek

The Cache Creek (CC) landfill accepts much of its waste from the Metro Vancouver region; only 3% of the waste disposed in 2014 came from other sources (Golder Associates 2015). MSW composition data were gathered via Metro Vancouver audit reports between 2010 and 2015. Phases 1–3 were topped with final cover, while approximately half of phase 4 had been covered as of 2014 (Golder Associates 2015). Leachate was applied to the surface to facilitate partial evaporation between 2005 and 2009. Average $CH_4$ and $CO_2$ for the study period were 53.6 and 36.9%, respectively. Average total flow rates ranged from 18,000 to 65,000 $m^3$/day since the landfill is active and expanded almost every year during the study period. Gas collection efficiencies were reported each year between 2011 and 2015 (ranging 65–87%), and a conservative estimate of 50% was assumed between 2005 and 2010 (Ahmed et al. 2015; Spokas et al. 2006) due to cases of model divergence in early work using values between 70 and 80%.

**Fig. 1** Average landfill gas composition for four western Canadian sites. **a** CC. **b** SK. **c** RE. **d** HL
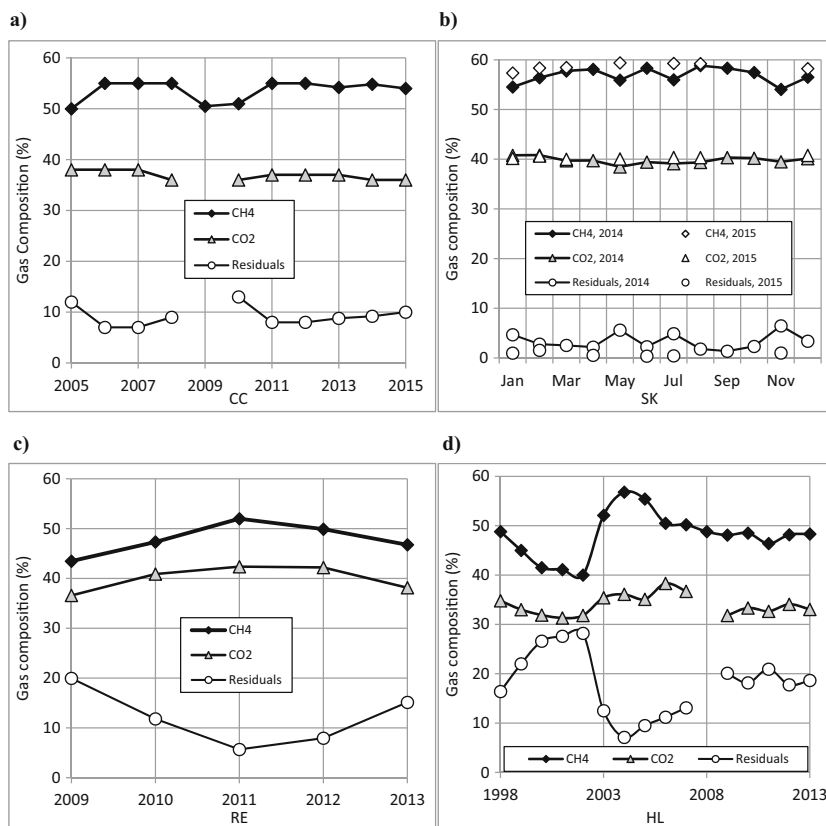


### Saskatoon

The Saskatoon (SK) landfill's well field is spread along the central and northern sections of old cells, which have had little disposal since 2004. Further disposal occurred in 2010 and 2013, with minimal disposal in 2015. Reported gas measurements were taken on average 2.3 days per month, with some missing months. Each well was sampled an average of once per month for $CH_4$, $CO_2$, $O_2$, and LFG flow. Average flow rates ranged between 15,740 and 20,390 $m^3$/day, and the average $CH_4$ and $CO_2$ for the study period were 57.7 and 40.1%, respectively. A collection efficiency of 70% was used, as the design basis report planned final cover to be constructed soon after the well field (Comcor Environmental Ltd. 2010).

### Regina

The Regina (RE) landfill's well field consists solely of vertical wells and did not change during the study period. The well field footprint covers 38% of phase 1 (1961–2011) and is spread across the northern cells. The final cover for this section was constructed in 2007 (Conestoga-Rovers and Associates 2006). Cells south of the well field remained uncovered until cell closure in 2011.

Composition data were based on a 12-month audit conducted by Canart and McMartin (2009) starting in October 2007. Uniquely among the four sites, per-minute gas data

were available for $CH_4$, $CO_2$, $O_2$, and total gas between August 2008 and December 2013 and daily $CH_4$ data in 2014. Annual gas collection was calculated using 2009 of 2198 days (91.4%), with values removed for negative composition measurements due to scheduled maintenance. LFG collection efficiencies were assumed to be 70 (for years 2009–2011) and 80% (2012–2014) in order to account for the varied cover over the cells south of the well field. Average daily flow rates for the entire system ranged between 12,380 and 15,130 $m^3$/day. Average $CH_4$ (48%) and $CO_2$ (40%) during the study period were on the low end of expected ranges from the literature for $CH_4$ (45–60%), but on the higher end for $CO_2$ (30–45%). This is likely due to the young age of the landfill and that the study period included both semi-open and closed years.

### Hartland

Hartland (HL) was privately owned and operated from the early 1950s (assumed 1951 in the present study) until 1985. Vertical LFG collection wells were installed in old cells in 1990. The well field and final cover area expanded during the study period, which was reflected in the increased collection efficiencies reported (range of 28.5–80.8%). A leachate recirculation pilot project was initiated in 2002. This coastal site currently covers 36 ha, with a projected final footprint of 46 ha.

**Table 2** Site conditions at four study landfills

| Site | Area (phase) (ha) | Lifetime (years) | CO₂ data | MSW composition data (years) | Final cover design | Collection efficiency (%) | Ave. precipitation (mm/year) |
|------|------|------|------|------|------|------|------|
| CC | 49.6 (1–4) 6.7 (5) | 1989–present | 2005–2015 | 2010–2015 (4) | 1 m low-permeability soil, 0.15 m topsoil with vegetation | 50 (2005–2010), 65–87 (2011–2015) | 328 |
| SK | 27 (1) 9 (2) | 1955–2015 | 2014–2015 | 2006, 2013, 2014 | 1–1.2 m low-permeability soil, 0.15 m topsoil | 70 | 358 |
| RE | 16 (wells) 43 (1) | 1961–2011 | 2009–2013 | 2007–2008 (1) | 1 m clay, 0.15 m topsoil with vegetation | 70 (2009–2011), 80 (2012–2014) | 411 |
| HL | 20 (1) 26 (2) | 1951 (assume)–present | 1998–2013 | 1990–2010 (5) | 1 m compacted clay, 0.15 m topsoil, geo-membrane | 28.5–80.8 | 886 |

Waste disposal data were limited to the period of 1980 to 2013. Estimates for the remaining period were based on an estimated waste in place value of 6.3 Mt at the end of 2011 (Fillipone et al. 2012) and an exponential growth relationship derived using waste trends between 1980 and 1989 ($R^2$ = 0.865, data not shown). Average total flow rates ranged from 19,720 to 48,160 m³/day. Average $CH_4$ and $CO_2$ for the study period were 48.1 and 34.0%, respectively.

### Formulas for $k$ and $L_o$

Some studies have supported formulas relating precipitation and the decay rate, $k$, in FOD models (Environment Canada 2014; McDougall and Pyrah 1999; Thompson et al. 2009). Thompson et al. (2009) used Eq. 2, which was developed by the Research Triangle Institute (RTI) based on US EPA default $k$'s, in their study:

$$k = 3.2 \cdot 10^{-5} \cdot (x) + 0.01 \tag{2}$$

where $k$ = decay rate (year$^{-1}$) and $x$ = annual average precipitation (mm). Environment Canada (2014) supported the use of a similar equation developed by the RTI:

$$k = 7 \cdot 10^{-5} \cdot (x) - 0.0172 \tag{3}$$

In this study, Eqs. 2 and 3 were used to calculate annual $k$ values with precipitation data gathered from Environment Canada's weather archives. If the main weather stations were discontinued, then the nearest station was used (HL in 2007, CC in 2015). All $k$'s were positive except for 2 years in CC using Eq. 3 (2009 and 2015). The two negative $k$ values were arbitrarily set to a low value (0.00001 year$^{-1}$) to use a positive number while representing the formula's low estimate. The equations yielded a wide range of $k$'s (Eq. 2: 0.016 to 0.048 year$^{-1}$; Eq. 3: 0.00001 to 0.067 year$^{-1}$).

The IPCC provided a common formula for calculating $L_o$ from waste audit data, which has since been used in some studies (IPCC 1996; Thompson and Tanapat 2005; Aguilar-Virgen et al. 2014; Environment Canada 2014):

$$L_o = F \times MCF \times DOC \times DOC_f \times 16/12 \tag{4}$$

where $L_o$ [Mg $CH_4$/Mg of waste], $F$ = average $CH_4$ content in LFG [fraction], MCF = $CH_4$ correction factor [1.0 for maintained landfills], $DOC_f$ = 0.50 or 0.77 [fraction, 2006 and 1996 defaults], 16/12 = molecular mass conversion factor [Mg $CH_4$/Mg C], and DOC = degradable organic carbon [Mg C/Mg of waste]. Equation 4 was used with $DOC_f$ values of 0.50 and 0.77 as lower and upper bounds, respectively (Thompson et al. 2009). Table 3 summarizes calculated $k$ and $L_o$ data for the four sites, including the $L_{o\text{-}CO2}$ values used in the ratio approaches (see "Simple ratios for $L_{o\text{-}CO2}$").

### Modified LandGEM formulas and optimization procedures

Two groups of variations on the existing FOD, Scholl Canyon-type formula were generated for study: subsections and annual $k$'s (Table 4). The subsections group differ only by the time increment selected, where $j$ = 1, 0.5, 0.25, and 0.1 (Thompson et al. 2009). Lower time increments tend to reduce the LFG estimates by less than 5% (Alexander et al. 2005), depending on site-specific $k$, which tends to be lower in the cold study climates.

Annual $k$'s are used in place of the lifetime $k$ values in order to account for annual differences in infiltration, using precipitation rates as an approximation. The same time increment "$j$" was kept as the current LandGEM formula to simplify comparison. It was assumed that $k$ would be equal for both $CH_4$ and $CO_2$ given that it represents the rate of biodegradation,

**Table 3** Site-specific data and values for LandGEM $CO_2$ modelling

| Site | $k$ (year$^{-1}$) | | CH$_4$ $L_o$$^a$ (m$^3$/t) | | 1.4 Ratio $L_{o\text{-}CO2}$ (m$^3$/t) | | 1.2 Ratio $L_{o\text{-}CO2}$ (m$^3$/t) | |
|---|---|---|---|---|---|---|---|---|
| | LandGEM default | Average $k$ (Eq. 3) $\pm0.001$ | $L_o$ (0.5) | $L_o$ (0.77) | $L_{o\text{-}CO2}$ (0.5) | $L_{o\text{-}CO2}$ (0.77) | $L_{o\text{-}CO2}$ (0.5) | $L_{o\text{-}CO2}$ (0.77) |
| CC | 0.020 | 0.006 | 82.8 | 127.6 | 59.2 | 91.1 | 69.0 | 106.3 |
| SK | 0.020 | 0.008 | 97.4 | 150.0 | 69.6 | 107.1 | 81.1 | 125.0 |
| RE | 0.020 | 0.012 | 86.2 | 132.7 | 61.6 | 94.8 | 71.8 | 110.6 |
| HL | 0.040 | 0.045 | 83.3 | 128.3 | 59.5 | 91.7 | 69.5 | 107.0 |

$^a$ CH$_4$ $L_o$ used as starting values in the optimization algorithms to determine ideal $k$

and not compound-specific measures. This assumption exists in the current LandGEM model. The non-optimized approaches (Default w/o Residuals, Default w/ Residuals, 1.2 Ratio, and 1.4 Ratio) tested the utility of the $k$ equations (2 and 3) in simple, non-optimized annual $k$ LandGEM.

The algorithms used to determine optimized parameters (Fig. 2) minimized the residual sums of squares (RSS) between model estimates and collected data using the Solver function in Excel. Multiple starting values for $k$ and $L_o$ were used to reduce bias in the resulting optimized values. CH$_4$ data were used to calculate optimized $k$'s for two reasons: (i) site-specific $L_o$ values were available and (ii) all sites except SK had more data for CH$_4$ than CO$_2$, thus improving the reliability of optimized $k$. Method A used two starting sets to determine preferred starting $k$ and DOC$_f$ values for each site. Method B used one starting set, as the dual-cell change algorithm was unaffected by starting values.

Methods C and D used four starting sets. The approaches differed in which variable ($k$, $L_o$) was changed first to identify the more accurate algorithm. Methods B and D were subject to

boundary constraints given difficulties with divergence in the literature (Amini et al. 2012), CH$_4$ $L_o$ values were bound between 90% of $L_o$ (0.5) and 110% of $L_o$ (0.77), due to the uncertainty of using default DOC$_f$ values. Meanwhile, $k$ values were conservatively bound between 0.0005 and 0.4 year$^{-1}$, values appropriate for more extreme climates (or leachate recirculation operations) than those in this study. These boundaries were selected in order to account for annual shifts in precipitation and the effect of the small leachate recirculation projects in CC and HL. According to Alexander et al. (2005), the LandGEM default $k$ for full-scale leachate recirculation landfills was 0.7 year$^{-1}$.
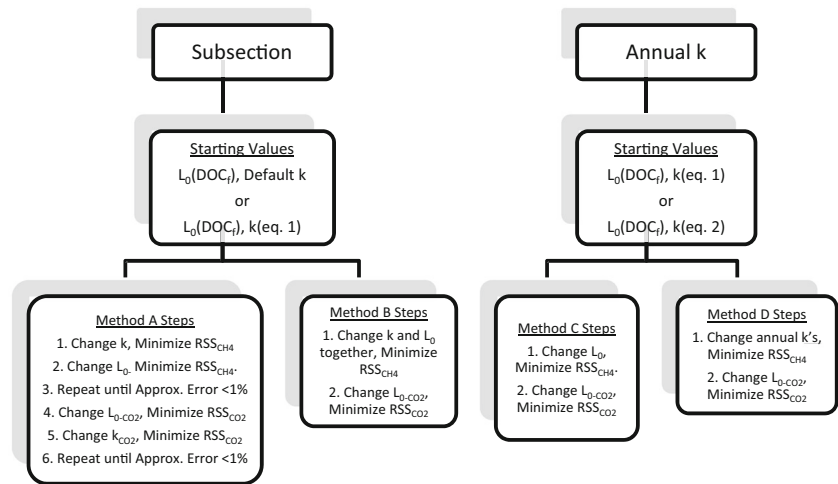
### Simple ratios for $L_{o\text{-}CO2}$

The four sites' CH$_4$ to CO$_2$ ratios were similar save for RE's (1.20:1), which was closer to the average predicted from empirical/lab methods (Table 1). CC (1.45:1), SK (1.44:1), RE (1.20:1), and HL (1.42:1) had an average ratio of 1.38:1, a result close to the average field study ratio (1.4 from Table 1)

**Table 4** Default and modified LandGEM $CO_2$ modelling formulas

| Formula modification | Derivation (names) | Formula | Terms |
|---|---|---|---|
| Subsection | LandGEM 3.02 (Sub-10) | $Q_{CO2} = \sum\limits_{i=1}^{n} \sum\limits_{j=0.1}^{1} k L_o \frac{M_i}{10} e^{-kt_{ij}}$ | $Q_{CO2}$ = CO$_2$ generation rate (m$^3$/year) <br> $i$ = 1-year time increment <br> $n$ = (year of calculation) − (initial year of waste acceptance) |
| | Thompson et al. (2009) (Sub-4, Sub-2) | $Q_{CO2} = \sum\limits_{i=1}^{n} \sum\limits_{j=0.25}^{1} k L_o \frac{M_i}{4} e^{-kt_{ij}}$ | $j$ = 0.1-year time increment <br> $k$ = methane generation rate (year$^{-1}$) <br> $k_i$ = $k$ for the $i$th year (year$^{-1}$) |
| | | $Q_{CO2} = \sum\limits_{i=1}^{n} \sum\limits_{j=0.5}^{1} k L_o \frac{M_i}{2} e^{-kt_{ij}}$ | $L_o$ = potential CO$_2$ generation capacity (m$^3$/Mg) <br> $M_i$ = mass of waste accepted in the $i$th year (Mg) <br> $t_{ij}$ = age of the $j$th section of waste mass $M_i$ accepted in the $i$th year (decimal years) |
| | LandGEM 2.01 (Sub-1) | $Q_{CO2} = \sum\limits_{i=1}^{n} k L_o M_i e^{-kt_i}$ | |
| Annual $k$ | This study (annual k) | $Q_{CO2} = \sum\limits_{i=1}^{n} \sum\limits_{j=0.1}^{1} k_i L_o \frac{M_i}{10} e^{-k_i t_{ij}}$ | |
| Current | LandGEM 3.02 (default) | $Q_{CO2} = Q_{CH4} \cdot \left( \frac{100}{P_{CH4}} - 1 \right)$ | $Q_{CO2}$ = CO$_2$ flow rate (m$^3$/year) <br> $P_{CH4}$ = methane content (%) |

**Fig. 2** RSS optimization modelling algorithms



in the literature. The formulas and starting values in "Formulas for $k$ and $L_o$" were thus repeated using conservative ratios of 1.2 and 1.4 in order to determine whether these simple, non-optimized approximations yielded $CO_2$ estimates more accurate than raw content LandGEM estimates. The method thus consists of scaling down known $L_o$ to produce $L_{o\text{-}CO2}$, which can be input into LandGEM formulas, according to the following simple equation:

$$L_{o\text{–}CO2} = \frac{L_o}{r} \qquad (5)$$

where $r$ = the pre-design collection ratio between $CH_4$ and $CO_2$.

## Results and discussion

Table 5 presents the comparison of the best starting set results from each combination of approaches and governing equations relative to the LandGEM default Sub-10 approach using field $CH_4$ content (the Default w/ Residuals set). Results were compared to this approach since it was expected to produce the highest RSSs due to overestimation and did so for sites with longer study periods (HL and CC). SK had exceptionally poor accuracy with the 1.2 Ratio approach, while RE resulted in low losses of accuracy in both the 1.2 Ratio and Default w/o Residuals approaches.

Overall, the optimization approach led to the most accurate results with an average RSS reduction of 60.2% for subsection formulas (Sub-1, Sub-2, Sub-4, and Sub-10) and 97.7% reduction for "annual $k$" models. This was expected due to the inherent advantage of numerical methods over assumed ratios; however, the application of optimized results is limited to improving collection forecasting, not pre-design. On the other hand, the 1.4 Ratio approach is more applicable for collection system pre-design given reliable $k$ and waste composition

data. The mean RSS decrease between the subsection formulas was 50.2% for this approach.

The SK results produced low improvements (aside from annual $k$ formulas) and tended towards high reductions in accuracy for Default w/o Residuals and 1.2 Ratio approaches. This was likely due to it having the shortest available study period (2 years), and monthly LFG flow experienced more fluctuation in 2014 (6.6 to 17.8 $m^3$/min) than in 2015 (12.3 to 13.0 $m^3$/min). By removing the SK results, the 1.2 Ratio approach (mean RSS reduction 22.9% for all formulas) becomes comparable with the Default w/o Residuals approach (19.2% RSS reduction). Overall, the optimized and 1.4 Ratio approaches were the first and second most accurate, and the annual $k$ formulas yielded the highest increases rather than reductions in accuracy. Thus, the Ratio approaches can be considered comparable or more accurate than assuming 0% residual gases. The results in Table 5 will be discussed in the following sections in more detail.

### Default approaches

As expected, the Default w/ Residuals approach led to overestimated $CO_2$ for all but 2 years (SK, Fig. 3b, and RE, Fig. 3c). The optimized subsection formulas for all sites (Table 5) ranged from 13.0 to 97.2% lower RSS than the existing approach. CC uniquely had the "optimized" (using the Sub-10 formula) and Default w/o Residuals approaches' results overlap each other closest to the actual data for most of the study period. SK, however, yielded a higher amount of actual gas in 2015 due to low total gas collection flow rates in 2014 (triangle symbols, Fig. 3b), thus skewing the results for optimized models due to the short study period.

As an exception, the Default w/o Residuals approach consistently underestimated the actual gas data in HL (Fig. 3d), although the resulting RSS was 30.7% lower than that of the Default w/ Residuals approach. The shift was likely due to the different preferred $k$'s (0.040 and 0.045 $year^{-1}$): Default w/

**Table 5** Lowest RSSs from studied $CO_2$ approaches and models

| Site | Approach | $CO_2$ (%) | Opt. $k$ (year$^{-1}$)[a] ±0.001 | $L_{o\text{-}CO2}$ (m$^3$/t) | Percent decrease in RSS (%)[b] | | | | |
|------|----------|-----------|----------------------------------|------------------------------|--------|--------|--------|---------|----------|
| | | | | | Sub-1 | Sub-2 | Sub-4 | Sub-10 | Annual $k$ |
| CC | Default w/o Residuals | 40.8 | 0.020 | (82.8)[c] | 84.8 | 84.8 | 84.8 | 84.8 | −23.2 |
| | Default w/ Residuals | 36.9 | 0.020 | | −7.8 | −3.4 | −1.3 | 0.0 | −208 |
| | Optimized | | 0.011 | 95.5 | 88.3 | 88.3 | 88.2 | 88.2 | 98.7 |
| | 1.4 Ratio | | 0.020 | 59.2 | 82.4 | 83.0 | 83.2 | 83.4 | 68.8 |
| | 1.2 Ratio | | 0.020 | 69.0 | 22.7 | 26.1 | 27.8 | 28.8 | −19.8 |
| SK | Default w/o Residuals | 41.0 | 0.020 | (150.0)[c] | −40.5 | −46.5 | −49.7 | −51.6 | −1441 |
| | Default w/ Residuals | 40.1 | 0.020 | | −5.9 | −2.5 | −0.9 | 0.0 | −2613 |
| | Optimized | | 0.007 | 115 | 13.1 | 13.0 | 13.0 | 13.0 | 95.4 |
| | 1.4 Ratio | | 0.020 | 81.1 | 10.6 | 9.5 | 8.8 | 8.4 | −2019 |
| | 1.2 Ratio | | 0.008 | 125 | −1182 | −1148 | −1132 | −1122 | −7033 |
| RE | Default w/o Residuals | 45.5 | 0.012 | (86.2) | −4.6 | −6.4 | −7.3 | −7.8 | −197 |
| | Default w/ Residuals | 40.0 | 0.012 | | −4.1 | −1.8 | −0.7 | 0.0 | −430 |
| | Optimized | | 0.016 | 66.6 | 42.2 | 42.2 | 42.2 | 42.2 | 98.6 |
| | 1.4 Ratio | | 0.020 | 61.6 | 26.6 | 28.8 | 29.8 | 30.4 | 17.0 |
| | 1.2 Ratio | | 0.020 | 71.8 | −5.2 | −7.0 | −7.9 | −8.5 | −195 |
| HL | Default w/o Residuals | 41.4 | 0.045 | (83.3)[c] | 40.0 | 38.9 | 38.2 | 30.7 | 47.4 |
| | Default w/ Residuals | 34.0 | 0.040 | | −44.2 | −37.5 | −34.3 | 0.0 | −13.8 |
| | Optimized | | 0.049 | 67.1 | 97.2 | 97.2 | 97.2 | 96.8 | 98.0 |
| | 1.4 Ratio | | 0.045 | 59.5 | 84.8 | 82.8 | 81.7 | 69.4 | 81.5 |
| | 1.2 Ratio | | 0.045 | 69.4 | 96.9 | 97.2 | 97.2 | 94.8 | 95.4 |
| Mean | Default w/o Residuals | 42.1 | | | 19.9 | 17.7 | 16.5 | 14.0 | −403.4 |
| | Default w/ Residuals | 37.7 | | | −15.5 | −11.3 | −9.3 | 0.0 | −816.3 |
| | Optimized | | | | 60.2 | 60.2 | 60.2 | 60.1 | 97.7 |
| | 1.4 Ratio | | | | 51.1 | 51.0 | 50.9 | 47.9 | −462.9 |
| | 1.2 Ratio | | | | −266.8 | −258.0 | −253.6 | −251.7 | −1788 |

[a] Optimized approach $k$ reported as average from all formulas, all other approaches were starting $k$

[b] Reported in terms of reduction from the Default w/ Residuals approach (Sub-10 model)

[c] Values in parentheses are $CH_4$ $L_o$

Residuals used the lower bound $k$ (Table 5) to offset the over-estimation tendency of the approach. Unlike HL, all other sites had equal $k$'s for both approaches, possibly due to shorter study periods.

For each site, the Sub-10 formula yielded the lowest RSS for the Default w/ Residuals approach. The mean RSS (considering all sites) increased by 7.1 to 64.3% using the subsection and annual $k$ modifications. This is consistent with Alexander et al. (2005), who reported that the higher divisor (Sub-10) yields 1–2% lower estimates than those using Sub-1 for $CH_4$. Since the Default w/ Residuals tends to overestimate $CO_2$, the Sub-10 formula's systematic lower estimates tend to be more appropriate.

### Optimized approach

The optimization process resulted in identical $k$ and $L_{o\text{-}CO2}$ values in preliminary work for starting sets with equal $L_o$

values ($k$ in the case of method C, Fig. 2). This is the result of the pseudo-second-order algorithm's first step, where one parameter, usually $k$, was changed before the other. This affected both subsection and annual $k$ models. The magnitude of the first changed parameter was insignificant, as the algorithm was dependent on the unchanged parameter. For instance, when $k$ was changed first in Method D, optimized $k$'s were equal for both starting equations for the same $L_o$ ($DOC_f$).

The Canadian field data (Fig. 1) suggests that 1:1 $CH_4$ to $CO_2$ ratios are uncommon in LFG collection, resulting in optimized $L_{o\text{-}CO2}$ values consistently lower than those of $L_o$. This may be partly due to significant differences in solubility (Jeong et al. 2015), as $CO_2$ is 22 times more soluble than $CH_4$ at 35 °C (Gevantman n.d.). Solubility is not a factor in LandGEM, as the model estimates LFG generation rather than collection, and the collected gas is often presented as an assumed fraction of generation. While this may be more appropriate for $CH_4$ estimates, the complexity incurred by $CO_2$'s

**Fig. 3** Comparison of default modelling approaches with actual data. **a** CC. **b** SK. **c** RE. **d** HL
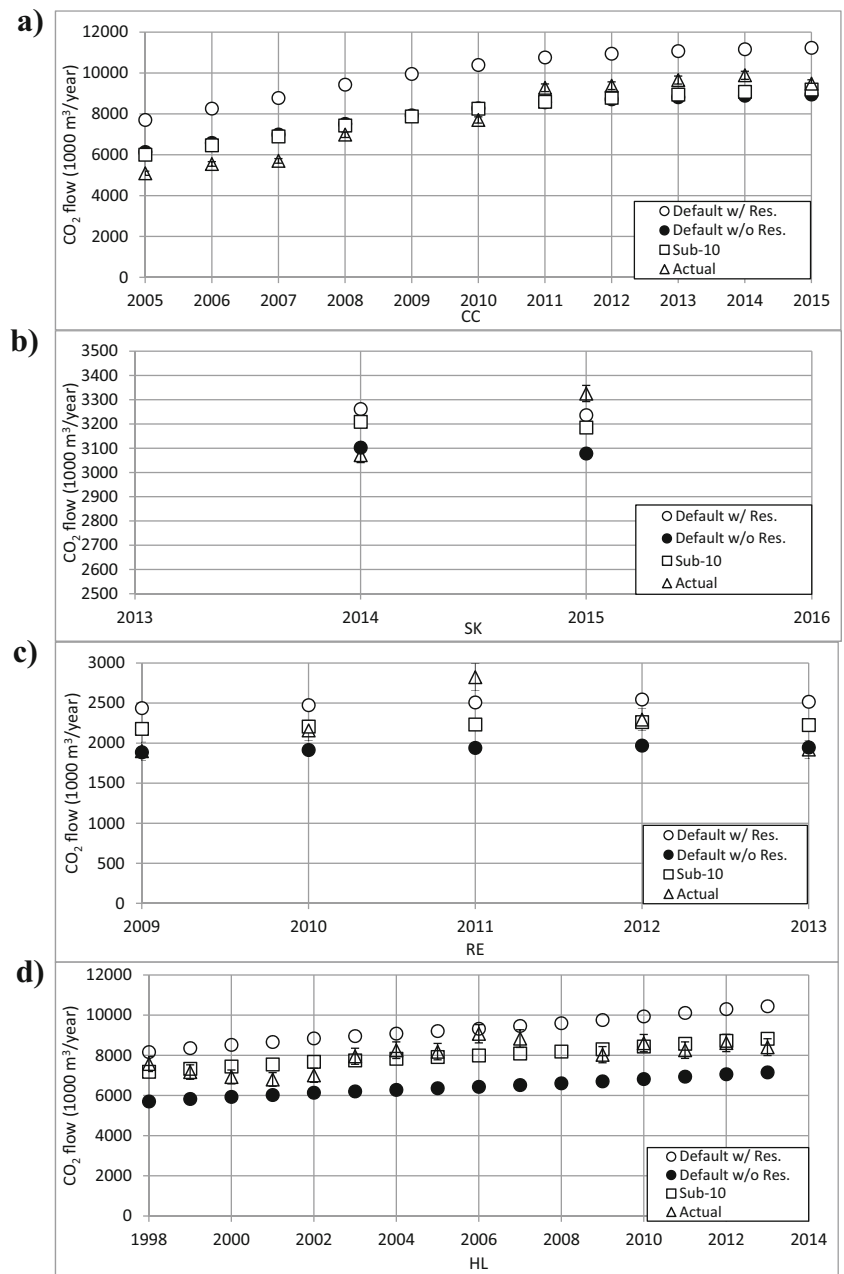


higher solubility makes it more difficult to forecast $CO_2$ using the current model. It is thus expected that landfills with higher precipitation and higher moisture content (such as HL, Table 2) will tend to collect gas with less $CO_2$ (Fig. 1d).

*Annual k models*

Figure 4 shows the unique, comparable results for methods C and D. Method D ($DOC_f$ = 0.5 and 0.77) tended to produce lower RSSs for sites with fewer data points, as SK (2 years) and RE (6 years) are invisible in Fig. 4 with RSSs of 2020 and 6426, respectively. Method D represented the optimal annual

$k$ results in Table 5 for every site as a result, in addition to the most optimized results for all approaches and formulas.

Results for Eq. 3 in method C are absent from Fig. 4 due to the tendency to produce enormous RSSs (from 2 to 59 times higher) for CC and HL and 6% higher RSS for RE. Equation 3 was less applicable for annual use than Eq. 2 in the present study, as it resulted in negative $k$ values for 2 years with low precipitation in CC (2009 and 2015, corrected to $k$ = 0.00001 year$^{-1}$). The least accurate optimized results tended to come from method C overall; thus, neither Eq. 2 nor Eq. 3 is recommended for use in annual $k$ models. Moreover, the annual $k$ models were less accurate than the sub-10 Default w/ Residuals approach for 11 of 20 results (Table 5) and the worst

**Fig. 4** Optimized annual $k$ RSSs
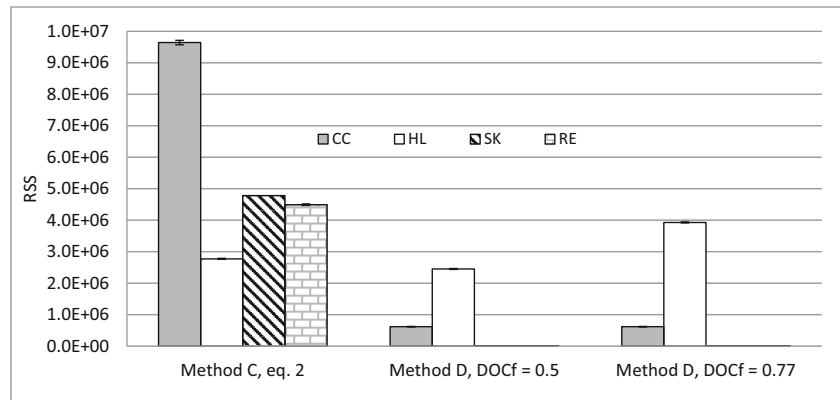


model for 12 of 20 approaches. Thompson et al. (2009), however, encountered mixed success using lifetime $k$ in multiple models based on average precipitation, among other assumptions.

Method D consistently yielded the lowest RSS values, meaning that the success of the annual $k$ governing formula rested on the iterative method rather than starting values. All other methods changed at most two reference cells at once (lifetime $k$ and $L_o$), and those cells were calibrated based on their applied fit across the study period. Method D, however, was able to calibrate the $k$ values on a per-year basis, unencumbered by poor fits with one or more data points, an issue which is reduced using the multi-phase approaches in the newest Afvalzorg and IPCC models.

Method D has potential to determine new annual $k$ formulas. Precipitation has been used as an independent variable in some studies to estimate $k$ using linear relationships (Garg et al. 2006; Karanjekar et al. 2015; Thompson et al. 2009). Of three sites in Fig. 5 (SK excluded due to its shortest study period), only RE yielded a significant linear relationship ($R^2 = 0.92$ and 0.93) between $k$ (optimized via collected $CH_4$) and precipitation. The slopes of the linear formulas for RE in Fig. 5b have the same magnitude as that of annual $k$ Eqs. 2 and 3. RE's slope magnitudes are also higher than those of CC and HL when one HL point ($k = 0.4$ year$^{-1}$) is removed. RE's stronger sensitivity between $k$ and precipitation was surprising considering RE had a partial final cover during the study period (and later a full cover), a feature intended to reduce moisture infiltration, although final covers can be highly susceptible to freeze-thaw processes common in cold climates, combined with desiccation processes in semi-arid climates (Sadek et al. 2007) like RE. Low precipitation is unlikely to be the lone reason for the relationship strength, as CC has lower precipitation rates than RE.

*Subsection models*

The optimized results of the subsection models yielded a smaller range of RSSs compared to the annual $k$ runs.

Percent difference between the lowest and highest RSSs among all subsections and starting sets ranged between 1.3% (RE) and 26.1% (CC), while the annual $k$ methods ranged between 75.8% (HL) and 199.8% (SK) by comparison (data not shown). Considering only the sets with the lowest RSS results (Table 5), the range of differences between optimized formulas reduce to <0.03% (42.2% for RE) to 12.30% (96.8 to 97.2% HL), with the other two sites lower than 0.3%.

Method B, which changes $k$ and $L_o$ together, yielded the best RSSs for both $CH_4$ and $CO_2$ datasets in CC, RE, and SK (data not shown). For HL, the optimized $CO_2$ results for method A were more accurate in terms of RSS, despite method B resulting in the lowest $CH_4$ RSS. This may be because HL notably had years where residual gases well exceed 15% (Fig. 1), which may suggest conditions affecting $CH_4$ $k$, such as aerobic pockets via intrusion.

**Simple ratio approaches**

This approach's lack of an optimization process resulted in higher RSSs across all formulas, starting values, and sites; however, both the 1.2 Ratio and 1.4 Ratio approaches were more accurate than the Default w/ Residuals approach in most cases (Table 5), and at least one of them was more accurate than the Default w/ Residuals approach at each of the four sites. This was expected given the uncertainty associated with default and calculated $k$'s. The results from simple ratio approaches were comparable to those from the Default w/o Residuals approach; thus, they may be appropriate for simple pre-design estimates. The 1.4 Ratio approach may be more applicable to semi-arid sites, as only HL (a coastal site with high precipitation, see Table 2) had higher accuracy using the 1.2 Ratio approach.

The higher accuracy for the 1.4 Ratio approach for three of the sites is supported by the field literature (Table 1) and average ratio in these sites' collection data (1.38). Although the ideal empirical formulas and results in Barlaz et al. (1989a) support the use of a 1:1 ratio still used in most recent studies (Calabro et al. 2011; Chalvatzaki and Lazaridis 2010;

**Fig. 5** Method D optimized k's and precipitation for three sites. **a** CC. **b** RE. **c** HL
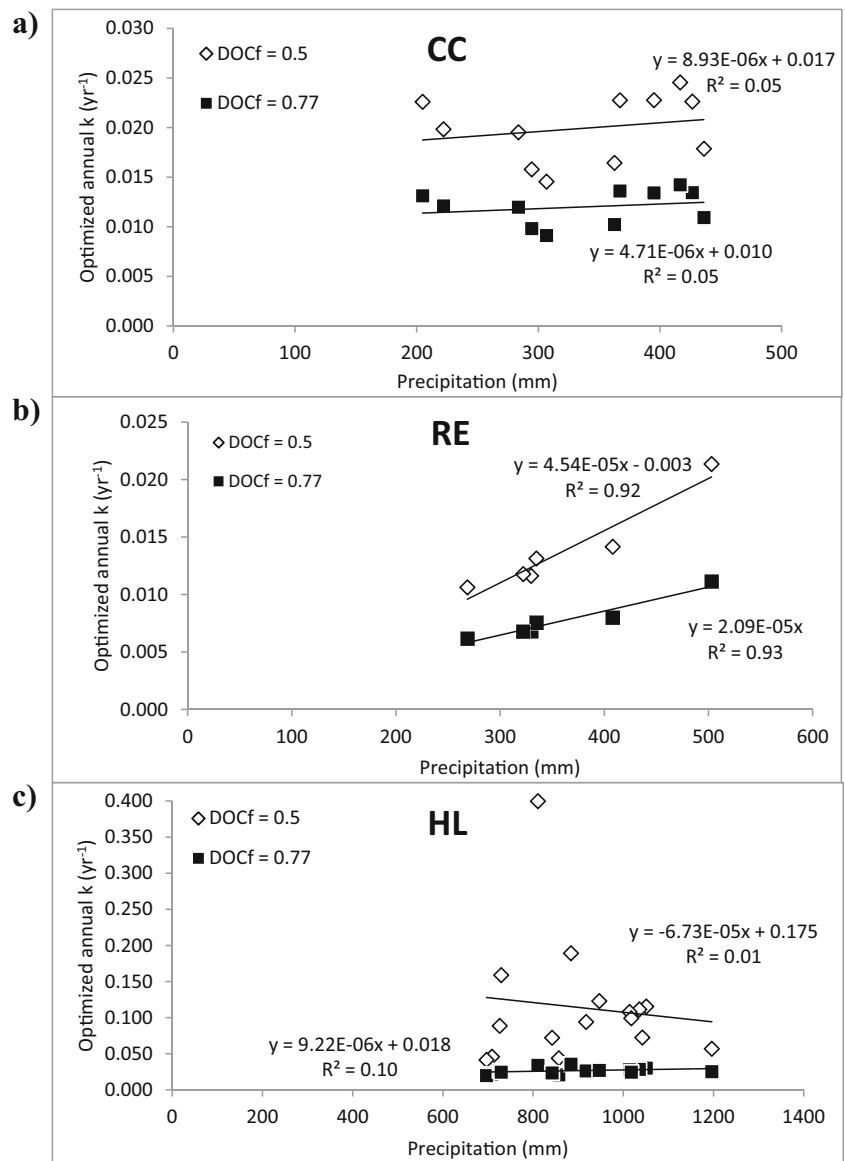


Goswami et al. 2011; Kumar and Sharma 2014; Marroni et al. 2010; Rezaee et al. 2014), this ratio is unreliable and too low for most observed field gas composition in the literature (Table 1). This is likely due to the higher solubility of $CO_2$ in water. The identified 1.2 and 1.4 $CH_4$ to $CO_2$ ratios may provide more accurate $CO_2$ generation forecasts during collection system design than the existing 1:1. However, these ratios may be less applicable to landfills with planned leachate recirculation as observed by Calabro et al. (2011).

**Modelled $CO_2$**

Figure 6 compares the formulas and approaches which yielded the lowest RSS for each site. The 1.2 Ratio approach tended to overestimate $CO_2$ generation in CC and SK and underestimate generation at RE. Slight differences in

subsection models resulted in Sub-1 (HL, CC) and Sub-10 (SK, RE) yielding the most accurate of the subsection models. The actual gas data for HL and CC are subject to some fluctuations throughout operations due to variable upgrade schedules for the well fields and variable cover over the well field. By comparison, the two sites in Saskatchewan (RE and SK) did not expand their well fields during the study periods. The British Columbia sites (HL and CC) are also active, with new wells installed in lifts with intermediate cover and old wells mostly under final cover. For instance, final cover at CC overlaid phases 1–3 and the slopes of phase 4 below the active cells.

The quoted collection efficiency for HL may have been underestimated in 2005 (33.0%), as the $CO_2$ peak occurs in 2005 well before 2013 (Fig. 6d), where the peak should appear due to the landfill's operating status. The optimized

annual $k$ value for HL (Fig. 5c) was 0.4 year$^{-1}$ for 2005 and is a clear outlier compared to all other points at HL and other landfills. The value peaked at 0.4 year$^{-1}$ due to an upper boundary constraint, as preliminary work yielded $k$ larger than 1 year$^{-1}$, well above values seen in the literature.

**High residual gases**

Field collection efficiencies are highly dependent on cover thickness (Barlaz et al. 2009). This can be difficult to include in modelling since reporting standards vary over landfill lives



**Fig. 6** Preferred $CO_2$ modelling approaches and actual data for four western Canadian sites. **a** CC. **b** SK. **c** RE. **d** HL
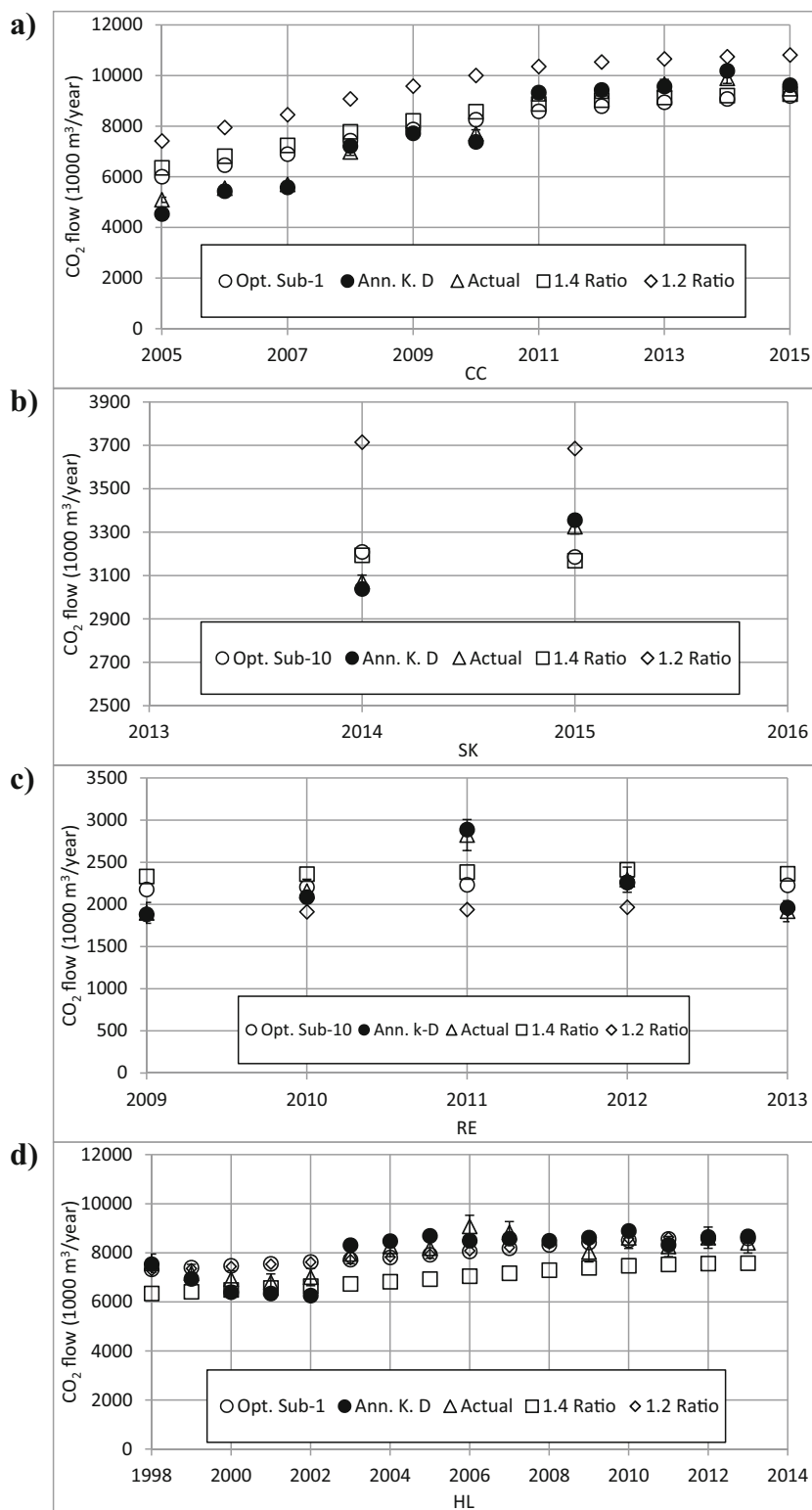
and reliable detailed records are often unavailable for onsite cover placement. Poorly managed LFG collection systems may also lead to excess atmospheric intrusion as seen in 75% of the vertical wells studied by Tolaymat et al. (2010). The freeze-thaw cycle in cold climates can wear down final covers as well, increasing air intrusion.

$N_2$ sample data were available for RE (7 days total, 6 prior to 2010) and HL (8 days total, 2007–2012). All but one sample had higher $N_2$:$O_2$ ratios than the atmospheric average, suggesting partial oxidation during intrusion and over-pressurized collection systems (Guter and Nuerenberg 1987). Another potential source of intrusion was in the pipes and connections. Hettiarachchi et al. (2013) reported condensate freezing in collection lines at a biocell in Calgary, another cold, western Canadian city. Freezing led to blockages and reduced flow rates. This is supported by the tendency in the monthly SK and daily RE data to experience significant drops in LFG collection flow rates during winter months between November and March. $N_2$ content data were unavailable to further support this observation.

## Conclusions

$CO_2$ estimates in LandGEM currently rest on the assumptions that $CO_2$ is a function of $CH_4$ and that the two gases make up nearly 100% of LFG content. This can lead to oversights in collection system design and faulty input estimates for LFG collection system pre-design modelling. Five approaches to LandGEM-based $CO_2$ modelling were compared in this study and concluded the following:

- The current form of the LandGEM equation (Sub-10) yielded the most accurate results for the existing approach to $CO_2$ modelling for all four sites due to tendency to produce lower estimates. The mean RSS increased by 7.1 to 64.3% using modified models. However, this approach was the least accurate compared to most other approaches and models at all four western Canadian sites studied.
- Using a $CH_4$ to $CO_2$ ratio of 1.4 yielded more accurate results for $CO_2$ in LandGEM than the existing approach (mean RSS reduction of 50.2% for all sites and subsection models), and to a lesser extent so did a ratio of 1.2 for two of the four western Canadian landfills. This may be due to significant differences in solubility between $CH_4$ and $CO_2$ affecting field gas and high residual gas content (range 0.4–28.2%) affecting the existing approach. The method may therefore serve as a better pre-design assumption when limited to traditional landfills with no leachate recirculation. The use of a 1:1 ratio in LFG collection modelling is not recommended.

- Annual $k$ models yielded the lowest accuracy in 12 of 20 approaches for the four sites, although they yielded the highest accuracy in all four sites using the "Optimized" approach. Optimized annual $k$'s yielded the lowest RSS values; however, strong relationships with precipitation were not observed in the present study.
- Method D produced the lowest RSSs for models with optimized parameters and greatest percent decrease across all approaches and formulas (mean 97.7%) due to per-year calibration. Methods A and B were dependent on starting values. Method B's $CO_2$ RSS may have been affected by the assumption of equal $k$ for $CH_4$ and $CO_2$. Sampling data suggested significant air intrusion in HL and RE, thus decreasing the field $CH_4$ $L_o$.
- Two existing empirical formulas (Eqs. 2 and 3) relating $k$ and precipitation produced worse estimates for model $CO_2$ when applied to a modified annual $k$ LandGEM. The models were worse than the existing approach and formula in 11 of 20 cases.
- The best approach for $CO_2$ modelling was optimization, which had the greatest reduction in RSS over the default approach (60.1 to 97.7%). However, this approach depends on available data for gas, waste, and collection efficiency. For pre-design, the 1.4 Ratio approach may be more appropriate, at least for the four Canadian landfills considered in this study.

**Compliance with ethical standards**

**Conflict of interest** The authors declare that they have no conflict of interest.

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