Characteristics of Gas and Leachate at an Elevated Temperature Landfill

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Abstract

Data from a municipal solid waste (MSW) landfill with elevated temperatures are evaluated to assess how characteristics of landfill gas and leachate evolved as the landfill temperature increased from ranges typically associated with methanogenic decomposition (40 – 50°C) to more than 100°C. The MSW landfill was deep (~ 100 m), unlined, and most of the waste was saturated and below the water table. Temporal trends in landfill gas temperature measured at the wellhead indicate that the gas temperatures increased abruptly and systematically following a shutdown to address a concern about the potential for subsurface combustion. Temperature profiles collected subsequently indicated that the highest temperatures were substantially below the leachate level, making combustion an unlikely mechanism for the elevated temperatures. Temporal trend analysis indicated that the primary gas ratio (CH₄:CO₂) decreased systematically and substantially when the gas temperature increased abruptly. Leachate chemistry also changed significantly after the gas temperature increased abruptly, with BOD, COD, and the BOD:COD ratio increasing rapidly, pH dropping more than one unit, and total suspended solids increasing more than two orders of magnitude.

INTRODUCTION

Exothermic reactions that produce heat and elevated temperatures have been observed in a limited number of municipal solid waste (MSW) landfills in North America over the last decade. Conditions in these so-called elevated temperature landfills (ETLFS) evolve without the presence of oxygen, aerobic microbial communities, or unusual reactive wastes. Mechanisms responsible for the heat and elevated temperatures in ETLFs are not well defined. Some have attributed the heat to smoldering combustion. However, in many cases, exothermic reactions are occurring deep in the waste under very wet or saturated conditions, precluding combustion reactions. Only some of the ETLFs have received significant quantities of industrial wastes with potential reactivity, precluding exothermic reactions associated with industrial waste as the cause of the elevated temperatures. The Environmental Research and Education Foundation (EREF) has commissioned a team of internationally recognized landfill experts to study and understand these reaction mechanisms so that ETLFs can be avoided in the future.

The ETLF described in this paper, which has experienced temperatures exceeding 100 °C, received municipal solid waste (MSW) from the mid 1980s until the mid 2000s. No significant quantities of industrial waste were disposed. The landfill is deep (~100 m), unlined, and has steep sidewalls. Base elevations are below the surrounding water table, and vertical leachate extraction wells in the waste lower the leachate surface below the phreatic surface in the surrounding geological environment. Consequently, most of the waste is very wet or saturated. Leachate collected from the wells is treated at an on-site treatment plant.
An active gas system with a flare was installed incrementally beginning in the 1990s. Gas temperature and composition are monitored at the wellheads, in leachate extraction wells, and at the flare. An earthen final cover consisting of a 0.6-m-thick barrier layer with an as-built saturated hydraulic conductivity $< 1 \times 10^{-7}$ m/s overlain by a 0.3-m-thick vegetated soil layer was constructed over a majority of the landfill in the mid 2000s. An exposed geomembrane cover was placed over the earthen cover in the early 2010s to provide more effective containment of gas and to control odors. A majority of the exposed geomembrane is a co-extruded ethyl vinyl alcohol (EVOH) geomembrane that has lower diffusion coefficients for methane and hydrogen sulfide than conventional geomembranes (Eun et al. 2016).

Elevated temperatures were observed in a limited number of gas wells in the landfill for several years. In some of these wells the temperature increased gradually and systematically, but without concern. However, as part of routine monitoring, CO concentrations in wells with temperatures above 55 °C were found to exceed 500 ppm, raising concern about the potential for subsurface combustion. The gas system was shut down temporarily in the portion of the landfill having wells with elevated temperatures and elevated CO to limit entry of oxygen that might contribute to combustion. When the system was re-started, sudden and large increases in gas temperature and changes in gas composition were observed at other gas wells. These conditions are attributed to an exothermic reaction, which propagated throughout a large area of the landfill over the next five years. This paper describes gas and leachate data from the landfill before and after the propagating exothermic reaction was discovered.

**LANDFILL GAS**

**Gas composition and temperature in MSW.** Landfill gas is generated as organic materials within MSW degrade in response to aerobic and anaerobic microbial activity. Oxygen within the waste is quickly consumed and exhausted by aerobic microbes before substantive decomposition occurs. Consequently, most of the organic matter in MSW landfills is degraded by anaerobic microbes (Bareither et al. 2013).

Cellulose and hemi-cellulose are the predominant degradable organic materials in MSW. Anaerobic degradation of cellulose $[(C_6H_{10}O_5)_n]$ follows the stoichiometry (Barlaz 2006):

$$
(C_6H_{10}O_5)_n + nH_2O \rightarrow 3nCO_2 + 3nCH_4
$$

where C is carbon, H is hydrogen, O is oxygen, H$_2$O is water, CO$_2$ is carbon dioxide (gas), and CH$_4$ is methane (gas). Anaerobic degradation of hemi-cellulose $[(C_5H_8O_4)_n]$ follows similar stoichiometry:

$$
(C_5H_8O_4)_n + nH_2O \rightarrow 2.5nCO_2 + 2.5nCH_4
$$

Eqs. 1 and 2 indicate that degradation of cellulose and hemi-cellulose in MSW results in a gas comprised of equal amounts of CO$_2$ and CH$_4$. Landfill gas also contains other non-methane organic compounds (NMOCs) derived from other processes such as volatilization of organic compounds within the waste, byproducts of sulfate reducing bacteria such as hydrogen sulfide (H$_2$S), siloxanes, etc. (Thomas and Barlaz 1998). For practical purposes, however, landfill gas can be considered roughly one-half CO$_2$ and one-half CH$_4$ when methanogenic decomposition is
occurring. In practice, each fraction typically will vary from 35-65% of the gas stream in a MSW landfill that is operating normally with methanogenic decomposition. Other “balance” gases will also present, with the sum of the gas fractions adding to 100% (LMOP 2015). The ratio of CH₄ to CO₂, referred to as the primary gas ratio, typically between 0.8 and 1.4 under normal conditions of anaerobic decomposition.

Landfills operating under methanogenic conditions typically have landfill gas temperatures in the range of 30 to 55 °C. At temperatures much greater than this range, the methanogenic microbial community diminishes, the stoichiometry in Eqs. 1 and 2 is no longer valid, and the primary gas ratio will deviate from 1. For example, if oxygen is drawn into the waste by the gas collection system, aerobic biodegradation can be initiated, which can generate higher temperatures and, in some cases, promote combustion. Elevated temperatures and elevated oxygen content can be indicative of aerobic conditions, and carbon monoxide (CO) concentrations in excess of 1000 ppm, combined with smoke and light can be indicative of combustion. For this reason, most state regulations require that gas wellhead temperatures be maintained below 55 °C, the oxygen content be less than 5%, and nitrogen content be less than 20% (40 CFR §60.753, EPA 1999, LMOP 2015).

Gas temperature. Sixty gas wells had been installed in the landfill in the area with elevated temperatures before the propagating exothermic reaction was discovered. Some of the wells had been in place for years, whereas others were newer or had been refurbished. In the year prior to the exothermic reaction being discovered, 18 of the 60 wells exhibited increasing temperature. However, of these 18 wells, 10 were undergoing a normal climb in temperature associated with methanogenic decomposition (3 were in place less than 1 yr), and 5 exhibited a very gradual trend of increasing temperature. Five wells had temperatures close to or exceeding 55 °C for years (four with no temporal trend).

Abrupt increases in temperature were observed in some of the wells when the gas system was re-started after the shutdown to address concerns regarding combustion, as illustrated for two different wells in Figure 1. The vertical line in each graph in Figure 1 corresponds to the date when the propagating exothermic reaction is recognized as being discovered, which was approximately the same time that the gas system was re-started. Increases in temperature of approximately 20-45 °C occurred in wells regardless of whether they were “hot” prior to the gas system being shut down. For example, the temperature in well shown in Figure 1b was approximately 45 °C prior to the gas system being shut down, and reached almost 90 °C within two weeks after the gas system was re-started.

Some wells exhibited the sudden jump in temperature immediately after the system was re-started (e.g., Figure 1), whereas the jump occurred later in other wells. However, within five years of the reaction being discovered, a substantial jump in temperature occurred in more than 85% of the well field (Figure 2) in the area originally associated with elevated temperatures. Thermocouple strings installed in the landfill indicated that the highest temperatures were approximately 20 m below the leachate surface where the waste was saturated, thereby precluding a combustion reaction.
Figure 1. Examples of wellhead temperatures before and after the gas collection system was shut down and re-started and the propagating exothermic reaction discovered.

**Gas Composition.** An example of changes in gas composition in response to the jump in temperature are shown in Figure 3. For this gas well, temperatures had been stable but elevated and close to 55 °C for more than 4 yr prior to the gas system being shut down (Figure 3a). Virtually no oxygen was present in the gas, suggesting that the vacuum was not too high and that air from outside the landfill was not being pulled into the well. The CH₄ and CO₂ fractions were similar and steady, suggesting methanogenic conditions and a stable well (Figure 3b). The primary gas ratio was also steady (~1.2-1.4, Figure 3b), suggesting stable methanogenic conditions. Balance gases were modest before the reaction was discovered.
Figure 2. Percentage of wells with jump in temperature vs. time since reaction was discovered.

Gas temperatures at the wellhead dropped precipitously when the gas system was shut down, which was expected as the wellhead equilibrated with the atmosphere. Approximately two weeks later, when the gas system was re-started, the wellhead temperatures were nearly 20 °C higher than immediately prior to shut down of the gas system. During that same period after the system was re-started, the CO₂ fraction increased dramatically. A concomitant drop in the CH₄ fraction and the primary gas ratio also occurred. These conditions persisted for three years after the system was re-started (the data set available extended only three years after the reaction was discovered).

The findings in Figures 1-3 suggest that shutting down the gas system may have been detrimental rather than beneficial, and accelerated or magnified the reaction. The decision to shut down was based on the assumption that the waste was undergoing combustion, which has been a standard operating procedure in the industry for years. One hypothesis for the detrimental effects of the shutdown is that heat continued to accumulate when the gas system was shut down, providing the energy to increase the kinetics and energetics of the exothermic reactions. Another is that absence of gas removal may have provided greater access to reactants in the gas phase, further exacerbating the reaction. These hypotheses are preliminary and are currently being explored.

LEACHATE

Major indicator parameters commonly used to characterize leachate include biological oxygen demand (BOD), chemical oxygen demand (COD), pH, specific conductance, and total suspended solids (TSS). BOD describes the amount of oxygen needed by aerobic biological organisms to degrade organic material present in water at standard temperature. COD is the oxygen equivalent of organic matter in water that is susceptible to oxidation by a strong chemical oxidant
Changes in BOD, COD, pH, specific conductance, and TSS of the leachate reflect a change in the chemical characteristics of the waste. Tracking these parameters provides an understanding of the state of decomposition of the MSW and biochemical changes occurring within the landfill.

Figure 3. Gas wellhead temperature vs. time for hot but steady well prior to shut down of the gas system (a), gas composition as a function of time (b), and primary gas ratio as a function of time (c).
BOD, COD, and BOD:COD of the leachate are shown as a function of time in Figure 4. The data in Figure 4 represent spatially commingled leachate from all extraction points in the landfill before and after the exothermic reaction was discovered (i.e., a mixture of leachate from areas where reactions were and were not occurring). The BOD and COD are relatively constant for the three years prior to initiation of the exothermic reaction. The BOD:COD ratio is also relatively constant, and representative of decomposed waste at least 10 yr old (Benson et al. 2007, Barlaz et al. 2010). The pH is constant over the same period (Figure 5a), and generally within the range of 6-8 that is characteristic of leachate from decomposed waste (Benson et al. 2007, Barlaz et al. 2010). Specific conductance and TSS are also constant over this same period (Figures 5b and 6). That is, prior to the exothermic reaction, the leachate data were consistent with stable decomposed refuse.

![Figure 4. BOD and COD (a) and BOD/COD (b) vs. time.](image-url)
Approximately one year after the onset of the reaction, BOD, COD, BOD:COD, TSS, and specific conductance increased substantially, and the pH dropped more than one unit (Figs. 4-6). Settlements of more than 5 m were also observed within one year of the abrupt increases in gas temperature. These abrupt changes in leachate chemistry, coincident with the substantial and abrupt increases in gas temperature, a plunge in primary gas ratio and the absence of oxygen in the gas, and substantial and rapid settlement suggest that decomposition of the waste was occurring due to an abiotic reaction. This reaction entrained dissolved constituents and suspended inorganic and organic solids in the leachate, resulting in the increase in BOD, COD, BOD:COD ratio, EC, and TSS shown in Figs. 4 and 5. There was no evidence of smoke, light, or oxygen in the landfill gas prior to this abrupt change in behavior, and the highest temperatures occurred meters below the leachate level in nearly saturated or saturated waste. This suggests that an exothermic reaction other than combustion was generating the heat responsible for the elevated gas temperatures. An exothermic pyrolysis reaction has been hypothesized as the source of elevated heat and temperature in this landfill. This reaction mechanism is being explored.

Figure 5. pH (a) and specific conductance (b) of leachate vs. time since the propagating exothermic reaction was discovered.
SUMMARY AND CONCLUSIONS

This paper has described gas and leachate data from an elevated temperature landfill (ETLF) where waste temperatures have exceeded 100 °C. A propagating exothermic reaction was discovered in the landfill after a portion of the gas system had been shut down temporarily to address concerns associated with the potential for combustion. CO concentrations exceeding 500 ppm at some wells with temperatures in excess of 55 °C were the basis for this concern. When the gas system was restarted, sudden increases in gas wellhead temperature as large as 45 °C occurred. Over the next 5 yr, the exothermic reaction propagated throughout a large area of the landfill, resulting in temperature jumps and elevated temperatures at more than 85% of the gas well heads in the affected area.

The sudden increases in temperature were associated with concomitant changes in the gas composition, namely a large increase in CO₂ fraction, a precipitous drop in CH₄ fraction, and a large drop in the primary gas ratio. BOD, COD, and the BOD:COD ratio of the leachate also increased dramatically after years of exhibiting conditions representative of stable decomposed refuse. pH of the leachate dropped, and large increases in specific conductance and total suspended solids occurred. Settlements of more than 5 m occurred within one year of the reaction being discovered.

The exothermic reaction may have been exacerbated by the shutdown of the gas system to address concerns regarding the potential for combustion. At the time, the decision to shut down the gas system was consistent with well-accepted standard operating procedures in the industry and is a practice that likely would have been followed at any site exhibiting similar conditions. Evidence available today, however, suggests that a reaction other than combustion
was occurring, and the kinetics and energetics of this reaction may have been exacerbated by the absence of gas removal during the shutdown. This hypothesis is preliminary and is currently being explored.

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