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Using a Gas-Phase Tracer Test to Characterize the Impact of Landfill Gas Generation on Advective-Dispersive Transport of VOCs in the Vadose Zone

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Abstract

A gas-phase tracer test (GTT) was conducted at a landfill in Tucson, AZ, to help elucidate the impact of landfill gas generation on the transport and fate of chlorinated aliphatic volatile organic contaminants (VOCs). Sulfur hexafluoride (SF₆) was used as the non-reactive gas tracer. Gas samples were collected from a multiport monitoring well located 15.2 m from the injection well, and analyzed for SF₆, CH₄, CO₂, and VOCs. The travel times determined for SF₆ from the tracer test are approximately two to ten times smaller than estimated travel times that incorporate transport by only gas-phase diffusion. In addition, significant concentrations of CH₄ and CO₂ were measured, indicating production of landfill gas. Based on these results, it is hypothesized that the enhanced rates of transport observed for SF₆ are caused by advective transport associated with landfill gas generation. The rates of transport varied vertically, which is attributed to multiple factors including spatial variability of water content, refuse mass, refuse permeability, and gas generation.

Keywords

gas-phase transport; landfill gas generation; VOCs; gas tracer test

INTRODUCTION

Landfill gas generation has long been of concern with respect to its impact on landfill operations and its potential risk for adjacent commercial and residential properties. More recently, concern over emissions of landfill gas have heightened due to their role in global climate change. There is also interest in the potential impact of landfill gas generation on the transport and fate of VOCs that are routinely present at landfill sites. Landfill waste often serves as a long-term source of VOCs in the vadose zone. In turn, this contamination can

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have a significant impact on groundwater and on residential or commercial indoor air quality through vapor intrusion.

In areas with shallow groundwater, the primary mode of transport for landfill contaminants is leachate generation and aqueous-phase transport to groundwater. The typical groundwater contaminant profile for this situation includes waste constituents that have low volatilities (e.g., antibiotics, pesticides, detergents, salts, etc.), in addition to VOCs. In arid and semi-arid regions such as the southwest US, groundwater can be up to hundreds of meters deep. However, many landfills in the southwest region are regulated contaminated sites with VOCs present in groundwater. For example, there are four landfill sites in Tucson at which groundwater is contaminated by VOCs. Local recharge of groundwater in these regions is often minimal due to limited precipitation and large evapotranspiration potential. Therefore, the contribution of leachate migration to groundwater contamination is typically negligible. This is supported by the observation that VOCs are the primary contaminants present at some of these sites, while the low-volatility contaminants present in humid regions are typically absent. This leads to the question: How do select VOCs (e.g., carbon tetrachloride, trichloroethene, tetrachloroethene) reach the groundwater in these regions?

In the absence of dissolved-phase transport, migration from the waste to groundwater must occur via gas-phase diffusive and advective transport processes. Density driven vapor-phase advective transport of VOCs is unlikely for many municipal landfill systems, given that the large quantities of solvent liquid required for such transport are generally not present. Gas-phase diffusion is anticipated to occur, but detection of VOCs in groundwater is often observed sooner than predicted based solely on diffusive transport. Therefore, it has been hypothesized that landfill gas generation is facilitating the transport of VOCs from the landfill to groundwater.

Gas tracer tests (GTT) have been used to characterize numerous properties for vadose-zone systems, such as water content (e.g., Nelson et al., 1999; Keller and Brusseau, 2003; Carlson et al., 2003; Han et al., 2006) and gas flow velocities and tortuosity (e.g., Kremer et al., 1988; Werner et al., 2004; Tick et al., 2007). Several GTT methods exist to characterize landfill gas generation, such as double tracer techniques (e.g., Scheutz et al., 2011), multiple tracer tests (e.g. Jung et al., 2012), tracer tests from leachate wells (e.g., Fredenslund et al., 2010), and gas push-pull tests (e.g., Gomez et al 2008; Streese-Kleeberg et al., 2011).

A gas-phase tracer test was conducted at a landfill in Tucson, AZ, to evaluate the impact of landfill gas generation on the transport and fate of chlorinated aliphatic volatile organic contaminants. A single injection-extraction well couplet was used, with sulfur hexafluoride (SF_6) serving as the non-reactive gas tracer. The tracer-test data were used to determine travel times, which were compared to values calculated using Fick's Law for diffusion-only transport.

MATERIALS AND METHODS

Site Description

The El Camino del Cerro Landfill is an unlined, alluvial capped landfill located in Tucson, Arizona, that was in operation from 1973 to 1977. No disposal records exist, but it is believed that the site contains municipal solid waste, paper, green waste products, scrap iron, construction debris, plastics, and miscellaneous industrial solvents and household cleaning products. The heterogeneous refuse extends from a few meters below land surface to approximately 26 meters deep. Depth to groundwater is approximately 40 m. An extensive perimeter well network was installed in 1994 for sampling and monitoring of landfill gas at the site. Shallow landfill gas monitor probes were installed in 1995 to monitor methane gas concentrations between the landfill and nearby businesses. Methane was observed at concentrations up to 60% by volume and has been detected in the shallow vadose zone perimeter wells. The site was placed on the Water Quality Assurance Revolving Fund (WQARF) list (i.e., deemed a State Superfund Site) in 1998. Volatile organic compounds, including tetrachloroethene, trichloroethene, 1,1-dichloroethene, and vinyl chloride, are present in soil gas at the site. Some of these compounds are present in groundwater at levels ranging from below detection to more than 300 ug/L. Note that VOCs are the only contaminants present in groundwater at levels above regulatory limits, which suggests that aqueous-phase migration of leachate to groundwater is likely minimal for the site.

Tracer Experiment

A single injection-extraction well couplet was used, with a separation distance of 15.2 m. The tracer injection well (I-6) and the observation well (O-5) are located in the south-central portion of the landfill with total depth of both wells being 25.9 and 26.2 meters below ground surface (bgs), respectively (Figure 1). The single injection well was constructed with five multi-depth “nested” probes screened at discrete intervals. The five screens are labeled as ports “a” through “e”. The ports are screened at 4.6 – 7.6, 9.1 – 12.2, 13.7 – 16.8, 18.3 – 21.3, and 22.9 – 26.0 m for ports “a” through “e”, respectively. The single observation well was constructed similarly to the single injection well with probes and intervals identical.

A truck mounted Central Mine Equipment Model No. 75 rotary drill rig was used to drill each borehole. The wells were constructed with Schedule 40 2-inch PVC casing, sealed with a silicon sealant to prevent gas leakage. Sand filter packs were placed at each screen interval, with bentonite placed between each set of packs. A grout mix was then used to complete the construction to ground surface. The wells were fitted with steel protective well covers. A Foxboro 128 organic vapor phase analyzer and MSA Passport trigas meter were used to monitor health and safety conditions during all drilling events. Moisture content ranged from 6 to 53% based on gravimetric measurement of subsamples collected during drilling (Table 1),

SF₆ was used for the tracer test, at a concentration of 100 mg/L mixed in a balance of nitrogen and stored in a pressurized cylinder (80ft³ @ 3000psi). At the start of the tracer test, SF₆ was released into the injection well at a total rate of approximately 200 psi per minute for 3 minutes into ports “a” through “d”. Tracer was not injected into port “e” due to

a broken wellhead petcock. After injection was completed, the tracer pulse moved under the influence of whatever advective-dispersive processes were present under natural conditions (no continued artificial gradient due to injection). Tracer transport was monitored for 624 hours.

The injection and observation wells were sampled using gas-tight syringes (30 cm³). The samples were stored in evacuated canisters (80 cm³). Samples were collected without well purging during the first two days of the experiment to minimize potential dilution under conditions for which concentrations were anticipated to be changing rapidly. Purging prior to sampling began on day three of the experiment.

Initial landfill gas (CO₂, CH₄, O₂) concentrations, well pressures, and ambient air temperatures were analyzed using a Landtek GA-90™ gas analyzer with calibration performed using a portable calibration gas kit containing cylinders of 50/50 (50% CH₄ and 50% CO₂) and 5/95 (5% O₂ and 95% CO₂) with data downloaded to a personal computer. Sample concentrations for the landfill gas constituents were quantitated using a gas chromatograph (GC) coupled to a thermal conductivity detector (TCD) at Tracer Research Corp. Background VOC concentrations were analyzed using a flame ionization detector (FID) with samples containing elevated levels taken to an off-site laboratory for quantitative chemical analysis using EPA Method 8240. Draeger detection tubes were used to monitor all field personnel exposure limits during experiment operations.

Data Analysis

Temporal moment analysis of the SF₆ tracer concentrations measured for the observation well was used to determine the mean travel times. Travel times for diffusive transport of SF₆ were determined with Fick's 2nd Law, using the solution $C/C_0 = \text{erfc } x/(2[D_0t/T])^{0.5}$, where C = SF₆ concentration detected, C_0 = initial SF₆ concentration, erfc = complementary error function, D_0 = gaseous diffusion coefficient (0.036 cm²/h, from Kremer et al., 1988), T = tortuosity (assumed value = 2), and X = travel distance (15.2 m). This approach is based on the assumption that there was no vapor sorption or retardation of the SF₆, which is to be expected based on its properties. It is recognized that factors such as refuse heterogeneity, spatial and temporal variability of temperature, barometric pressure, and soil moisture, which are not explicitly accounted for with the analysis, may influence gas transport.

RESULTS AND DISCUSSION

Results

High levels of CH₄ and CO₂ were observed in vapor samples collected from the injection and observation wells, ranging from 43–80% and 20–39%, respectively (Table 2). These high values indicate that there is significant landfill gas generation occurring at the site. In general, methane concentrations were higher in the shallow and piezometer wells versus the deep wells. Wells located directly within the landfill refuse generated higher gas concentrations than those wells located on the periphery of the landfill or the unsaturated soil boundary areas. Background results prior to the tracer experiment indicate that the southern half of the landfill consistently produces slightly higher concentrations of methane

than the northern half. The presence of representative concentrations of methane in all wells (shallow, piezometer, deep) suggests the wells provide a robust network for the sampling of landfill gases and VOCs.

The injection and observation wells contained only trace amounts of chlorinated compounds, with concentrations ranging from 0.001 to 0.4 ug/L for select VOCs (Table 2). Significantly larger concentrations were observed for ethylbenzene, toluene, and xylene. The concentrations were highest for port “a” for most VOCs. The results indicate that both landfill gas and VOC concentrations are higher at the shallower depth.

A 3-day diurnal analysis was performed during the asymptotic phase of the GTT to evaluate the potential impact of environmental conditions on SF₆ concentration. Samples were collected 3 times a day for 3 days (morning, noon, and afternoon). The results show that no significant variability in concentration was observed for the injection well (Figure 2). Weather data (Figure 3) shows that barometric pressure was relatively constant throughout the test. Temperature oscillated from approximately 28 C to 40 C for the 3-day test. The results indicate that pressure and temperature had no measureable impact on tracer gas concentrations. No precipitation was recorded during the test.

The measured SF₆ concentrations (normalized to the initial concentration of 6 µg/L) for ports “a” through “d” of the injection well are presented in Figure 4. Inspection of the figure shows that there is a general exponential decay (attenuation) trend of SF₆ for all ports, as would be expected. Concentrations of SF₆ at port “d” exhibited the fastest attenuation, whereas the slowest attenuation was observed for port “a”. Concentrations of SF₆ attenuated at similar rates for ports “b” and “c”. These observed differences in attenuation rates indicate that tracer dissipation via transport away from the injection well was vertically nonuniform.

The SF₆ concentrations measured for samples collected at the observation well are presented in Figures 5–9. The concentrations are observed to fluctuate over time, and exhibit multi-peak behavior for some ports. Such behavior may indicate the impacts of temporally and/or spatially non-uniform gas generation as well as permeability heterogeneity. The detection of SF₆ in observation well port “e” is a possible indication of vertical movement, given that injection only occurred in ports “a” through “d”.

The mean travel times and pore-gas velocities determined for transport of SF₆ to ports “a” through “e” of the observation well are reported in Table 1. The fastest SF₆ velocities (0.14 m/hr) are obtained for ports “d” and “e”, the two deepest. In contrast, significantly slower SF₆ velocities ranging from 0.03 to 0.04 m/hr are reported for the shallow ports “a” through “c”. These data indicate that SF₆ traveled to the observation well at two significantly different velocities between the shallower and deeper ports. The faster velocities observed for SF₆ transport to the deeper ports of the observation well is consistent with the results observed for the injection well, wherein the fastest tracer attenuation was observed for port “d”.

The vertical variability in the observed rates of tracer transport could be the result of several factors, such as vertical non-uniformity in landfill gas generation, water content, and refuse permeability. As noted previously, landfill-gas concentrations are generally higher for the

shallower depth, perhaps indicating great rates of generation. However, the fastest tracer velocities are observed for the deeper depth. The water content for the deepest port was the lowest, 7%, of the five measured values. The lower water content would translate to a larger relative air permeability, which would result in faster rates of advection. However, the water content for the shallowest depth is lower than that observed for the port “d” depth. This suggests that water content is not the only variable that influenced transport. Of course, it must be recognized that the water content data represent local conditions, and that water content may vary between the two wells. Permeability of the waste material is also expected to be a significant factor influencing transport. The complexity of landfill refuse can make the identification of lower and higher permeability areas quite difficult. The presence of higher-permeability zones in the deeper depths and lower-permeability zones in the shallower depths would be consistent with the observed disparity in transport rates.

The estimated diffusive travel times for SF₆ are 1100 hours for ports “a” through “c” and 1990 hours for ports “d” and “e” (Table 2). The mean travel times determined from the tracer test are significantly smaller than the estimated travel times based on diffusion-only transport. The difference is an order of magnitude for the deeper ports, and ranges from a factor of 2–3 for the shallower ports. The great disparity between the measured and diffusion-based travel times indicates that advective transport due to landfill gas generation contributed significantly to overall tracer transport.

Conclusion

A gas-phase tracer test (GTT) was conducted at a landfill in Tucson, AZ, to help elucidate the impact of landfill gas generation on the transport and fate of chlorinated aliphatic VOCs. The travel times determined for SF₆ from the tracer test are approximately two to ten times smaller than estimated travel times that incorporate transport by only gas-phase diffusion. In addition, high concentrations of CH₄ and CO₂ were measured, indicating production of landfill gas. Based on these results, it is hypothesized that the enhanced rates of transport observed for SF₆ are caused by advective transport associated with landfill gas generation. The rates of transport varied vertically, which is attributed to multiple factors including spatial variability of water content, refuse permeability, and gas generation.

Currently, landfills within the southwestern US, that is California, Nevada, Utah, Arizona and New Mexico have numerous landfills that are actively generating methane and other landfill gases (<http://www.epa.gov/outreach/lmop>). Depths to groundwater in the southwest typically range from 50–200 meters below ground surface, whereas landfills measure 20–30 meters below ground surface. In Tucson, The Los Reales, Silverbell, El Camino del Cerro, and Broadway/Pantano Landfills have a depth to groundwater of 65, 44, 40, and 107 meters bgs, respectively. The difference of many tens to hundreds of meters between landfill depth and groundwater is significant when examining the movement of VOCs from the landfill to the groundwater. The results of this study suggest that gas-phase advective and diffusive transport enhanced by landfill gas generation likely mediates the movement of VOCs to groundwater when leachate migration is limited, as it may often be in the southwest US. The advective transport induced by landfill gas generation may also result in faster and greater migration of VOCs to the land surface, thereby enhancing the potential for vapor intrusion.

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Figure 1.
Injection/Extraction monitoring well system used for tracer gas test.

Figure 2.
Three-day diurnal SF₆ tracer gas data for the injection well (I-6).

Figure 3.
Weather data during the 3-day diurnal SF₆ tracer gas test for injection wells.

Figure 4.
SF₆ concentrations for gas samples collected from the injection well (I-6).

Figure 5.
SF₆ Concentrations for Observation Well Port A.

Figure 6.
SF₆ Concentrations for Observation Well Port B.

Figure 7.
SF₆ Concentrations for Observation Well Port C.

Figure 8.
SF₆ Concentrations for Observation Well Port D.

Figure 9.
SF₆ Concentrations for Observation Well Port E.

Table 1

Measured and Expected Diffusive Travel Times to observation well for F₆ Tracer.

Screened Interval (m bgs)	Measured Mean Travel Time (hrs)	Expected Diffusive Travel Time (hrs)	Velocity (m/hr)	Moisture Content (% by wt.)
a: 5–8	452	1107	0.03	15.7
b: 9–12	374	1107	0.04	22.1
c: 14–17	525	1107	0.03	51
d: 18–21	108	1989	0.14	30.4
e: 23–26	108	1989	0.14	7.3

Table 2

Summary of vapor-phase constituent concentrations.

Background Summary of "Fixed" Gas Concentrations and Gas-Phase VOCs											
Well ID	port : depth (meters bgs)	[CH ₄] (%/vol.)	[CO ₂] (%/vol.)	[O ₂] (%/vol.)	PCE (ug/L)	TCE (ug/L)	cis 1,2 - DCE (ug/L)	VC (ug/L)	Ethylbenzene (ug/L)	Toluene (ug/L)	Xylene Total (ug/L)
O-5	a: 5-8	70.8	30.5	0	ND	ND	ND	ND	21.0	7.5	18.8
O-5	b: 9-12	43.4	22.9	2.7	ND	ND	ND	ND	18.8	3.3	15.0
O-5	c: 14-17	75.6	25.8	0	ND	ND	ND	ND	0.42	ND	1.07
O-5	d: 18-21	80.2	19.6	0	ND	ND	ND	ND	0.05	ND	0.24
O-5	e: 23-26	57.4	39.3	0	ND	ND	0.003	0.004	0.05	0.01	0.19
I-6	a: 5-8	63.9	34.2	0.2	0.161	ND	ND	0.376	17.2	14.5	51.0
I-6	b: 9-12	66.2	34.6	0	0.001	0.0002	0.001	ND	0.04	0.05	0.17
I-6	c: 14-17	61.2	37.3	0	NA	NA	NA	NA	NA	NA	NA
I-6	d: 18-21	56.1	37.4	0	0.001	0.0002	0.002	0.001	0.08	0.09	0.44
I-6	e: 23-26	BROKEN WELL HEAD PETCOCK									