

1 **Organic Compound Emissions from a Landfarm Used For Oil and Gas Solid Waste**  
2 **Disposal**

3 Seth N. Lyman<sup>a,b,\*</sup>, Marc L. Mansfield<sup>a,b</sup>

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5 <sup>a</sup>Bingham Entrepreneurship & Energy Research Center, Utah State University, 320 Aggie  
6 Boulevard, Vernal, Utah 84078, U.S.A.

7 <sup>b</sup>Department of Chemistry and Biochemistry, Utah State University, 4820 Old Main Hill, Logan,  
8 Utah, U.S.A.

9 \*Corresponding author, seth.lyman@usu.edu

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11

12 **Abstract**

13 Solid or sludgy hydrocarbon waste is a byproduct of oil and gas exploration and production.  
14 One commonly-used method of disposing of this waste is landfarming. Landfarming involves  
15 spreading hydrocarbon waste on soils, tilling it into the soil, and allowing it to biodegrade. We  
16 used a dynamic flux chamber to measure fluxes of methane, a suite of 54 non-methane  
17 hydrocarbons, and light alcohols from an active and a remediated landfarm in eastern Utah,  
18 U.S.A. Fluxes from the remediated landfarm were not different from a PTFE sheet or from  
19 undisturbed soils in the region. Fluxes of methane, total non-methane hydrocarbons, and  
20 alcohols from the landfarm in active use were 1.41 (0.37, 4.19) (mean and 95% confidence  
21 limits), 197.90 (114.72, 370.46), and 4.17 (0.03, 15.89) mg m<sup>-2</sup> h<sup>-1</sup>, respectively. Hydrocarbon  
22 fluxes were dominated by alkanes, especially those with 6 or more carbons. A 2 ha landfarm  
23 with fluxes of the magnitude we observed in this study would emit 95.3 (54.3, 179.7) kg day<sup>-1</sup> of  
24 total hydrocarbons, including 11.2 (4.3, 33.9) kg day<sup>-1</sup> of BTEX (benzene, toluene, ethylbenzene,  
25 and xylenes).

26

27

28 **INTRODUCTION**

29 The climate and air quality impacts of oil and gas development have been studied extensively  
30 (Ahmadov et al., 2015; Field et al., 2015; Helmig et al., 2014; Lyman and Tran, 2015; McDuffie  
31 et al., 2016; Oltmans et al., 2014; Prenni et al., 2016; Warneke et al., 2014). Field-wide and  
32 basin-wide measurements of emissions from the oil and gas sector are usually higher than  
33 emissions predicted by official inventories (Ahmadov et al., 2015; Brandt et al., 2014; Karion et  
34 al., 2013). One reason for this discrepancy could be that some oil and gas-related emission  
35 sources are not included in current inventories (Brandt et al., 2014; Caulton et al., 2014). One  
36 source category that has been excluded from oil and gas inventories is waste disposal (WRAP,  
37 2015), including produced water and solid waste. Very few measurements of emissions related  
38 to oil and gas waste disposal have been collected (Pardue and Valsaraj, 2000; Thoma, 2009).

39 Solid and sludgy hydrocarbon waste from the oil and gas industry is often disposed of in  
40 landfarms. Landfarms are sites where hydrocarbon-contaminated waste from the oil and gas  
41 industry is applied to and mixed with surface soil as a method of bioremediation (Gurska et al.,  
42 2009). Oil and gas solid waste disposed of at landfarms may include drilling waste, solid  
43 material accumulated in liquid storage tanks, or other materials. Often, fertilizers or other  
44 additives are used to amend landfarm soils (Callahan et al., 2002) and the soil is periodically  
45 tilled (Ausma et al., 2001) to promote decomposition of hydrocarbons. Volatile hydrocarbons  
46 tend to be emitted from landfarms into the atmosphere, rather than being degraded (Hejazi et al.,  
47 2003). Emissions from landfarms decrease rapidly after application of solid waste as the supply  
48 of volatiles becomes depleted (Ausma et al., 2002; Pardue and Valsaraj, 2000). Tilling increases  
49 emissions of hydrocarbons from landfarm soils (Ausma et al., 2003; Ausma et al., 2001).  
50 Emissions from landfarms have been modeled by Pardue and Valsaraj (2000), Dupont (1986),  
51 and Thibodeaux and Hwang (1982).

52 We report below on measurements of organic compound emissions from a landfarm facility in  
53 the Uinta Basin of eastern Utah. We present information about the magnitude, distribution and  
54 speciation of measured emissions.

55 **METHODS**

56 **Study Location**

57 We measured fluxes from a landfarm facility in the Uinta Basin during fall 2014, spring 2015,  
58 and spring 2016 (21 total flux chamber measurements). The facility consisted of two  
59 landfarms—one that had been remediated and another that was actively receiving oil and gas  
60 solid waste. The remediated landfarm did not receive any applications of clean soil or other  
61 additives after disuse. It contained some natural vegetation. The operator did not disclose the  
62 date of the last waste application to the remediated landfarm. For comparison with the  
63 landfarms, we measured fluxes from undisturbed soils in oil and gas-producing areas throughout  
64 eastern Utah.

65 **Flux Chamber**

66 We measured fluxes with a polycarbonate dynamic flux chamber, which was constructed  
67 according to the design of Eklund (1992), except that (1) we used a mixing fan with a  
68 polyethylene blade to ensure that air inside the chamber was well mixed, and (2) instead of using  
69 purified sweep gas, the chamber pulled in ambient air from a 1.2 cm hole in one side. We  
70 determined flux as:

71 
$$F = ( \Delta C \times FR ) / S \quad (1)$$

72 where F is the soil-air flux in  $\text{mg m}^{-2} \text{h}^{-1}$ ,  $\Delta C$  is the difference in concentrations of the compound  
73 of interest inside versus outside the chamber in  $\text{mg m}^{-3}$ , FR is the flow rate in  $\text{m}^3 \text{h}^{-1}$ , and S is the  
74 surface area covered by the chamber in  $\text{m}^2$  (Denmead, 2008). We sampled air inside and outside  
75 the chamber using PFA tubing with PTFE filters to avoid particle contamination. The flow rate  
76 through the chamber was  $10 \text{ sL min}^{-1}$  (standard conditions of  $25^\circ\text{C}$  and 1 atm). Each flux  
77 measurement was collected over 60 min.

78 Additional discussion about the chamber method, including possible interferences from nearby  
79 emission sources and the results of several different quality control experiments, are available  
80 from Lyman et al. (2017) and Lyman et al. (2018)

## 81 **Methane and Carbon Dioxide**

82 We measured methane and carbon dioxide with a Los Gatos Research (LGR) Ultra-Portable  
83 Greenhouse Gas Analyzer. Flux detection limits were 0.06 and 18.9 mg m<sup>-2</sup> h<sup>-1</sup> for methane and  
84 carbon dioxide (3 times the standard deviation of blanks). We checked the LGR analyzer against  
85 NIST-traceable compressed gas calibration standards daily. Calibration standard recovery was  
86 98 (97, 99)% (mean (lower confidence limit, upper confidence limit)) for methane and 98 (96,  
87 100)% for carbon dioxide.

## 88 **Non-methane Hydrocarbons**

89 We collected whole-air samples from inside and outside the chamber with silonite-coated 6 L  
90 stainless steel canisters. We analyzed the canisters for 54 non-methane hydrocarbons (NMHC),  
91 methanol, ethanol, and isopropanol using a method similar to guidance provided by the U.S.  
92 Environmental Protection Agency for Photochemical Assessment Monitoring Stations (EPA,  
93 1998). We used stainless steel Alicat mass flow controllers to regulate flow into the canisters.  
94 We used cold trap dehydration (Wang and Austin, 2006) with an Entech 7200 preconcentrator  
95 and a 7016D autosampler to preconcentrate samples. We analyzed samples with two Shimadzu  
96 GC-2010 gas chromatographs, a flame ionization detector (for C2 and C3 NMHC), and a mass  
97 spectrometer (for all other compounds). Compounds were separated with a Restek rtx1-ms  
98 column (all compounds; 60 m, 0.32 mm ID), a Restek Alumina BOND/Na<sub>2</sub>SO<sub>4</sub> column (C2  
99 and C3 NMHC; 50 m, 0.32 mm ID), and another Restek rtx1-ms column (all other compounds;  
100 30 m, 0.25 mm ID). Calibration checks had an average recovery of 104.6 (103, 104)%.  
101 Duplicate samples were -2 (-3, 0)% different. Flux detection limits for individual NMHC were  
102 0.05 (0.04, 0.06) mg m<sup>-2</sup> h<sup>-1</sup>.

103 We measured fluxes from a 3 mm PTFE sheet as a blank (n = 8). Methane, carbon dioxide, and  
104 individual NMHC fluxes from the PTFE sheet were 0.02 (-0.01, 0.04), 16 (-31, 109), and 0.01  
105 (0.00, 0.02), respectively.

## 106 **Meteorology**

107 We used a New Mountain NM150WX to measure temperature, relative humidity, wind  
108 conditions, and barometric pressure at 6 m above ground. We used a Campbell CS300 to  
109 measure solar radiation and a Campbell CS655 to measure soil moisture.

## 110 **Statistical Analysis**

111 We calculated 95% confidence limits in R using bootstrapping and the BCa method (DiCiccio  
112 and Efron, 1996). We present results as bootstrapped means with lower and upper confidence  
113 limits in parentheses.

## 114 **RESULTS AND DISCUSSION**

115 Methane, total NMHC, and alcohol fluxes from the remediated landfarm were -0.54 (-2.41,  
116 0.10), -0.10 (-0.78, 0.34), and -0.03 (-0.63, 0.31) mg m<sup>-2</sup> h<sup>-1</sup>, respectively. In a student's t-test ( $\alpha$   
117 = 0.05), these values were not different from fluxes from a 3 mm thick PTFE sheet (n = 8) and  
118 from undisturbed soils in eastern Utah (n = 28). Undisturbed soils had methane and total NMHC  
119 fluxes of -0.06 (-0.09, -0.02) and 0.50 (-0.05, 1.05) mg m<sup>-2</sup> h<sup>-1</sup>, respectively (Lyman et al., 2017).

120 Fluxes from the active landfarm were extremely spatially variable. Some measurements were  
121 collected on soils that were black with visible oil, while others were collected on soils that were  
122 indistinguishable in color from natural soil. Figure 1 shows the distribution of total NMHC  
123 fluxes from the active landfarm.

124 Table 1 shows summary statistics for measurements from the active landfarm. The highest  
125 measured fluxes were of carbon dioxide, likely indicating that soil bacteria were actively  
126 decomposing organic compounds that were added to the soil. Carbon dioxide flux was well  
127 correlated with TNMHC flux ( $r^2 = 0.72$ ), but poorly correlated with methane flux ( $r^2 = 0.13$ ).  
128 TNMHC fluxes from the active landfarm were correlated with solar radiation ( $r^2 = 0.40$ ) and  
129 with air temperature inside the chamber ( $r^2 = 0.25$ ), but not with any other of the meteorological  
130 variables measured, including ambient air temperature ( $r^2 = 0.01$ ) and soil temperature ( $r^2 =$   
131 0.01). Air temperature inside the chamber (26.8 (23.2, 30.3) °C) was higher than air temperature  
132 outside (16.5 (14.0, 19.1) °C), and air temperature inside the chamber was correlated with solar  
133 radiation ( $r^2 = 0.37$ ). It may be that higher temperature inside the chamber led to increased  
134 volatilization of NMHC from the soil (Eklund, 1992).

135 Figure 2 shows the speciation of measured organic compound fluxes. Alkanes made up 77% of  
136 the total organic compound flux from the active landfarm, and aromatics accounted for 20%,  
137 while methane accounted for only 1%. Methane fluxes from the active landfarm were not  
138 significantly higher than at the remediated landfarm ( $p = 0.37$ ) and were very low compared to

139 total NMHC. Light alkane fluxes were also relatively low, with C1-C5 alkanes comprising only  
140 2% of the average total organic compound flux. Also, benzene and toluene (C6 and C7  
141 aromatics in Figure III.C.1), comprised a minority of the aromatic flux, which was dominated by  
142 C8 and C9 aromatics.

143 The NMHC flux speciation from the active landfarm is in contrast to the speciation of crude oil  
144 in the Uinta Basin (ERT, 2015). Excluding compounds in crude oil with more than 11 carbons  
145 (since our analytical system only measured C2-C11 NMHC), the percent of NMHC flux from  
146 the active landfarm that was C3-C4 compounds was only 48% as much as that in crude oil, while  
147 the percent of flux that was C6-C8 compounds was 3.4 times greater than in crude oil, and the  
148 percent of flux that was C10-C11 compounds was roughly equivalent to the makeup of crude oil.  
149 The underrepresentation of light compounds in the flux relative to the composition of crude oil  
150 was likely due to the fact that these compounds rapidly volatilize, and the solid waste/soil  
151 mixtures we sampled had likely already been depleted of these compounds before we sampled  
152 them. We expect that the over-representation of C6-C8 compounds in the flux relative to crude  
153 oil was due to the fact that these compounds are of intermediate volatility—they survive in oily  
154 waste long enough to still be there when we sampled, but they are volatile enough that they were  
155 actively emitting from the waste at the time of sampling. As the number of carbon atoms  
156 increases, NMHC become less and less volatile and are more likely to remain in the soil and  
157 eventually be decomposed (Hejazi et al., 2003).

158 Landfarms in the Uinta Basin commonly cover 1-3 ha. Assuming the fluxes shown in Table 1  
159 occur over the area of a 2 ha landfarm, that landfarm would emit 95.3 (54.3, 179.7) kg day<sup>-1</sup> of  
160 total NMHC. Fluxes of methanol, benzene, and total BTEX (benzene, toluene, ethylbenzene,  
161 and xylenes) would be 0.10 (-0.25, 0.36), 0.4 (0.12, 0.84), and 11.2 (4.3, 33.9) kg day<sup>-1</sup>,  
162 respectively. For comparison, extrapolation of measurements by Pardue and Valsaraj (2000) at  
163 active landfarms in Louisiana over a 2 ha area would result in total BTEX emissions of 3.2 (1.8,  
164 6.9) kg day<sup>-1</sup>.

165 It is impossible to accurately scale up our measurements to the level of the entire Uinta Basin,  
166 since the total acreage of landfarms in the Basin is not known, we didn't collect any  
167 measurements in summer or winter when fluxes could be higher or lower, we only sampled at  
168 one facility, the measurements we collected showed high variability, etc. If we make the

169 assumption that the measurements we collected at this facility are representative of other  
170 seasons, we calculate annual emissions of total NMHC of 35 tonnes, or annual BTEX emissions  
171 of 4 tonnes. Emissions are highest immediately after oil and gas solid waste is exposed to the  
172 atmosphere (Ausma et al., 2002; Pardue and Valsaraj, 2000), and for safety reasons we were not  
173 able to sample waste as it was being applied to the landfarm. Thus, we expect that our  
174 measurements significantly underestimate true landfarm emissions at this facility.

## 175 **CONCLUSIONS**

176 We measured fluxes of methane, NMHC, and alcohols at a facility that contained a remediated  
177 and an active landfarm. Fluxes from the remediated landfarm were low and similar to  
178 undisturbed soils in the region, indicating that most of the added volatile organic compounds had  
179 decomposed or volatilized. Fluxes from the active landfarm were spatially variable. Fluxes  
180 were dominated by alkanes, especially compounds with 6 or more carbons. Fluxes of aromatics  
181 were dominated by compounds with 8 and 9 carbons. Total organic compound flux from the  
182 active landfarm was 216.3 (126.8, 391.7) mg m<sup>-2</sup> h<sup>-1</sup>. We estimate that a 2 ha landfarm would  
183 emit at least an average of 95.3 (54.3, 179.7) kg day<sup>-1</sup> of total NMHC, including 11.2 (4.3, 33.9)  
184 kg day<sup>-1</sup> of BTEX compounds.

## 185 **ACKNOWLEDGEMENTS**

186 This work was funded by the Department of Energy and the Research Partnership to Secure  
187 Energy for America (Contract No. 12122-15). We are very grateful to the private company that  
188 allowed us access to their landfarm for this project.

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283 **Table 1. Summary of fluxes measured from the active landfarm.**

<b>mg m<sup>-2</sup> h<sup>-1</sup></b>	<b>Average</b>	<b>Lower limit (95% conf.)</b>	<b>Upper limit (95% conf.)</b>
Methane	1.41	0.37	4.19
Carbon dioxide	3258	1642	7367
Total NMHC	197.9	114.7	370.5
Alkanes	156.2	93.27	271.3
Alkenes	1.66	0.42	4.06
Aromatics	39.90	19.14	102.8
Alcohols	4.17	0.01	16.30

284

285

286 Figure 1. Map showing the location of measured TNMHC fluxes from the active landfarm (circle  
287 coloration is by quantile). Flux magnitude, in units of  $\text{mg m}^{-2} \text{h}^{-1}$ , is shown for each  
288 measurement location. The shape of the landfarm has been stylized and the orientation changed  
289 to protect the identity of the owner. The scale and the measurement positions relative to each  
290 other are accurate.

291 Figure 2. Fractions of emission flux from an active landfarm that were due to alkanes, aromatics,  
292 alkenes, alcohols, and methane. Fractions of alkane and aromatic emissions are also broken  
293 down by carbon number.

294 **Seth Lyman** and **Marc Mansfield** are scientists affiliated with Utah State University's Bingham  
295 Research Center. Seth is a Research Associate Professor in USU's Department of Chemistry and  
296 Biochemistry, and Marc is a Research Professor in the same department.

297