FIELD MEASUREMENT OF NON-METHANE ORGANIC COMPOUND EMISSIONS FROM LANDFILL COVER SOILS

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SUMMARY: Emissions of methane (CH_4) and more than 30 non-methane organic compounds (NMOCs) were quantified at two French landfills: Lapouyade (near Bordeaux) and Grand'Landes (near Nantes). At Lapouyade, three areas were investigated: the Phase I final cover area; the Phase II temporary cover area; and a forest control area. At Grand'Landes, emissions from three areas were measured: the cell 25B final cover area with a geomembrane overlying an innovative gas collection layer; the cell 25A final cover area; and a field control area. Methane emissions from the final cover areas had high spatial variability including both positive and negative fluxes; however, all of the methane fluxes from cell 25B (geomembrane) were negative, indicating atmospheric uptake. Speciated NMOC fluxes from final cover areas were very small with both positive and negative fluxes on the order of 10^{-8} to 10^{-5} $g \cdot m^{-2} \cdot d^{-1}$. Methane emissions from the temporary cover area at Lapouyade were higher (78.2 g CH_4 $m^{-2} \cdot d^{-1}$) with mainly positive NMOC fluxes on the order of 10^{-5} to 10^{-4} $g \cdot m^{-2} \cdot d^{-1}$. In general emitted compounds consisted of species recalcitrant to aerobic degradation (especially higher chlorinated compounds such as perchloroethylene and CFCs), while negative emissions (uptake from the atmosphere) were observed for more degradable species such as the aromatics. To investigate methanotrophic oxidation capacities for both CH_4 and NMOC species, microcosms containing Lapouyade soils were incubated aerobically with CH_4; these are discussed in a separate paper (Scheutz et al., 2003a). This study demonstrated that: (1) both the oxidation of CH_4 and the co-oxidation of NMOCs in cover soils lowers emissions to the atmosphere; and (2) cover soils can function both as sources and sinks for atmospheric CH_4 and NMOCs.

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1. INTRODUCTION

Landfilled refuse decomposes anaerobically with production of methane and carbon dioxide (CO_2). Methane from landfills is strongly implicated in global change scenarios as a major source of anthropogenic CH_4 in many developed countries. Landfill gas also contains trace quantities of many other hydrocarbons, including C_2 and higher alkanes, aromatics, halogenated hydrocarbons, and organic sulfur compounds. The NMOC species are either volatilized directly from waste materials or produced through biochemical reactions during waste degradation. Even though individual NMOC species are typically quantified at ppmv to pptv levels in composite landfill gas collected from headers, there is environmental concern with respect to their net emissions because some species are known carcinogens (benzene and vinyl chloride) while the chlorofluorocarbons (CFCs) contribute to ozone depletion in the upper atmosphere (Rowland and Molina, 1974).

Several laboratory and field studies (Kightley et al., 1995; Czepiel et al. 1996; Bogner et al., 1997b; Liptay et al., 1998; Chanton et al., 1999; Christophersen et al., 2000, 2001; Scheutz, 2003) have documented that landfill cover soils exposed to CH_4 can develop high capacities for CH_4 oxidation by indigenous methanotrophic microorganisms. Critical variables include soil texture and moisture content, temperature, CH_4 and O_2 concentrations, and nutrients (Whalen et al., 1990; Börgesson et al., 1998; Boeckx and Van Cleemput, 1996; Scheutz and Kjeldsen, 2003). Recently, landfill ”biocover” designs engineered to optimize CH_4 oxidation have also been developed (Humer and Lechner, 1999; DeVisscher et al., 1999; Hilger et al., 2000). Because of the broad substrate specificity of the methane monooxygenase (MMO) enzyme, methanotrophs can also co-metabolize a variety of aliphatic compounds, including some halogenated hydrocarbons. However, there have been very few studies addressing either oxidation rates or net emissions of non-methane hydrocarbons in landfill settings. Kjeldsen et al. (1997) showed co-metabolic degradation of trichloroethylene and 1,1,1-trichloroethane in the presence of CH_4 in landfill gas-affected soil. Bogner et al. (1997c) measured very low emissions (10^{-6} to 10^{-4} g m^{-2} d^{-1}) for selected NMOC species at a northeastern Illinois landfill under “worst case” conditions (thin interim soil cover over recently landfilled waste). The study summarized below and in Scheutz et al. (2002, 2003a) provide the first complementary measurements of speciated NMOC emissions in parallel with laboratory quantification of soil attenuation capacities.

2. FIELD SITES

Lapouyade is located near Bordeaux in southwestern France. The Phase I area received more than 300,000 tonnes of waste from 1996-1998. Final cover was placed in 1998 and consists of 40 cm coarse sand overlain by 80 cm of clayey silt and topsoil; this area was fully vegetated (mixed grasses and legumes) during the field campaign in September, 2001. The upper 35 cm were very dry with a water content below 6.5% (w/w). The Phase II area was initiated in 1998; this area includes the current operational zone and a temporary cover zone where the refuse is overlain by 40 cm coarse sand. In addition, a forested control area immediately outside of the landfill was investigated.

Grand’Landes is located in western France near Nantes. This 26 ha landfill has been accepting approximately 185,000 tonnes waste yr^{-1} since 1989. The field campaign in September, 2002, focused on two experimental cells, 25A and 25B. Each cell has a surface area of 10,000 m^2; cell 25A contains 69,000 m^3 of waste and 25B contains 59,000 m^3 of waste. Cell 25A has five conventional vertical wells and a cover design consisting of a coarse-grained leveling layer overlain by compacted clay and topsoil. The total thickness of the compacted clay/topsoil sequence is approximately 1 m. Cell 25B has an innovative gas collection system consisting of a 30 cm thick gravel layer containing two horizonal perforated pipes. The two pipes are placed perpendicular to each other with a common collection point at one corner of the cell. The collection layer is overlain, in turn, by a geotextile layer, a 1.5 mm thick geomembrane, a second geotextile layer, 70 cm compacted clay, and 30 cm topsoil. Both cells 25A and 25B are vegetated. A grassy field on the landfill property functioned as a non-landfill control.

Herein we report on emission measurements conducted at Lapouyade at four locations on the fully vegetated Phase I final cover area (LP1, LP2, LP4, and LP6); one location on the Phase II temporary cover area (location LP5); and the forest control area (LP3). For Grand'Landes, we report on a subset of the emissions measurements conducted there, including area 25B with the geomembrane (F3); area 25A with conventional soil cover (F5, F7, F8, and F9); and the field control area (BG F).

3. MATERIALS AND METHODS

Emission rates of CH_4, CO_2, and speciated NMOCs were measured using static flux chambers. At each measurement point, two circular iron collars functioned as bases for two static chambers (A and B). The collars were placed adjacent to each other at a depth of 4-5 cm in the cover soil and, during sampling periods of 20 minutes (CH_4,CO_2) or 120 minutes (NMOCs), each was topped by a chamber consisting of a stainless steel (SS) hemisphere that exactly fit a circular trough welded to the top of each collar. During monitoring periods, the trough was filled with distilled water and the chambers secured with hand clamps. Each chamber top was fitted with a single SS Swagelok sampling port for sampling using a gas-tight syringe[for CH_4 and CO_2] or direct connection to an evacuated 2 L electropolished canister [for speciated NMOCs]. Chamber volume was 31830 cm^3, and the enclosed surface area was 1217 cm^2 resulting in a volume/area ratio (cm^3 cm^{-2}) of 26.

Emission measurements for CH_4 and CO_2 relied on five gas samples of 25-50 mL withdrawn over 20-30 minutes using syringes and stored in pre-evacuated serum bottles. Based on the results of a pilot study conducted at Lapouyade in May, 2001, three 2 L stainless steel canister samples/test were taken for NMOCs in a subsequent chamber test: the first sample was taken at time zero in chamber A; after 120 minutes an additional sample was taken in both chambers A and B. The fluxes reported herein rely on the initial value from chamber A and final value from chamber B (adjacent). The second 120 minute sample from chamber A acted as a maximum check on the observed flux from the adjacent chambers. Fluxes were calculated from the product of the change in concentration over time (dc/dt) and the [chamber volume/chamber area] ratio (Rolston, 1986). In general, the CH_4 concentration vs. time curves showed good linear fits (r^2>0.9) without any change in slope for the final sampling times. Furthermore, when a sampling interval of 200 minutes was tested at a location with relatively high CH_4 emissions (7.9 g m^{-2} d^{-1}), the final data points indicated only a minor flattening of the dc/dt slope with r^2 >0.98.

Following the flux measurements using chambers, soil gas profiles were determined at the same points via gas probes installed at different depths in the soil cover. We used stainless steel tubes (10 mm ID) which were closed at the bottom and had slits at the lower 3 cm; these were hammered into the ground at depths ranging from 10 to 100 cm. Using gas-tight syringes, 25-50 mL samples for major gases (CH_4, CO_2, O_2, and N_2) were withdrawn and stored in pre-evacuated glass serum bottles. For NMOCs, samples were taken thereafter at depths ranging from 10 to 100 cm and were retained in pre-evacuated 2 L stainless steel canisters.

Concentrated source gas samples for both major gases and speciated NMOCs were taken from the main header lines to the flare. Stable carbon isotopes ($\delta^{13}C$) were determined on CH_4 in the source gas, ambient air, soil gas profile samples, and the final chamber sample.

Major gases and stable carbon isotopes ($\delta^{13}C$) were analyzed at Florida State University. Methane and CO_2 concentrations below 1% were determined on a Shimadzu 14A gas chromatograph with a flame ionization detector and a methanizer, a 1 mL sampling loop, and a 2-m 0.32 cm diameter stainless steel column packed with Carbosphere. N_2 and O_2 + Ar were determined on a Shimadzu 8A gas chromatograph with a thermal conductivity detector. Scott Specialty gases were used as standards. Stable carbon isotopic ratios were determined using a Finnigan Mat Delta S-Gas Chromatograph Combustion Isotope Ratio Mass Spectrometer (GCC-IRMS) following methods adapted from Merrit et al. (1995). For air samples, a cryogenic focusing device was used on the front end of the gas chromatograph. The standard deviation of replicate analyses is approximately 0.15‰. The high CH_4 source gas samples were diluted to 1% CH_4 with nitrogen, and 0.1 to 0.5 mL were directly injected into the GCC-IRMS inlet system.

All 2L-canister samples were analyzed by the Blake-Rowland Laboratory at the University of California–Irvine. This laboratory has two separate high-resolution analysis systems capable of identifying and quantifying over 100 non-CH_4 hydrocarbons and halocarbons from whole gas samples using multidetector GC (gas chromatography) and combined GC-MS (mass spectrometry). The analytical apparatus utilizes three gas chromatographs (GCs) and five detectors. Each whole air sample is cryogenically trapped with liquid N_2, warmed, and injected into a helium flow stream. This stream is then split into five, with each stream feeding a separate GC column. One DB-1, one PLOT $A1_2O_3$/KCl, one Restek-1701, and two DB-5MS columns are used. One of the DB-5MS columns is plumbed into an electron capture detector (ECD) to separate C_1-C_2 halocarbons while the other DB-5MS is plumbed to a mass spectrometer. The Restek-1701 column is used for alkyl nitrate separation and is connected to an ECD. The DB-1/FID combination separates C_3-C_8 hydrocarbons. The DB5MS/mass spectrometer combination quantifies C_5-C_{10} hydrocarbons. The PLOT column, also plumbed to an FID, is used for separating C_2-C_5 hydrocarbons, some of which are not resolved adequately by the DB-1.

The preparation of standards for the halocarbons has been discussed thoroughly in previous publications (Colman et al., 2001). The range for halocarbon standards is 0.5-600 pptv, and concentration accuracies range between 1-20%. Calibration of the other NMOC compounds has been achieved by employing Scott calibration gases available in the 1-100 ppmv mixing ratio range. The measurement precisions for the halocarbons, hydrocarbons, and alkyl nitrates are in the 1-10% range.

4. RESULTS AND DISCUSSION

Source gas composition. Table 1 gives source gas composition and comparative fluxes for CH_4, CO_2, and NMOC species at Lapouyade; Table 2 summarizes results from Grand'Landes. The composite landfill gas in the collection header at Lapouyade averaged 49% CH_4 while the source gas at Grand'Landes averaged 33 V% CH_4; both sites had some air intrusion. All NMOCs were detected and quantified in the source gas at both Lapouyade and Grand'Landes. Toluene, xylenes, ethylbenzene, n-nonane, n-decane, perchloroethylene, and dichloromethane exhibited the highest concentrations ($>10\ \mu g\ L^{-1}$) at Lapouyade while n-nonane and n-decane were elevated in the 25A header at Grand'Landes, and selected aromatics were elevated in both 25A and 25B headers. In general, the NMOC concentrations in the source gas tended to be lower than reported by Brosseau and Heitz (1994) and values for seven landfills in the UK receiving both municipal waste and hazardous waste reported by Allen et al. (1997). The data were reasonably comparable to values from Eklund et al. (1998) for the Fresh Kills Landfill, New York, USA.

Table 1. Source gas concentrations (μg L^{-1} except CH_4 and CO_2) and surface emissions (g m^{-2} d^{-1}) at Lapouyade.

	Chemical formula	Source gas conc. μg·L^{-1}	LP1 Flux g·m^{-2}·d^{-1}	LP2 Flux g·m^{-2}·d^{-1}	LP4 Flux g·m^{-2}·d^{-1}	LP6 Flux g·m^{-2}·d^{-1}	LP5 Flux g·m^{-2}·d^{-1}	LP3 Flux g·m^{-2}·d^{-1}
Cover/Location			Final, soil	Final, soil	Final, soil	Final, hotspot	Temporary	Forest control
Methane	CH_4	48.5% v/v	0.0084	-0,0095	-0,0104	10.0	49.9	-0.0033
Carbon dioxide	CO_2	33.7% v/v	8.0	13.1	15.6	77.3	107.4	19.3
Oxidation (%)*			8.5	6.6	12.3	33.5	5.1	ND**
Alkanes								
Ethane	C_2H_6	1.5	$1.42\cdot10^{-6}$	$6.94\cdot10^{-6}$	$4.69\cdot10^{-6}$	$2.13\cdot10^{-5}$	$1.60\cdot10^{-4}$	$8.60\cdot10^{-7}$
Propane	C_3H_8	2.6	$-6.45\cdot10^{-5}$	$1.74\cdot10^{-5}$	$7.91\cdot10^{-6}$	$5.87\cdot10^{-5}$	$7.36\cdot10^{-5}$	$2.48\cdot10^{-6}$
n-Butane	C_4H_{10}	5.5	$-1.45\cdot10^{-4}$	$7.18\cdot10^{-6}$	$3.47\cdot10^{-6}$	$1.72\cdot10^{-5}$	$1.69\cdot10^{-4}$	$-1.64\cdot10^{-7}$
n-Pentane	C_5H_{12}	1.7	$-3.64\cdot10^{-6}$	$1.03\cdot10^{-5}$	$1.42\cdot10^{-5}$	$7.74\cdot10^{-6}$	$7.04\cdot10^{-5}$	$5.55\cdot10^{-7}$
n-Hexane	C_6H_{14}	0.8	$6.26\cdot10^{-7}$	$2.68\cdot10^{-6}$	$2.60\cdot10^{-6}$	$1.40\cdot10^{-6}$	$6.55\cdot10^{-5}$	$3.74\cdot10^{-8}$
n-Heptane	C_7H_{16}	1.2	$-2.91\cdot10^{-6}$	$1.21\cdot10^{-6}$	$1.66\cdot10^{-6}$	$-9.75\cdot10^{-6}$	$4.33\cdot10^{-4}$	$-7.60\cdot10^{-8}$
n-Octane	C_8H_{18}	0.7	$-1.66\cdot10^{-6}$	$7.00\cdot10^{-8}$	$8.44\cdot10^{-7}$	$-9.44\cdot10^{-6}$	$2.34\cdot10^{-4}$	BDL***
n-Nonane	C_9H_{20}	28.1	$-1.47\cdot10^{-5}$	$-7.86\cdot10^{-8}$	$-8.87\cdot10^{-7}$	$-2.56\cdot10^{-5}$	$8.14\cdot10^{-5}$	$-3.06\cdot10^{-7}$
n-Decane	$C_{10}H_{22}$	11.2	$-7.18\cdot10^{-5}$	$3.49\cdot10^{-7}$	$-1.94\cdot10^{-6}$	$-3.19\cdot10^{-5}$	$3.21\cdot10^{-5}$	$-1.23\cdot10^{-7}$
i-Butane	C_4H_{10}	5.0	$-1.15\cdot10^{-4}$	$5.79\cdot10^{-6}$	$3.00\cdot10^{-6}$	$3.70\cdot10^{-5}$	$1.18\cdot10^{-4}$	$2.02\cdot10^{-7}$
i-Pentane	C_5H_{12}	7.0	$-2.57\cdot10^{-5}$	$3.39\cdot10^{-6}$	$3.51\cdot10^{-6}$	$1.91\cdot10^{-5}$	$2.23\cdot10^{-4}$	$7.82\cdot10^{-8}$
2-methylpentane	C_6H_{14}	3.0	BDL	BDL	BDL	$1.19\cdot10^{-6}$	BDL	$-3.08\cdot10^{-7}$
3-methylpentane	C_6H_{14}	1.0	$-9.33\cdot10^{-7}$	BDL	BDL	$1.78\cdot10^{-7}$	BDL	$-2.33\cdot10^{-7}$
Alkenes								
Ethene	C_2H_4	5.2	$1.04\cdot10^{-5}$	$2.92\cdot10^{-6}$	$1.84\cdot10^{-5}$	$5.19\cdot10^{-6}$	$1.68\cdot10^{-5}$	$-2.28\cdot10^{-8}$
Propene	C_3H_6	5.5	$7.19\cdot10^{-7}$	$5.37\cdot10^{-6}$	$3.65\cdot10^{-6}$	$-3.85\cdot10^{-6}$	$1.72\cdot10^{-4}$	$-5.26\cdot10^{-8}$
t-2-Butene	C_4H_8	0.8	$-1.94\cdot10^{-7}$	$1.19\cdot10^{-6}$	BDL	BDL	$2.42\cdot10^{-5}$	BDL
1-Butene	C_4H_8	0.3	$-6.01\cdot10^{-7}$	$1.99\cdot10^{-6}$	$5.92\cdot10^{-8}$	$1.59\cdot10^{-6}$	$5.35\cdot10^{-5}$	$-3.65\cdot10^{-8}$
i-Butene	C_4H_8	2.7	$3.16\cdot10^{-6}$	$9.44\cdot10^{-6}$	$1.20\cdot10^{-6}$	$-7.15\cdot10^{-7}$	$7.96\cdot10^{-5}$	$3.04\cdot10^{-7}$
c-2-Butene	C_4H_8	0.4	$-2.67\cdot10^{-8}$	$5.33\cdot10^{-7}$	BDL	$-4.56\cdot10^{-7}$	$1.84\cdot10^{-5}$	BDL
Isoprene	C_2H_2	0.4	$4.86\cdot10^{-7}$	$1.08\cdot10^{-6}$	$-3.27\cdot10^{-7}$	$-1.34\cdot10^{-6}$	$1.54\cdot10^{-5}$	$-3.23\cdot10^{-6}$
Ethyne	C_5H_8	0.6	$-1.08\cdot10^{-7}$	$-6.34\cdot10^{-7}$	$1.34\cdot10^{-7}$	-3.50E-07	$-4.65\cdot10^{-7}$	$-3.89\cdot10^{-7}$
Halogenated hydrocarbons								
CFC-11	CCl_3F	2.0	$-7.92\cdot10^{-5}$	$5.18\cdot10^{-6}$	$2.24\cdot10^{-6}$	$7.63\cdot10^{-5}$	$2.08\cdot10^{-5}$	$5.21\cdot10^{-7}$
CFC-12	CCl_2F_2	5.7	$-1.68\cdot10^{-5}$	$2.17\cdot10^{-6}$	$1.84\cdot10^{-7}$	$1.04\cdot10^{-5}$	$2.56\cdot10^{-5}$	$-7.86\cdot10^{-8}$
HCFC-22	$CHClF_2$	0.0	$-4.89\cdot10^{-6}$	$5.03\cdot10^{-7}$	$-4.06\cdot10^{-8}$	$2.26\cdot10^{-5}$	$5.74\cdot10^{-5}$	$-1.50\cdot10^{-7}$
H-1211	$CBrClF_2$	0.8	$-5.90\cdot10^{-5}$	BDL	BDL	BDL	BDL	BDL
Trichloromethane	$CHCl_3$	0.0	$4.26\cdot10^{-7}$	$6.58\cdot10^{-7}$	$7.84\cdot10^{-7}$	$4.56\cdot10^{-6}$	$1.02\cdot10^{-6}$	$7.56\cdot10^{-6}$
Dichloromethane	CH_2Cl_2	10.3	$-2.10\cdot10^{-5}$	$2.08\cdot10^{-7}$	$4.98\cdot10^{-8}$	$-1.06\cdot10^{-5}$	$-3.22\cdot10^{-7}$	BDL
Chloromethane	CH_3Cl	0.1	$-2.24\cdot10^{-6}$	$-4.98\cdot10^{-7}$	$1.64\cdot10^{-6}$	$-3.90\cdot10^{-6}$	$-2.04\cdot10^{-6}$	$-3.05\cdot10^{-6}$
Perchloroethylene	C_2Cl_4	47.4	$-2.37\cdot10^{-7}$	$1.37\cdot10^{-6}$	$1.75\cdot10^{-7}$	$2.03\cdot10^{-6}$	$2.30\cdot10^{-5}$	$7.19\cdot10^{-8}$
Trichloroethylene	C_2HCl_3	0.8	BDL	BDL	BDL	$-1.08\cdot10^{-6}$	BDL	BDL
Vinyl chloride	C_2H_3Cl	2.4	BDL	BDL	BDL	$-1.03\cdot10^{-6}$	$1.03\cdot10^{-5}$	BDL
Aromatics								
Benzene	C_6H_6	1.8	$5.67\cdot10^{-7}$	$5.14\cdot10^{-7}$	$2.25\cdot10^{-7}$	$-3.92\cdot10^{-6}$	$3.41\cdot10^{-5}$	$-2.88\cdot10^{-7}$

Toluene	C_7H_8	76.8	$6.97\cdot10^{-6}$	$5.56\cdot10^{-6}$	$1.03\cdot10^{-6}$	$-3.57\cdot10^{-5}$	$-2.18\cdot10^{-5}$	$-3.20\cdot10^{-7}$
Ethylbenzene	C_8H_{10}	24.8	$6.70\cdot10^{-7}$	$2.75\cdot10^{-6}$	$-1.05\cdot10^{-6}$	$-2.78\cdot10^{-5}$	$-6.96\cdot10^{-5}$	$-1.38\cdot10^{-7}$
Xylenes Σ[m,p,o]	C_8H_{10}	85.5	$1.24\cdot10^{-5}$	$1.45\cdot10^{-5}$	$2.8\cdot10^{-6}$	$-3.24\cdot10^{-5}$	$3.71\cdot10^{-4}$	$1.28\cdot10^{-6}$

*Determined by isotopic analysis of the emitted CH_4 (Liptay et al., 1998; Chanton et al., 1999; Chanton and Liptay, 2000) **ND=not determined or not applicable.
***BDL = below detection limit (20 pptv for most species).

Table 2. Source gas concentrations ($\mu g\ L^{-1}$ except CH_4 and CO_2) and surface emissions ($g\ m^{-2}\ d^{-1}$) at Grand'Landes.

	25A Source Gas Conc. $\mu g\cdot L^{-1}$	25B Source Gas Conc. $\mu g\cdot L^{-}$	25B-F3 Flux $g\cdot m^{-2}\cdot d^{-1}$	25A-F5 Flux $g\cdot m^{-2}\cdot d^{-1}$	25A-F7 Flux $g\cdot m^{-2}\cdot d^{-1}$	25A-F8 Flux $g\cdot m^{-2}\cdot d^{-1}$	25A-F9 Flux $g\cdot m^{-2}\cdot d^{-1}$	BG F Flux $g\cdot m^{-2}\cdot d^{-1}$
Cover/Location			Final,geombn	Final, soil	Final, soil	Final, soil	Edge, hotspot	Field control
Methane	37.5 % v/v	29.1 % v/v	-0.0022	0.0	-0.0011	0.0001	29.0	4.8
Carbon dioxide	25.1 % v/v	25.0 % v/v						
Oxidation (%)*			ND	ND	ND	ND	7.1	ND
Alkanes								
Ethane	2.3	2.06	$3.73\cdot10^{-6}$	$3.45\cdot10^{-6}$	$4.64\cdot10^{-6}$	$4.11\cdot10^{-6}$	$3.22\cdot10^{-5}$	$8.99\cdot10^{-6}$
Propane	3.48	2.7	$-3.03\cdot10^{-6}$	$2.33\cdot10^{-6}$	$2.08\cdot10^{-6}$	$3.92\cdot10^{-6}$	$5.98\cdot10^{-5}$	$5.24\cdot10^{-6}$
n-Butane	6.21	3.94	$-2.39\cdot10^{-6}$	$1.73\cdot10^{-6}$	$7.77\cdot10^{-7}$	$2.53\cdot10^{-6}$	$4.73\cdot10^{-5}$	$2.15\cdot10^{-6}$
n-Pentane	1.73	3.26	$7.77\cdot10^{-6}$	$6.23\cdot10^{-6}$	$6.19\cdot10^{-6}$	$5.77\cdot10^{-6}$	$3.47\cdot10^{-5}$	$1.28\cdot10^{-5}$
n-Hexane	0.75	0.67	$1.76\cdot10^{-7}$	$1.18\cdot10^{-6}$	$4.80\cdot10^{-7}$	BDL	$2.03\cdot10^{-6}$	$1.19\cdot10^{-6}$
n-Heptane	5.27	3.75	$-3.92\cdot10^{-7}$	$-1.41\cdot10^{-7}$	$-1.30\cdot10^{-7}$	$-5.87\cdot10^{-6}$	$3.44\cdot10^{-7}$	$1.21\cdot10^{-6}$
n-Octane	3.51	1.21	$-4.76\cdot10^{-7}$	$-1.16\cdot10^{-7}$	$-9.55\cdot10^{-8}$	$-3.30\cdot10^{-6}$	$1.01\cdot10^{-5}$	$-1.01\cdot10^{-6}$
n-Nonane	10.60	2.37	$-1.20\cdot10^{-5}$	$9.31\cdot10^{-6}$	$1.19\cdot10^{-8}$	$-2.68\cdot10^{-5}$	$2.71\cdot10^{-7}$	$-8.69\cdot10^{-6}$
n-Decane	16.27	2.74	$-4.71\cdot10^{-6}$	$-2.56\cdot10^{-7}$	$-1.27\cdot10^{-5}$	$-3.76\cdot10^{-4}$	$1.63\cdot10^{-5}$	$-1.94\cdot10^{-5}$
i-Butane	5.02	2.96	$3.11\cdot10^{-7}$	$1.88\cdot10^{-6}$	$1.84\cdot10^{-7}$	$-1.10\cdot10^{-6}$	$7.29\cdot10^{-5}$	$6.69\cdot10^{-7}$
i-Pentane	3.94	8.19	$-1.23\cdot10^{-7}$	$5.14\cdot10^{-7}$	$4.22\cdot10^{-7}$	$-1.71\cdot10^{-6}$	$2.11\cdot10^{-4}$	$1.15\cdot10^{-6}$
2-methylpentane	1.5	1.76	$2.20\cdot10^{-8}$	$2.70\cdot10^{-7}$	$2.24\cdot10^{-7}$	$1.59\cdot10^{-8}$	$5.00\cdot10^{-6}$	$4.22\cdot10^{-7}$
3-methylpentane	0.81	0.99	$-2.13\cdot10^{-7}$	$2.70\cdot10^{-8}$	$1.60\cdot10^{-8}$	$-3.50\cdot10^{-7}$	$7.72\cdot10^{-6}$	$1.33\cdot10^{-7}$
Alkenes								
Ethene	2.76	1.74	$4.80\cdot10^{-5}$	$7.35\cdot10^{-5}$	$3.50\cdot10^{-5}$	$2.84\cdot10^{-5}$	$6.87\cdot10^{-6}$	$1.99\cdot10^{-4}$
Propene	7.77	5.00	$2.24\cdot10^{-5}$	$3.56\cdot10^{-6}$	$1.62\cdot10^{-6}$	$3.05\cdot10^{-6}$	$9.25\cdot10^{-6}$	$4.22\cdot10^{-6}$
t-2-Butene	0.26	0.24	$-7.68\cdot10^{-7}$	$2.20\cdot10^{-8}$	$4.69\cdot10^{-8}$	$-1.82\cdot10^{-6}$	$1.56\cdot10^{-6}$	$2.27\cdot10^{-7}$
1-Butene	0.25	0.24	$-5.53\cdot10^{-7}$	$-6.46\cdot10^{-7}$	$2.50\cdot10^{-7}$	$-5.74\cdot10^{-7}$	$4.91\cdot10^{-6}$	$6.70\cdot10^{-7}$
i-Butene	0.29	0.42	$1.42\cdot10^{-6}$	$-3.25\cdot10^{-7}$	$1.05\cdot10^{-6}$	$5.64\cdot10^{-7}$	$7.40\cdot10^{-6}$	$3.28\cdot10^{-6}$
c-2-Butene	0.25	0.20	$-5.49\cdot10^{-7}$	$-8.79\cdot10^{-8}$	$5.21\cdot10^{-8}$	$-2.27\cdot10^{-6}$	$1.03\cdot10^{-6}$	$1.69\cdot10^{-7}$
Isoprene	0.12	0.11	$4.11\cdot10^{-7}$	$5.39\cdot10^{-7}$	$4.30\cdot10^{-7}$	$6.16\cdot10^{-6}$	$-2.22\cdot10^{-7}$	$-4.27\cdot10^{-7}$
Ethyne	0.17	0.11	$6.84\cdot10^{-7}$	$5.12\cdot10^{-7}$	$-3.99\cdot10^{-7}$	$-3.14\cdot10^{-8}$	$-1.78\cdot10^{-6}$	$6.56\cdot10^{-7}$
Halogenated hydrocarbons								
CFC-11	3.05	1.62	$6.54\cdot10^{-7}$	$3.73\cdot10^{-5}$	$1.33\cdot10^{-6}$	$7.86\cdot10^{-7}$	$4.36\cdot10^{-7}$	$2.66\cdot10^{-6}$
CFC-12	0.67	4.90	$-2.16\cdot10^{-7}$	$-2.27\cdot10^{-7}$	$5.39\cdot10^{-7}$	$6.02\cdot10^{-7}$	$-2.13\cdot10^{-8}$	$-1.56\cdot10^{-7}$
HCFC-22	1.84	1.25	$-1.54\cdot10^{-7}$	$-6.10\cdot10^{-8}$	$1.85\cdot10^{-7}$	$-2.39\cdot10^{-8}$	$4.64\cdot10^{-6}$	$-5.20\cdot10^{-8}$
H-1211	BDL	BDL	$-8.44\cdot10^{-9}$	$-3.89\cdot10^{-9}$	$4.61\cdot10^{-9}$	$-3.05\cdot10^{-9}$	$4.37\cdot10^{-9}$	$2.84\cdot10^{-9}$
Trichloromethane	BDL	0.02	$-5.03\cdot10^{-6}$	$3.14\cdot10^{-5}$	$4.46\cdot10^{-5}$	$1.80\cdot10^{-5}$	$-1.29\cdot10^{-6}$	$-6.15\cdot10^{-6}$
Dichloromethane	0.18	0.41	$-2.75\cdot10^{-7}$	$-3.13\cdot10^{-7}$	$-2.60\cdot10^{-7}$	$-6.74\cdot10^{-7}$	$-8.53\cdot10^{-7}$	$-6.42\cdot10^{-7}$
Chloromethane	0.57	0.69	$-8.08\cdot10^{-7}$	$3.37\cdot10^{-7}$	$-4.41\cdot10^{-7}$	$-2.36\cdot10^{-6}$	$2.64\cdot10^{-7}$	$-2.71\cdot10^{-7}$

Perchloroethylene	1.65	0.63	$-1.41\cdot10^{-7}$	$-6.24\cdot10^{-7}$	$1.85\cdot10^{-7}$	$1.38\cdot10^{-7}$	$4.38\cdot10^{-7}$	$3.85\cdot10^{-7}$
Trichloroethylene	0.19	0.10	$-2.24\cdot10^{-9}$	$1.85\cdot10^{-8}$	$-4.76\cdot10^{-8}$	$-4.30\cdot10^{-7}$	$4.02\cdot10^{-7}$	$1.32\cdot10^{-7}$
Aromatics								
Benzene	0.74	0.30	$-1.43\cdot10^{-6}$	$-8.87\cdot10^{-7}$	$-2.03\cdot10^{-7}$	$-1.53\cdot10^{-6}$	$8.26\cdot10^{-8}$	$-7.38\cdot10^{-8}$
Toluene	20.59	6.87	$-5.71\cdot10^{-6}$	$-1.43\cdot10^{-6}$	$-3.24\cdot10^{-6}$	$-4.68\cdot10^{-5}$	$-1.69\cdot10^{-6}$	$4.05\cdot10^{-6}$
Ethylbenzene	33.8	13.49	$-2.16\cdot10^{-6}$	$-3.08\cdot10^{-7}$	$-3.33\cdot10^{-6}$	$-8.56\cdot10^{-5}$	$5.92\cdot10^{-6}$	$-3.31\cdot10^{-6}$
Xylenes Σ[m,p,o]	52.84	15.47	$-6.76\cdot10^{-6}$	$-9.48\cdot10^{-7}$	$-3.27\cdot10^{-5}$	$-4.05\cdot10^{-4}$	$3.37\cdot10^{-5}$	$-3.26\cdot10^{-5}$

See notes on Table 1.

Methane and speciated NMOC emissions. Emissions of CH_4 and selected NMOC species from the final cover, temporary cover, and adjacent forest area (control) at Lapouyade are shown in Table 1. Observed CH_4 fluxes from the final cover area varied between –0.01 and 0.008 g·m^{-2}·d^{-1}; however, LP6 was a hot spot with high CH_4 flux (10.0 g m^{-2} d^{-1}). The negative fluxes (LP2 and LP4) indicate oxidation of atmospheric CH_4 with no landfill CH_4 emissions, as reported in previous field studies (Bogner et al., 1995, 1997b, 1999; Börjesson and Svensson, 1997). In a parallel field investigation at Lapouyade using dynamic flux chambers placed randomly on the final cover area, the average CH_4 flux from the finished cell was 1.97±0.88 g m^{-2} d^{-1} with hot spots exhibiting high fluxes of 3.7-16.2 g m^{-2} d^{-1}; for the temporary cover cell, CH_4 emissions were higher, averaging 37.8±14 g m^{-2} d^{-1} with a maximum flux of 78.2 g m^{-2} d^{-1}. Using a stable carbon isotopic method described in previous publications (Liptay et al., 1998; Chanton et al., 1999; Chanton and Liptay, 2000), the fractional CH_4 oxidation ranged from 7-34% for the final cover cell and 5% for the temporary cover cell. Due to differences in CH_4 flux from the anaerobic zone, design of cover soils and CH_4 oxidation rates, a wide range of CH_4 emission rates have been reported in the literature for landfill settings, varying between 0.0004 and 4000 g m^{-2} d^{-1} with negative emissions also possible (Bogner et al., 1997a). The CO_2 fluxes at Lapouyade using an opaque chamber quantified the dark respiration of the soil-plant community. The observed values ranged from similar to higher than natural soils; the high values were associated with hot spots (high CH_4 flux). We speculate that the high CO_2 fluxes were a combination of increased soil respiration due to higher temperatures, oxidized landfill CH_4, and directly-emitted landfill CO_2.

At Lapouyade, the speciated NMOC fluxes from the final cover area were all very small with positive and negative fluxes on the order of 10^{-7} to 10^{-5} g m^{-2} d^{-1}. Species with negative fluxes included aromatics, n-nonane, n-decane, and ethyne. Previously, Bogner et al.(1997c) had also observed negative emissions for selected aromatics. NMOC flux rates in the forest control area were generally lower (order of 10^{-8} to 10^{-7} g m^{-2} d^{-1}) and negative. Higher and mainly positive fluxes in the order of 10^{-5} to 10^{-4} g·m^{-2}·day^{-1} were obtained from the temporary cover area. In the previous Illinois study (Bogner et al., 1997c), emissions of most NMOC species from an area with temporary cover (35 cm stony clay) were generally 10^{-5} to 10^{-3} g m^{-2} d^{-1}, thus comparable to emissions from the temporary cover area (LP5) at Lapouyade. Selected NMOCs are also emitted naturally from soil surfaces and plants; for example, Fukui and Doskey (1998) measured isoprene emissions from a grassland site ($4.3 \cdot 10^{-5}$ g m^{-2} d^{-1}) that were higher than the isoprene fluxes at Lapouyade. It cannot be excluded that surface emissions of isoprene and selected other species at Lapouyade are from natural sources. In addition, some NMOC species in the ambient air were elevated above observed levels in landfill gas [data not shown]. At the forest control station LP3, not affected by landfill gas, negative fluxes for benzene, toluene and ethylbenzene were measured. The elevated atmospheric concentrations of aromatics and other NMOCs above the soil surface at the background site were likely due to vehicular exhaust from the nearby road, which had heavy traffic from waste trucks, or its downwind location from the active tipping area.

Air samples downwind from the active cell had significant greater CH_4 concentrations (17 to 36 ppmv) compared with upwind samples (1.92 ppmv). Moreover, air samples collected across the landfill before dawn under a nocturnal inversion had CH_4 concentrations up to 394 ppmv.

At Grand'Landes, CH_4 fluxes from a 6-point static chamber transect across cell 25B were all negative, ranging from –0.2 to –2.2 mg m^{-2} d^{-1}. [Data not shown.] Methane was thus being oxidized out of the atmosphere by methanotrophic microorganisms in the soil above the geomembrane. Five of the six fluxes from a similar transect across cell 25A also resulted in a narrow range of negative fluxes from –0.3 to –2.4 mg m^{-2} d^{-1}; one flux was essentially zero (time plot resulted in a horizontal line). Flux F9 (Table 2) was the highest CH_4 flux observed among known hot spots identified by French colleagues in dynamic chamber tests being conducted as part of the same field campaign; their mean values were 0.001 (25A; n=42) and 0.62 (25B, n=75) g CH_4 m^{-2} d^{-1} (Baroudi and Jodart, 2003).

Speciated NMOC fluxes at Grand'Landes (Table 2) were similar to observed ranges at Lapouyade with positive and negative fluxes on the order of 10^{-8} to 10^{-6} g m^{-2} d^{-1}. The highest positive fluxes on the two cells were observed for ethene (10^{-5} g m^{-2} d^{-1}), but these were exceeded by the ethene flux from the control area, suggesting a natural source. At the F9 hot spot, the highest positive flux was for i-pentane at $2.1 \cdot 10^{-4}$ g m^{-2} d^{-1}. In general, the positive fluxes at F9 were in a higher range of 10^{-7} to 10^{-4} compared to the F3, F5, F7, and F8 fluxes. Fluxes of aromatics from both the landfill cells and the control area, but not the hot spot, were predominately negative, consistent with the discussion above for Lapouyade.

*Soil gas concentration profile***.** Soil gas concentration profiles are influenced by numerous microbial, gaseous transport, and meteorological processes. If we compare three soil gas profiles from the final cover area (LP1, LP2 and LP6) and one from the temporary cover area (LP5) at Lapouyade, one can observe significant differences in concentrations among NMOC species but similar trends among chemically related compounds at the same location. This is especially true for the aromatics and the alkanes. Most NMOC species increased in concentration over several orders of magnitude from the ground surface to the top of refuse. However, some species (e.g., PCE, TCM, CFC-12) had relatively constant soil gas concentrations with depth, while others (aromatics) showed increasing gas concentrations toward the surface, consistent with atmospheric uptake often observed for individual aromatics. Figure 1 shows soil gas profiles for major gases and selected NMOCs at location LP6 (final cover) at Lapouyade. At 80 cm, the soil was fully anaerobic with high concentrations of CH_4 and CO_2. Between 80 and 60 cm, the shift in CH_4 and CO_2 concentrations and the increase in C^{13}-CH_4 together with the decrease in C^{13}-CO_2 indicates CH_4 oxidation. Note that all of the aromatics and vinyl chloride show a similar trend at 60-80 cm depth. These compounds may be readily degraded under aerobic conditions.

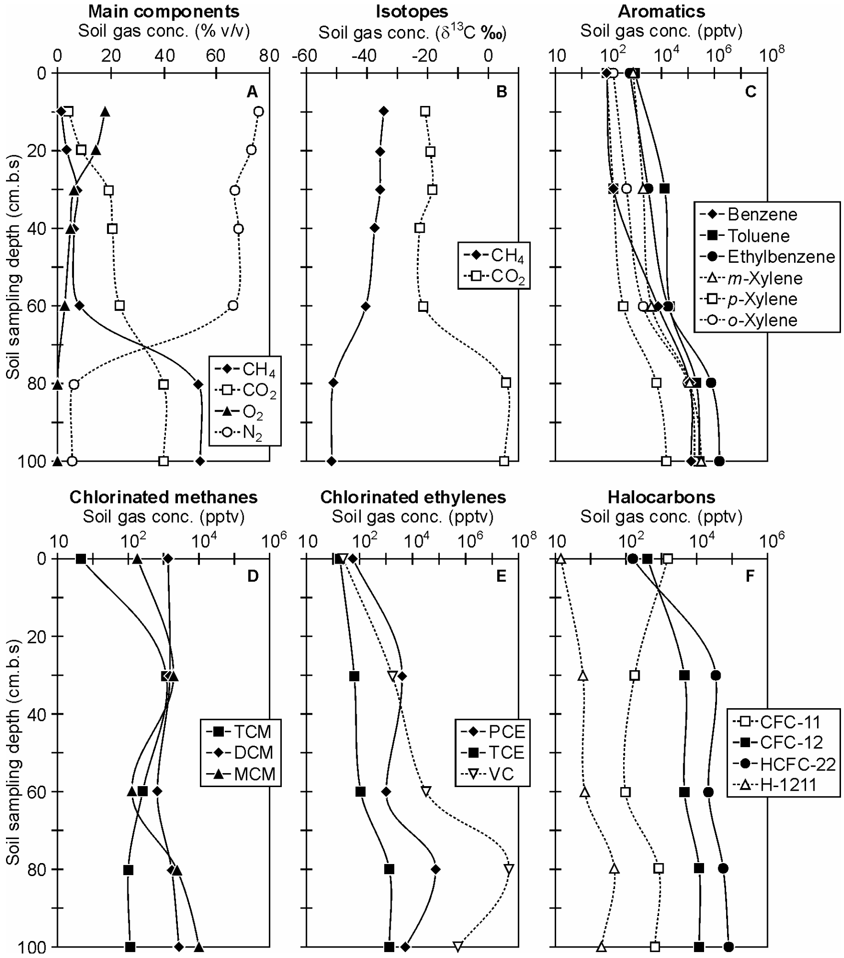

Figure 1. Soil gas concentration profiles at LP6 (final cover area, Lapouyade).

Based on the emission measurements discussed above and complementary laboratory experiments (Scheutz et al., 2003a,b), a general coherence was observed between emissions and biodegradability of various NMOCs. The emissions consisted mainly of compounds which are not degradable or slowly degraded under aerobic conditions (e.g., CFCs and higher chlorinated compounds), while low to negative emissions were observed for compounds more readily degradable under aerobic conditions: especially the aromatics and lower chlorinated compounds.

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