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Effect of subsurface soil moisture variability, and atmospheric conditions on methane gas migration in shallow subsurface

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Gas transport Shallow subsurface

A major concern resulting from the increased use and production of natural gas has been how to mitigate fugitive greenhouse gas emissions (predominantly methane) from natural gas infrastructure (e.g., leaky shallow pipelines). Subsurface migration and atmospheric loading of methane from pipeline leaks is controlled by source configurations and subsurface soil conditions (e.g., soil heterogeneity and soil moisture) and are further affected by atmospheric conditions (e.g., wind and temperature). How-ever, the transport and attenuation of methane under varying subsurface and atmospheric conditions are poorly understood, making it difficult to estimate leakage fluxes from methane concentration measurements at and above the soil surface. Based on a series of controlled bench-scale experiments using a large porous media tank interfaced with an open-return wind tunnel, this study investigated multiphase processes controlling migration of methane from a point source representing a buried pipeline leaking at fixed flow rate (kg/s) under various saturation and soil-texture conditions. In addition, potential effects of atmospheric boundary controls, wind (0.5 and 2.0 m s⁻¹) and temperature (22 and 35 °C), were also examined. Results showed the distinct effects of soil heterogeneity and, to a varying degree, of soil moisture on surface methane concentrations. In addition, results also showed the pronounced effects of wind and, to a lesser degree, of temperature on surface methane concentrations in the presence of varying soil and moisture conditions. The observed subsurface methane profiles were simulated using the multiphase transport simulator TOUGH2-EOS7CA. Observed agreement between measured and simulated data demonstrates that for the conditions studied, multiphase migration of a multicomponent gas mixture (including methane) under density-dependent flow can be adequately represented with a Fickian advection-diffusion (or dispersion) model (ADM) framework. The dominant effect of saturation over the soil texture, could also be inferred from numerical characterization.

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1. Introduction

Despite continuing efforts to reduce greenhouse gas loading to the atmosphere to mitigate climate change, anthropogenic emissions of greenhouse gases has accelerated during the last decade (2000–2010) compared to the preceding decade (Intergovernmental Panel for Climate Change; IPCC, 2014). The energy supply sector (inclusive of all energy extraction, conversion, storage, and transmission to final-users), the largest contributor to global greenhouse gas emissions, was responsible for 35% of the total anthropogenic emissions in 2010 (IPCC, 2014). Consequently, the energy sector has recently received renewed attention with the perspective of reducing future emissions by adopting low-carbon technologies in energy production. Fossil fuel-switching, in particular, is considered a promising strategy to reduce greenhouse gas emissions from the electricity-generation sector. Natural gas, in this regard, plays a role as a “transition fuel” when shifting from the high-carbon fossil fuels (coal and oil) to more environmentally-friendly renew-able energy substitutes in the future (Levi, 2013). Notably, natural gas has a considerably smaller greenhouse gas footprint compared to that of coal in terms of greenhouse gas emissions per unit of energy produced in fossil fuel chains; natural gas is less intensive (290–930 gCO₂eq/kWh) than oil (510–1170 gCO₂eq/kWh) and coal

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Natural flexible pipelines under differing near-surface atmospheric conditions paid to the controlling mechanisms of fate and transport of natural gas/methane in the shallow subsurface around the recommendations in the 2014 IPCC report. Estimates are particularly incomplete and involve a wide range of uncertainties including invalidated emission factors (Karion et al., 2013; Lamb et al., 2015), undercounted “super-emitters” (Brandt et al., 2014), underrepresented high-emission production technologies (e.g., liquid unloading: API/ANGA, 2012), and unsolved issues in emission estimation methods (e.g., Pétron et al., 2012; Levi, 2012), etc. Several pipeline emission surveys conducted along major U.S. urban city roads also revealed surprisingly high CH₄ leaks (Jackson et al., 2013; Phillips et al., 2013) which were generally attributed to the aging underground gas infrastructure (USEPA, 2014) where more recently, Lamb et al. (2015) reported decreasing methane emissions. Importantly, all estimates, predictions, and recommendations in the USEPA (2014) study were made based on above-ground concentration measurements, with very limited attention paid to the controlling mechanisms of fate and transport of natural gas/methane in the shallow subsurface around the leaking pipelines under differing near-surface atmospheric conditions.

Natural gas gathering, transmission and distribution pipelines range in material composition from high strength steel or copper to flexible plastic and can range in size from 40 to 6 in. in diameter, depending on the location and use (Folga, 2007: Transportation of Natural Gas, 2016). Although new steel and plastic pipelines are less prone to leaks than their older counterparts, aged pipelines, made

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**Nomenclature**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>D_K</td>
<td>Molecular diffusivity of component K in phase (m²·s⁻¹)</td>
</tr>
<tr>
<td>g</td>
<td>Gravitational acceleration vector (m·s⁻²)</td>
</tr>
<tr>
<td>k</td>
<td>Intrinsic permeability (m²)</td>
</tr>
<tr>
<td>kr</td>
<td>Relative permeability (−)</td>
</tr>
<tr>
<td>P</td>
<td>Total pressure (Pa)</td>
</tr>
<tr>
<td>q_1</td>
<td>Mass flux (kg·m⁻²·s⁻¹)</td>
</tr>
<tr>
<td>T</td>
<td>Temperature (°C)</td>
</tr>
<tr>
<td>V</td>
<td>Wind velocity (m·s⁻¹)</td>
</tr>
<tr>
<td>x</td>
<td>Mass fraction with phase subscript and component</td>
</tr>
<tr>
<td>Y</td>
<td>Surface area (m⁻²)</td>
</tr>
<tr>
<td>Z</td>
<td>Volume (m⁻³)</td>
</tr>
</tbody>
</table>

Greek symbols

- \( \alpha \): 1/Po in van Genuchten’s capillary pressure function (Pa⁻¹)
- \( \phi \): Porosity (m³·m⁻³)
- \( \mu \): Dynamic viscosity (kg·m⁻¹·s⁻¹)
- \( \rho \): Density (kg·m⁻³)
- \( \tau \): Capillary pressure (Pa)
- \( \varepsilon \): Tortuosity (−)

Subscripts and superscripts

- \( \text{g} \): gas components (superscript)
- \( \text{l} \): liquid
- \( \text{w} \): water
- \( \text{s} \): saturation
- \( \text{max} \): maximum
- \( \text{res} \): residual
- \( \text{r} \): reference value

---

(675–1689 gCO₂eq/kWh) (IPCC, 2011) and therefore is a promising energy surrogate for the coming decades (Cathles et al., 2012).

However, fugitive atmospheric emissions from leaky natural gas infrastructure may largely offset intended environmental benefits of natural gas usage since methane (CH₄), the predominant component of natural gas, has a global warming potential 86 times greater than CO₂ on a 20-year basis and 25 times greater over a 100-year time horizon (Jackson et al., 2013). (Note that the greenhouse gas potency is sensitive to the time frame of interest because CH₄ is converted to CO₂ in decadal scales.) Recent studies revealed that coal-to-gas switching would bring meaningful climate forcing benefits (in both short-term and long-term scenarios) provided fugitive CH₄ emissions are maintained below about 3% of gas production (Lelieveld et al., 2005). Nevertheless, the latest statistics on atmospheric methane leaks in gas supply chains are not conclusive and vary between 5%–1% (e.g., Howarth et al., 2011; Cathles et al., 2012). The present inventory-based leakage estimates are particularly incomplete and involve a wide range of uncertainties including invalidated emission factors (Karion et al., 2013; Lamb et al., 2015), undercounted “super-emitters” (Brandt et al., 2014), underrepresented high-emission production technologies (e.g., liquid unloading: API/ANGA, 2012), and unsolved issues in emission estimation methods (e.g., Pétron et al., 2012; Levi, 2012), etc. Several pipeline emission surveys conducted along major U.S. urban city roads also revealed surprisingly high CH₄ leaks (Jackson et al., 2013; Phillips et al., 2013) which were generally attributed to the aging underground gas infrastructure (USEPA, 2014) where more recently, Lamb et al. (2015) reported decreasing methane emissions. Importantly, all estimates, predictions, and recommendations in the USEPA (2014) study were made based on above-ground concentration measurements, with very limited attention paid to the controlling mechanisms of fate and transport of natural gas/methane in the shallow subsurface around the leaking pipelines under differing near-surface atmospheric conditions.

Natural gas gathering, transmission and distribution pipelines range in material composition from high strength steel or copper to flexible plastic and can range in size from 40 to 6 in. in diameter, depending on the location and use (Folga, 2007: Transportation of Natural Gas, 2016). Although new steel and plastic pipelines are less prone to leaks than their older counterparts, aged pipelines, made
of cast iron or unprotected steel, often leak due to earth movement, breakdown of joints and corrosion of unprotected steel, and graphitization (i.e., natural degrading to softer elements over time) of iron pipelines. Pipelines are usually placed in a trench 1.5–4 feet below ground. In the U.S., the depth is specified according to federal regulations (Transportation of Natural Gas, 2016) and depends on the pipe diameter, soil or rock type, terrain characteristics, etc. Upon laying the pipe, the trench is backfilled with excavated material. As backfilling often involves mechanical compaction (causing a low-permeability zone), additional materials, sometimes of different soil types, are used to achieve a level surface. If the excavation was done in rocky formations, a layer of broken rocks may be placed above the pipeline thus forming a high-permeability layer. In brief, differently-characterized layers are often found within the backfill zone above a leaky pipeline, which may markedly affect the subsurface methane migration.

Transmission mains typically carry natural gas at high-pressure conditions (3500–9600 kPa) whereas low pressure conditions (1.5–2000 kPa) prevail in distribution systems. Consequently, in the event of a leak, the mechanism of gas emission into the surrounding porous medium, as well as the domain of influence of the leak, will vary depending upon the pressure conditions of the pipelines. In practice, high-pressure (advection-controlled) leaks are relatively easy to detect and fix up, while detecting small and diffusion-controlled leaks, as is the case presented in this study, is challenging.

The methane migration length scales from a leaky distribution pipeline may vary broadly in the range of 2–10 m (Okamoto and Gomi, 2011, Yan et al., 2015, and Xie et al., 2015) depending upon the burial depth, soil properties and moisture status (discussed below) as well as on the gas composition. With regard to gas composition, natural gas is composed almost entirely of methane (>94%) in transmission and distribution systems, however less methane content may present in production (79%) and processing (87%) stages (USEPA, 2003).

Fate and transport of methane in soil are primarily controlled by subsurface conditions such as heterogeneity, soil moisture, temperature, and pressure gradients (Poulsen et al., 2003). For example, gas migration in a texturally heterogeneous soil system (e.g., in the presence of a low-permeability clay lens embedded in a sandy formation) will be markedly different from that of a homogenous soil system due to the different texture- (or porosity-) induced tortuosity effects (Ho and Webb, 2006). Similarly, soil moisture
controls methane migration in variably-saturated soils because of attenuation due to methane solubility, oxidation, and water-induced tortuosity (Smith et al., 2003) and simultaneous water vapor transport in the soil gas phase. Furthermore, temperature affects molecular diffusion and thereby the diffusive flow of gases while the presence of pressure gradients leads to advective gas flow. Methane flow regimes are typically dominated by advective flow near the leak (due to high pipeline pressure) and become diffusion-controlled as gas migrates away from the point of leak-age. Moreover, the wind- or temperature-induced near-surface fluctuations may also affect the subsurface migration and atmospheric emission of methane (Poulsen et al., 2003). However, to our knowledge, only a limited number of combined experimental and numerical studies investigating subsurface methane migration under different subsurface conditions and atmospheric controls have been reported in literature.

Previous laboratory-based studies investigating methane (or other natural gas-related gases; e.g., CO₂, propane) migration in soil/porous media mainly involved one-dimensional column experiments (Hibi et al., 2009, 2012; Karl-Heinz et al., 2004). In most studies, a single- or multi-component gas (or a gas mixture) was applied through a uniformly packed soil column under a constant-flow rate (or a pressure gradient) and gas concentrations were measured at selected ports along the column. A few field-scale two- or three-dimensional natural gas transport studies have also been performed to investigate gas (CH₄ or CO₂) concentration profiles across differently-characterized soil profiles (e.g., Okamoto and Gomi, 2011; Yan et al., 2015), however, literature on the effect of the degree of saturation on methane concentration profiles could not be found.

Experimental gas transport studies in coupled domains of porous media and free flow are particularly rare in the literature. Based on a recent two-dimensional study in a sand-packed tank interfaced with a free flow (atmospheric) domain, Basirat et al. (2015) investigated CO₂ migration from a buried point source in both porous media and free flow domains, but did not consider the effects of any atmospheric controls on surface-subsurface gas transport processes. The experimental results, in general, commonly identified the effect of gas density on gas flow characteristics; high-density gases (e.g., CO₂, propane) were found to migrate downward and accumulate in the zone below the gas source while low-density gases (e.g., CH₄, H₂) preferentially moved upward by buoyancy. Further, laboratory-based experimental studies on heterogeneous and variably saturated porous media distinguished differences in gas-transport behavior of different zones e.g., the preferential flow regions or low-permeability zones (Volckaert et al., 1995; Delahaye and Alonso, 2002; Okamoto et al., 2014) in relation to advective and diffusive gas migration.

Most numerical approaches presented in the literature on modeling the migration of multicomponent gas mixtures in porous media are primarily founded on the classical advection-diffusion (or dispersion) model (ADM), which combines Darcy’s law (advection) and Fick’s law (ordinary diffusion) (Abriola and Pinder, 1985a,b; Slough et al., 1999; Okamoto and Gomi, 2011). The models varied widely depending on whether they consider or ignore the effects of gas density, non-isothermal flow properties, real-gas mixture properties (e.g., temperature dependence on gas density, viscosity, and solubility), compressibility etc. A wide range of studies using the concept of the Dusty Gas Model (DGM) (Cunningham and Williams, 1980; Mason and Malinauskas, 1983) for simulating multicomponent gas flow can also be found in the literature (e.g., Webb, 1998; Hibi et al., 2009). The applicability of the DGM concept was found to be essentially important in low-permeability formations (e.g., graphite k ~ 10⁻¹⁸ m²; Webb, 1996) where molecular-wall interactions and thermal Knudsen diffusion play an important role in diffusive mass transport. However, for high-permeability porous media (k > 10⁻¹⁵ m²), as is the case in the present study, the results from AD and DG models were observed to be comparable (Oldenburg et al., 2004; Hibi et al., 2009), and a good agreement was often observed between AD-modelled and measured data (e.g., Okamoto and Gomi, 2011). Nevertheless, the applicability of the Fickian diffusion approach for multicomponent gas transport modeling needs to be further verified with experimental observations. In addition, limited numerical studies were found for simulating CH₄ (or CO₂) migration in coupled porous media and atmospheric domains (e.g., Oldenburg and Unger, 2004; Basirat et al., 2015) which also motivates validation with experimental observations.

Based on a controlled bench-scale experimental and numerical study, we investigated methane (mixed with air and water vapor) migration under carefully controlled subsurface and atmospheric conditions. The key hypotheses of this study are: (i) the subsurface migration of multicomponent gas mixtures and the development of the atmospheric concentration boundary layer are essentially controlled by subsurface conditions (e.g., soil moisture, heterogeneity) and atmospheric controls (near-surface wind and temperature); (ii) the degree to which the controlling parameters (described in hypothesis (i)) affect the subsurface methane flow and transport can be experimentally evaluated in a coupled two-dimensional porous media-wind tunnel test facility at a bench-scale; and (iii) experimentally identified effects (described in hypothesis (ii)) can be numerically simulated by modeling density-dependent flow of a multicomponent real-gas mixture (CH₄, air and water vapor) by the Fickian ADM under constrained atmosphere boundary conditions. We emphasize that the results are based on the length scales of the experimental and numerical domains which may not truly mimic more realistic soil-atmosphere continuum scales at specific pipeline-soil systems.

2. Materials and methods

2.1. Experimental set-up and porous media

A series of controlled bench-scale laboratory experiments was conducted using a low-velocity open-return wind tunnel interfaced with a 2D soil tank of dimensions 35 cm 55 cm 10 cm (height length width) as illustrated schematically in Fig. 1.

The ductwork forming the wind tunnel was connected to the tank to channel heated air flow across the test section. To control the wind velocity, the wind tunnel was equipped with an in-line ducting control and a variable velocity controller, and a galvanized 15-cm circular mirror (Davaranzi et al., 2014), The wind velocity was continuously measured by means of a pre-calibrated pitot static tube (Dwyer Instruments, Inc. Model 167-12, 0.32 cm diameter, 30.48 cm insertion length, accuracy 65%) connected to a differential pressure transmitter (Omega Engineering, Inc. Model PX653-0.1D5V, Range 50–0.1 in. of H₂O, 0–25 Pa). For high-temperature (35–40°C) measurements, a constant heat pulse was applied by means of an infrared heater located upstream (80 cm from the test section) of the wind tunnel. The heater assembly consisting of five infrared heater units (Mor Electric Heating Assoc., Inc. Infrared Salamander Model FTE 500-240) was placed in parallel within a reflector. The heater and the fan were operated a minimum of 3 h ahead of the experiment to allow sufficient time to reach steady state in terms of air and heat flow by the onset of the experiment. Temperature/relative humidity sensors (Decagon Devices Inc.
EHT RH/Temperature, accuracy 62% from 5 to 90% RH, 63% from 90 to 100% RH, temperature accuracy 0.25 °C) were placed immediate upstream and downstream of the test section at two levels above the surface: 0 cm (i.e., at the surface) and 17 cm. Methane
concentrations within the wind tunnel test section were measured using an atmospheric fast flame ionization detector (fast FID) system (HFR 400, Cambustion Ltd, UK, relative sensitivity to methane, 0.97). The sampling head assembly (2 Nos.) was mounted on a laterally movable frame to facilitate length-wise measurements (i.e., along the soil surface). Provisions were also made for vertical movements to measure methane concentrations from 0 to 18 cm above the soil surface. A 20-cm long capillary tube was attached to each sampling head to extract methane gas (50 cm³/min) at a pre-set pressure gradient of 150 mm Hg (response time = 1–2 ms). The operating combustion temperature at steady state was maintained at 250–300 °C (per manufacturer’s recommendation) and remained constant throughout the experiments. Prior to each experiment, a calibration test was conducted (at the above temperature) using five standard methane gas concentrations (in ppm): 10, 100, 1000, 10000, and 50000. A strong linear relation (r² > 0.98) between the concentration (ppm) and the response signal (mV) was observed in all cases. In addition, background methane concentrations were monitored and considered in the measurements/calculations (on average 2 ppm). All above-surface measurements including wind velocity, temperature, relative humidity, and methane concentrations were monitored continuously, real-time, using LabVIEW software (National Instruments Corp.).

An integrated sensor network embedded into the soil tank was used to measure soil temperature (15 Nos, Decagon Devices Inc. RT-1, accuracy ±0.5 °C from 5 to 40 °C) and moisture (24 Nos, Decagon Devices, Inc. ECH2O EC-5, measurement frequency 570 MHz, accuracy 63%). The soil moisture sensors were individually calibrated before measurements following Sakaki et al. (2008). A point gas source (50,000 ppm CH₄ and 950,000 ppm N₂, flow rate = 0.5 lpm) was placed 2 cm above the bottom of the tank (at the centerline) to mimic a diffusion-dominant leak from a shallow underground pipe. Note that the selected flow rate (0.5 lpm) is within the rage of field-estimated leakage rates (0.002–12.9 lpm) from distribution systems reported in the literature (e.g. GRI/EPA, 1996; GTI, 2013; EPA, 2014; Lamb et al., 2015). No noticeable temperature change was recorded in the vicinity of the gas source at the onset of injection and the ambient temperature conditions, as controlled by the surface temperature, prevailed near the source.

The tank was packed uniformly with a silica sand # 30/40 (identified by the effective sieve number) (Accusand, Unimin Corporation, Ottawa, MN) to represent a homogenous system. To represent a layered system, a 5-cm thick coarse-textured (#12/20) sand layer was sandwiched 15 cm below the surface. The layered test configuration was selected based on its simplicity (simple soil heterogeneity) and the locations of the soil moisture sensors and sampling ports (discussed below). Also, the location of the coarse-textured layer allows for the development of variable soil water saturation based on the soil’s hydraulic properties (discussed below).

Experiments were conducted for near-dry (i.e., at residual saturation) and partially saturated (i.e., wet-packed and drained to 35 cm H₂O with respect to the center of the tank) conditions. Basic thermo-physical properties of the two porous media are given in Table 1, which also presents van Genuchten parameters used for numerical simulations. The high chemical purity and the presence of only a trace amount of organic carbon (0.03%) is a notable feature of both sand types, which limited the potential for methane oxidation due to microbiological activities during the experiments. For further details on the two sand types, measurement methods and packing conditions, see Smits et al. (2012).

To confirm that steady-state conditions were achieved prior to the start of the experiments, surface methane concentrations were continuously monitored at two selected locations on the soil surface until the time-invariant concentrations were recorded for 1–2 h using the Fast FID system. At steady state, gas samples (1 ml) were extracted (in triplicates) from selected ports at depths (measured from the surface) of 0.5, 7.5, 17.5, 30 cm using a high-precision analytical syringe (VICI Precision Sampling, Inc., LA) and methane concentrations were measured (with no time lag) using gas chromatography (Agilent Technology, 6890). The sampling ports are located vertically along the centerline of the tank and at the rear side of the tank (not shown in Fig. 1).

Table 2: 2 soil states (homogenous and layered) × 3 wind velocities (0, 0.5, 2.0 ms⁻¹) × 2 atmospheric temperatures (22 °C and 35 °C) were used to test the control of subsurface conditions (e.g. soil moisture, heterogeneity) and atmospheric controls (wind and temperature) on

Fig. 1. Schematic of the experimental set-up (not to scale). The arrows show the wind direction. The symbols (  &  ) denote sensor locations for soil moisture (in the front side) and temperature (in the back side), respectively.
The subsurface migration of methane gas. Three experiments were duplicated (not shown in Table 2) to confirm the repeatability of the measurements. In the following text, we follow the notations in Table 2 (presenting saturation-soil state-temperature combinations) to describe results relevant to the different scenarios.

All the experiments were conducted within a barometrically controlled building under invariant barometric pressure conditions.

Table 1
Physical properties and transport parameters of the two porous media.

<table>
<thead>
<tr>
<th>Porous media # (D50)</th>
<th>Particle diameter (μm)</th>
<th>Particle density (g cm⁻³)</th>
<th>Total porosity (&lt;I&gt;)</th>
<th>Bulk density (g cm⁻³)</th>
<th>Saturated hydraulic conductivity (Ksat)</th>
<th>Permeability (cm⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12/20</td>
<td>0.53</td>
<td>2.665</td>
<td>0.33</td>
<td>1.82</td>
<td>0.376</td>
<td>0.056</td>
</tr>
<tr>
<td>30/40</td>
<td>0.31</td>
<td>2.665</td>
<td>0.33</td>
<td>1.77</td>
<td>0.106</td>
<td>0.017</td>
</tr>
</tbody>
</table>

Table 2
Experimental schedule and the notations used to describe different experimental conditions.

<table>
<thead>
<tr>
<th>Controls</th>
<th>Low Temp. (20–24 °C)</th>
<th>High Temp. (35–38 °C)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>u = 0 ms⁻¹</td>
<td>u = 0.5 ms⁻¹</td>
</tr>
<tr>
<td>Near dry Ground</td>
<td>Homogeneous</td>
<td>Layered</td>
</tr>
<tr>
<td>Partially-sat</td>
<td>Homogeneous</td>
<td>Layered</td>
</tr>
</tbody>
</table>
| ND, Near-dry; PS, Partially-saturated; HG, Homogenous; LY, Layered; LT, Low temperature; HT, High temperature

The experimental set up was located within the laboratory in such a way that the induced pressure gradients (e.g., due to opening/closing of doors) minimally influence the experimental results. The passersby were restricted within the experimental area.

Component flux: advective and diffusive

\[ \text{adv} = \chi^a \mathbf{F} \]

such that during the experiments to avoid likely pressure fluctuations.

Relative permeability (van Genuchten, 1980)

\[ \frac{\text{diff}}{\text{adv}} = \sum_{i=1}^{n} \frac{1}{T_D} = \frac{f}{\sqrt{T}} \]

if \( S < k_c = S^* 1 - \left\{ S^* \right\}^{\lambda} \)
2.2. Numerical model

Numerical simulations were performed using the multiphase transport simulator TOUGH2 \citep{Pruess1999} combined with the equation of state module EOS7CA \citep{Oldenburg2015}. TOUGH2/EOS7CA simulates the subsurface flow and transport of aqueous and gas phases containing five components (i.e., H$_2$O, brine, non-condensable gas (e.g., CH$_4$), gas tracer, and air) under capillary pressure \citep{vanGenuchten1980} and Henry’s law if $S_l \geq S_{lr} k_r = 1$ if $S_{gr} = 0 k_r = 1 - k_r$

\[
S^* = (S_l - S_{lr}) / (S_{lr} - S_c)
\]

subject to $P_{max} \leq P_c \leq 0$

\[
P_K = KH_{Hm}
\]

isothermal or non-isothermal conditions. The model approximates air as a mixture of 79\% N$_2$ and 21\% O$_2$ by volume. TOUGH2/EOS7CA uses thermodynamic properties in real gas mixtures (i.e., density, enthalpy departure and viscosity) calculated using the Peng–Robinson equation of state model \citep{Peng1976}. A temperature-dependent Fickian molecular diffusion coefficient is used to model gas diffusion. Henry’s law models dissolution of gaseous methane in the aqueous phase, with Henry’s coefficients estimated using the method presented by \citet{Cramer1982}. The Henry’s law approach in the model is accurate for low pressure (less than approx. 1 MPa) conditions and hence the model is applicable to relatively shallow subsurface porous media systems \citep{Oldenburg2015} as considered in this study. The reader is referred to \citet{Pruess1999} and \citet{Oldenburg2015} for the full model description; however, the governing equations relevant to the present study are given in Table 3.

The simulations were done using a 2-D Cartesian computational domain discretized into 63 “porous” elements representing the porous (subsurface) system with an additional “atmospheric” element (characterized with unit total porosity, infinitely large volume, and high permeability) coupled with the surface porous elements to represent the wind tunnel region (atmosphere). To simulate gas flow in partially-saturated conditions, a pre-simulation (without gas flow) was first performed using measured saturation data from different depths. The pre-simulation allowed for the model to reach saturation-capillary pressure equilibrium at the steady state. This was followed by a complete steady-state simulation using the pre-simulation data together with gas flow. Due
have been used in literature to characterize velocity-induced atmospheric mass transfer behaviors (e.g., Veranth et al., 2003).

\[ C(x) = C_{so} \]

\[
\begin{align*}
P &= P_{atm} \\
T &= T_{surf,exp} \\
C &= C_{surf,exp} \\
P_{g,0} &= P_{atm} \\
S_{l,0} &= S_{exp,0} \\
T_{0} &= T_{exp,0} \\
q_l &= q_g = q_h = 0 \\
q_l &= q_g = q_h = 0
\end{align*}
\]

Fig. 2. Initial and boundary conditions used for the porous medium (subsurface) computational domain in TOUGH2/EOS7CA simulations. \( q \) denotes the liquid flux, \( q_g \) the gas flux, \( q_h \) the heat flux, respectively. The subscript \( \text{exp} \) denotes the experimental data. The dotted lines show the free flow domain. The source/inflow of CH4 and N2 gas mixture (CH4: 50,000 ppm, 2.9425E–07 kg/s; N2: 950,000 ppm, 1.0200E–05 kg/s) is also shown.

to the high purity of the two sands (Schroth et al., 1996), the biological attenuation of methane could be reasonably neglected. The adopted initial and boundary conditions are shown in Fig. 2.

3. Results and discussion

3.1. Experimental results

3.1.1. Surface methane concentrations

The surface \( \text{CH}_4 \) concentrations measured (0.5–1.0 mm above the surface) along the soil surface using the Fast FID method are shown in Fig. 3 for selected control conditions. Wind, in particular, showed a strong effect on surface \( \text{CH}_4 \) concentration variations as compared to the effect of temperature. In all cases, the no-wind condition \( (i.e., u = 0 \text{ m s}^{-1}) \) showed the highest surface \( \text{CH}_4 \) concentrations which decreased with increasing wind velocity. Notably, the observed surface \( \text{CH}_4 \) concentration profiles for the no-wind condition were mostly irregular without a mathematically expressible pattern, except for a few profiles (e–h) showing near-constant concentrations along the surface. The observed irregular behavior could be attributed to the mixing and circulation of the differently dense air and methane within the test section. Nevertheless, the corresponding profiles for higher velocities \( (u = 0.5 \text{ and } 2.0 \text{ m s}^{-1}) \) could be adequately described with a simple nonlinear parametric power function (Eq. (1)). Similar empirical power-law functions where \( L \) (cm) is the length of the tank, \( C_{so} \) (ppm) is the maximum surface concentration (at the downstream edge), and \( n \) (−) is the shape factor representing the shape of the surface \( \text{CH}_4 \) concentration buildup at different wind conditions. The fitted models are also illustrated in Fig. 3 in corresponding lines. Generally, for the wind velocities of 0.5 and 2.0 m s\(^{-1}\), \( C_{so} \) ranged between 2530 and 5150 ppm and 1190–2190 ppm, respectively, implying the wind-induced erosion of the surface methane concentration boundary layer. The \( n \) value, on the other hand, varied in the ranges of 0.2–0.3 and 0.5–0.65, respectively. Note that \( n = 0 \) in Eq. (1) mathematically represents the no-wind condition with a constant concentration \( (C-C_{so}) \) along the surface as also observed with some no-wind concentration profiles (Fig. 3e–h). Further observations, however, are required to derive a statistically meaningful relationship between \( n \) and wind velocity \( (u) \).
The temperature generally increased the surface methane concentration but to varying degrees, as shown in Fig. 3. This is likely due to the changes in the thermodynamic properties of methane at high temperature, causing decreased density and increased diffusivity and thermal conductivity, resulting in increased temperature-induced diffusive methane fluxes to the atmosphere. High temperature effects were expected at high-wind conditions due to the heated-up air passing over the soil surface, enhancing diffusive emissions. Indeed, at partially-saturated conditions, the heated air flow heats up the soil surface over time and tends to dry out the near-surface soils. This increases air-filled pore spaces and facilitates methane emissions. The temperature effects were more evident in no-wind conditions, although the results were somewhat obscured by the marked fluctuation in observed data (note that the error bars show one standard deviation from the mean value). Despite the expected higher temperature effects at high-wind conditions, the wind effects were dominant over the temperature effects in the methane concentration profiles. Note that the temperature-induced effects, to some extent, were evident also at \( u = 0.5 \text{ ms}^{-1} \), however were largely masked at \( u = 2.0 \text{ ms}^{-1} \) by the pronounced wind effects.

3.1.2. Depth-wise variation of temperature and water saturation

The depth-wise temperature variations are shown in Fig. 4 for all the subsurface and atmospheric conditions. In addition to experimental observation, the temperature profiles are needed to estimate the heat loss (which was very small) out of the plexiglass walls of the tank in the model. Note that the distinctly low temperatures in Fig. 4(a) for low-temperature measurements were due to the low ambient air temperature at the time of the experiments. As expected, the temperature at the surface and in the topsoil (0 to \(-10\) cm) was highest for high-air temperature, high-velocity conditions due to heated air movement along the soil surface. The temperature below 15 cm depth, however, remained mostly unaffected by above-surface temperature variations, though a slight subsurface ‘cooling effect’ could be seen at high surface temperature conditions particularly for partially-saturated soils due to the latent heat transfer for soil-water vaporization. Importantly, no effect due to the presence of the coarse-textured layer could be seen on temperature profiles presumably due to the similar thermal conductivity values for fine-textured (#30/40) and coarse-textured (# 12/20) media for both dry and partially-saturated conditions (see Table 1).

Fig. 5 shows the depth-wise variation of water saturation under partially-saturated conditions for (a) homogenous and (b) layered soil systems. In contrast to \( \tau_w \), monotonically increasing saturation
Fig. 3. Measured surface methane concentration (in ppm) as a function of the distance (in cm) along the surface (measured from the downstream edge of the test section) for 24 experimental conditions. Measurements for three wind velocities (0, 0.5, 2.0 ms\(^{-1}\)) are shown in corresponding colors in each sub-figure. The arrow (in 3a) shows the wind direction. The solid lines (in corresponding colors) for 0.2 ms\(^{-1}\) and 0.5 ms\(^{-1}\) show the parameterized model (Eq. (1)) descriptions while the lines for 0 ms\(^{-1}\) show point-to-point linear interpolation. Error bars denote one standard deviation (±) from the average concentration (n ≥ 20).

Fig. 4. Depth-wise variation of soil temperature (°C) for the 24 experimental conditions. Error bars denote one standard deviation (±) from the mean value (n ≥ 20). The coarse-textured layer in the layered systems (4b and c) is demarcated by the two horizontal lines.

With depth in homogenous systems, the layered systems showed a distinct high-saturation (20%) region within the capillary barrier (approx. 2.5-cm above the textural interface; shown by the dotted line in Fig. 4b) and was underlain by the course-textured layer with relatively low saturation under steady state conditions. The capillary barrier effect is due to capillary tension that limits the water’s downward movement from a finer soil into a coarser soil. A coarser soil layer underlying a finer soil layer causes temporary water storage in the upper (finer) soil layer that can ultimately be removed by evaporation, lateral drainage or percolation into the lower layer (Stormont and Anderson, 1999). In fact, the steady state condition with respect to saturation could not be ideally maintained under partially-saturated conditions as the surface evaporation, moisture movement and redistribution continuously evolved (at...
a decreasing rate) with time throughout the experiment. Clearly, the high-temperature, high-wind conditions at the surface expedited the subsurface moisture dynamics (Fig. 5). Nonetheless, for a given experiment, the subsurface moisture distribution (and hence presumably the moisture induced effects on methane migration) remained largely unchanged with respect to the selected surface-subsurface controls.

3.1.3. Depth-wise variation of methane concentration

Methane concentration profiles in the soils are shown in Fig. 6 for all 24 scenarios studied. Note that the three profiles shown in each subfigure (a through h) correspond to the three wind velocities (0, 0.5, 2.0 m s\(^{-1}\)). Evidently, subsurface methane concentration profiles for both the homogeneous and layered systems depicted no distinct effects of wind (when other parameters remained unchanged), suggesting that wind has made no noticeable control on subsurface migration of methane. Importantly, even for the measurements immediately below the surface (i.e., at 0 to 0.5 cm depth) where some wind-induced effects could be expected within a penetrative boundary layer, methane concentrations for different wind speeds were mostly within the range of intrinsic uncertainty (within one SD from the mean as shown by error bars) indicating no clear (or macroscopically distinguishable) effects of wind on subsurface methane migration. A similar observation was also made by Kimball and Lemon (1971) whose results for mixing heptane in fine textured-media under induced surface wind conditions were also affected by the large scatter in measured data. The results corroborate the observations from past studies, for example, that of Ishihara et al. (1992) who noted that for fine-textured media of 0.2–0.5 mm size (which resembles the grain size of #30/40), the mixing depth does not extend beyond a magnitude of 1–10 times grain diameter. They further noted that a considerable mixing depth could be observed for high permeable coarse-textured media with a diameter > 10 mm. In fact, both air permeability (\(k_a\), \(\mu\)m\(^2\)) and pore size of the medium may control the permissible air penetration depth under a given surface wind condition. Based on a field study, Maier et al. (2012) reported that the ratio of air permeability to air content (\(k_a/e\)) should exceed 1000 \(\mu\)m\(^2\) in order for the turbulence-induced near-surface gas transport (referred to as pressure/barometric pumping) to be statistically significant. For this study, the maximum \(k_a/e\) ratio (for dry #30/40 sand) does not exceed approx. 350 \(\mu\)m\(^2\) and hence confirms that wind-induced barometric pumping may not play a significant role in the soil-atmosphere gas exchange process (note that barometric pressure differential within and outside the laboratory was found to be negligible). Thus, the overall results suggest that for fine-textured media,

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**Fig. 5.** Depth-wise variation of saturation for the partially-saturated (a) homogenous and (b) layered systems. Error bars denote one standard deviation \(\pm\) from the mean value \((n = 20)\). The coarse-textured layer in the layered systems (5b) is demarcated by the two horizontal solid lines. The horizontal dotted line in 5(b) denotes approximately the upper limit of the observed capillary barrier above the coarse layer.

**Fig. 6.** Observed depth-wise methane concentration profiles for the 24 experimental conditions. Measurements for three wind velocities (0, 0.5, 2.0 m s\(^{-1}\)) are shown in corresponding colors in each sub-figure. Error bars denote one standard deviation \(\pm\) from the mean value \((n = 3)\).
(grain size < 0.5 mm) the gas exchange across the soil-atmosphere interface is predominantly diffusion-controlled which reaffirms the conclusions of Kimball and Lemon (1971).

3.1.4. Effect of textural heterogeneity

The effect of soil layering, was discernable in the CH₄ concen- tration profiles as seen in Fig. 7. For example, the variation of CH₄ concentration with depth for near-dry, layered systems was markedly lower with a steeper concentration gradient than those for near-dry, homogenous systems (Fig. 7a). Similar CH₄ concen- tration profiles in variable permeability soil systems have been observed in past experimental and numerical studies (e.g., Jiránek, 2010). We attribute the relatively low CH₄ concentration in the lay ered system to improved mixing and migration of CH₄ in the underlying fine-textured layer compared to the fine-textured layer. Notably, the average pore size in the coarse-textured (# 12/20) sand is almost twice as large as that in the fine-textured (# 30/40) sand and the former (# 12/20) shows four-fold permeability as compared to the latter (# 30/40) (see Table 1). This essentially implies the presence of large, well-connected and less tortuous pore space in the coarse-textured (# 12/20) sand in comparison to the fine- textured sand as also noted by Chamindu Deepagoda and Elberling, 2015. Consequently, rapid mixing and migration (both in lateral and vertical directions) of CH₄ within the coarse-textured layer is expected, causing decreased CH₄ concentrations within the layer. Since mixing and migration are higher for the low-density CH₄ compared to the high-density N₂, the gas mixture consisting of CH₄, N₂ and air in the pore space in the underlying fine-textured layer also becomes less CH₄-dominant, causing decreased CH₄ con- centrations in the bottom fine-textured layer as well. Nonetheless, near-surface methane concentrations for both homogenous and layered systems become comparable in order to have balanced inflow (at the point of leakage) and outflow (diffusive emissions off the surface) rates at the steady state (assuming advective flow is not likely a mechanism at the surface as discussed before). We note, however, that the data for the layered systems showed marked variation compared to those for the homogenous systems (shown by the error bars) due to the effects of localized mixing within the coarse layer. For high-temperature conditions (Fig. 7b), however, the effect of layering was not apparent and likely normalized due to the large variability in the measured data.

3.1.5. Effect of saturation

The degree of saturation is a key controller for gas migration in variably saturated porous media since both air permeability and gas diffusivity are strongly moisture-dependent parameters (Sallam et al., 1984; Springer et al., 1998). Previous laboratory-based methane transport studies highlighted the effect of saturation on steady-state methane migration (e.g., changes in the dispersion coefficient; Batterman et al., 1995). Revisiting Fig. 6, the effect of saturation is not immediately visible (within the measurement uncertainty) from the measured subsurface CH₄ concentration data for partially-saturated homogenous systems. A few measurements at partially-saturated conditions (e.g., low-temperature, high-wind conditions; Fig. 6e) showed lower CH₄ concentration at 30 cm depth, but this behavior was not consistent with other data. However, the effect of saturation was apparent in partially-saturated layered systems, especially for the bottom layers (e.g., ~10 to 30 cm) where high CH₄ concentrations were observed compared to dry conditions. This is due to the increased water-induced tortuosity for methane diffusion as will be discussed later. Notably, the presence of the high-saturation layer within the capillary barrier did not cause any appreciable change in the steady-state CH₄ concentration that was discernable from the measured data within the range of uncertainty. Literature presenting the effect of saturation on methane concentration profiles could not be found for direct comparison with our data, hence justifying the need for numerical simulations (below). In fact, the effect of saturation could be better described based on the simulated data as will be discussed later.

3.2. Numerical simulations

Fig. 8 shows the TOUGH2/EOS7CA-simulated methane concen- tration profiles for different subsurface conditions together with the measured data for no-wind conditions. The above-surface wind effects on subsurface methane migration were not accounted for in the numerical simulations since the TOUGH2/EOS7CA code is not formulated to simulate wind-induced surface-atmosphere inter- actions. However, we did account for the effects of temperature (and thereby also the changes in temperature-dependent proper ties of methane) on subsurface flow and transport of methane. For no-wind conditions, we observed a good agreement between mea sured and simulated data (note that the simulations involved no fitting or inversely-estimated parameters), showing the promis ing applicability of the model for simulating subsurface methane migration. The observed good agreement between the measured and simulated data also implies the successful verification of our hypothesis that multiphase migration of multicomponent gas mix tur e (including methane) under density-dependent flow can be adequately represented with a Fickian ADM framework, as also evi-
by Okamoto and Gomi (2011). It should, however, be noted that the effect of the presence of a temperature gradient on Fickian diffusion-based multicomponent gas migration is not tested herein since the temperature gradient within the soil is small under the
no-wind conditions (see Fig. 4) and the Okamoto and Gomi (2011) study was isothermal. We await further experimental data to test the model applicability for fully-coupled heat and multicomponent gas flow simulations with larger temperature gradients. We further note that no methane oxidation was accounted for within the ADM framework considering high chemical and biological purity of the porous media as discussed previously.

As shown in Fig. 9, the measured and simulated data for saturation agreed very well for both (a) homogenous and (b) layered soil systems. Note that in homogenous systems, the data under-estimated by the model (encircled by a solid line in Fig. 9a) correspond to the saturation values in the vicinity of the point of the pipe leakage, implying that gas flow caused water to displace from the soil near the point source pore spaces to more distant pores where the simulated saturation increased over the measured values (encircled by a dotted line in Fig. 9a). This is more clearly illustrated graphically in Fig. 9(b) together with the complete saturation profile across the depth. Corresponding profiles for a partially-saturated layered system are shown in Fig. 9(c) and (d), respectively while Fig. 9(d) illustrates the capillary barrier overly-ing the coarse-textured layer (demarcated by the horizontal white lines).

The model-derived steady-state contour maps showing the migration of the gaseous methane plume are shown for four selected scenarios in Fig. 10. Note that due to the low specific grav- ity, methane shows a tendency to preferentially move upward by buoyancy while lateral and downward movements are tortuosity led primarily by diffusion. As a result, methane contours appear as asymmetric and upward-bulging curves (see Fig. 10b–d). At close proximity to the point of leakage, both advective (pressure-driven) and diffusive (concentration-driven) transport occurs, however the flow becomes more and more diffusion-controlled as methane moves away from the source (Okamoto and Gomi, 2011). Since the diffusive methane fronts become nearly horizontal towards the surface and thus become weakly linked to the source, track-ing down the source/leak based on near-surface concentration measurements is challenging in many practical situations, and is further complicated by the different subsurface conditions. In the homogenous system under dry conditions (Fig. 10a), buoyancy-induced upward methane movement is readily supported by the lateral diffusion (due to a high diffusion coefficient at dry con-ditions; Table 1) and therefore the source concentration prevails in the domain at steady state. However, in the layered system under the same conditions (Fig. 10b), both the buoyancy-driven upward movement and the diffusion-driven lateral movement are more pronounced (due to the presence of the high permeability coarse-textured layer), causing the steady-state methane to sta- bilize with a distinct vertical concentration gradient across the system. In the presence of water, methane solubility affects the gaseous phase methane distribution while gaseous methane move- ment is further retarded by limited air-filled pore space (since part of the total pore space is occupied by water), simultaneous movement of water vapor in the gaseous phase, and increased (water-induced) gaseous phase tortuosity and discontinuity. Con- sequently, under partially-saturated conditions, (Fig. 10c and d), the diffusion-controlled lateral movement is more hindered than the buoyancy-induced upward movement of methane as demon- strated by the bulb-shaped distribution profiles. Note that the differences in the two partially-saturated systems (Fig. 10c and d) are not as great as the corresponding profiles for dry condi-

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Conditions</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Homogenous – Low temp.</td>
<td>CH₄ concentration (ppm)</td>
<td>0, 22°C, 0 ms⁻¹</td>
</tr>
<tr>
<td>Homogeneous-High Temp.</td>
<td></td>
<td>35°C, 0 ms⁻¹</td>
</tr>
<tr>
<td>Layered – Low temp.</td>
<td></td>
<td>TOUGH2/EOS7CA</td>
</tr>
<tr>
<td>Layered – High Temp.</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Fig. 9. Scatterplots showing measured and TOUGH2/EOS7CA-simulated saturation for the partially-saturated (a) uniform and (b) layered systems. Error bars denote one standard deviation (±) from the mean value (n = 3). The under-simulated data encircled in solid line correspond to the saturation in the neighborhood of the gas source and while those over-simulated by the model are encircled in dotted lines. The corresponding saturation contours for the two systems are shown graphically in fig. (c) and (d), respectively. The dotted horizontal white lines in (d) demarcate the coarse-textured layer.

Fig. 10. Concentration maps derived from and TOUGH2/EOS7CA simulations for four selected soil systems. The horizontal white lines (in b and d) demarcate the coarse-textured layer in layered systems.

Finally, we emphasize that the results of the present study are essentially applicable to the selected boundary conditions and length scales in experimental and numerical domains which may not be truly representative of more realistic and practically important soil-atmosphere continuum scales under field conditions. For example, the results may significantly vary under barometrically influenced field conditions since pressure induced advective emissions of gases may potentially dominate over diffusion-controlled emissions (Elberling et al., 1997). Also, methane oxidation in microbiologically active field soils may typically range between 4.7 and
16.7 μg CH₄ m⁻² h⁻¹, depending upon soil type/texture, land use and soil moisture status (Boeckx et al., 1997) and hence could be a key mechanism controlling methane emissions. Therefore, care needs to be taken in generalizing the present results and apply- ing them to (or comparing them with) field-scale conditions where additional complexities may also play important roles at larger scales. The study thus provides valuable experimental and numer- ical insights for future field-scale studies on methane dynamics to further investigate atmospheric and subsurface control mech- anisms and thereby to find appropriate strategies to mitigate methane migration from buried shallow leaky gas infrastructure.

4. Conclusions

Based on a series of controlled laboratory experiments using a two-dimensional bench-scale porous media test facility cou- pled with an open-return wind tunnel, this study investigated the effects of near-surface atmospheric controls (wind and tempera- ture) and soil conditions (textural heterogeneity and soil moisture) on methane transport in the shallow subsurface. Wind velocity showed a more pronounced effect on steady-state surface methane concentration as compared to temperature. The observed decreases-ing surface methane concentration with increasing wind speed could be expressed by a two-parameter nonlinear power function. Temperature effects at no- wind conditions were affected by large variability in measured data while the wind effects obscured the temperature effects at high-wind conditions. Yet, the wind effects were not particularly evident in subsurface methane concentra- tion measurements due likely to limited mixing/penetration depth combined with low permeability in fine-textured porous media.

Clear effects of soil textural heterogeneity and, to a varying degree, of soil moisture were observed in steady-state subsur- face methane concentration profiles. The distinctly low methane concentration across the depth in layered systems was attributed to the rapid mixing and migration of methane within the high-permeability coarse-textured layer. The subsurface methane concentration profiles for no-wind conditions could be adequately simulated using the TOUGH2/EO57CA simulator based on mul- tiphase, density-dependent flow and transport of methane, air, and water vapor with a Fickian ADM framework. Temperature effects were not pronounced in observed and simulated subsurface methane concentration profiles under no-wind conditions. Some important effects, for example, the dominant effect of saturation over soil texture, could also be inferred from numerical characteri- zation. We emphasize that the experimental and numerical results are essentially valid for the selected length scales of the study and should be extended to larger scale applications with caution.

Acknowledgements

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References


Intergovernmental Panel on Climate Change (IPCC), 2011. In: Edenhofer, O.


Table 1
Physical properties and transport parameters of the two porous media.

<table>
<thead>
<tr>
<th>Porous Media</th>
<th>Particle Diameter</th>
<th>Particle Density (ρp)</th>
<th>Total Porosity (Φ)</th>
<th>Bulk Density (ρb)</th>
<th>Saturated Hydraulic Conductivity (Ksat)</th>
<th>Thermal Conductivity (λ)</th>
<th>Gas Diffusivity (Dg/Do)</th>
<th>van Genuchten Parameters</th>
<th>Permeability</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(D95)</td>
<td>mm</td>
<td>g cm⁻³</td>
<td>cm³ cm⁻³</td>
<td>g cm⁻³</td>
<td>cm s⁻¹</td>
<td>W m⁻¹ K⁻¹</td>
<td>(cm³) (cm⁻³) (cm⁻³)</td>
<td>n</td>
</tr>
<tr>
<td>12/20</td>
<td>1.04</td>
<td>2.665</td>
<td>0.33</td>
<td>1.82</td>
<td>0.376</td>
<td>0.314</td>
<td>2.948</td>
<td>0.201</td>
<td>0.056</td>
</tr>
<tr>
<td>10/40</td>
<td>0.53</td>
<td>2.665</td>
<td>0.336</td>
<td>1.77</td>
<td>0.166</td>
<td>0.287</td>
<td>2.887</td>
<td>0.184</td>
<td>0.017</td>
</tr>
</tbody>
</table>

* Measured independently using one-chamber diffusion apparatus (Taylor, 1949), see Chant and Deepak et al. (2016).

* Estimated from Ksat assuming fluidity of 0.81 x 10⁻¹² m⁻¹ s⁻¹ at 20°C.

Table 2
Experimental schedule and the notations used to describe different experimental conditions.

<table>
<thead>
<tr>
<th>Above Ground Controls</th>
<th>Low Temp. (20–24°C)</th>
<th>High Temp. (35–38°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>u = 0 m s⁻¹</td>
<td>u = 0.5 m s⁻¹</td>
<td>u = 2.0 m s⁻¹</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Below Ground Controls</th>
<th>ND-HG-LT</th>
<th>ND-HG-HT</th>
<th>ND-HG-HT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ground</td>
<td>Homogeneous</td>
<td>Layered</td>
<td>Homogeneous</td>
</tr>
<tr>
<td>Controls</td>
<td>Partially-sat</td>
<td>Homogeneous</td>
<td>Partially-sat</td>
</tr>
</tbody>
</table>

ND, Near-dry; PS, Partially-saturated; HG, Homogenous; LY, Layered; LT, Low temperature; HT, High temperature

the subsurface migration of methane gas. Three experiments were duplicated (not shown in Table 2) to confirm the repeatability of the measurements. In the following text, we follow the notations in Table 2 (presenting saturation-soil state-temperature combinations) to describe results relevant to the different scenarios.

All the experiments were conducted within a barometrically closed building under invariant barometric pressure conditions. The experimental setup was located within the laboratory in such a way that the induced pressure gradients (e.g., due to opening/closing of doors) minimally influenced the experimental results. The passersby were restricted within the experimental area during the experiments to avoid likely pressure fluctuations. Pressure sensors installed within the building were randomly checked to ensure constant pressure conditions.

2.2. Numerical model

Numerical simulations were performed using the multiphase transport simulator TOUGH2 (Pruess et al., 1999) combined with the equation of state module EOS7CA (Oldenburg, 2015). TOUGH2/EOS7CA simulates the subsurface flow and transport of

Table 3
Governing equations pertinent to this study as solved in TOUGH2/EOS7CA.

<table>
<thead>
<tr>
<th>Description</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass and energy balance</td>
<td>( \frac{\partial}{\partial t} \int_{V} M \phi_{k} \mathbf{v}<em>{k} , dV + \int</em>{S} \mathbf{f}_{e} \cdot \mathbf{n} , dS )</td>
</tr>
<tr>
<td>Mass accumulation</td>
<td>( M_{k} = \mathbf{p} \cdot \mathbf{S}<em>{k} + \Delta \Sigma</em>{i} p_{i} k_{i} )</td>
</tr>
<tr>
<td>Phase flux</td>
<td>( F_{p} = -K_{p} \phi_{p} \left[ P_{p} - P_{w} \right] )</td>
</tr>
<tr>
<td>Component flux: advective and diffusive</td>
<td>( F_{k}^{adv} = -K_{k} \phi_{k} \mathbf{S}_{k} \cdot \mathbf{v} )</td>
</tr>
<tr>
<td>Relative permeability (van Genuchten, 1980)</td>
<td>( \frac{S_{h}}{S_{s}} = \left{ 1 - \left[ 1 - \left( \frac{S_{w}}{S_{w}} \right)^{n} \right]^{1/n} \right}^{2} )</td>
</tr>
<tr>
<td>Capillary pressure (van Genuchten, 1980)</td>
<td>( \Delta P_{c} = P_{c} \left[ \left( \frac{\mathbf{S}^{1/3}}{1 - \mathbf{S}^{1/3}} \right)^{1/3} \right] )</td>
</tr>
<tr>
<td>Henry's law</td>
<td>( P_{e} = K_{s} \phi_{w} )</td>
</tr>
</tbody>
</table>