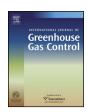
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Contents lists available at ScienceDirect

International Journal of Greenhouse Gas Control

journal homepage: www.elsevier.com/locate/ijggc



Characteristic length measurement of a subsurface gas anomaly—A monitoring approach for heterogeneous flow path distributions



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ARTICLE INFO

Article history: Received 3 August 2015 Received in revised form 29 January 2016 Accepted 8 February 2016

Keywords:
Monitoring
CO₂
Carbon capture and storage
Leakage
Quantification
Subsurface

ABSTRACT

Geogenic gases from natural sources, carbon dioxide (CO_2) from a geological repository (carbon capture and storage—CCS) or a leaking gas pipeline can present serious risks in industrial and urban areas. To extend the lead time for risk treatment in such critical regions, reliable detection of gases within the shallow subsurface is required to observe critical gas accumulations before degassing into the atmosphere.

A near real-time monitoring approach is introduced to determine the volumetric expansion of a leaking gas in the subsurface. Considering the pressure relaxation with the ambient air pressure, the approach enables the forecasting of the final size of a pressurized gas body in terms of characteristic lengths. According to theoretical basics, such a characteristic length, which enables us to perform a gas (safety) measurement based on a purely geometrical measure, behaves independently of subsurface properties, i.e., it enables a reliable quantification of the escaping gas irrespective of its heterogeneous or changing flow path distribution. A field test for a 10 l/min pinhole leakage injected into a 10 m long, 0.4 m wide, 0.95 m deep soil-filled trench that was equipped with linear sensors shows the lateral-vertical volumetric gas expansion along these sensors, and demonstrates the applicability of the characteristic length approach.

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1. Introduction

Gas components such as carbon dioxide (CO_2) , carbon monoxide (CO), methane (CH_4) , and hydrogen (H_2) , or mixtures such as natural gas, city gas, and dump gas, are becoming increasingly important for various environmental and technical processes or urban use. This results in a growing need to monitor these gases, for instance within the subsurface, to enhance safety, protect the environment and human health, or to estimate (greenhouse) gas flows into the atmosphere. Measurement techniques have to be capable of integrating over locally fluctuating gas concentrations from heterogeneous natural systems, such as soils, aquifers, and surface water bodies, or over leakage pathways from infrastructure (e.g. pipelines, repositories, and reactors).

The geological storage of CO_2 as an option for CCS (carbon capture and storage), which is an ambitious technology to decrease anthropogenic greenhouse gas loading to the atmosphere, contains

risks caused by leakage (Gale, 2004; Pawar et al., 2015). Leakage from such a repository may occur along faults, fractures, or faulty, poorly plugged, corroded, or old abandoned wellbores (Liu, 2012; Tsang et al., 2008). Preferential gas flow paths can be activated by changing stress, earthquakes, or geochemical reactions, whereas structural deformations and fractures could also be a consequence of the CO₂ injection itself (Liu and Smirnov, 2009; Tsang et al., 2008). In addition, near the surface, the leakage behavior will be influenced by meteorological factors, e.g., variations in temperature and atmospheric pressure, wind close to the surface, and rainfall (Annunziatellis et al., 2008; Guo et al., 2008; Neeper, 2001; Nilson et al., 1991; Viveiros et al., 2009; Wyatt et al., 1995).

Besides a leakage from a deep repository, leakage could also occur by corrosion of a shallow pipe or tension fissure degassing from small associated pin-hole leaks. Analyzing the pipeline transportation process from a coal-fired power plant to a storage site, Witkowski et al. (2014, 2013) simulated a leak-influenced region. They concluded that the most important safety issue is rigorous and robust hazard identification and recommended pipe segmentation using safety valves in combination with reliable leak detection systems—especially in populated areas.

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Nomenclature a_0, a_1, a_2 polynomial coefficients (mbar, mbar/s, mbar/s²) linear pressure change with time (mbar/s) Α surface area of membrane (m²) b width, m coefficient of order "k" β_k positive root of order "k" λ_k α relative diffusivity concentration (mol/m³ or vol%) C C_0 , C_δ concentration in the outer and inner membrane faces (mol/m³) mole fraction of gas "j" χi Ď diffusion coefficient (m²/s) δ membrane thickness (m) selectivity Φ_g gas-filled porosity geometric factor (m⁻²) g h Heaviside step function Η height (m) gas flux density (mol/m²/s) k_0^j, k_1^j offset (vol%) and slope (vol% s/mbar) for concentration measurement length of a line-sensor cell (m) L_{CO_2} characteristic length of gas impacted region (m) $p_{\rm in}^{j}$ partial pressure outside and inside the sensor cell p^j , (mbar) \bar{p}^j averaged partial pressure (mbar) \tilde{p}^{j} relaxed partial pressure (mbar) pressure in the measurement chamber (bar) $p_{\rm in}$ non-influenced ambient phase pressure (bar) p_n permeability (m²/s) log-mean radius of a tubular membrane (m) $r_{\rm ln}$ R gas constant (8.314 J/mol/K) R_i, R_o inner and outer radius of a tubular membrane (m) ROI radius of influence (m) S solubility t time (s) time constant (s) t_0 temperature (K) T dimensionless time parameter τ_s ı9 ratio of mole numbers mole number of a gas (mol) ν_m , $\nu_{\rm in}$ volume of measurement chamber (m3) coordinate directions (m) X, ZSubscripts, superscripts bgr background below ground level bgs within measurement chamber in index, e.g., of a line-sensor or an installation depth

A comprehensive review of developments in understanding potential environmental impacts of CO₂ leakage is given by Jones et al. (2015). This review analyzes onshore as well as offshore ecosystems. Near or at the soil surface (onshore), the environmental impacts caused by CO₂ leakage include (Chadwick, 2008): (i) hazards for living organisms, i.e. near-surface conditions that could permit leaking CO₂ to accumulate locally, and high concentrations of CO₂ may be attained in depressions and confined spaces, and (ii)

inj

j, s

m

injection

gas component

within the membrane

leakage of CO_2 will impact on the biodiversity of ecosystems, as shown, e.g., for agro-ecosystems by Patil et al. (2010).

This paper focuses on the early detection of CO2 leakages at an initial "pinhole-status" in the shallow subsurface. A large number of potential technologies for CO₂ monitoring already exist, as summarized in the literature (e.g. Chadwick, 2008; Chadwick et al., 2009; Jenkins et al., 2015; Litynski et al., 2012; Liu, 2012). Technologies and methods available for near-surface and surface monitoring include: groundwater monitoring (e.g. de Caritat et al., 2013), discrete and continuous vadose zone gas sampling (e.g. Beaubien et al., 2013; Romanak et al., 2012; Schacht and Jenkins, 2014; Schloemer et al., 2013), soil flux monitoring based on accumulation chambers and micrometeorological eddy flux monitoring (e.g. Anderson and Farrar, 2001; Elío et al., 2013; Madsen et al., 2009), and combined observations (e.g., Quattrocchi et al., 2009; Voltattorni et al., 2009). Besides such direct monitoring methods that consider the concentration(s) of the target gas component(s), tracers (e.g. Koike et al., 2014; Myers et al., 2013; Stalker et al., 2015; Wells et al., 2007), and geoelectric measurements (e.g. Flechsig et al., 2010; Lamert et al., 2012) have also been applied in the shallow subsurface (viz. the unsaturated zone, shallow groundwater).

Monitoring above the surface in the low atmosphere could also provide spatial information of a leakage, as shown for example by Turnbull et al. (2014), who examined the point source CO₂ emission of a gas treatment plant up to an altitude of about 100 m above ground. However, several challenges exist for this approach. At the small (meter) scale of a typical leakage, gas mixing causes a highly variable dispersion and dilution of the escaping CO₂. Furthermore, several sources of CO₂ emissions exist, such as soil and vegetation, and those on a larger scale, e.g., urban traffic, combustion, and other industrial processes that could mask a leakage due to their magnitude and temporal variability (Chadwick et al., 2009). For direct gas monitoring at the shallow surface, in turn, a dense sensor network would be necessary to detect and monitor CO₂ leakages from a deep CO₂ source (i.e. a repository, natural source). This spatial sampling problem with point-wise monitoring is discussed, e.g., by Jenkins et al. (2015).

Direct monitoring within the shallow subsurface requires greater effort for installation with respect to monitoring at the surface. However, it has also a number of advantages, e.g.: (i) it has a higher probability of detecting a leak due to the longer residence time of the gas anomaly in the subsurface than in the atmosphere above the leak, (ii) the expected position of the gas discharge can be estimated with a comparatively high accuracy with respect to gas movement detected in the deeper ground, and (iii) shallow subsurface monitoring can enhance the time for hazard management.

To monitor gases in the subsurface, membranes (e.g. tubes or membrane-coated chambers) have often been used for gas phase separation. Microporous hydrophobic membranes, e.g., those made from polypropylene (Accurel), enable fast sampling due to a more rapid equilibration of the gas phase within a measurement chamber; meanwhile, dense membranes e.g. polydimethylsiloxane (silicon), cause comparatively little interaction with an ambient gas phase due to a comparatively slow diffusive equilibration (for applications, see e.g., DeSutter et al., 2006; Jacinthe and Groffman, 2001; Loose et al., 2009; Ooki and Yokouchi, 2008; Panikov et al., 2007; Schloemer et al., 2013; Seethapathy and Górecki, 2012).

Most direct monitoring systems for the shallow subsurface are designed to provide local information. Schloemer et al. (2013) developed a space-resolved unsaturated zone monitoring technique based on spatially distributed closed wells (using Accurel membranes) for soil gas baseline monitoring with respect to CCS.

An alternative monitoring technology was developed that could gather information from larger areas. Here, membrane-tube based linear sensors (line-sensors) are used, which could be installed directly in the subsurface into refilled holes and horizontal slots,

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