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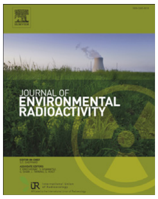

Field study of using naturally occurring radon to assess the dense non-aqueous phase liquid distribution in saturated zone

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ABSTRACT

The concept of radon deficiency such as the ratios of radon concentrations to the maximum measured value of a sample batch was employed as the survey methodology for this study to investigate contamination sources in an industrial zone that was suspected of causing subsurface dense non-aqueous phase liquid (DNAPL) contamination. The results showed that radon concentrations in certain wells were significantly lower than that in uncontaminated regions. Radon concentrations in groundwater are influenced by the in situ bioremediation of vegetable oil, which causes abnormal reductions of the radon in groundwater because radon partitions into vegetable oil and results in more variable for the radon deficit method to showing the impacts of remediation. Six contaminated regions were identified by integrating radon concentration ratios (divided into low (L), middle (M), and high (H) levels) and DNAPL concentrations (divided into low (L) and high (H) levels). Contaminated regions in the LH, MH, and HH categories are located in the vicinity of the contamination source, and those in the HL category are located far from the source zone. The ML and LL categories indicate the involvement of unknown factors, and that additional analyses are required to uncover the facts that affect radon and DNAPL concentrations.

1. Introduction

Non-aqueous phase liquids (NAPLs) are organic liquids that are immiscible with water, such as gasoline, diesel fuel, and chlorinated organic solvents. Inappropriate disposal of these chemicals or leakages of subsurface storage tanks and pipes cause subsurface contamination. Chlorinated solvents are the major contaminants in groundwater supplies and at disposal sites and can remain subsurface over an extended period, resulting in long-term contamination sources of groundwater (Cohen and Mercer, 1993). To investigate the contaminated source regions and to quantify the contaminants are crucial for taking appropriate remediation measures.

Radon-222 (hereafter referred to as radon) has been used as a tracer to locate contaminated sources and to quantify contaminants through direct exposure with contaminants at numerous NAPL-contaminated site investigations. Changes of radon concentration in groundwater or soil gas have been applied at sites contaminated by NAPLs (Davis et al., 2003; Fan et al., 2007; Galhardi and Bonotto, 2012; García-González et al., 2008; Grossmann et al., 2007; Höhener and Surbeck, 2004; Hunkeler et al., 1997; Schubert et al., 2011, 2007a; 2007b, 2007c; 2005, 2001; Starr, 2007).

Using radon as a tracer to locate or quantify subsurface NAPL is based on comparing the distributions of radon concentration of groundwater. If NAPL is present in subsurface, the radon concentration in the groundwater surrounding contaminated source regions significantly reduces as radon distributes from the groundwater into the NAPL. This reduction in radon concentration is referred to as radon deficiency (Semprini et al., 2000). The radon deficiency level is correlated to the content of NAPL; thus, comparing the radon concentrations in the groundwater of monitoring wells in different regions enables evaluating the locations of potential NAPL sources. In addition, the relative residual saturation of NAPL (S_{NAPL}) in soil can be estimated using the following formula (Semprini et al., 1993):

$$C_{\mathrm{w(NAPL)}}/C_{\mathrm{w(background)}} = 1/(1+S_{\mathrm{NAPL}}(K_{\mathrm{c}}-1)) \quad (1)$$

where $C_{\mathrm{w(NAPL)}}$ represents the radon concentration in groundwater of the regions that contain NAPL, $C_{\mathrm{w(background)}}$ represents the radon

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concentration in the groundwater of the region that does not contain NAPL, S_{NAPL} represents the relative saturation of residual NAPL in the soil of saturated zone, and K_c represents the radon partition coefficient between NAPL and water. Based on this equation, when soil does not contain residual NAPL ($S_{\mathrm{NAPL}} = 0$), radon concentration ($C_{\mathrm{w(NAPL)}}$) equals the background concentration ($C_{\mathrm{w(background)}}$). As the S_{NAPL} increases, the $C_{\mathrm{w(NAPL)}}/C_{\mathrm{w(background)}}$ (i.e., radon deficit in this paper) decreases.

Radon deficiency is highly sensitive to small amounts of saturated residual NAPL. Semprini et al. (2000) employed Soltrol 220 at a residual saturation of 5%, and indicated that radon concentration decreased to 31% of the original level. Hunkeler et al. (1997) surveyed a diesel fuel-contaminated site, and found that radon concentration in the contaminated site was approximately 60% of that in groundwater upstream of the site. Schubert et al. (2007c) investigated an abandoned gasoline station and found that the radon concentration measured in known contaminated source regions was the lowest (i.e., approximately 50% of the background concentration in local groundwater). Fan et al. (2007) examined a petrochemical refinery plant and showed that radon concentration in contaminated source regions was approximately 31% of that in upstream groundwater.

Although a naturally occurring subsurface radon tracer has been extensively applied in NAPL contamination site investigations, it has rarely been employed at DNAPL-contaminated sites (Davis et al., 2003; Semprini et al., 2000; Starr, 2007). Davis et al. (2003) and Semprini et al. (2000) indicated that radon deficiency exhibited superior ability in predicting the amount of existing DNAPL. However, in a field investigation, Starr (2007) did not obtain expected results, potentially because of the heterogeneity in the radon concentration of the ground layer or because of other unknown factors.

Studies related to radon deficiency in DNAPL-contaminated sites are scarce, additional field studies and applications are required. This study focused on an industrial area suspected of originating groundwater DNAPL contamination and investigated contamination sources in this area based on radon deficiency. In addition, this study integrated radon concentration ratios and DNAPL concentrations to classify the types of contamination at contaminated sites. The study results may contribute relevant knowledge regarding radon deficiency levels in DNAPL-contaminated site investigations and clarify current understanding of this topic.

2. Site description

2.1. History of industrial area

The study site is an industrial zone in Central Taiwan, which began operating in the 1970s. Since 1980, various industries (e.g., optical products, glass and grinding, and metal) have flourished, and subsequently, the electronic and optical product industries have become the two main industries in this area. These two industries were associated with using organic solvents. Before 1997, approximately 10 factories in the industrial zone had been using trichloroethylene (TCE) and tetrachloroethylene (PCE) as cleaning solvents; however, the factories stopped using chlorinated organic solvents in 1997, when the regulation and control of chlorinated organic solvent use went into effect. In 2001, investigations by the local government revealed that traces of chlorinated organics had been detected in the groundwater of several regions in the city. In 2010, following a long-term follow-up investigation, the industrial zone was identified as a potential source of the chlorinated organic contaminants for groundwater. After an extensive groundwater quality survey of the industrial zone, the results indicated that two regions contained high concentrations of TCE and PCE, which were located on the south side of the industrial zone (between Nos. 3 and 4) and in the vicinity of No. 2 (Fig. 1).

2.2. Site hydrogeology

The research site is composed of Holocene alluvium, and the main geological material is gravel mixed with fine sand and silt. The particle size and content of gravel approximately 50–60 m below the ground surface (bgs) gradually increase with depth. Additional examinations showed that at 200 m bgs, the layer still consists of gravel mixed with sand or silt; thus, the actual thickness of the topmost aquifer could not be determined. Groundwater table in the southern part of the perimeter of the industrial zone was found at approximately 60–67 m bgs. The levels of groundwater table go deeper gradually from west to east, where gravel content increases with depth, and silt or clay content decreases correspondingly. This phenomenon indicates that deeper layers possess a superior geological condition of permeability. Based on a slug test result of the monitoring wells, the hydraulic conductivity of this site was approximately 10^{-4} to 10^{-2} cm s^{-1}, which is a characteristic of a typical aquifer medium that contains moderately fine sand and silty sand, and is highly permeable. Groundwater flows from north to south, slightly tending toward the southeast direction (Fig. 1). The average hydraulic gradient for the overall flow field was approximately 0.7%, and the groundwater flow velocity was approximately 1–60 cm d^{-1}.

Both geological materials and rainfall infiltration affect variations in radon concentrations. Although no emanation studies have been conducted using aquifer samples, a previous study of radon levels in the same groundwater regions in which the study site located reported that in a region measuring 5 km (in the groundwater flow direction) × 1 km (in width), the radon concentration ranged between 15.63 and 20.85 Bq l^{-1}, with a relative standard deviation of 8.7%. The results were regarded as the natural variability in radon concentrations for the case study.

2.3. Remediation measures

The remediation zone at the perimeter of the south side of the industrial zone was divided into two regions: the area between well Nos. 3 and 4, and the area in the vicinity of well No. 2. Screens for remediation wells are 9 m long and located beneath the groundwater table to improve the surrounding aquifer of the wells to at least 9–10 m deep. Between June and July 2012, the two regions underwent bioremediation by injection of EcoClean and EcoClean-E. EcoClean is composed of food ingredients, such as hydrocarbons and amino acids, which are a source of nutrients for soil microorganisms. EcoClean-E is an emulsion-like substance primarily composed of food-grade vegetable oil and surfactants; it is also a nutrient source for microorganisms and is able to maintain the remediation effect for months.

2.4. Batches of groundwater sampling

The groundwater for radon analysis was sampled in November 2011, June 2012, and October 2012 by using the diffusion bag approach to sample various depths concurrently. A portion of the sampled water underwent volatile organic compound (VOC) analysis at a certificated environmental analysis institution. The analysis results for both groundwater radon and VOCs were used for contamination source investigation. Data of the wells employed for each sampling campaign are shown in Table 1. The purpose of obtaining samples in November 2011 was to verify, according to the radon deficiency, that Wells 2, 3, and 4 were located in the primary areas of groundwater contamination, and to determine whether