

11th Applied Isotope Geochemistry Conference, AIG-11 BRGM

Using natural radon as a tracer of gasoline contamination

Gabriele De Simone^a, Gianfranco Galli^b, Carlo Lucchetti^a, Paola Tuccimei^{a,*}

^aUniversità "Roma Tre", Dipartimento di Scienze, Largo San Leonardo Murialdo 1, 00146 Roma, Italy

^bIstituto Nazionale di Geofisica e Vulcanologia, Sezione Roma 1, Via di Vigna Murata 605, 00143 Roma, Italy

Abstract

Radon concentration in soil gas can be employed to trace the presence of gasoline contamination because it is very soluble in hydrocarbons. Radon available in the soil pores accumulates in the contaminated volume, producing a local deficit at shallow depth that can be easily mapped. The suitability of this method is here checked by field simulation of a gasoline spill and by laboratory experiments where known amounts of a volcanic tuff contaminated with increasing gasoline show proportional drops of radon exhalation rates.

© 2015 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Peer-review under responsibility of the scientific committee of AIG-11

Keywords: Radon; Natural tracer; NAPLs contamination,

1. Introduction

Spills of gasoline, or other Non-Aqueous Phase-Liquids (NAPLs) can lead to significant contamination if not handled quickly and properly. Any appropriate actions need a good knowledge of the contaminant extension in the subsurface. Standard practice is to excavate visible contamination or to drill monitoring-wells for the purpose of collecting samples for analysis. However, it is expensive and difficult to satisfactorily trace the shape of subsurface NAPL pollution this way. In order to obtain a sufficiently detailed picture of ground contamination, a complementary detection method which allows a denser network of monitoring points should be desirable [1].

The radioactive noble gas radon, a naturally occurring component of soil gas, exhibits very good solubility in a wide range of NAPLs [1, 2, 3]. Consequently, subsurface NAPL contamination partly traps the soil gas radon,

* Corresponding author. Tel.: +39-06-5733-8092; fax: +39-06-5733-8201
E-mail address: paola.tuccimei@uniroma3.it

resulting in a local reduction of the soil gas radon concentration in the vicinity of the contaminated soil volume (Fig. 1).

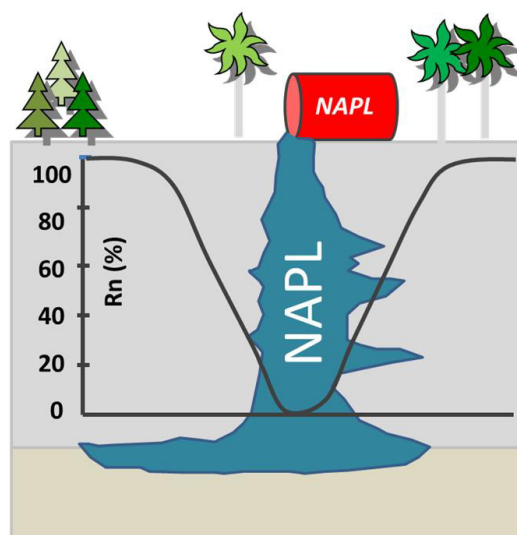


Fig. 1. Local reduction of soil radon concentration in the area surrounding the contaminated soil volume. Modified after Schubert et al. [1]

Provided this “NAPL-Effect” is significant, a NAPL contamination could be indirectly localized by carrying out a straightforward radon survey on a sampling grid covering the suspected area.

The aim of this investigation is to verify the principle, reproducing a gasoline spill within a volcanic bedrock and monitor soil radon concentration over following weeks. Laboratory tests are also presented to quantify radon exhalation decrease as a function of volumes of added gasoline.

2. Simulation of gasoline spills

In order to verify the principle, gasoline spills were reproduced in the laboratory and in the field, choosing a volcanic bedrock because of its high radon activity concentration ($100 - 200 \text{ kBq/m}^3$). Radon in soil gas was then monitored over following weeks to assess its evolution.

2.1 Laboratory simulation of a gasoline spill

A large garbage bin was filled with “Tufo di Villa Senni” ignimbrite, a volcanic product available in large quantities in the city of Rome. A PVC tubing (evidenced in blue in Fig. 2a) allowed us to inject gasoline at the bottom of the container. A hollow probe was driven into the terrain at a depth of 40 cm, about 10-15 cm above the contaminated soil volume and connected via vinyl tubing to a drying unit (identified as drierite in Fig. 2a) and to RAD7 radon monitor (DurrIDGE Co.).

Soil radon was measured twice before the gasoline injection, immediately after the spill, and five times more in the following weeks. Fig. 2b shows clearly the abrupt drop of concentration right after the NAPL input and a gradual increase of soil radon values over time due to progressive volatilization of the gasoline. The recovery of

pristine concentration was not obtained, indicating the presence of residual NAPL that still accumulates radon, producing a radon deficit. This demonstrates the potential of radon in soil gas to detect the presence of residual NAPL in the vadose zone of an aquifer.

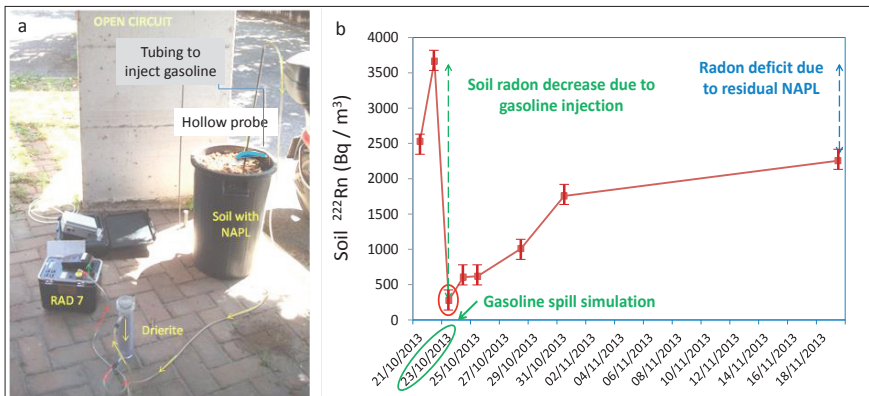


Fig. 2. Experimental set-up used to simulate a gasoline spill and monitor related soil radon concentration evolution (a). Radon activity concentration in soil gas during the test (b).

2.2 Field simulation of a gasoline spill

A second spill was reproduced in Vallerano town (Lazio Region, Italy), where pyroclastic flow deposits (Ignimbrite D) from Vico volcano outcrop. Two liters of gasoline were poured in the center of the area at 80 cm depth and soil radon was monitored at 15 sampling stations using a grid of 3x5 m, before, just after the spill and over the following months. Soil radon concentration at the spill point fell from about 700 to 7 kBq m⁻³ (few hours after the contamination event). A week later the concentration was still the same, but increased up to about 60 kBq m⁻³ four months later. It is worth noting that the deficit effect was closely restricted to the contaminated station, because sites located just 3 meters apart just showed slight decreases around half or one third of the starting value. Stations at five meters distance did not display any departure from initial condition (Fig. 3).

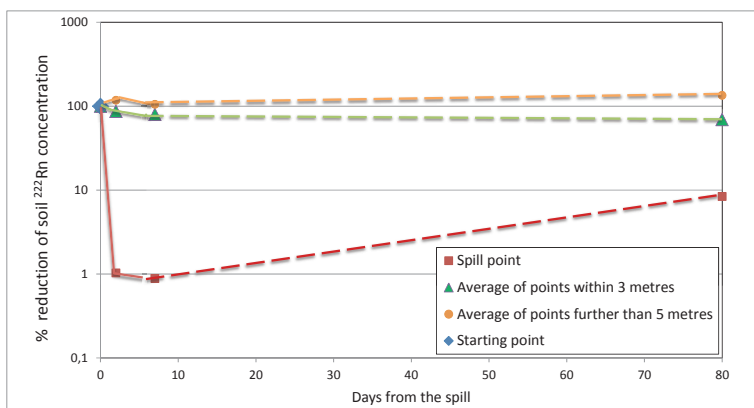


Fig. 3. Percent reduction of soil radon concentration in Vallerano (Lazio Region, Italy), following a simulated gasoline spill at different monitoring stations located over a 3 x 5 m grid of 15 measurements points.

The presence of gasoline is very well indicated by high concentration of H₂S (up to about 370 ppm at the spill point), due to the residual sulphur component in the gasoline after the oil refining process.

3. Decrease of radon exhalation rate of a volcanic material induced by addition of gasoline

Seven hundreds grams of a volcanic material (Villa Senni tuff from Colli Albani volcano, Roma, Italy) have been grounded, dried at 100°C and placed in a stainless steel accumulation chamber, connected to a drying device (drierite) and to the radon monitor in a closed-loop circuit. The chamber is located in a thermo-refrigerating bath, set at 35°C to keep constant temperature during the measurement and enhance the radon exhalation [4].

The different experiments were carried out adding different amount of gasoline (from 0 to 25 mL) to the tuff, resulting in gasoline/material ratios (mL/kg) from 0 to 50. Each tests had a duration of 24 hours, with 12 two hours long cycles, keeping relative humidity below 10% inside the radon monitor.

As reported in Fig. 4, radon mass exhalation rates exponentially reduce with increasing gasoline volumes, probably because more radon is progressively dissolved in gasoline. These experiments need a further control in order to assess if drierite absorbs vaporized gasoline where radon is partially dissolved.

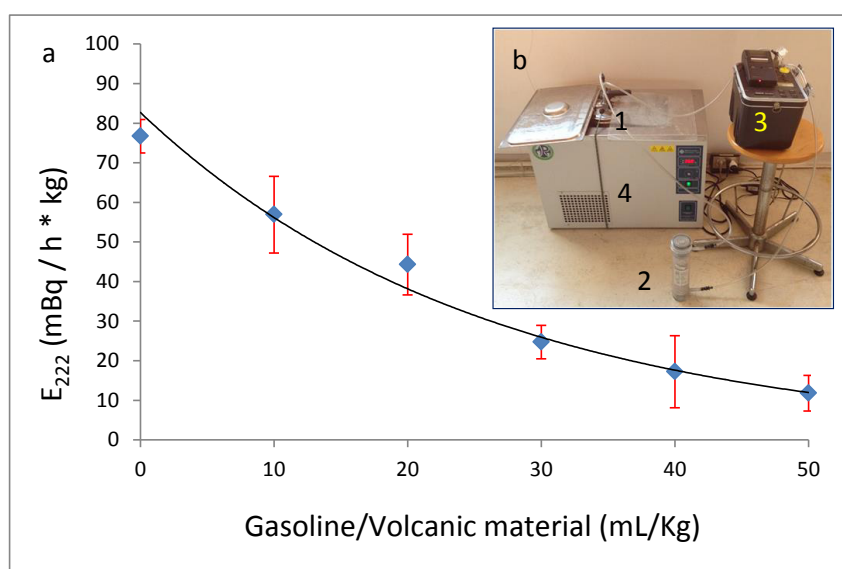


Fig. 4. Decrease of radon exhalation rate of Villa Senni tuff induced by the addition of different volumes of gasoline (a). The experimental set-up consists of: the accumulation chamber containing the contaminated volcanic material (1), the drierite (2), the radon monitor (3), the thermo-refrigerating bath (4), connected by vinyl tubing in a closed-loop circuit (b).

References

- Schubert M, Freyer K, Treutler HC, Weiss H. Using radon-222 in soil gas as an indicator of subsurface contamination by non-aqueous phase-liquids (NAPLs). *Geofisica Internacional* 2002; **41**: 433-437.
- Barton AFM. *Handbook of solubility parameters and other cohesion parameters*. 2nd ed. Boca Raton/FL/USA: CRC; 1991.
- Lewis C, Hopke PK, Stukel J. Solubility of radon in selected perfluorocarbon compounds and water. *Industrial Engineering & Chemical Research* 1987; **26**: 356-359.
- Tuccimei P, Castelluccio M, Moretti S, Mollo S, Vinciguerra S, Scarlato P. Thermal enhancement of radon emission From geological materials. Implications For laboratory experiments on rocks under increasing deformation. In: Benjamin V, Szigethy J, editors. *Horizons in Earth Science Research*, Volume 4, New York: Nova Science Publishers; 2011. p. 247-256.