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## **Radiation Measurements**

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## Radon measurements over a natural-gas contaminated aquifer

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#### HIGHLIGHTS

- ▶ High Radon/Thoron ratios were localized near the natural-gas emanations in a river.
- ► Natural gases are ascending trough a deep geological fault.
- ▶ Apparently, the radon anomaly shows the site where natural gas enters the aquifer.
- ▶ Natural gas source may be related to leaks in the structure of abandoned gas wells.

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#### ABSTRACT

Radon and thoron concentrations in soil pores in a gas production region of the Anzoategui State, Venezuela, were determined by active and passive methods. In this region, water wells are contaminated by natural gas and gas leaks exist in the nearby river. Based on soil gas Radon data surface hydrocarbon seeps were identified. Radon and thoron concentration maps show anomalously high values near the river gas leaks decreasing in the direction of water wells where natural gas is also detected. The area where the highest concentrations of <sup>222</sup>Rn were detected seems to indicate the surface projection of the aquifer contaminated with natural gas. The Radon/Thoron ratio revealed a micro-localized anomaly, indicating the area where the gas comes from deep layers of the subsoil. The radon map determined by the passive method showed a marked positive anomaly around abandoned gas wells. The high anomalous Radon concentration localized near the trails of ascending gas bubbles at the river indicates the zone trough where natural gases are ascending with greater ease, associated with a deep geological fault, being this the main source of methane penetration into the aquifer. It is suggested that the source of the natural gas may be due to leaks at deep sites along the structure of some of the abandoned wells located at the North-East of the studied area.

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#### 1. Introduction

According to the inhabitants of a locality in Anzoategui state, Venezuela, there have been intense natural gas leaks (mostly methane) in several sites in the nearby river for about 10 years. Moreover, during the drilling of two water wells for public consumption there was methane gas leaks and the wells had to be sealed. Isotopic analyses indicated thermogenic origin of methane. The village is located in an area where there are many, mostly

inactive, gas wells. The penetration of natural gas into the aquifer can occur through naturally occurring fault conduits and fracture systems that connects with deep underground gas layers or with a breakdown somewhere along the structure of a gas well.

According to Kristiansson and Malmqvist (1982), nondiffusive transport of Radon can be easily explained by carrier-gas transport, so this non-reactive gas is an ideal tracer for gas transport from deeper layers to the soil surface, and its detection is one of the indicators of hydrocarbon leaks from subsoil (Li et al., 2006; Dyck and Jonason, 2000; Ghahremani, 1985). The advantage of using Radon as tracer of light hydrocarbons lies in that the latters might be involved in shallow biological reactions (Fleischer and Turner, 1984). The location of active faults can be achieved by detecting Radon over a large area. The Radon anomalies make it possible to

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highlight more emissive zones that are either related to main faults or to secondary fractures acting as migration pathways (Landrum et al., 1989; Hus et al., 1999; Patrick et al., 2011). If a strong Radon anomaly is not associated with uranium mineralization, most likely this anomaly is due to the ease of gas migration through highly permeable deep fracture zones possibly caused by geological faults. Irrespective of the origin, the geogases tend to migrate towards the surface due to pressure and buoyancy effects creating areas of anomalous degassing. When the geogas microflow crosses the groundwater may form a stream of bubbles (Sikka and Shives, 2001). Bubble flows related to faults may occur in different geological environments and have been theoretically and experimentally recognized as a mechanism for rapid gas migration.

How to characterise hydraulic connectivity of faults/fractures with a river and alluvial aquifers and how to locate these permeable structures are becoming very critical questions for groundwater risk assessment and water management in oil and gas producing areas. The aim of this study was to determine the areal distribution of Radon and Thoron concentrations in soil gas on a large region that includes the village, for the purpose of detecting the location of structures (fault zones) and suggest which is or may be the source responsible for the occurrence of natural gas in the river and aquifer.

#### 2. Materials and methods

The study was conducted in Freites municipality, Anzuategui state, Venezuela (Fig. 1). The local topography is flat with little variation in the soil properties. Data were collected on a grid composed of eight sets of rectangular rings centered on the locations of the two contaminated water wells, four abandoned gas wells suspected of gas leaks and the river location where gas was observed to emanate. A total of 210 locations, 250 m apart, covering approximately 22 km², were analyzed for soil gas Radon. Measurements were carried out during the dry season.

In the active measurement, soil gas was sampled through steel sampling tubes at a depth of 60 cm. Radon and Thoron measurements were performed with radiation monitors (AB-5 Pylon, Ottawa, Canada) coupled with 150-cm<sup>3</sup> Lucas cells. The monitors were adapted with a simple external filtering system to eliminate

moisture and small soil particles. Flow meters were also employed to ensure the cells were completely full with soil gas at the end of the pumping cycle and for Thoron corrections. The pumping cycle was for 3 min followed with nine 1-min counting periods. Radon and Thoron concentrations were calculated from the recorded count rates. Radon in soil gas was also monitored for long time periods (passive method) using LR-115 type II track detectors from Dosirad Co., France, fixed at the bottom of cylindrical diffusion chambers (65 mm diameter and 75 mm height high density plastic cups). The top of the diffusion chambers were covered with a 30  $\mu m$ thick polyethylene foil to keep aerosols, Radon daughters and Thoron gas out of the measuring volume. Three cups with Radon detectors inside were located at the bottom of each hole drilled at 60 cm depth. The holes were covered with the extracted soil. The time of exposure was 45 days. Detector etching was done under standard conditions (10% NaOH solution at 60 °C during 120 min). After etching, films were washed with distilled water and dried in a dust free chamber. The alpha tracks recorded on the films were counted using an optical microscope coupled to a digital camera. Track images were processed by the ImageJ free Internet access software. Relative Radon concentrations were given by the average track densities since the exposure time, etching conditions and method of track reading were the same for all detectors. Radon and Thoron contour maps were created in the GS+ program (Robertson, 2008).

#### 3. Results

#### 3.1. Active radon measurements

Fig. 2 shows the spatial distribution of soil gas Radon concentrations in the studied area. In zone **A** there are no oil or gas wells, active or inactive, and as far as we know it is not considered a prospective area. The Radon maximum concentration values are near the location where gas emanates from the river and decrease in the direction to the location of the contaminated water wells. Since the aquifer is unconfined, the observed behavior seems to indicate the ascending flux of natural gas dissolved in groundwater, and hence the superficial projection of the gas-contaminated aquifer and its hydraulic connection with the river emanations.

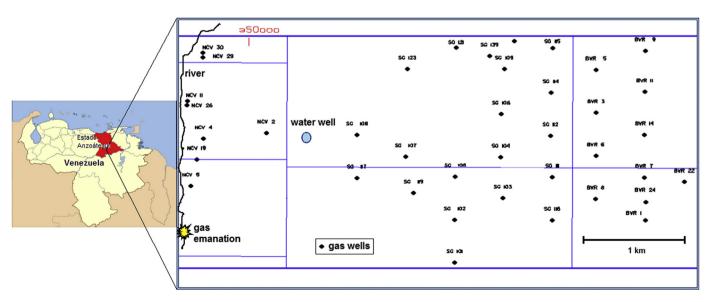


Fig. 1. Map of Venezuela showing where the study took place (left). Location of a water well, where methane gas leak occurred, respect to adjacent gas wells and river gas emanations (right).

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