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Effect of water vapour condensation on the radon content in subsurface air in a hypogeal inactive-volcanic environment in Galdar cave, Spain

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HIGHLIGHTS

- We monitored the weather-driven processes controlling gas exchange on subsurface.
- Anomalies on radon signal were statistically clustered.
- Relative humidity over 70% triggers the vapour effective condensation (EC).
- Reduction of air-filled porosity of rock by EC hides the radon exchange by diffusion.
- Microclimatic conditions were parameterized to predict radon concentration anomalies.

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ABSTRACT

Fluctuations of trace gas activity as a response to variations in weather and microclimate conditions were monitored over a year in a shallow volcanic cave (Painted Cave, Galdar, Canary Islands, Spain). ^{222}Rn concentration was used due to its greater sensitivity to hygrothermal variations than CO_2 concentration. Radon concentration in the cave increases as effective vapour condensation within the porous system of the rock surfaces inside the cave increases due to humidity levels of more than 70%. Condensed water content in pores was assessed and linked to a reduction in the direct passage of trace gases. Fluctuations in radon activity as a response to variations in weather and microclimate conditions were statistically identified by clustering entropy changes on the radon signal and parameterised to predict radon concentration anomalies. This raises important implications for other research fields, including the surveillance of shallow volcanic and seismic activity, preventive conservation of cultural heritage in indoor spaces, indoor air quality control and studies to improve understanding of the role of subterranean terrestrial ecosystems as reservoirs and/or temporary sources of trace gases.

1. Introduction

In volcanic near-surface subterranean systems, by studying variations in the flux of trace gases as radon (^{222}Rn) across the soil/rock–air interface, certain geodynamic processes associated with shallow volcanic activity can be assessed (Eff-Darwich et al., 2002; Viñas et al., 2007; among others). However, anomalies in trace gas concentrations caused by geological events may be hindered by or present irregularities due to certain physical and meteorological factors. Other studies that consider the temporal variations of radon in caves, tunnels and bore-holes focus mainly on the convective processes induced by inside/outside temperature differences and variations in atmospheric pressure (Fernandez-Cortes et al., 2009; Kowalczk and Froelich, 2010; Perrier and Richon, 2010; among others).

The gaseous exchange between surface/underground and atmosphere is linked to the presence of pores, macro-pores and subsurface cavities, which act as storage and/or temporary sources of radon (^{222}Rn), CO_2, H_2O_v and CH_4. Thus, a similar pattern of CO_2 and Rn gas exchange, as controlled by the degree of moisture

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present in the pore and fissure system in the external soil and epikarst, has been described for very stable subterranean atmospheres under vapour-equilibrium pressure near saturation (Cuezva et al., 2011; Fernandez-Cortes et al., 2011).

Subsurface trace gases may be divided into three states: in caves/voids and pore air, dissolved in pore water, and attached by sorption to soil grains (Nazaroff, 1992). In particular, a portion of radon content of pore air is then exhaled from the pore space into microcracks, fractures and the cave atmosphere, mainly by molecular diffusion or advective transport, due to differences in air pressure between the air that fills the cave and the porous rock that surrounds it. Diffusion-induced subsurface radon movement is notable in capillaries and small-pored rocks, compared with advection transport that may play an exclusive role in larger pores or fractured media (Etiope and Martinelli, 2002).

In this paper, water condensation in porous volcanic materials is studied in detail in order to understand the variation in and anomalous levels of radon gas in a shallow cave (Painted Cave, Galdar, Canary Islands, Spain). This is a suitable study site, as it is isolated from direct rainfall and solar radiation, air ventilation with the outer atmosphere is restricted by an artificial barrier, and there is no upper soil cover, which means that the gas exchange between subsurface-atmosphere is mainly limited through the homogeneous volcanic host-rock with a condensable pore structure. We test the hypothesis that condensation/evaporation cycles in inner substrates of host-rock govern the short-term and seasonal isolation and opening of the subterranean atmosphere and, consequently, play a key role in the gas exchange with the outer atmosphere. The cave's microclimate and weather conditions were monitored exhaustively, gas anomalies were statistically identified by clustering the changes in gas signal entropy, and a detailed study of effective condensation was carried out in terms of microclimate conditions and pore structure properties.

2. Materials and methods

2.1. Site and rock description

The town of Galdar is located on the subtropical latitudinal belt with a Mediterranean pluviseasonal-oceanic climate. The Painted Cave is part of a larger complex of six caves that were excavated in the volcanic toba by the island's former inhabitants. The cave complex of Galdar was settled at 99 m above sea level in an isolated volcanic geological structure located well above the current water table. The host rock of the cave complex outcrops without any covering soil layer, and is a sequence of high porous pyroclastic layers (lapilli tuff) with an average thickness of 10–40 cm and softer mm-thick beds (Sanchez-Moral et al., 2002). The volcanic host-rock has a complex porous media: high values of porosity (~36%, with high proportion of connected porosity (28%) and polymodal pore-size distribution (Benavente et al., 2009). Tuff presents two important fractions of small pores in the pore radius ranges of 1 nm–0.01 μm and 0.1–1 μm, which are related to pyroclastic minerals such as zeolites, micritic calcite and argillaceous minerals. The largest pores are defined by pyroclasts in ranges of 1–10 μm and 0.1 μm–1 mm which should therefore favour pore fluid displacement.

The monitoring site is a rectangular decorated chamber (159 m^3) excavated in the host rock (Fig. 1) with the ceiling (average thickness of 2.4 m), floor and three walls of tuff and an artificial, non-airtight closure near the built exterior. The study site is isolated from direct rainfall and solar radiation by a raised structure made from aluminium with polyurethane foam insulation covering an area of 5500 m^2, including the complete footprint of the cave on the surface. This construction is not closed off by any walls, allowing for partial ventilation from surface air. A concrete wall also surrounds the archaeological complex to avoid runoff from allochthonous water. These features make Galdar cave a unique subterranean laboratory in volcanic rock and the conditions help to simplify the study as water vapour is the only source of water supply, and therefore the only hydrological factor to consider.

2.2. Micro-environmental monitoring system and time series analysis

The subterranean laboratory has an eleven-sensor network designed for the narrow range of measurements of air and rock-surface parameters: temperature, relative humidity, and carbon dioxide and radon (^{222}Rn) content. Data were collected every 5 min over a period of 13 months (1/03/2004–31/03/2005) using a

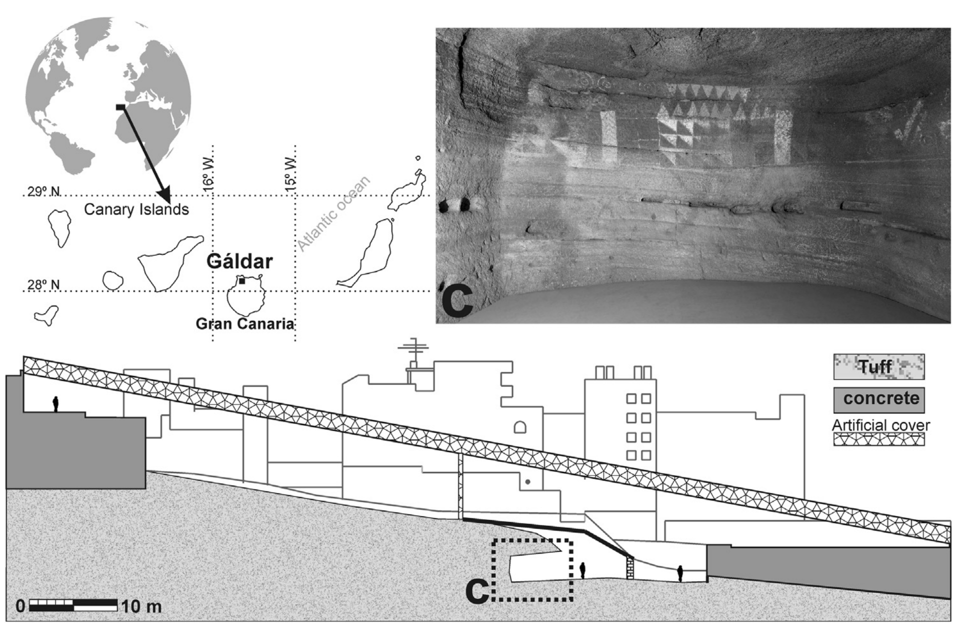

Fig. 1. Location and cross-section of the volcanic outcrop of the Galdar cave complex. C: View of inner surfaces of the Galdar cave.