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# RADON CONCENTRATION IN CAVES AS A PROXY FOR TECTONIC ACTIVITY IN THE CANTABRIAN MOUNTAINS (SPAIN)

# KONCENTRACIJE RADONA V JAMAH KOT KAZALNIK TEKTONSKE AKTIVNOSTI V KANTABRIJSKEM GOROVJU (ŠPANIJA)

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**Abstract** UDC 551.44:546.296(460)

***Daniel Ballesteros, Sergio Llana-Fúnez, Mónica Meléndez-Asensio, Ismael Fuente Merino, Carlos Sainz, Luis Quindós & Irene DeFelipe: Radon concentration in caves as a proxy for tectonic activity in The Cantabrian Mountains (Spain)***

Radon (Rn) constitutes a good geochemical tracer for neotectonic activity in faults since associated fracturing near the surface favours fluid escape to the atmosphere. In this contribution, we measured the Rn concentration in the air inside karst caves to constraints the recent fault activity in the Cantabrian Mountains (N Spain). Rock formations exhumed during the uplifting of the Cantabrian Mountains record a long history of fracturing, which has the potential to connect deeper sources of Rn with the surface. In this regional study, we correlate Rn measurements with cave survey data and geological structures using a Geographic Information Systems. Thirty-four Rn average concentration was recorded by CR-39 detectors during 8 integrated months. The method is applied to the central part of the Cantabrian Mountains that is built on sedimentary and low-grade metamorphic rocks relatively poor in U. Dominant tectonic structures and Rn concentration are examined in 28 cavities. The concentration of Rn values is higher than 0.5 $kBq \cdot m^{-3}$ in caves developed preferably following fractures with the direction N30ºW, being the concentration greater than 0.8 $kBq \cdot m^{-3}$ in cavities located less

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***Daniel Ballesteros, Sergio Llana-Fúnez, Mónica Meléndez-Asensio, Ismael Fuente Merino, Carlos Sainz, Luis Quindós & Irene DeFelipe: Koncentracije radona v jamah kot kazalnik tektonske aktivnosti v Kantabrijskem gorovju (Španija)***

Prenos radona iz globokih virov na površje velikokrat poteka vzdolž dobro prevodnih neotektonskih razpoklinskih struktur. Zato je povečana koncentracija radona na površju eden od potencialnih kazalnikov tektonske aktivnosti v nekem masivu. V raziskavi smo ugotavljali povezavo med koncentracijo radona v kraških jamah in tektonskimi strukturami v Kantabrijskem gorovju v severni Španiji. Zaradi tektonskega dvigovanja Kantabrijskega gorovja, so formacije močno razpokane, zato so povezave med globokimi viri radona in površjem zelo verjetne. Z detektorji CR-39 smo na 34 točkah izmerili povprečno koncentracijo radona v obdobju osmih mesecev. Meritve koncentracije radona so bile izvedene v 28 jamah v centralnem delu Kantabrijskega gorovja. Območje je pretežno iz sedimentnih in nizko metamorfoziranih kamnin z nizko vsebnostjo urana. Korelacijo med koncentracijo radona v jamah ter položajem tektonskih struktur smo določali z orodji geografskih informacijskih sistemov. Koncentracije radona nad 0,5 $kBq \cdot m^{-3}$ smo izmerili v jamah, ki so nastale ob tektonskih strukturah v smeri sever–zahod. Med temi smo koncentracijo nad 0,8 $kBq \cdot m^{-3}$ izmerili v jamah, ki so v bliži-

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than 200±50 m from subvertical faults with such orientation. Rn anomalies point to relative high connectivity along subvertical fault zones NW-trending, preserving fracture connectivity in the most recent structures in the Cantabrian Mountains. Finally, in the study area there is a low but significant radioactive hazard which is associated to fault zones in a fractured rock massif. It contrasts with other active tectonic settings where the radioactive hazard may come from fault movements.
**Key words**: Active fault, geoindicator, karst cave, radon.

ni (< 200 ± 50 m) subvertikalnih prelomov. Visoke koncentracije radona kažejo na dobro razpoklinsko povezanost masiva, ki jo v najmlajših strukturah Kantabrijskega gorovja vzdržujejo subvertikalni prelomi v smeri sever–zahod. Študija je pokazala, da je na območju raziskave majhno, a značilno tveganje povečane radioaktivnosti v povezavi s prelomnimi in razpoklinskimi conami. V nasprotju s tem je v drugih tektonsko aktivnih okoljih radioaktivno tveganje povezano s premiki aktivnih prelomov.
**Ključne besede:** aktivni prelom, geokazalniki, kraška jama, radon.

## INTRODUCTION

Three physical properties of radon (Rn) make this element a reasonable geochemical tracer to identify active geological structures (e.g., Barbosa *et al*. 2015; Baskaran 2016): 1) Rn is a gas that has a relative high mobility, even through low porosity rock formations; 2) Rn is a noblre gas that does not combine with other elements, thus it is not influenced by biotic processes; and 3) the half-life of the Rn is short (3.8235 days for $^{222}$Rn, the most common isotope), therefore its presence is indicative of a nearby source (e.g., rocks with high Rn concentration) and/or a quick transport from the source.

The occurrence of the Rn depends on its emanation fraction linked to rocks and the permability of the bedrock. The emanation fraction is an intrinsic property of the rocks and controls the escape of Rn from the solid grains to the porous media (Sakoda *et al*. 2010; Lee *et al*. 2018). To be transported to the surface, Rn requires a carrier that can be either water, $CO_2$, $CH_4$ or magma, and a high permeability pathway. Thus, the deeper the source is located, the higher the permeability is required (Katsanou *et al*. 2010; Sciarra *et al*. 2015). The geometry and arrangement of fractures or discontinuities in the crust are key in order for the gas to be transported the surface. Additionally, the permeability of an intact rock increases two to ten orders of magnitude with the presence of organised fracture systems, but also by the intersection between them, particularly when the intersecting fractures are subvertical (Sibson 1992, 1996). For these reasons, Rn has been used extensively as a tracer for seismic activity of fault zones (e.g., Ghosh *et al*. 2011; Briestensky *et al*. 2014; Koike *et al*. 2014), detecting Rn anomalies in active faults as the San Andreas fault (see Morrow *et al*. 2014). However, Woith (2015) found that in continuous monitoring of Rn concentration, anomalies associated with non-tectonic origin produced similar anomalies to those associated with faults. The influence of weather, groundwater conditions and/or the lithology can be of similar magnitude to tectonic causes (Girault *et al*. 2012; Zarroca *et al*. 2012).

Rn can accumulate in karst caves due to their low ventilation, representing anomalies in Rn (Elío *et al*. 2017). Annual average Rn concentrations in karst caves ranges typically from 0.2 to 15 kBq·m$^{-3}$ (Kobal *et al*. 1986, 1987; Hakl *et al*. 1997; Vaupotič 2010; Somlai *et al*. 2011; Bourges *et al*. 2014; Nguyet *et al*. 2018; Smith *et al*. 2019), being 2.8 kBq·m$^{-3}$ the average concentration measured in 220 caves around the World (Hakl *et al*. 1997). However, higher concentrations have been reported, for instance: 155 kBq·m$^{-3}$ in the UK (Gunn *et al*. 1991), 23-168 kBq·m$^{-3}$ in Spain (Dueñas *et al*. 1999; Lario *et al*. 2005; Fernandez-Cortes *et al*. 2009; Alvarez-Gallego *et al*. 2015), 4-123 kBq·m$^{-3}$ in China (Wang *et al*. 2019), 88 kBq·m$^{-3}$ in Greece (Papastefanou *et al*. 2003), 84 kBq·m$^{-3}$ in USA (Kowalczk and Froelich 2010) and 27-54 kBq·m$^{-3}$ in Slovenia (Gregorič *et al*. 2013).

The concentration of Rn varies over time and space one to two orders of magnitude in karst caves (Fernandez *et al*. 1984, 1986). Its accumulation in caves is ultimately controlled by the interaction between the cave system, the atmosphere, the groundwater system and the geological bedrock, which necessarily must include the fracture network. Thus, Rn concentration is sensitive to cave ventilation, groundwater conditions, cave sediment distribution and/or uranium content in the bedrock and karst deposits (Hakl *et al*. 1997). The ventilation depends on wind dynamics within the cave (exhalation or inhalation flux, stratified air), controlled by differences in temperature between the outside and inside of the cavity, atmospheric pressure oscillations the geometry of the karst cave, as well as anthropic factors (Xie *et al*. 2015; Dumitru *et al*. 2016; Pérez-López *et al*. 2017). In general, caves in Atlantic areas show peak concentrations of Rn in winter and minimum values in summer, when the ventilation is usually higher (e.g., Fernandez-Cortes *et al*. 2011; Lu *et al*. 2011). Groundwater conditions control the presence of Rn differently, being higher during humid seasons. This is caused by the role of the water as an important carrier of Rn and the increment of vapour condensation in porous media in bedrocks and sediments, which reduce the air-filled porosity (Fernandez-Cortes *et al*. 2011, 2013). This is common in winter in temperate climate regions. Recent studies revealed that sediments rich in clay can control Rn diffusion from the bedrock to the cave passages (Gillmore *et al.* 2002; Gregorič *et al.* 2013). At the same time, these sediments may constitute an additional source of Rn, which may include U as a tracer (Porstendörfer 1994; Gillmore *et al.* 2000).

In spite of the complex microclimatic characteristics of cave systems, Rn measurements in karst caves have often been correlated with seismicity and crustal deformation, requiring the continuous monitoring of Rn concentration in cavities (Garavaglia *et al.* 1998; Šebela *et al.* 2010; Briestensky *et al.* 2011) as well as other gasses that may be released during tectonic activity such as $CO_2$ (e.g., Chiodini *et al.* 2011; Smeraglia *et al.* 2016). Preliminary results of these works indicate that tectonic activity in convergent settings produce higher Rn anomalies, while extensional settings do not generally seem to affect the concentration of this gas, although high values in extensional setting have been also measured (Steinitz *et al.* 2003).

In the Cantabrian Mountains (northern Iberian Peninsula), Rn concentration have been documented in dwellings (e.g., García-Talavera *et al.* 2013), groundwater (González-Díez *et al.* 2009), karst caves (see Sainz Fernández *et al.* 2017) and in two regional structures, named Sabero and León faults (Fig. 1; Künze *et al.* 2012). Both faults are relevant from a regional geological point of view and have similar accumulated displacement (~5 km). The main difference between them resides in their seismicity record, as the Ventaniella fault shows an associated low magnitude clustering of earthquakes (López-Fernández *et al.* 2004, 2018), absent in the Sabero fault. Radon concentration in soil covering the fault resulted to be four times higher in the Sabero fault (up to 441 $kBq{\cdot}m^{-3}$) with respect to the Ventaniella fault (up to 106 $kBq{\cdot}m^{-3}$), despite the former being regarded as aseismic. Motivated by this, we carried out the measurement of Rn gas in 28 karst caves with the aim of identifying the regional fracture system that facilitates the current ascent of Rn through the upper crust. For this purpose, we combine in a Geographic Information System (GIS) three essential pieces of information: Rn measures, speleological cave survey data and structural data in geological maps.

## GEOLOGICAL SETTING

### TECTONIC EVOLUTION OF THE CRUST

The Cantabrian Mountains and the Pyrenees formed in the Alpine tectonic cycle during the Paleogene collision between African and European tectonic plates, when the plate boundary was located to the north of the Iberian plate (Fig. 1A) (e.g., Srivastava *et al.* 1990; Teixell *et al.* 2018; DeFelipe *et al.* 2019). Specifically, the Cantabrian Mountains were exhumed by the reactivation of Variscan faults E-W oriented, and therefore subperpendicular to the main N-S direction of compression of the Alpine orogeny (Alonso *et al.* 1996). Fig. 1B shows the main structures of the west-central Cantabrian Mountains, highlighting the Alpine faults over previous structures. Most of these existing structures affecting the alpine basement formed during the Variscan Orogeny, mostly Carboniferous in age in the study area (Pérez-Estaún *et al.* 1991), and during extensional events from the late Permian to Cretaceous, which led to thick sedimentary basins such as the Basque-Cantabrian basin or the offshore Asturian basin (e.g., Tugend *et al.* 2015; DeFelipe *et al.* 2017, 2018, Cadenas *et al.* 2018).

The exhumation of the Cantabrian Mountains during the Alpine convergence implied a huge deformation of the crust. The main alpine structure corresponds to a S-vergent thrust system dipping gently to the N that uplifts the Cantabrian Mountains over the Duero basin (Fig. 1B) (Alonso *et al.* 1996; Gallastegui *et al.* 2016; Acevedo *et al.* 2019). The major period of exhumation took place in the Eocene and the Oligocene, producing the exhumation of a minimum of 1.7-3 km of rocks (Alonso *et al.* 1996; Martín-González *et al.* 2011; Fillon *et al.* 2016). The amount of Alpine shortening has been established in 96-98 km in the eastern Cantabrian Mountains (Pedreira *et al.* 2015; Gallastegui 2016). In terms of the amount of finite shortening and the dominant style of structures that accommodate deformation, two crustal segments in the western Cantabrian Mountains have been differentiated: an Asturian segment to the E, dominated by frontal or longitudinal thrusts; and a Galician segment to the W, characterised by conjugate strike-slip faulting (Llana-Fúnez & López-Fernández 2015). Both crustal segments show different relief evolution and crustal seismicity in agreement with the geometry of the dominant type of structures.

The main structures in the Cantabrian Mountains that play a role in the current crustal architecture are (1) reverse faults or thrusts and, (2) strike-slip faults. The main reverse faults described in the literature are E-W oriented, longitudinal to the orogenic belt, and are named as Llanera, Cabuérniga, Sabero and León (Fig. 1) (Alonso *et al.* 1996). The Llanera and Cabuérniga faults formed originally as normal faults in the Mesozoic and were overprinted during the Alpine Orogeny; while the Sabero and León faults are Variscan thrusts that were reactivated during the Alpine convergence (Alonso *et al.* 1996). The other type of structures that accommodate N-S shortening are strike-slip faults (Fig. 1B). One of these structures, the As Pontes fault, is a NW dextral strike-slip structure that uplifted the Xistral Range from the Oligocene to the Miocene (Grobe *et al.* 2014). Similar strike-slip faults formed in the Asturian segment, for instance the Ventaniella fault (Julivert 1967; Tavani 2012; Fernández-Viejo *et al.* 2014), a dextral strike-slip structure with an offset of ~5 km (Fig. 1B). Currently, low-magnitude seismic activity is associated to the Ventaniella fault (López-Fernández *et al.* 2018) that elevated an erosional flat surface along the Cantabrian coast (López-Fernández *et al.* 2020). Faults with similar orientation are significantly more abundant to the E of the Ventaniella fault (Fig. 1B). Northwesterly faults constitute the boundaries of minor Permian sedimentary basins (Lepvrier and Martínez-García 1990) while northeasterly faults coincide with the structural orientation of an aborted rift branch (Arche & López-Gómez 1996; Cadenas & Fernández-Viejo 2017).

In general, karst caves in the Asturian segment show a strong structural control since they are developed in highly fractured massifs where cave conduits usually follow bedding and up to seven families of joints, as well as their mutual intersections (Ballesteros *et al.* 2014, 2015a). The arrangement of the karst cave segments depict the structural features within the basement but develop more strongly along those fractures with higher connectivity for aqueous fluids dissolving the rock.

## RN CONCENTRATIONS IN KARST CAVES

More than 2 km of Paleozoic basement rocks involved in the Alpine uplift of the Cantabrian Mountains are limestones, which are Cambrian, Devonian or Carboniferous in age. All of these rock formations are karstified since at least the Pliocene (e.g., Ballesteros *et al.* 2019). Recent systematic studies of Rn concentration in dwellings in NW of Spain, within the study area, indicate relative local high concentrations of Rn (> 0.3 kBq·m$^{-3}$) that cannot be directly related to the lithology exposed at the surface (García-Talavera *et al.* 2013; Sainz Fernández *et al.* 2017). Recent studies in tourist karst caves in the Cantabrian Mountains measured frequently average concentrations

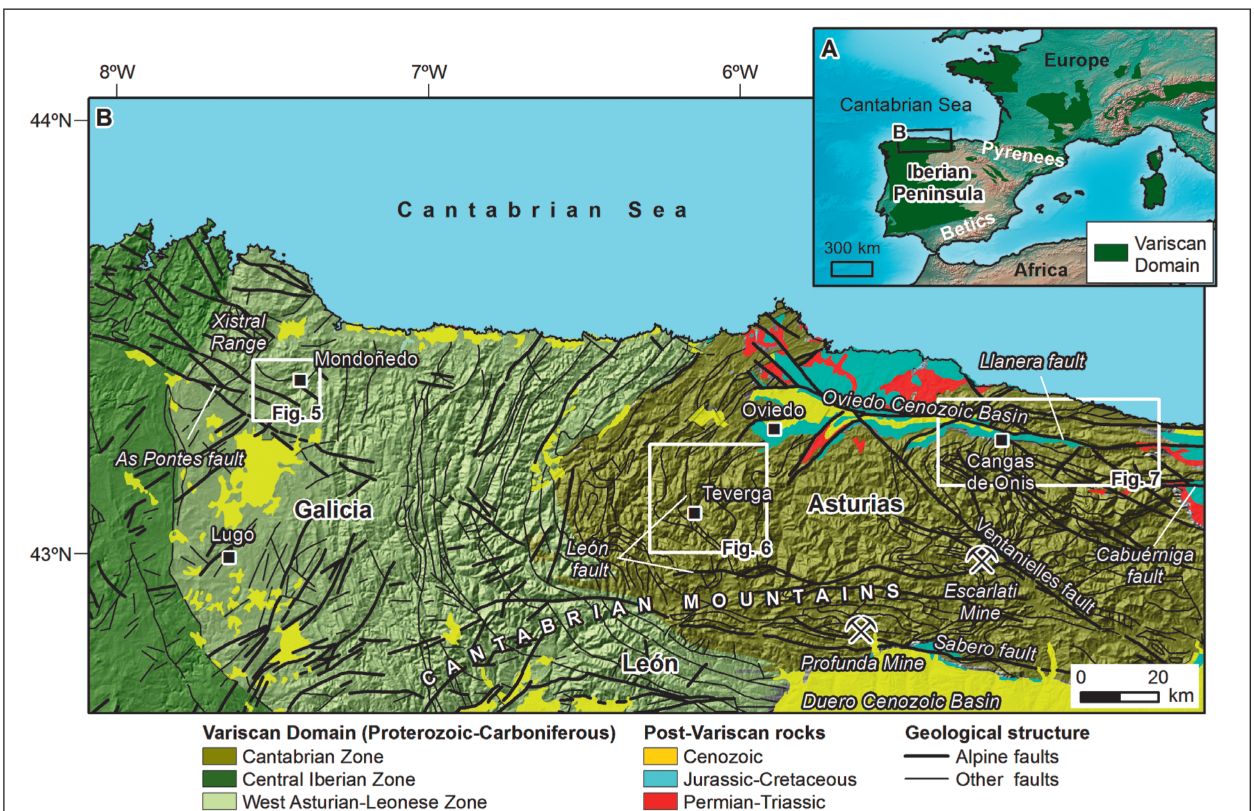


*Fig. 1: (A) Location of the Cantabrian Mountains in the N of the Iberian Peninsula. (B) Synthetic geological map of the Eastern and central Cantabrian Mountains. Geology was extracted from the Spanish Geological Survey continuous geological maps (Rodríguez Fernández et al. 2015). The seismic events up to 4.1 moment magnitude were recorded between 1984 and 2014 by the Spanish Geological Survey (Instituto Geográfico Nacional, www.ign.es).*

of 0.1-1.5 kBq·m$^{-3}$ during annual campaigns, although through continuous monitoring natural peaks are up to 7.1 kBq·m$^{-3}$, and artificial peaks to 15.9 kBq·m$^{-3}$ due to the anthropic closure of the cavities (Hoyos *et al.* 1998; Poncela *et al.* 2004; Lario *et al.* 2005; Sainz *et al.* 2007). The presence of Rn in groundwater was also recognized as significant, generally related to thermal springs and spas since González-Díez *et al.* (2009) reported Rn concentrations in groundwater up to 0.8 kBq·m$^{-3}$, without a clear relationship with the lithology or tectonic structure in the bedrock exposed at the surface.

## METHODS AND STUDIED CAVES

The work method includes: (1) the measurement of Rn concentration in karst caves air; (2) the analyses of the main directions of the caves and their relation to tectonic and other geological structures, and (3) the projection of the cave topography on a geographical information system (GIS) in order to establish relationships between Rn concentration, cave direction and the geological tectonic structure.

Twenty-eight caves were selected for this study in the Cantabrian Mountains, divided into three areas: Mondoñedo, Teverga and Cangas de Onís (Fig. 1B). The areas were selected according to the presence of key regional structures. The first area is located in the Galician segment of the Cantabrian Mountains and comprises four caves to the SE of Mondoñedo (Fig. 1B). These cavities were developed in metamorphosed Cambrian limestone affected by minor faults with a NW-SE direction. The cavities are located at the southeastern lateral termination of the As Pontes fault (Santanach *et al.* 2005). The Teverga area is located to the SW of Oviedo (Asturias) and includes five cavities developed in Carboniferous limestone, some of them nearby the León fault (Fernández *et al.* 2018). The Cangas de Onís area is located to the E of the Oviedo Cenozoic Basin, where twenty-two caves were studied. The basin shows an elongated shape in the E-W direction, controlled by the Llanera fault to the N, which elevates the northern block. The Llanera fault is cut across by the Ventaniella fault with a northwesterly trend (Alonso *et al.* 1996; Pulgar *et al.* 1999).

Rn concentration was measured using CR-39 detectors following Sainz *et al.* (2007), Somlai *et al.* (2011), Dumitru *et al.* (2016) and Smith *et al.* (2019). These detectors were fastened under the cap of a cylindrical polypropylene container that prevents Rn decay products and $^{220}$Rn from entering. One detector was placed inside each cavity, except in the Rei Cintolo cave (two detectors), Güerta cave (five detectors) and Vegalonga cave (two detectors) because they are kilometric caves. The location of the detector within the cavity followed three criteria: ~5 m from the cave entrance, cave passages should have little or absent clay deposits, relative dry walls and lack evidences of water condensations such as water drips or moonmilk.

The detectors were recording from winter to autumn 2016, covering dry and wet periods. The accumulated radiation by alpha particles from $^{222}$Rn was determined by counting the tracks produced in track-etched detectors (Poncela *et al.* 2004; Sainz *et al.* 2007) in the Laboratory of Environmental Radioactivity of the University of Cantabria (Spain), accredited by the Spanish National Entity (ENAC, ISO/IEC 17025). The error of the measurements was 6-9% and the limit of detection was 6 Bq·m$^{-3}$. Rn concentration was calculated from the ratio of the accumulated radiation by the exposition time.

The main directions for the twenty-eight caves were calculated using rose-diagrams, extracting lengths and orientation of galleries from cave surveys. The speleological groups GE Polifemo, GES Montañeiros Celtas, SIS CE Terrassa, GE Diañu Burlón, GE Gorfolí, SEB Escar and CADE-FESPA provided the original survey data for twelve cavities. Another eight caves were surveyed using the DistoX laser distanciometer and restored the survey data of eight cavities (Favre 1978; L'Esperteyu Cavernícola-Espéleo Club 1987; Puch 1998) following Ballesteros *et al.* (2014).

The orientation of cave entrances and their conduits extracted from the geological map were projected in a rose diagram to establish the link between Rn concentrations, cave main directions and the geological structure. Cave speleological surveys and Rn concentration values were later introduced in ArcGIS 10, together with the digital and continuous geological map GEODE carried out by the Geological Survey of Spain (compiled by González Menéndez *et al.* 2008 in the West-Asturian Leonese Zone and by Merino-Tomé *et al.* 2013 in the Cantabrian Zone). The spatial analysis in GIS allowed us to establish the correlation between concentration values and the orientation of major dominant discontinuities for each measurement locality, and to calculate the distance between each cave to the nearest fault. The relationship of these parameters was inferred using polynomical equations of second degree.

*Tab. 1: Rn concentrations in cave air (measured from winter to autumn 2016), cave geometry (length, vertical range, verticality index and main direction), relative cave ventilation and the distance between the cavities and the northwesterly trending faults (the distance is zero if the faults intercept the cave). The vertical index is the quotient between the vertical range and the cave length (Piccini 2011). Rn concentration values in Arenas and Les Escobes caves are below the detection limit. Determination of relative cave ventilation is qualitative and based on the number of entrances, vertical range of the cave and between entrances, and air flux (mainly inhalant or exhalant during measuring, wind velocity).*

| Study cave | Rn concentration ($Bq \cdot m^{-3}$) | Cave geometry | | | | | Relative cave ventilation | Distance (m) to faults with N-S and NE-SW trending |
|---|---|---|---|---|---|---|---|---|
| | | Length (m) | Vertical range (m) | Verticality index | Main direction (º) | Number of known entrances | | |
| Alda | 1155 ± 82 | 394 | 28 | 0.07 | 175 | 1 | Low | 350 |
| Arangas | 156 ± 17 | 42 | 4 | 0.10 | 77 | 1 | Low | 300 |
| Arenas | <21 | 27 | 4 | 0.15 | 104 | 3 | High | 300 |
| Berdayes | 825 ± 58 | 288 | 15 | 0.05 | 25 | 1 | Low | 50 |
| Canes | 1344 ± 83 | 837 | 71 | 0.08 | 163 | 1 | Low | 80 |
| Carmona | 1709 ± 118 | 15 | 14 | 0.93 | 34 | 1 | Low | 100 |
| Collía | 3816 ± 262 | 268 | 6 | 0.02 | 5 | 1 | Low | 1050 |
| Cuerres | 315 ± 25 | 107 | 22 | 0.21 | 105 | 1 | High | 1800 |
| Cave X | 882 ± 62 | 1810 | 60 | 0.03 | 60 | 1 | Low | 300 |
| Güerta 1 (N-S gallery) | 1539 ± 105 | >23350 | 281 | 0.01 | 165 | 2 | High | 0 |
| Güerta 2 (E-W gallery) | 1109 ± 78 | | | | | | | |
| Güerta 3 (N-S gallery) | 1610 ± 110 | | | | | | | |
| Güerta 4 (N-S gallery) | 1616 ± 112 | | | | | | | |
| Güerta 5 (N-S gallery) | 1628 ± 113 | | | | | | | |
| Güeyos del Riu | 1022 ± 70 | 10 | 2 | 0.20 | 160 | 1 | Low | 50 |
| La Peruyal | 2429 ± 167 | 294 | 33 | 0.11 | 95 | 1 | Low | 40 |
| La Porquera | 155 ± 14 | 365 | 15 | 0.04 | 125 | 1 | Low | 800 |
| La Trapa | 397 ± 28 | >1000 | 100 | 0.10 | 35 | 1 | Low | 250 |
| Les Cámares | 320 ± 26 | 89 | 8 | 0.09 | 135 | 1 | Low | 450 |
| Les Escobes | <13 | 409 | 15 | 0.04 | 95 | 1 | High | 300 |
| Les Paraes | 239 ± 20 | 16 | 7 | 0.44 | 145 | 1 | Low | 850 |
| Lobos | 161 ± 14 | 170 | 22 | 0.13 | 35 | 1 | Low | 550 |
| Los Covazones | 49 ± 9 | 4219 | 171 | 0.04 | 45 | 1 | High | 860 |
| Marabio | 27 ± 6 | 10 | 1 | 0.10 | 90 | 1 | High | 1400 |
| Obar | 2636 ± 181 | 3563 | 197 | 0.06 | 74 | 1 | Low | 0 |
| Osu | 87 ± 11 | 3600 | 220 | 0.06 | 120 | 3 | High | 500 |
| Peches | 1741 ± 122 | 32 | 12 | 0.38 | 71 | 1 | Low | 150 |
| Pedraces | 332 ± 27 | 5 | 0 | 0.00 | 85 | 1 | Low | 400 |
| Rei Cintolo 1 | 1830 ± 126 | >4000 | 100 | 0.03 | 60 | 1 | Low | 150 |
| Rei Cintolo 2 | 1914 ± 131 | | | | | | | |
| Santos | 1832 ± 125 | 40 | 16 | 0.40 | 150 | 1 | Low | 350 |
| Vegalonga 1 | 221 ± 21 | 5900 | 212 | 0.04 | 45 | 2 | High | 600 |
| Vegalonga 2 | 46 ± 10 | | | | | | | |
| Vistulaz | 136 ± 12 | 3050 | 85 | 0.03 | 30 | 1 | High | 1600 |

## RESULTS

The main characteristics of the selected twenty-eight caves and the results of average Rn concentration over the length of the study are listed in Tab. 1. This table shows Rn concentrations in sixteen caves to be lower than 500 Bq·m$^{-3}$, the air of ten cavities shows values between 825-1914 Bq·m$^{-3}$, and in three caves the Rn content exceeds 2400 Bq·m$^{-3}$. Concentrations below 13-21 Bq·m$^{-3}$, the resolution limit of the technique, were found in two caves.

The general ventilation in twenty caves is relatively low since the caves are mainly formed by horizontal galleries, their vertical range is less than 100 m, cave streams are absent, and the cavities have only one reported entrance. In relative terms, these caves represent natural traps with relative poor ventilation. In contrast, there are eight caves in which the ventilation is relatively high. In four of them, the Les Escobes, Covazones and Marabio caves, the air flow is mainly towards the inside of the cavity, thus tending to reduce the concentration of Rn gas. The entrances of the other four caves, Güerta, Osu, Veigalonga, Vistulaz caves, are usually exhalant, with air flow velocities that can exceed 4 m·s$^{-1}$. Accordding to these results, the cave ventilation can not explain the Rn measures carried out in caves. In all limestone massifs, whether Cambrian in the Mondoñedo area or Carboniferous in age in the remaining areas, the U content is almost irrelevant (González-Díez *et al*. 2009) and the emanation power of the hosting rocks is relatively low,

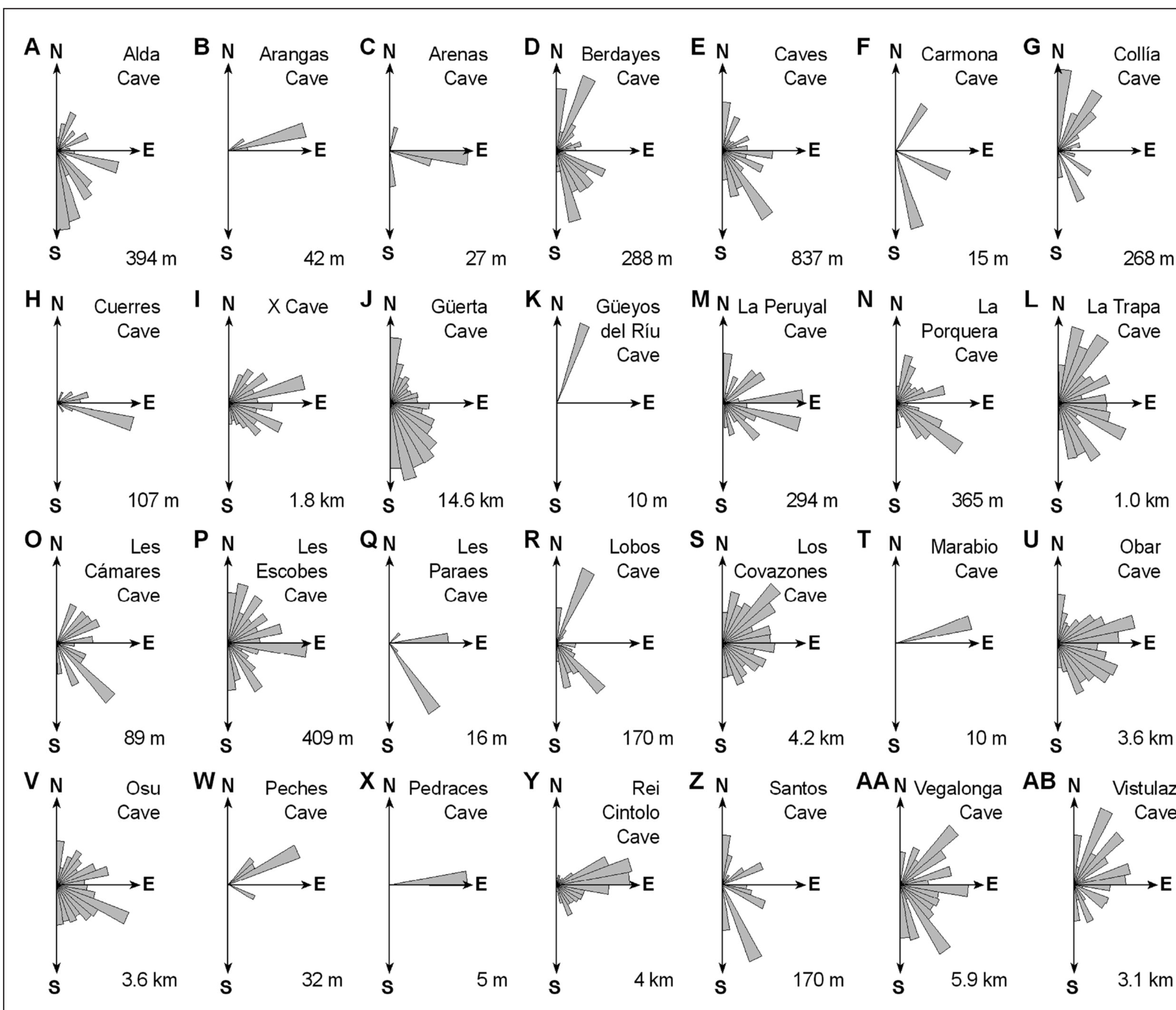


*Fig. 2: Rose diagrams for each cave segment showing the main direction of cavity segments (see Tab. 1). The full length of the caves is also indicated.*

estimated in 70.1 ± 2.0 Bq·m$^{-3}$ (García-Talavera *et al.* 2013). The contribution to Rn concentration in cavities by the host rock can be regarded as relatively low.

The studied caves range in length from 5 m to 23 km of conduits, being the vertical range up to 281 m (Tab. 1). The orientation of cavity segments in a rose diagram shows their main orientations to be uniformly distributed from N0º-180ºE (Fig. 2). In general, Rn levels are higher than 500 Bq·m$^{-3}$ in caves orientated following the directions N150º-180ºE, although an increment in this gas is also detected in caves with the N0º-30ºE main direction (Fig. 3). The exceptions to this are the caves of La Peruyal, Obar, Peches, Rei Cintolo and Cave X, with Rn concentrations from 882 to 2636 Bq·m$^{-3}$. Such high values are related to the presence of nearby faults, as described in the following paragraph. Besides, the five detectors installed in the Güerta Cave indicate that the Rn content is 1.5 times higher in galleries with N-S direction (1539-1628 Bq·m$^{-3}$) than in conduits with a W-E direction (1109 ± 78 Bq·m$^{-3}$).

The Rn concentration data shows a dependence on the distance between the caves and faults with NW-SE orientation, except the case of Güerta Cave that intercepts a N-S trending fault (Fig. 4) (Llana-Fúnez and Ballesteros 2020). The concentration is greater than 825 Bq·m$^{-3}$ in cavities located less than 100±50 m from a fault, and it is lower than 332 Bq·m$^{-3}$ in caves more than ca. 400 m away from a cartographic fault. In the latter group of cavities, the Rn concentration approaches the regional background values in the study area. Despite the general trend described, the Collía Cave presents the largest measured Rn concentration (3816 Bq·m$^{-3}$) in spite of its location 1050 m away from a NW-SE orientated fault.

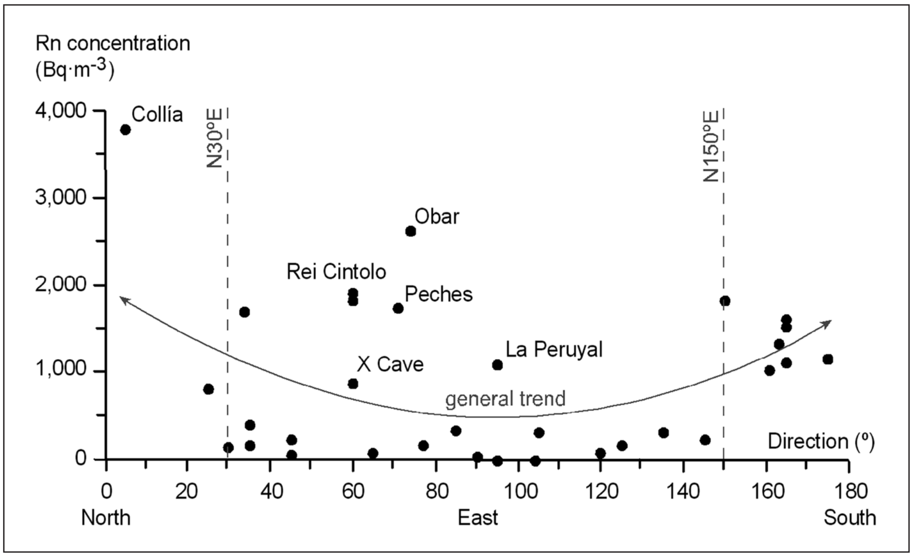


*Fig. 3: Rn concentration with respect to the dominant orientation for each cave (data detailed in Tab 1).*

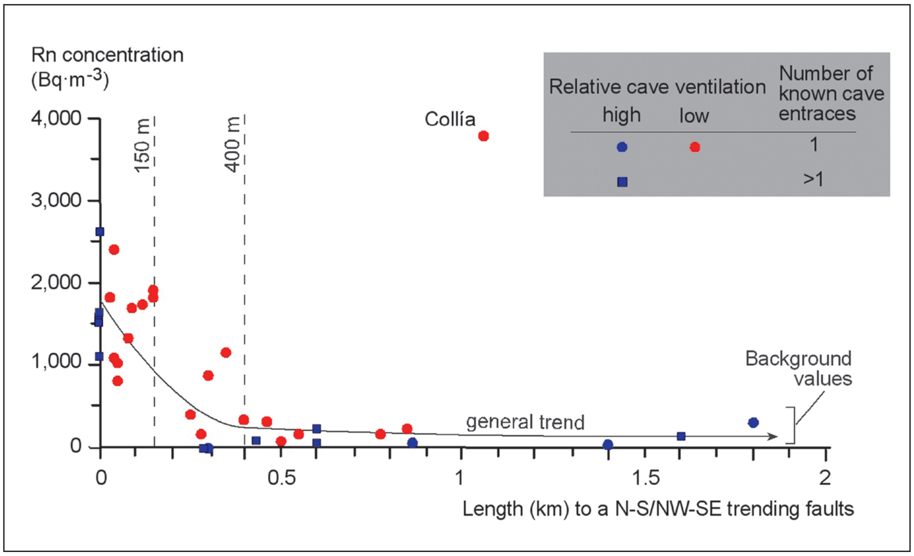


*Fig. 4: Rn concentration in cave air with respect to distance to NE-SW trending faults (N-S directions in the case of the Güerta Cave). Further details in Tab. 1.*

# DISCUSSION

## GEOLOGICAL INTERPRETATION OF THE RN ANOMALIES

The concentration of Rn in air within karst caves in the cavities selected shows a clear geometric relation to the orientation of cartographic faults. In the Mondoñedo area (Fig. 5), the high values of Rn concentration (1.8-1.9 kBq·m$^{-3}$) identified in the Rei Cintolo and Santos caves are related to northwesterly faults. In fact, the small fractures depicted in Fig. 5 represent the lateral termination of the As Pontes fault, which is associated with the rise of the Xistral Range in northern Galicia (Grobe *et al.* 2014).

In the Teverga area (Fig. 6), the Güerta Cave (>23.3 km long), following partly the León fault, shows the highest Rn concentrations in the area (1.1-1.6 kBq·m$^{-3}$) despite the relatively high ventilation in the cave. The León fault formed during the Variscan Orogeny, but parts of this major structure were likely reactivated during the Alpine Orogeny (Alonso *et al.* 2007, 2009; Fernández *et al.* 2018). The high Rn values associated with this structure indicate that the fault zone remains an effective pathway to the ascent of fluids through the crust.

The major fault in the Cangas de Onís area is the Llanera fault, trending W-E (Fig. 7) (Alonso *et al.* 1996). In general, the caves located near this fault have low Rn concentration, with the exception of the Gueyos del Río and Carmona caves with 1.0-1.7 kBq·m$^{-3}$. Our results show that the highest concentrations of Rn, above 1.1 kBq·m$^{-3}$, in the Alda, Canes, Carmona, Colía, Obar, Peches, and Peruyal caves are associated with minor faults trending NW, very likely overprinting the Llanera fault system. The high values recorded in the Alda Cave (1.1 kBq·m$^{-3}$) can be related to the nearby Cu ore depos-

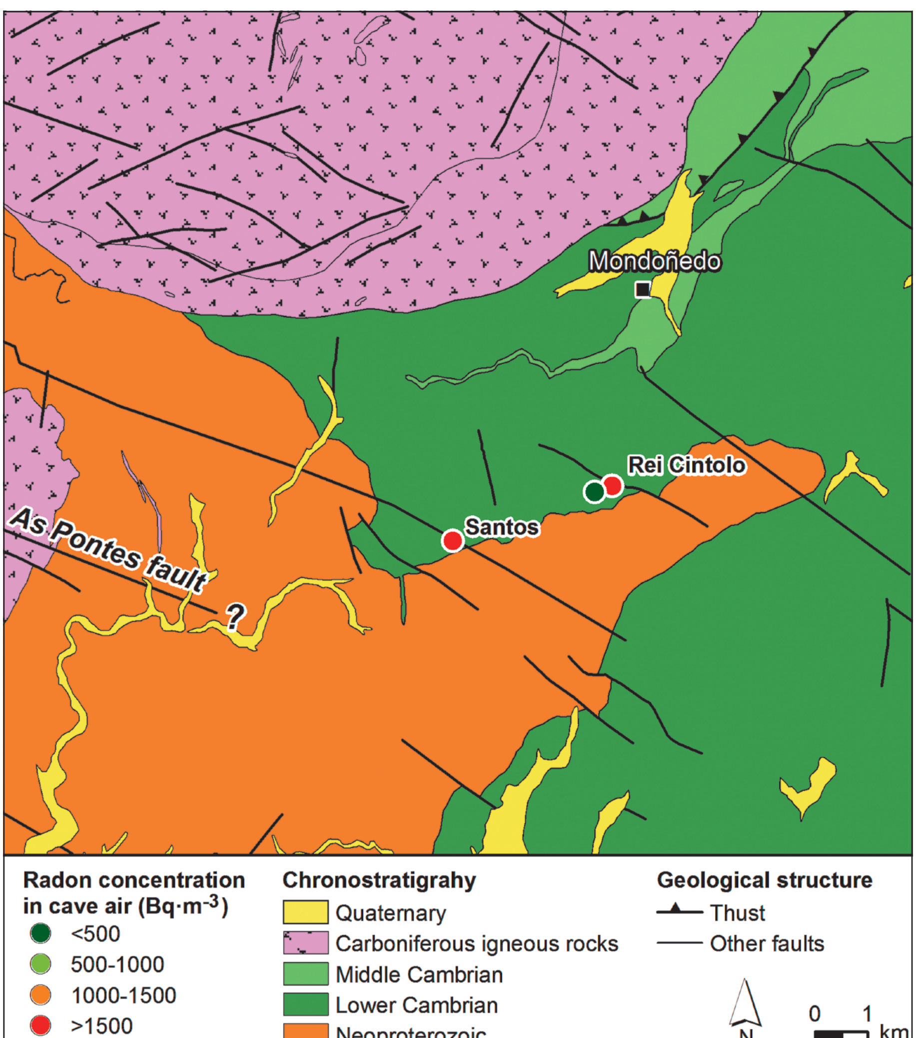


*Fig. 5: Geological map of the Mondoñedo area (geology from González Menéndez et al. 2008). The location of Rn analyses in the three caves studied is indicated with dots, coloured according to concentration.*

its (Rodríguez-Terente *et al.* 2006), while the concentrations of 2.6 kBq·m$^{-3}$ in Obar cave-spring may be related to thermal springs (Ballesteros *et al.* 2015b). The highest Rn concentrations in the area were measured in the Collía Cave with 3.8 kBq·m$^{-3}$, located 3.6 km away from a cartographic tectonic structure. However, this cave is located in the lateral prolongation of the trace of a northwesterly fault, thus the Rn anomaly may suggest that the fracture system extends laterally beyond the cartographic structure as shown in geological maps (Fig. 7).

The correlation between Rn concentration and faults oriented NW-SE indicates that the permeability at depth for Rn flow in these fault systems is higher than in other fault systems. These structures overprint all earlier structures, as evidenced in geological maps (e.g., Alonso *et al.* 1996). For these structures to remain relatively open, and in the absence of criteria evidencing current tectonic activity, no sealing has taken place since the last tectonic event. According to the current location of the plate boundary in the Iberian Peninsula, in the S of the peninsula, no tectonic activity is expected in these set of structures since it migrated in the Miocene from N to S (Teixell *et al.* 2018). However, these set of fractures remain an effective pathway communicating a source of Rn at depth with the surface. This agrees with the reported presence of Rn anomalies in aseismic scenarios (Ghosh *et al.* 2011; Woith 2015). It also suggests that reported Rn anomalies in dwellings in the study area may be related to the presence of similar structures oozing Rn to near surface geological formations, which can be either recent sediments or soils covering tectonically fractured bedrock.

## POTENTIAL RN SOURCES

Despite of the emanation of Rn from the surface exposed rocks is minoritary, their contribution should be evalu-

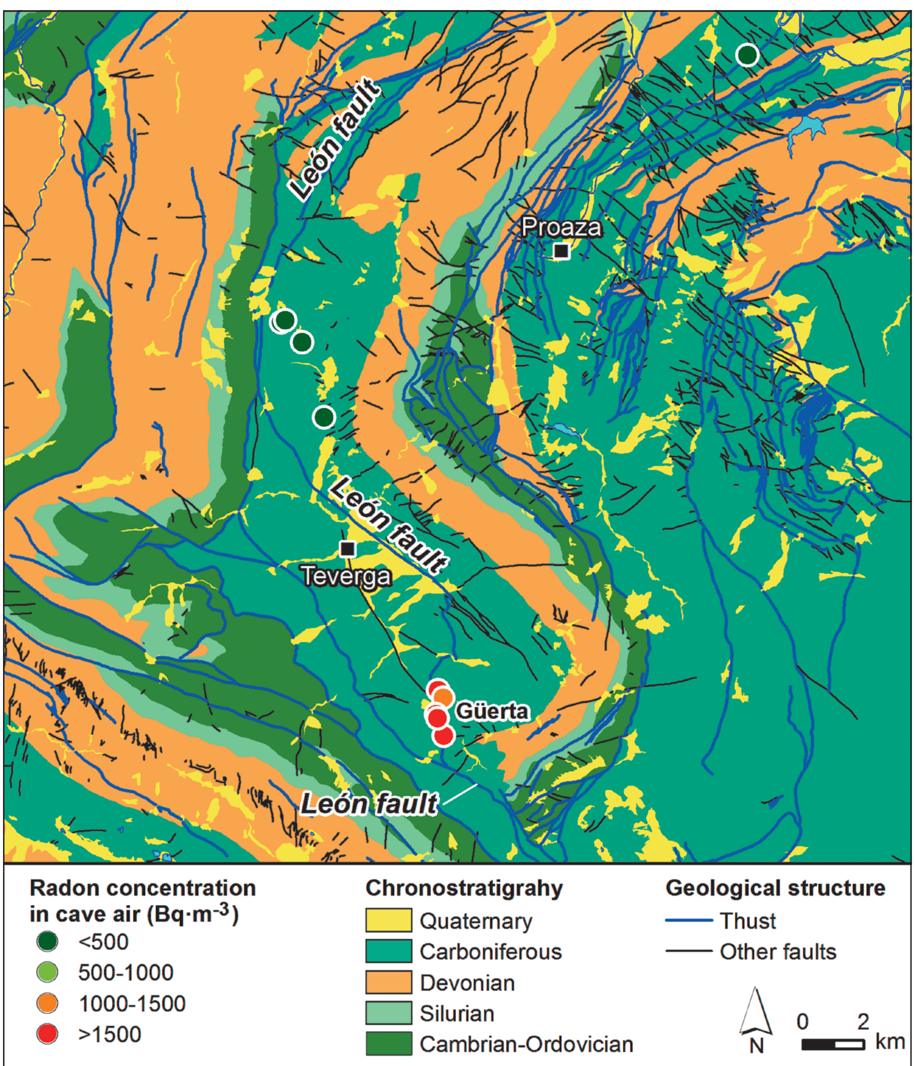


*Fig. 6: Geological map of the Teverga area (geology from Merino-Tomé et al. 2013). Based on geometric criteria, Variscan thrusts are shown in blue, while later fractures (very likely Alpine in age) are indicated in black. The trace of the León fault is according to Alonso et al. (2009). The location of nine analysis spots for Rn measurements in four caves are indicated with dots, coloured according to concentration (details in Tab. 1).*

ated. The specific mineral sources for Rn gas in the study area are yet unclear since the presence of igneous rocks and uranium-rich minerals in the study areas is generally low (Corretge & Suarez 1990), in fact, none of them associated to the caves studied. Many porphyritic subvolcanic and diabasic intrusions with associated hydrothermal systems were originated in the study area following northwesterly faults (Suárez *et al.* 1993, 1999; Fernández-Suárez *et al.* 1998; Gallastegui *et al.* 2001; Ballesteros *et al.* 2011). The few plutonic rocks reported in the Cantabrian Mountains have relatively low uranium, between 0.2 and 8 ppm, reaching up to 11.1 ppm in acid rocks (Cepedal *et al.* 2013; Martínez-Abad *et al.* 2015; and unpublished data courtesy of the Petrogenesis Group of the University of Oviedo).

Northwesterly faults are in most cases polyphasic and polyorogenic in the study areas, particularly those of large entity. Close to the coast, faults with such orientation bound small Permian sedimentary basins (Martínez-García *et al.* 1991), and to the E of the study area, towards the Vasco-Cantabrian basin, Espina *et al.* (1994) reported a similar relation to Permian sedimentary rocks. In either case, the faults formed as normal extensional faults. There are two separate sets of intrusions reported in the W of the Asturian segment: at 285 Ma porphyritic subvolcanic intrusives, with an associated hydrothermal system, and at 255 Ma a generation of diabasic dykes that also have an associated hydrothermal system (Martín-Izard *et al.* 2000). Structures with NW orientation have significant ore deposits associated with hydrothermal fluids, the ages of the majority of the intrusions and associated ore bodies yield Permian ages (Martín-Izard *et al.* 2000; Martínez-García *et al.* 2004).

Overall, Permian normal faults have been interpreted to form in relation to an aborted rift branch from the Tethys (Arche & López-Gómez 1996). To the E of the Asturias segment, there are several ore bodies that have been associated with Permian hydrothermal systems (Martínez-García *et al.* 2004). In the Escarlati deposit (Fig. 1), veins rich in Sb-Hg found with a northwesterly orientation, are interpreted in relation to an N-S shortening and have also been related to Permian hydrothermalism (Martín-Izard *et al.* 2009). There is one documented ore deposit with uranium minerals exposed at the surface: the Profunda Mine (Fig. 1). This deposit is characterised by Cu-Ni-Co-U-As-S mineralisations (Paniagua *et al.* 1987). Although there are no reported cartographic northwesterly faults in relation to the Profunda Mine, mineralizing fluids precipitated along veins oriented following NW-SE. Yet, there is no indication of any other hydrothermally formed deposit of regional occurrence showing similar mineralogy and a potential candidate for the source of Rn gas at each of the caves studied. This fact supports that the Rn sources are related to the depth crust, connected to the topographic surface via fault zones.

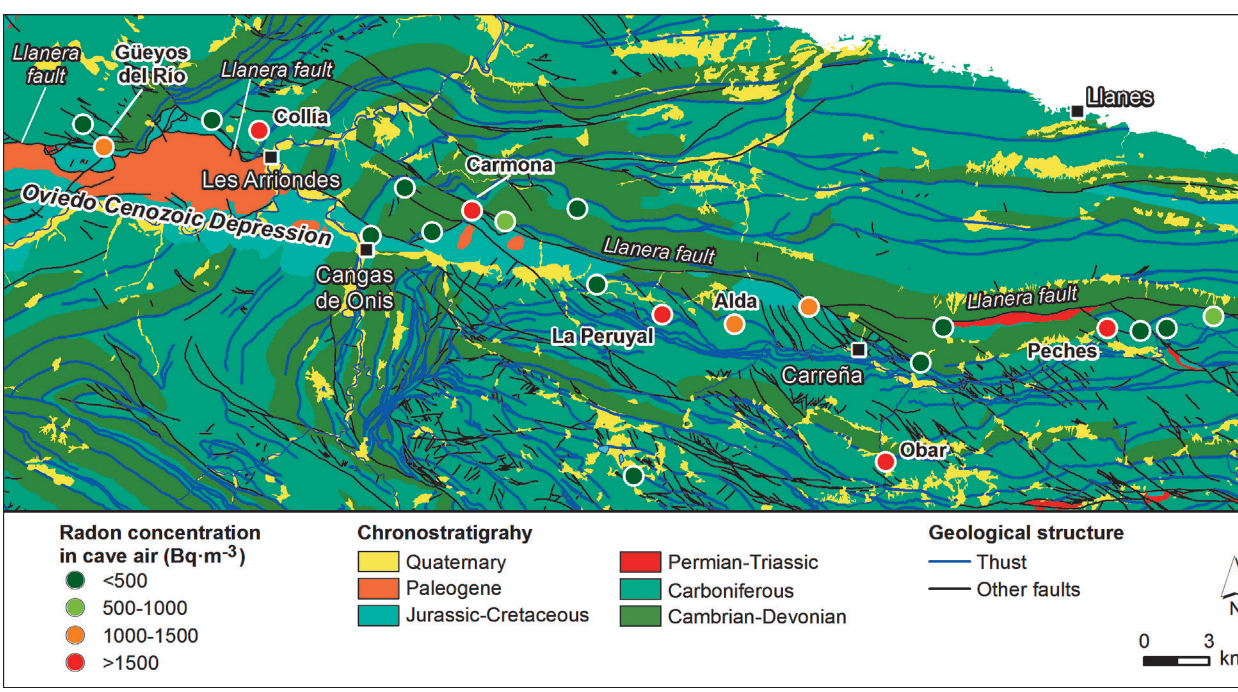


*Fig. 7: Geological map of the Cangas de Onís area (geological map based in the compilation by Merino-Tomé et al. 2013). The trace of the Llanera fault is highlighted. The location of Rn concentrations in 22 caves are indicated and colour coded according to concentration (details in Tab. 1).*

## CONCLUSIONS

We present thirty-four measurements of Rn concentration in air in twenty-eight karst caves from the Cantabrian Mountains. We show that cavity conduits parallel to NW-SE trending subvertical faults have higher content in Rn (>0.5 kBq·m$^{-3}$) than caves more than 100±50 m away from faults from that particular system. Consequently, the permeability for the NW-SE trending fault is higher than other faults systems present, indicating that the northwesterly structures remain relatively open. This suggests that NW-SE trending faults are related to the last tectonic event in the western and central part of the Cantabrian Mountains, as they often overprint earlier Alpine structures.

The dataset presented in this contribution provides a robust correlation among Rn concentration measurements, cave survey data (length, orientation and vertical range) and fault orientation data. The methodology was applied successfully in a region with Atlantic climate, with minor intraplate seismicity but a very complex tectonic history due to the presence of structures formed and overprinted by the superposition of two orogenies, Variscan and Alpine, separated by several extensional events during the Permian and Mesozoic. Despite the intrinsic tectonic complexity, the results of this work support the use of Rn in karst caves as an indicator of fault activity as they help identifying those with higher permeability.

## ACKNOWLEDGMENT

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