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Preliminary results on $^3He/^4He$ isotopic ratios in terrestrial fluids from Iberian peninsula: seismoctectonic and neotectonic implications

Resultados preliminares sobre relaciones isotopicas $^3He/^4He$ en fluidos terrestres de la Peninsula Iberica: Implicaciones seismotectonicas y neotectonicas

N. M. Pérez (*,), S. Nakai (*), H. Wakita (*), J.F. Albert-Bertrán (***), and R. Redondo (**,***)**

(*) Laboratory for Earthquake Chemistry THE UNIVERSITY OF TOKYO Bunkyo-ku, Tokyo 113, Japan
(**) TERRANOSTRA Research Institute P.O. Box 225, 38400 Puerto de la Cruz, Tenerife Canary Islands, Spain
(***) Laboratorio de Isotopos estables, S.I.D.I. UNIVERSIDAD AUTONOMA DE MADRID Ciudad Universitaria de Canto Blanco, 28049 Madrid, Spain
(****) Font Vella, S.A., Avda. Diagonal 615, 5º- 08028 Barcelona, Spain

RESUMEN

La sistemática de isotópos de helio está siendo aplicada en la península Ibérica con el fin de obtener información adiccional sobre las características neotectónicas y sismotectónicas del área de estudio. Resultados preliminares sobre la distribución geográfica de las relaciones isotópicas $^3He/^4He$ en fluidos terrestres reflejan variaciones de los niveles de emisión de helio-3 significativas, y estas están intrinsecamente ligadas a las características sismotectónicas y neotectónicas de la península Ibérica.

Key words: *Rare Gases, Isotopes, Iberian peninsula, Seismotectonicas, Neotectonicas*

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Introduction

Helium isotopic composition in natural systems varies by more than three orders of magnitude. The decay series of U and Th are mainly responsible for the production of ^4He in terrestrial fluids while ^3He is primarily a tracer of mantelic component (Clarke *et al.*, 1969, Mamyrim *et al.*, 1969). Helium isotope signature from MORB sources has a well constrain value of 8±1 Ra, being Ra (helium isotope atmospheric ratio) = 1.4×10^{-6} (Craig and Lupton, 1976; Kurz and Jenkins, 1981; Lupton, 1983). In the case fluids escaping from arc volcanic environments, $^3He/^4He$ ratios range from 6-8 Ra reflecting a helium magmatic source from the upper mantle (Craig *et al.*, 1978; Sano and Wakita, 1985; Poreda and Craig, 1989). Higher helium isotope signatures has been characterized from mantle plume regions such as Hawaii, Iceland, Yellowstone, Reunion, and the Canary Islands where $^3He/^4He$ ratios range from 9 to 30 Ra indicating a helium degassing source from the lower mantle (Craig *et al.*, 1978; Kaneoka and Takaoka, 1980; Sano *et al.*, 1985; Marty *et al.*, 1993, Pérez *et al.*, 1994). On the contrary, the isotopic signature of helium from crustal fluids is around 0.02 Ra (Sano and Wakita, 1985; Oxburgh *et al.*, 1986; Hiyagon and Kennedy, 1992).

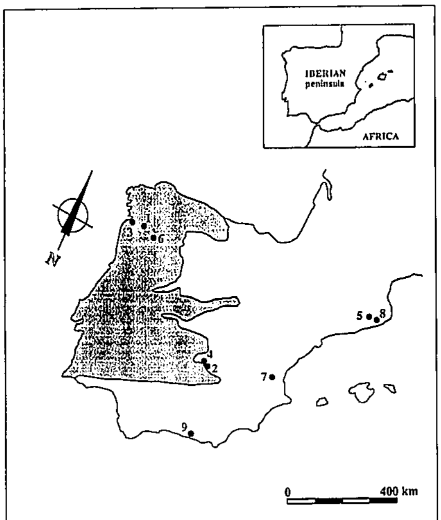

Fig.1 Sampling sites location in the Iberian peninsula.

Fig. 1.- Localización de los puntos de toma de muestra en la peninsula Ibérica

Spatial variations of $^3He/^4He$ ratios and ^3He flux can provide important and additional insight about regional tectonic environments (Sano and Wakita, 1985; Sano *et al.*, 1987; Marty *et al.*, 1992). Stable areas should be mainly characterized by ^4He generated by radioactive decay processes within the continental crust while zones of active extension or young volcanism show everywhere mantle-derived helium. Therefore, migration of deep-mantelic fluids towards the surface environment in these active areas is mainly controlled by neotectonics. The aim of this study is to evaluate spatial variations of $^3He/^4He$ ratios in terrestrial fluids from the Iberian peninsula to search for additional insights about its complex neotectonics framework.

Sampling and analytical procedures

Sampling sites are shown in Figure 1. Gas samples were collected in lead-glass containers fitted with high-vacuum stopcocks at both ends during 1995 and 1996. In the laboratory, a sample of about 0.5 cm^3 STP was admitted to the purification

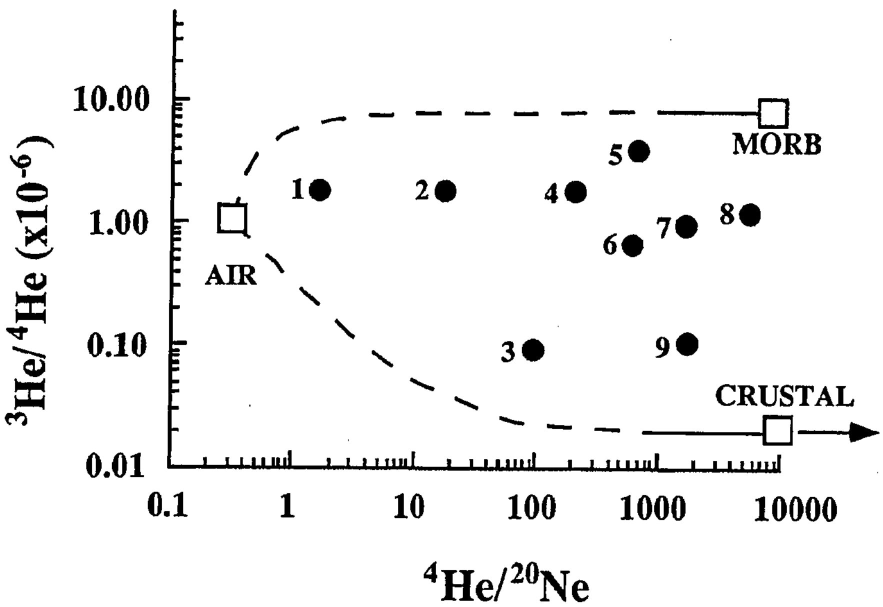

Fig.2 Correlation between the $^3He/^4He$ and $^4He/^{20}Ne$ ratios in terrestrial fluids from Iberian peninsula. Dashed lines represent the mixing lines between MORB-type He, atmospheric He and crustal-type He.

Fig. 2.- Relación entre las relaciones isotópicas $^3He/^4He$ y $^4He/^{20}Ne$ en fluidos terrestres de la peninsula Ubérica. Las lineas discontinuas reflejan lineas de mezcla entre He-MORB, Hc-atmosferico y Hc-cortical

vacuum line. Purification and separation of helium and neon from other components such as water, carbon dioxide, and nitrogen were carried out by using a hot Ti-Zr getter held at 550°C, a SAES getter at room temperature, and three charcoal traps held a 77K. The $^4He/^{20}Ne$ ratios were measured by a quadropole mass spectrometer (QMG 112, Blazers) using atmospheric air as standard. Errors for $^4He/^{20}Ne$ ratio are estimated about 10%. Helium was completely separated from neon by a cryogenic charcoal trap held at 40K and transfer to a high-precision gas mass spectrometer (VG5400, VG Isotopes). The helium isotope measurements were carried out under static conditions. Ion beams of ^3He and ^4He were detected simultaneously with a double collector system. Resolving power of about 550 at 5% peak height was attained for complete separation of $^3He^+$ from those of H_3^+ and HD^+. Atmospheric helium was used as a running standard. Uncertainty for R/Ra ratios is about 1%. All helium measurements were carried out at the Laboratory for Earthquake Chemistry of the University of Tokyo. A detailed procedure of the helium isotopic ratio measurement has been described elsewhere (Sano and Wakita, 1988)

Sple.	Location	Type	T (°C)	$^3He/^4He$ (x 10^{-6})	$(^3He/^4He)_{obs}$ (Ra)	$^4He/^{20}Ne$	$(^3He/^4He)_{cor}$ (Ra)	Date
1	Melgaço	CO_2	15	2.604±0.094	1.86	1.6	2.07	09/95
2	Calatrava	CO_2	14.5	2.534±0.020	1.81	17.3	1.82	05/93
3	Caldas de Tui	H_2S	49	0.128±0.006	0.09	96.7	0.09	09/95
4	Calatrava 1	CO_2	17	2.534±0.02	1.81	21.7	1.81	05/93
5	Amer	CO_2	15	5.446±0.050	3.89	683.8	3.90	09/95
6	Cabreiroa	CO_2	19.2	0.962±0.012	0.69	622.5	0.69	09/95
7	Cofrentes	CO_2	14	1.338±0.012	0.95	1650	0.95	01/96
8	Gerona-2	CO_2	36.8	1.736±0.020	1.24	5500	1.24	01/96
9	Lanjarón	CO_2	-	0.147±0.030	0.10	1780	0.10	01/96

CO_2: rich-CO_2 fluids; H_2S: rich-H_2S fluids

Table1.- Observed $^3He/^4He$, $^4He/^{20}Ne$, and corrected $^3He/^4He$ ratios of terrestrial fluids from Iberian peninsula

Tabla 1.- Relaciones isotópicas $^3He/^4He$ observadas, $^4He/^{20}Nc$ y $^3He/^4He$ corregidas en los fluidos terrestres de la Peninsula Ibérica

Results and discussion

Observed $^3He/^4He$ and $^4He/^{20}Ne$ ratios from terrestrial fluids of the Iberian peninsula are listed in Table 1. Errors related to $^3He/^4He$ measurements are one standard deviation. The $^3He/^4He$ and $^4He/^{20}Ne$ ratios vary from 3.89 Ra to 0.09 Ra, and from 1.6 to 5500, respectively.

The observed $^3He/^4He$ ratios of the gas samples are plotted against $^4He/^{20}Ne$ ratios (Fig.2). This distribution of all data in this diagram can be generally explained by a three component mixing: mantle-derived He, radiogenic He, and atmospheric He. Based on a simple mixing equation presented by Sano and Wakita (1985) it is possible to estimate the helium's fractions from each geochemical reservoir using the following equations:

$$(^3He/^4He)_s = (^3He/^4He)_a \times A + (^3He/^4He)_m \times M + (^3He/^4He)_c \times C \qquad (1)$$

$$1/(^4He/^{20}Ne)_s = A/(^4He/^{20}Ne)_a + M/(^4He/^{20}Ne)_m + C/(^4He/^{20}Ne)_c \qquad (2)$$

$$A + M + P = 1 \qquad (3)$$

where subscripts s, a, m, and c stand for sample, atmospheric, MORB-type, and crustal-type, respectively, and A, M and C are the fractions of atmospheric, MORB-type, and crustal-type helium, respectively. Taking values for $(^3He/^4He)_a = 1.4 \times 10^{-6}$, $(^3He/^4He)_m = 11 \times 10^{-6}$, $(^3He/^4He)_c = 0.01 \times 10^{-6}$, $(^4He/^{20}Ne)_a = 0.318$, $(^4He/^{20}Ne)_m = 1000$, and $(^4He/^{20}Ne)_c = 1000$ (Sano 1983, Sano and Wakita, 1985), helium's fractions of Iberian samples can be estimated (Table 2).

Assuming that the $^4He/^{20}Ne$ ratios of magmatic and crustal components are significant higher than that of air, it is possible to correct atmospheric helium contamination as follows:

$$(^3He/^4He)_{cor} = [(^3He/^4He)_{obs} - r] / (1-r) \qquad (4)$$

$$r = (^4He/^{20}Ne)_{air} / (^4He/^{20}Ne)_{obs} \qquad (5)$$

where $(^3He/^4He)_{cor}$, $(^3He/^4He)_{obs}$ indicate the corrected and observed $^3He/^4He$ ratios, and $(^4He/^{20}Ne)_{air}$ and $(^4He/^{20}Ne)_{obs}$ are the atmospheric $(^4He/^{20}Ne)$ ratio = 0.318, and observed $(^4He/^{20}Ne)$ ratio, respectively. This method is similar in principle to that reported by Craig *et al.* (1978). The corrected $^3He/^4He$ ratios are also listed in Table 1. High $^4He/^{20}Ne$ ratio measurements in these Iberian samples clearly reflects very low air contamination with the exception of the sample collected at Melgaço (Portugal).

Nine terrestrial fluid samples were collected in relationship with the Iberian Hercynian belt and the Mediterranean tectonic depressions or Neogene grabens. Most of the samples showed $^3He/^4He$ ratios close or below to 1 Ra, but higher values were clearly observed in samples collected from Quaternary volcanic areas such as Calatrava (Ciudad Real) and near Olot (Gerona).

Terrestrial fluid samples collected in Calatrava as well as in Galicia and Northern part of Portugal are related to Iberian Hercynian belt which have experienced significant tectonic activity during Neogene and Quaternary. Samples collected in the Northwestern part of the Iberian peninsula showed $^3He/^4He$ ratios with an important crustal component (up to 98.8% crustal helium), but helium-3 emission is still high for some samples collected from this granitic and metamorphic area. Observed $^3He/^4He$ ratio in Caldas de Tui was 0.09 Ra, value very similar to crustal fluids' helium isotopic signature which have an average of 0.02 Ra. On the contrary, other samples from this region showed values up 0.68 Ra which is significant higher than typical crustal-helium. This relatively anomalous observed $^3He/^4He$ ratio in the sample collected at Cabreiroa reflects that 8.35% of the degassed helium is mantelic in origin. This relatively high $^3He/^4He$ ratio might suggest magma emplacement in the subsurface of this region. It has been argued elsewhere (O'Nions and Oxburgh, 1988) that the most pausible means of scavenging and concentrating He which occurs in very low concentrations in the mantle, is the process of partial melting and that the presence of mantle volatiles in near-surface fluids may provide evidence of sub-surface magmatism when other indicators are lacking (O'Nions *et al.*, 1989). In these cases, mantelic helium released during melting are emplaced in the crust and reach the near surface. Tectonic activity has been quite intensive in this region of Galicia and Northern part of Portugal during the Neogene and Quaternary. Anomalous seismic activity have mainly occurred in relation to the movement of active NNE trending faults in this area and where occurs a significant number of rich-CO_2 ground water wells along this fault system. The highest seismic event during the last year was recently registered in this region where two 4.6

Sample	He (%) mantelic	He(%) crustal	He(%) atm.
1. Melgaço (Portugal)	20.62	59.53	19.8
2. Calatrava 2 (Ciudad Real)	22.21	75.98	1.81
3. C. de Tui (Pontevedra)	0.86	98.84	0.30
4. Calatrava 1 (Ciudad Real)	22.42	77.46	0.12
5. Amer (Gerona)	48.49	51.49	0.02
6. Cabreiroa (Orense)	8.35	91.62	0.02
7. Cofrentes (Valencia)	11.73	88.27	0.00
8. Gerona-2 (Gerona)	15.29	84.71	0.00
9. Lanjaron (Granada)	1.06	98.94	0.00

Table 2.- Estimated helium fractions(%) of mantelic, crustal and atmosferic origin

Tabla 2.- Estimaciones sobre los niveles de He (%) procedentes de los reservorios mantelicos, cortical y atmosferico

magnitude earthquake occurred during the last period of 1995 (Julio Mezcua, IGN, personal communication). This mechanism of magma emplacement might be also a pausible explanation to increase pore pressure in the subsurfac region, and this last process might help to trigger seismic activity in active tectonic areas (Nur, 1996; Sato, 1996). It has been previously postulated that major steep faults are channels for the escape of mantle-derived CO_2 (Irwin and Barnes, 1980). Carbon isotope measurements of rich-CO_2 discharges along NNE trending faults in the Northern part of Portugal, Chaves and Villarelho de Raia, showed $\delta^{13}C$ values between -5 to -8‰ suggesting the released of deep-seated CO_2 (Almeida, 1982; Aires-Barros, *et al.*, 1995). This carbon isotope data in addition to the helium isotopic signature at Cabreiroa revealed clearly a significant mantelic degassing component suggesting that rich-CO_2 discharges along this NNE trending faults might be related to a mixing process of mantelic and marine carbonate reservoirs. Carbon isotopes measurements in the CH_4 fraction released from Cabreiroa well show clearly a thermogenic origin ($\delta^{13}C(CH4)$ = -45.1‰), and this value is in good agreement with our hypothesis. Rich-CO_2 discharges from active volcanic environments such as La Palma, Canary Islands, showed also similar $\delta^{13}C(CH4)$ values, -44.8‰ (Pérez, in preparation). $^3He/^4He$ ratio in fluids from Melgaço is to high, 1.86 Ra, for this granitic environment, and isotopic fractionation during sampling might have occurred. Significant level of atmospheric contamination suggest and support also this hypothesis (up to 19.8 % of helium of atmospheric origin). Therefore, no discussion about this value will be taking in consideration. Gas samples collected in the Calatrava region showed the highest mantelic signature among sampling sites related to the Iberian Hercynian belt. $^3He/^4He$ ratios are around 1.8 Ra in Calatrava where mantelic helium fraction can reach values up to 22%. Quaternary volcanic activity in this region (Hernández Pacheco, 1932) is mainly responsible for this relatively enriched helium-3 isotopic signature, but a significant radiogenic helium dilution process seems also responsible to keep relatively low this $^3He/^4He$ ratio. Seismic studies revealed that Mohorovic'c discontinuity is located at a depth of 36 km; being the continental crust in this central region the thickest one in the Iberian peninsula (Payo and Ruiz, 1977).

Terrestrial fluid samples from Northeastern part of Spain showed the highest observed $^3He/^4He$ ratio in the Iberian peninsula, 3.89 Ra, and where a significant Neogene-Quaternary volcanic activity had occurred (Araña *et al.*, 1983). An additional helium isotopic signature from this region is related to the Ampurdan's area where a $^3He/^4He$ ratio of 1.24 Ra was observed in Gerona-2 well. The difference between these two $^3He/^4He$ ratios seems to have a close relationship with the age of the volcanic activity in the Northeastern part of Iberian peninsula. Radiometric datations revealed that the older eruptions are situated in the Ampurdan graben while the Quaternary volcanism is around Olot (Araña *et al.*, 1983). Both values are still the highest helium-3 emission levels detected in the Iberian peninsula, and they show a very good relationship with terrestrial heat flow data which is also the highest one detected in the Iberian peninsula (Albert Beltrán, 1979a, 1979b). The observed $^3He/^4He$ ratio of 3.89 Ra in Amer's well reflects that 48.49% of the degassed helium is mantelic in origin while a 15.29% is estimated for Gerona-2. These observed $^3He/^4He$ ratios do no show such a good correlationship with crustal thickness in this region where Mohorovic'c discontinuity is estimated to be between 15 and 23 km in the Ampurdan area while is around 15 to 30 km in the tectonic depression of Olot. Two additional rich-CO_2 discharges were collected in the Iberian peninsula, Cofrentes and Lanjarón which are related to the Iberica and Betic cordilleras, respectively. The observed $^3He/^4He$ ratio in Cofrentes is 0.95 Ra and this value reflects a mantelic helium fraction about 11.73%. This value show a relatively enrichment of helium-3 emission which migh be also related to recent tectonic activity in this region where deep-seated fluids can reach the surface environment through deep faults. CO_2 discharges at Cofrentes are quite spectacular like that at Cabreiroa where mantelic helium fraction is 8.35%. Seismic hazards (frequency of seismic activity) in this area might be low, but seismic risk is quite high at Cofrentes and its surroundings since a Nuclear Power Plant is located nearby this rich-CO_2 discharge of significant mantelic component, and therefore related to a deep fault where an increase of pore pressure might be responsible to trigger earthquakes in the region. Rich-CO_2 discharge at Lanjarón showed a $^3He/^4He$ ratio of 0.10 Ra which reflects only a 1.06% of mantelic helium degassing for this site.

Conclusions

Significant spatial variations of $^3He/^4He$ ratios have been observed in the Iberian peninsula. High $^3He/^4He$ ratios are close related to Quaternary volcanic areas, but relatively enrichment of helium-3 emission in non-volcanic regions might reflect subsurface magmatism which is not always related to rich-CO_2 discharges in the surface environment. Geographical distribution of $^3He/^4He$ ratios are mainly related to seismic and neotectonic characteristics of the Iberian peninsula, and additional data to complete an intensive survey is needed to have a better understanding of its complexity. $^3He/^4He$ ratio measurements in terrestrial fluids from Iberian peninsula seem also to be an efficient geochemical parameter to detect gas-flow through deep faults which might be sensible geological structures for earthquakes.

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