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Noble gases in mantle derived xenoliths from Eastern Australia and their implications for the tectonic evolution: Summary

György Czuppon
Institue for Geochemical Research, Budapest

Takuya Matsumoto
IAEA

Jun-ichi Matsuda
Dep. of Earth and Space Science, Osaka University, Osaka

Monica R. Handler
School of Geography, Victoria University of Wellington, Wellington

John Everard
Tasmanian Mineral Resources, Tasmania

Lin Sutherland
Ausrtalian Museum, Sydney

Noble gas elemental and isotopic compositions of mantle derived xenoliths have been measured from different localities along eastern margin of Australia. The observed noble gas composition of xenoliths samples from northernmost locality (Mt. Quincan, North Queensland) seems to be very homogeneous, and it is very similar to those reported from mid-ocean ridge basalts (MORBs). The studied xenoliths from Tasmania indicate more heterogeneous noble gas isotopic compositions with the presents of MORB-like and radiogenic components. The contributions of MORB-like noble gas component to the xenoliths from both regions indicate a large-scale occurrence of this component in subcontinental lithospheric mantle (SCLM) beneath eastern Australia. This MORB-like fluid addition is likely linked with rifting tectonic settings when asthenospheric fluid (and magma) could rise into the subcontinental lithospheric mantle. Eastern margin of Australia has been controlled by rifting tectonic setting during Mesozoic and Cenozoic when opening of Tasman Sea and subsequent extension phase took place. The MORB-like component found xenoliths might reflect these tectonic events. The presence of radiogenic component in xenoliths from Tasmania might reflect subduction events when the mantle wedge and SCLM could be metasomatised by U, Th rich fluids. Subduction processes have played important role in the evolution of Eastern Australia during the Paleozoic; the radiogenic component likely associated with this event. Although this Paleozoic subduction was extend from Tasmania to Northeastern Australia, the samples from North Queensland do not indicate contribution of subduction related (i.e. radiogenic) noble gas component showing pure MORB-like noble gas composition. Thus, the fluid-entrapment responsible for this MORB-like signature likely overwrites any pre-existing signatures or even replaces it with "MORB"-like mantle by physical removal of previously metasomatised lithospheric mantle (i.e. delamination).

Address of corresponding author: Gy. Czuppon: H-1112 Budapest, Budaörsi út 45, Hungary
e-mail: czuppon@geochem.hu

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Helium isotopes distribution in NW Iberian peninsula: evidences of a local neotectonic activity

Fausto Grassa, Giorgio Capasso
Istituto Nazionale di Geofísica Vulcanologia, Sezione di Palermo, Palermo

Paula M. Carreira
Instituto Tecnológico e Nuclear, Sacavém

Maria R. Carvalho
Universidade de Lisboa, Faculdade Ciencias, Dpt. Geologia, Lisboa

Jose M. Marques
Instituto Superior Técnico, Lisboa

Manuel Antunes da Silva
UNICER Bebidas, S.A. S. Mamede de Infesta

In this work we report new data on He abundances and isotope ratios ($^3He/^4He$) from gas associated to some thermal and CO_2-rich mineral waters in N-Portugal. Collected gas samples are mainly CO_2-dominant except two sites where gas is N_2-rich. All the sampling sites are characterized by exceptionally high helium contents with $^3He/^4He$ ratios, corrected for air contamination, varying considerably from 0.09 to 2.68 Ra. In all sites, the $^3He/^4He$ ratios are higher than that typical for stable continental areas thus indicating a variable but not-negligible (up to 30%) contribution of mantle-derived primordial He. In all the CO_2-rich waters, $CO_2/^3He$ ratios and $\delta^{13}C_{CO2}$ are comparable with mantle values, thus suggesting a magmatic origin also for CO_2. On the contrary, in the N_2-rich waters He is mainly radiogenic, and CO_2 is organic in origin. Since no recent volcanic activity is observed in NW Iberia, high $^3He/^4He$ values could be due, at least, to three processes:

a) releasing of gas from the local upper mantle through deep extensional fault systems; b) releasing of magmatic volatiles from crustal reservoir(s) formed during past volcanic activity; c) degassing of a subsurface emplaced magma body.

Mantle He flux in N-Portugal has been estimated to be up to 3 orders of magnitude higher than that typical for stable continental areas, thus suggesting, in this area, the presence of a tensional tectonic regime. This implies that mantle gases could migrate upward probably through inherited tectonic structures reactivated by neotectonic activity. The third possible scenario seems to be less plausible since seismic surveys carried out in NW Iberian did not find any significant evidence of mantle intrusion in the crust. The observed spatial variability in mantle-derived contribution could reflect the geometry of the granitic plutons in this area, thus supporting the hypotheses of an upper mantle degassing. Alternatively, it could be the result of a lateral migration of magmatic volatiles stored in a crustal reservoir.

Address of corresponding author: F. Grassa: Via Ugo La Malfa 153, 90146 Palermo, Italy
e-mail: grassa@pa.ingv.it

Deep groundwater age dating with noble gases (He) and chlorine-36: The Continental Intercalaire aquifer from the Sahara basin

Abdelhamid Guendouz
Engineering and Science Faculty, Blida University, Blida-Algeria

Adnane Souffi Moulla
Dating and Isotope Tracing Dept. Algiers Nuclear Research Centre, Algiers

Jean-Luc Michelot
Paris-Sud University, Orsay

In the northern part of the Sahara desert, is located a very large aquifer system, the deep "Continental Intercalaire" (C.I.), extending over an area of approximately 1,000,000 km^2. This arid zone vital resource is considered as being "fossil", i.e. mainly inherited from previous climatic conditions, that were more humid than at present.

Sampled groundwaters show a quite wide range of ^{36}Cl contents, ranging from 8 to 99 10^{-15} at-at^{-1}, expressed as ^{36}Cl/Cl atomic ratio. The space distribution of ^{36}Cl contents fits fairly well with what is known about the piezometric contours of the aquifer: a decrease is observed from recharge to discharge zones. If this decrease is result of radioactive decay, it can be interpreted in terms of groundwaters transit time. Maximum time intervals of about 3 half-lives (900 Ka) may be computed using ^{36}Cl specific activities (at.l^{-1}).

In order to evaluate the epigene production, measurements were performed on chloride extracted by leaching from a soil profile. The results are the same order of magnitude (10^{-15} at-at^{-1}) as for groundwater chloride sampled near recharge areas and could reflect the true value of initial ^{36}Cl input.

The residence times determined on the main flow line where the radio decay is observed are expressed in terms of minimum ages (16 to 500 Ka) and maximum ages (25–1200 Ka).

Noble gas data are presented to improve the palaeoclimatic and residence time interpretation for the Continental Intercalaire aquifer system. The groundwater recharge temperatures (RT) were estimated from the averaged amounts of noble gases (Ne, Kr, Xe) corrected for the excess air effect. The RTs for most groundwaters are generally lower than the present day recharge temperatures. Along the main flow direction (south-east from the Atlas mountains), the CI Paleowaters (ages 20 to 40 ka BP) have an average RT of 16.9 °C which is some 5 °C cooler than at the present day. Recharge temperatures calculated in four samples from the CT aquifer (30–150 m depth) average 19.7 °C, close to the present day mean annual temperature of 21 °C.

Address of corresponding author: A. Guendouz: PO Box. 270. Route de Soumâa Blida 09000 Algeria, e-mail: ah_guendouz@hotmail.com

Study of the behavior of radon isotope in Pál-völgy show cave (Budapest, Hungary)

Hedvig Éva Nagy
Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös University, Budapest

Csaba Szabó
Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös University, Budapest

Department of Atomic Physics, Eötvös University, Budapest

Ákos Horváth
Department of Atomic Physics, Eötvös University, Budapest

Attila Kiss
Danube–Ipoly National Park Directorate Pál-völgy show cave, Budapest

We have been doing radon measurements in Pál-völgy cave (Budapest, Hungary) for one and a half year. The cave is situated in the Buda Hills, which is the NE part of the Transdanubian Central Range. The wall rock of the cave is dominantly Eocene Szépvölgy Limestone Formation. Above the limestone Eocene Buda Marl and Oligocene Tard Clay are deposited. The main aim of this study is to determine the time dependent radon concentration and the sources of the radon in the cave.

The radon concentration in the cave air has been measured continuously by AlphaGUARD radon monitor and outside the cave meteorological parameters were collected simultaneously. The radon concentration of the air in the cave varies between 104–7776 Bq/m^3 during the measurements, the average value is 1920 Bq/m$_3$. These data strongly depend on the outside air temperature. If the temperature outside is higher than inside (11 °C) the radon concentration increases. The correlation coefficient between the radon concentration of the air in the cave and the outside air temperature is 0.75. The spatial distribution of radon concentration in the cave air was measured simultaneously by active radon detectors, which shows values as high as 1000 Bq/m^3 where Buda Marl is the surrounding rock, and 500 Bq/m^3 where Szépvölgy Limestone is the wall rock.

To define the source of radon, besides the wall rock limestone and marl, clayish cave sediments have been collected. The radioactive isotope (^{226}Ra, ^{232}Th, ^{40}K) content of clayish cave sediments shows results typical for soils: 26–37 Bq/kg ^{226}Ra, 21–31 Bq/kg ^{232}Th, 265–386 Bq/kg ^{40}K. However, the radon and thoron exhalation rates of these samples, 2–12 Bq/kg for ^{222}Rn and 1–12 Bq/kg for ^{220}Rn, are higher than expected based on the ^{226}Ra content. These results can be related to high percentage of fine grain size fraction corresponding to high specific surface which provides high possibility of exhalation. Our results suggest that the most likely radon source is the Buda Marl.

Address of corresponding author: H. É. Nagy: 1117 Budapest, Pázmány P. sétány 1/C, Hungary
e-mail: jadzsa@gmail.com

Uranium, radium and ^{222}Rn isotopes in thermal waters from Podhale Trough (Polish Inner Carpathians)

Chau Dinh Nguyen, Jakub Nowak, Pawel Jodlowski
AGH University of Science and Technology, Faculty of Physics and Applied Computer Science, Kraków

The Podhale Trough belongs to Polish Inner Carpathians; it is situated between the Tatras in the south and the Pieniny Klippen Belt in the north. The Trough is the most important geothermal water system in Poland. Thermal water has been reported in over a dozen boreholes, but only nine of them are exploited. Currently the water is used for heating and for recreational purposes.

From all exploited boreholes, water samples were collected. For each sample ^{238}U, ^{234}U, ^{226}Ra, ^{228}Ra and ^{222}Rn activity concentrations were determined. Uranium isotopes were measured by α-spectrometer after separation of them by adequate procedure. The radium isotopes were separated from the water sample as sulfate compounds and measured with α/β liquid scintillation spectrometer (LSC). The ^{222}Rn activity concentrations were determined by LSC method. The chemical composition of water sample was measured using an ICP-AES spectrometer.

The obtained results show that the activity concentrations of ^{234}U and ^{238}U vary from 4.1 mBq/L to 303 mBq/L and from 2.8 mBq/L to 324 mBq/L, respectively. Radium isotopes activity concentrations vary from 29 mBq/L to 590 mBq/L for ^{226}Ra and from 17 mBq/L to 360 mBq/L for ^{228}Ra. The ^{222}Rn concentrations of the samples range from <0.2 Bq/L to about 80 Bq/L. The highest activity concentrations of uranium and radon isotopes were found in the water sample from the Szymoszkowa GT-1 borehole. The highest activities of radium isotopes were observed in the water sample from the Bukowina Tatrzańska PIG/PGNIG borehole. The maximum activities of radium and uranium isotopes contained in the mineral waters of the similar mineralization from the Outer Carpathians are significantly lower and amount to 170 mBq/L and 56 mBq/L respectively. Such a high level of natural radioactivity in the thermal waters from the investigated region could be resulted by the interaction of water with the crystalline, igneous and metamorphic rocks of the Tatra Mts. as the thermal waters likely have strong leachability.

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Address of corresponding author: J. Nowak: Al. Mickiewicza 30, 30-059 Kraków, Poland
e-mail: jnowak@novell.ftj.agh.edu.pl

^{222}Rn concentration measurements of soil gas in Hungary

Katalin Zsuzsanna Szabó
Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös University, Budapest

Ákos Horváth
Department of Atomic Physics, Eötvös University, Budapest

Csaba Szabó
Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös University, Budapest

^{238}U is one of those terrestrial radioisotopes that can be found on Earth since its formation. The ^{222}Rn radioactive noble gas is a daughter product of uranium decay chain. Its parent isotope is ^{226}Ra, which is present in the rocks and soils. Alpha decay of ^{226}Ra provides the ^{222}Rn which emanate out from rock or soil mineral to the pore space by diffusion or alpha recoil. Exhalation coefficient shows that how many percent of radon got to the soil gas or the surface. Because radon is an inert gas and has half life of 3.8 days it can migrate in subsurface and soil environment getting into the indoor air. It can be accumulated in high concentrations in rooms and can cause lung cancer. Soil gas radon concentration is the main source of indoor radon. Geogenic radon risk can be determined from soil gas radon values taking into account the permeability of soil.

We studied the radon exhalation rate in the upper 150 cm of soils for better understanding of the importance of depth from radon point of view in Pest County. Five–five soil samples of 11 soil profiles (down to depth of 150 cm) were examined by gamma spectroscopy to have radium content. We also measured the radon exhalation of soil samples and then the radon exhalation coefficient was calculated. *In situ* soil gas radon and soil permeability measurements were made at 47 sites in 80 cm depth to assess the geogenic radon risk.

Results show that the average of exhalation coefficient is 0.15 (min. and max. values are 0.03 and 0.62, respectively) in the upper 90–100 cm and the deeper layer has 10 times lower values (average is 0.01, min. and max. values are 0.003 and 0.03, respectively). The reason of this decrease is the increase of compactness with depth where the radon cannot escape easily. This indicates the relevant depth for radon is the upper 90–100 cm in general case where there are no special geological features (e.g. uranium deposit). This depth (90–100 cm) also means the measurements depth for soil gas radon measurements.

Radon risk of the 47 sample sites of Pest County was determined from *in situ* soil gas radon and permeability measurements. It shows that 2 sites have high, 16 sites have medium and 29 sites have low radon risk in the study area.

Address of corresponding author: K. Zs. Szabó: H-1117 Budapest, Pázmány P. sétány 1/C, Hungary
e-mail: sz_k_zs@yahoo.de

Testing of fast ^{222}Rn and ^{220}Rn exhalation measurements of high number of Hungarian adobe building material samples

Zsuzsanna Szabó, Csaba Szabó
Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös University, Budapest

Ákos Horváth
Department of Atomic Physics, Eötvös University, Budapest

^{222}Rn and ^{220}Rn are responsible for 52% of the total annual effective dose to humans, therefore it is important to know the exhalation properties of different building materials. The main aim of this work is to test a ^{222}Rn exhalation rate measurement technique and to study ^{222}Rn and ^{220}Rn exhalation rates of Hungarian adobe considering that this is a common building material at certain parts of the country and a potential source of ^{220}Rn.

Adobe samples were taken from settlements in three areas: Békés County (SE-Hungary), Sajó and Hernád Rivers Valleys (NE-Hungary) and E-Mecsek Mts (S-Hungary).

^{222}Rn exhalation rates were estimated in laboratory by two different techniques, both using Al-accumulation chambers and RAD7 detectors. One technique is based on measuring the growth curve of ^{222}Rn concentration in the chamber for ten days, and the other on measuring for four hours the ^{222}Rn equilibrium concentration after three weeks (about $5(T_{1/2})$) closing. The latter one is faster for high number of samples. For ^{220}Rn only the equilibrium concentrations can be used ($5(T_{1/2}) \approx 5$ min).

The second method shows less ^{222}Rn exhalation rates but correlation ($R=0.85$) is found between the ratio of results and the degree of leakage. It is shown that when leakage is below $\alpha \approx 0.006$ h^{-1}, based on the growth curve, the two methods give the closest results. Accepted as correct results of ^{222}Rn and ^{220}Rn exhalation rates are similar (average values are 8 ± 2, 7 ± 2 s^{-1} kg^{-1}, respectively), Hungarian adobe building materials can exhale short half-lived ^{220}Rn almost in the same amount as ^{222}Rn.

It can be concluded that a tightly gas proof circuit is required for the best estimation of both of the methods; however, measurements for high number of samples can be made faster. Hungarian adobe is a potential ^{220}Rn exhaling building material; therefore indoor measurements are necessary to evaluate its effect.

Address of corresponding author: Zs. Szabó: H-1117 Budapest, Pázmány P. s. 1/C, Hungary
e-mail: zsszabo86@gmail.com

Isotopic constraints on genesis of the multistacked CO_2-CH_4-N_2 Répcelak gas field (Pannonian Basin System, W Hungary)

István Vető
Consultant geochemist, Budapest

László Palcsu
Nuclear Research Institute, Debrecen

István Futó, Gergely Vodila, László Papp, Zoltán Major
Nuclear Research Institute, Debrecen

Several multistacked CO_2-CH_4-N_2 gas fields have been found in different parts of the Neogene-Quaternary Pannonian Basin System. These fields are characterised by drastic upward decrease of CO_2 content and very low CH_4/N_2 ratio (< 4).

The 22 reservoirs (mostly sandy) of the multistacked Répcelak gas field (W-Hungary) are developed between 1300 to 800 m depth below sea level (BSL). In this depth interval the CO_2 content gradually decreases upward from 95 to 1–2%. This gradual but drastic change in gas composition is the result of mixing of CO_2 with a CH_4-N_2 fluid that probably arrived later.

Here results of an stable and noble gas isotope study of five reservoirs among the deeper ones (1300–1000 m BSL) of the Répcelak field are presented and discussed.

$^3He/^4He$ ratios prove the predominantly mantle origin of the helium and suggest the same origin for CO_2. The most likely source of these two gases is a basic magmatic body intruding the basement during the late Neogene. The rather high N_2/noble gas (Ne, Ar) ratios confirm that the N_2 cannot be of atmospheric origin. The very low CH_4/N_2 ratios (<3) suggest late thermogenic origin of the CH_4 and N_2. Anchimetamorphic Paleozoic shales of the basement are the most likely source of the CH_4-N_2 fluid.

Within the 1300–1000 m depth interval CO_2 content decreases from 94 to 72.5%, this decrease is accompanied by a moderate increase of $\delta^{13}C_{CO2}$ (from –3.28 to –2.61‰).

In four of the five reservoirs CH_4/N_2 ratio shows upward increase from 1.16 to 2.65, while $\delta^{13}C_{CH4}$ and δD_{CH4} show upward decreases from –41.9 to –49‰ and from –181 to –214.5‰, respectively. These depth related changes in composition and isotopic compositions of the CH_4-N_2 fluid likely reflect a moderate and upward increasing contribution of isotopically light (bacterial?) CH_4.

Address of corresponding author: I. Vető: H-1026 Budapest, Balogh Á. u. 18/c, Hungary
e-mail: vetoie3840@gmail.com

Examination of radiogenic isotopes and chemical composition of coal slag and fly ash bearing building materials

Péter Völgyesi, Hedvig Éva Nagy, Csaba Szabó
Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös University, Budapest

Building materials or additives of building materials can contain relatively high levels of natural radionuclides, which increases the indoor radiation exposure of residents. By residential request we made *in situ* radiometric measurements in 14 houses in the central region of Hungary and we analyzed the building materials (gas silicate, containing fly ash; slag concrete and slag samples) from the studied houses using physical and chemical methods. It is important to know the background of the coal's radioactivity which the coal slag and fly ash was formed of. Laboratory studies have shown that coal can contain primarily uranium and a small amount of thorium. In Hungary the uranium content of coals varies in a wide range (~10 g/t–1 kg/t). The usage of such large U-bearing by-products as building material can cause serious risk to the health of residents, mainly due to radon exhalation. Based on the microscopic, scanning electron microscopic and X-ray diffraction analysis on coal slag, slag concrete and gas silicate samples, the most frequently identified mineralogical phases are quartz, calcite, metallic phases that have iron as their primary constituent, Ca-Mg-rich alumosilicates and gypsum. Also, 2–3 μm uranium-bearing grains in the coal slag sample were identified in large number by scanning electron microscope. The indoor radon concentration was measured by using active detector (short-term measurements) (40–226 Bq/m^3) and we also measured the radon concentration (by using track-etched detectors) and the equilibrium factor on a long term scale (3–6 months). The gamma dose rate was measured with FH 40G L10 detector. The results show values between 70–1000 nSv/h in the 14 studied houses. The ^{226}Ra, ^{232}Th and ^{40}K content of the samples was determined by HPGe Gamma-spectrometry. These results were use to classify with indices the building materials studied. Radium equivalents (Ra_{eq}) vary between 33–2812 Bq/kg and activity concentration indices (I) vary between 0.1–9.4. The calculated radon exhalations (RE) vary between 0.3 and 61.3 Bq/kg. The results show that the fly ash containing building materials (Ra_{eq}: 63–128 Bq/kg; I: 0.2–0.5; RE: 0.4–4.8 Bq/kg) do not have any risk, however in some cases the slag containing materials can achieve or significantly exceed the threshold values (Ra_{eq}: 33–2812 Bq/kg; I: 0.1–9.4; RE: 0.3–61.4). Our study draws the attention to the need of complex physical and chemical methods.

Address of corresponding author: P. Völgyesi: H-1117 Budapest, Pázmány P. s. 1/c, Hungary
e-mail: petervolgyesi11@gmail.com