Underground radon gas concentrations related to earth tides (*)(**)

A. KIES (1)(***), J. MAJERUS (1) and N. D'OREYE DE LANTREMANGE (2)(*$_*$*)

(1) *Centre Universitaire - 162a avenue de la Faïencerie, L-1511 Luxembourg*
(2) *European Centre for Geodynamics and Seismology - Walferdange, Luxembourg*

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Summary. — Over several years, radon concentrations have been recorded in a former gypsum mine near Walferdange (Luxembourg). Because of the exceptional quality of the site (thermal stability better than 0.01 °C/year, no running water, easy access, far enough from the oceans), today the mine hosts an underground laboratory for geodynamics and seismology with more than 25 permanent instruments that continuously record earth tides, earth quakes and meteorological parameters. One of the main interests in monitoring radon concentrations in this mine was to check the ability of earth tide effects on radon concentrations in the mine atmosphere. First results show that besides outside temperature and atmospheric air pressure, radon concentrations seem to be influenced by earth tides. Power spectra, calculated for different time series of radon concentrations, show the presence of both O_1 (lunar declination) and M_2 (principal lunar) tides. The increase in vertical extension and the decrease in gravity induced by earth tides in the bulk of the rocks may open supplementary pathways for radon and thus induce an increase of the radon transport through the rocks.

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1. – Introduction

Only a few years after Dorn discovered, in 1900, the radioactive gas radon emanating from the radium salts, geological applications of radon were found, for instance in the detection of underground inhomogeneities. Probably the first to mention radon anomalies associated to earthquakes was Shiratoi (1927). Presently monitoring of radon concentrations in water or gas phase has become a common tool in geophysics. Whenever mentioned, "radon" refers to the ^{222}Rn isotope.

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(**) The authors of this paper have agreed to not receive the proofs for correction.
(***) E-mail: kies@cu.lu
(*$_*$*) E-mail: ndo@ecgs.lu

Stress and strain build-up prior to an earthquake may trigger changes of underground radon concentrations in the water and gas phase. Unfortunately, neither the tectonic mechanisms, nor the meteorological and the hydrological mechanisms that influence the natural radon emissions are completely understood. Therefore, radon as earthquake precursor technique remains problematic. The present contribution discusses the possibility of an influence of earth tides on radon. The strain variations induced by earth tides are very small, less than 10^{-8}, but their appearances are periodic and of known magnitude.

Tidal forces deform the earth; effects induced on fluids near the surface of the earth are documented by the observations of water level changes in wells (for extensive bibliography see Westerhaus, 1996). These changes are driven by alterations of the pore pressure induced by tidal deformations of porous and fluid-saturated crustal material.

A great advantage of tidal signals is the well-known long-range character that allows their transformation into the frequency domain. By using spectral methods, disturbances due to transient meteorological events like rainfall and even atmospheric influences can be avoided; temperature effects, air pressure variations and moisture are restricted to certain constituents of the tidal spectrum and can be excluded from interpretation, or data can be corrected for.

Recently H. Woith (personal communication and 1998) could identify a tidal-induced radon signal in groundwater. A spectral analysis of the available time series revealed that at least 2 groundwater systems are sensitive to earth tidal strain: daily O_1 and half-daily M_2 lunar tides. This means strain sensitivity at these sides down to 10^{-8} for periodic strain variations.

Sides which are sensitive to stress/strain variations down to 10^{-8} could be especially propitious as natural sensors for earth tides identification and also for earthquake prediction. But the extrapolation from periodic variations to more abrupt changes prior to earthquakes may be problematic.

Until now, very few reports mention the possibility of showing tidal effects in air-borne radon of an underground reservoir, for instance a mine or a cave. The transport of radon is affected either by diffusion along concentration gradients or by motion of a carrier fluid along pressure gradients. While diffusion is responsible for the transport on a small scale, the motion of fluids along pressure gradients affects the transport on a large scale (Schery, 1984).

The response characteristic of the radon signal depends on two classes of rock parameters:

– Elastic properties and porosity in the vicinity of the galleries and the compressibility of the pore fluids. These poroelastic properties may define the magnitude of the strain-induced variations (Holub, 1981).

– Fluid flow conditions and the permeability of different geologic formations. The radon specific mechanism has to be considered: due to the short half-life of ^{222}Rn (3.82 days) its mobility in the rocks by diffusion is limited to small distances in the meter range. To be effective, radon needs a carrier fluid: the air in the microfractures and the pores in the present case.

Currently there is great interest in the interrelationship between rock dynamic deformation and fluid flow. Radon tidal effects could play an important role in the investigation on this problematical subject.

2. – The Walferdange mine

In order to find a possible correlation between radon and earth tides, continuous monitoring of radon was performed over years in a former gypsum mine in Walferdange situated 6 kilometres North of Luxembourg town. In an important gypsum deposit, formed in the hard mottled marls (Steinmergelkeuper) of Upper Trias, gypsum was mined in a maze of horizontal galleries (section $2\,\mathrm{m} \times 8\,\mathrm{m}$) extending up to 700 meters under 80 meters of triassic marls and Jurassic sandstone. Today mining has stopped. In the most distant part of the main gallery, an important laboratory of geophysics and geodynamics is installed.

Radon concentrations are measured continuously since the end of 1993 with different techniques. All revealed that radon concentrations experience only very low short-time fluctuations. They are little affected by atmospheric pressure and diurnal outside temperature variations (fig. 1), but change smoothly from very stable 2.2 kBq/m^3 in the

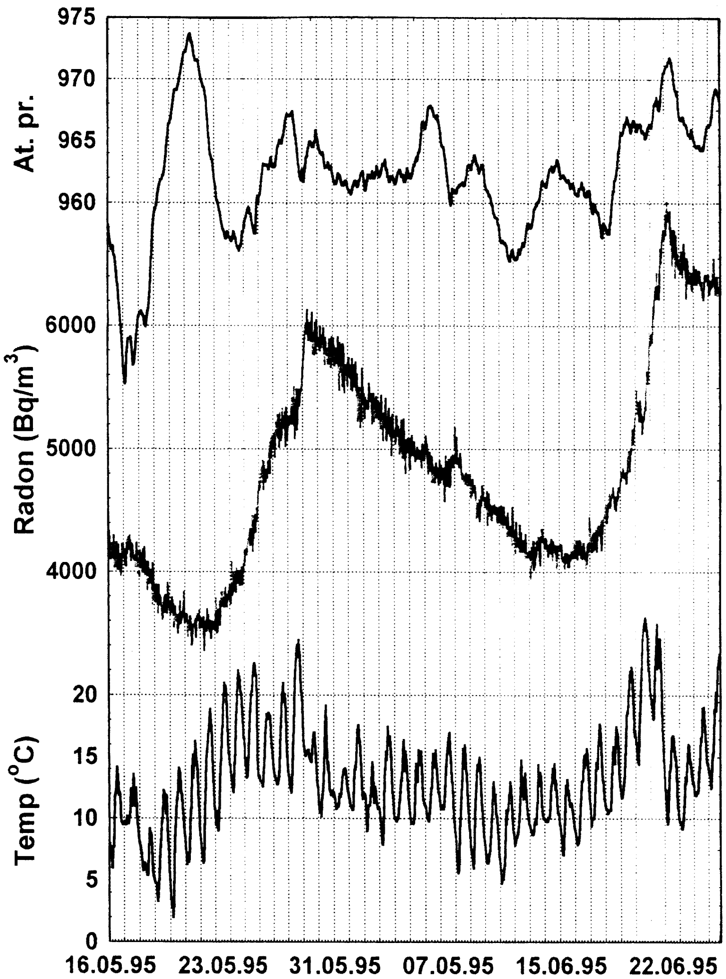

Fig. 1. – Gypsum mine in Walferdange: detail of variations in radon concentrations, atmospheric pressure and outside temperature.

cold months up to more than 8 kBq/m^3 in the hot summer periods. Radon concentrations are nearly independent of the measuring place inside the mine (Kies, 1997).

The variations of seasonal radon concentrations may be explained by differences between winter-summer air exchange rates in the mine. As temperature and moisture inside the mine are constant, external temperature and external air density fluctuations govern the air exchange rate. In winter, cold air moves in at the bottom and warmer air moves out at the top of the entrance gallery. In the warm season, when the mean outside density is lower than the inside density, air movements are reduced or even stopped.

The radium content of the bedrock is medium to very low so that the measured radon concentrations are higher than could be expected. The well-pronounced horizontal layers of gypsum and marls give rise to diffusion and pressure-induced horizontal transports out of the bulk of the rock. The lack of major cracks and the scarce air movements induce very small short-order variations in radon concentration.

A first study of radon concentrations measured in a borehole showed a possible correlation between radon-pore concentrations and earth tides (Lenzen, 1995). The interpretation proposed by Lenzen was the great inhomogeneity in the horizontal and vertical radon diffusion of the bulk material ($D_{\parallel} = 0.5 \cdot 10^{-9}$ m^2/s and $D_{\perp} = 2 \cdot 10^{-9}$ m^2/s) and the absence of disturbing processes.

Recently higher radon concentrations are monitored in summer, compared to those in the 1993 to 1996 period. The possible reason is the opening of supplementary pathways to the exterior, due to earth gliding near the entrance. The effect of these new pathways is an increase of mean radon concentrations especially in summer time due to air movements induced by the classical chimney effect.

3. – The experimental set-up

Radon-gas measurements were done over years by an Alphanuclear silicon-based radonmonitor. This instrument was well suited for long-time measurements but the sensibility of the instrument was not high enough to show small short-time variations. When available, Alphaguard ionisation chambers (from Genitron Instruments) were exposed in the mine. Over 4 years, most of the continuous radon monitoring was done by an autonomous measuring system based on a Lucas-scintillation cell (Lenzen, 1997). By a system of electrical valves, air is sucked into the ionisation chamber and each hour the chamber is evacuated and refilled with a fresh probe. For each air probe, the counts after 0.5, 1, 2, 4, 8 and 60 minutes are registered. This time sampling, because of the decay profile registered, offers the possibility to filter also the information on the thoron content in the mine.

4. – Treatment of the data

A first pre-treatment was performed which consisted in a data refinement using deglitching and presmoothing against noise. One of the main algorithm used to perform this kind of treatment was a non-local spline-smoothing as proposed by Reinsch (Reinsch, 1967, 1970), combined with rough statistics to discriminate against outliers. The spline-smoothing algorithm also allows direct interpolation of the corrected data. This feature was used to compute a data set with constant step as needed to perform discrete Fourier transforms. Consistent with the Nyquist theorem, a sampling rate of one hour was generally used.

Finally, to get the effective signal of interest, a spline-smoothed background was subtracted; first, in order to avoid low-frequency interferences with the signal of interest, the so-called leakage effects, and second, to minimise Gibbs oscillations in the spectral domain due to the strong cut-off of the time-limited data set. Note that the quality of this data pre-treatment may be refined and optimised, through a posterior criteria on the relative pic-intensities in the frequency domain.

This effective signal was finally used to compute an approximation of the power spectrum through discrete Fourier transform (DFT). To do so, an edge weighting of the data set was performed; the used weighting function was applied to the first and last 2.5% to 25% of the data set. At least double-length of zeros was added to eliminate alias-effects and to fill up the data set to $N = 2^{\alpha}$ points ($\alpha = 12$, $N = 4096$). The DFT of the weighted data was computed by means of the discrete FHT (Fast Hartley Transform) for real data.

Finally, the weight-corrected periodogram estimation was used to get an approximation of the spectral density $G(f)$ of the power spectrum, where f is the spectral frequency. The frequency-weighted power spectrum G^*f^2, computed for a given time span interval, is shown in fig. 2.

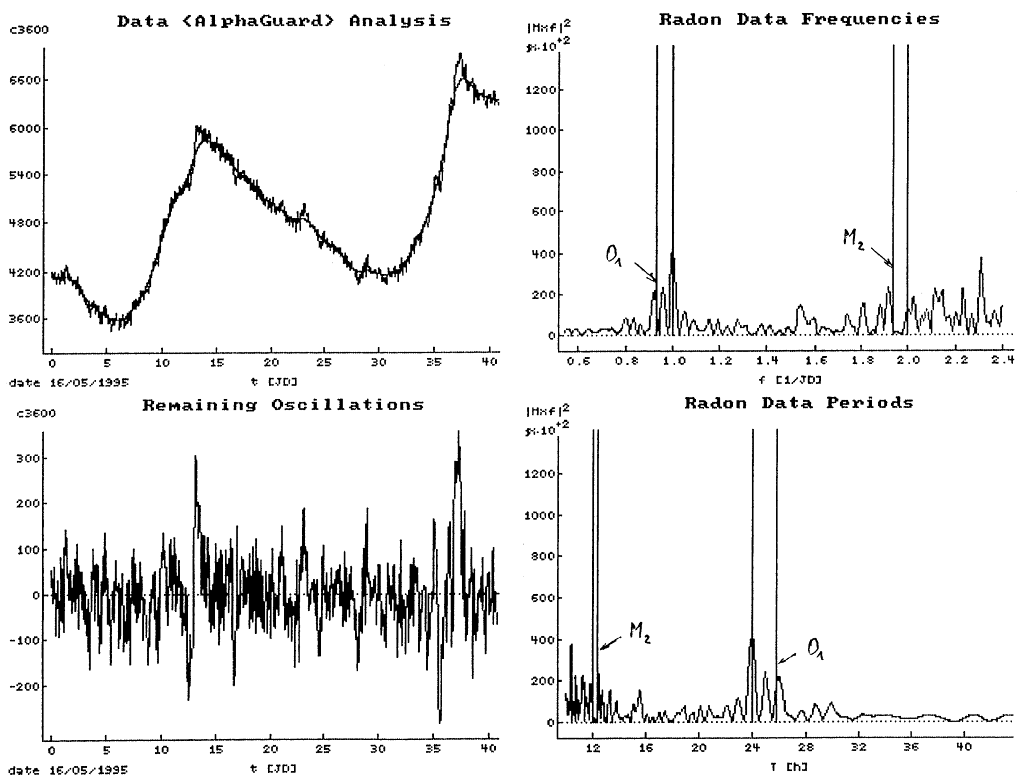

Fig. 2. – An example of the data measured over a given period is given, together with its treatment up to the final frequency-weighted power spectrum G^*f^2.

The periods which we looked for are

- $T = 12.4$ h; the principal lunar period M_2,
- $T = 25.8$ h, the lunar declination period O_1,
- $T = 12$ h, the principal solar period S_2.

The last period can be due to diurnal temperature and pressure influences; this period was not retained as a clue to decide for the influence of earth tides on the radon signal.

With a good accuracy, the periods or frequencies of the O_1 and M_2 earth tides can be seen. Analysis of other time series, related to different dates and instruments, gave similar results.

In order to initiate a possible interpretation of this influence of earth tides on radon concentrations in the Walferdange mine, some particularities of this mine will be discussed in the next section.

5. – Particularities of the Walferdange mine

Gypsum $CaSO_4$ $2H_2O$ has proved to be an ideal mineral for experimental study aimed at understanding the movement of volatiles during crustal deformations (Stretton, 1997). The highly anisotropic behaviour of gypsum minerals in a randomly oriented polycrystalline aggregate leads to stress-strain incompatibilities. That may generate void spaces, which could lead to considerable increases in rock porosity. Gypsum is essentially a layered structure bound by hydrogen bonds. In between the horizontal gypsum layers, clay layers of variable thickness in the millimetre range are embedded. The moderate to low radium content of the gypsum rocks is due to this clay.

The poorly permeable overburden of Keuper marls helps to confine radon in the underlying void or in the underlying gypsum lens. Radon can accrete and become mobile during active transport processes in the horizontal layers. Even if the rocks have moderate to low amounts of radium source material, the amounts are sufficient for radon to exhale in significant amounts from the surrounding walls into the voids of the gallery.

A possible influence on radon concentrations, due to a rise of the water table, may be rejected because the gypsum lens is sandwiched by impermeable marls.

The geological inhomogeneities, marls layers in gypsum, can be viewed as a contrast of structures with different poroelastic properties. The pressure response of jointed media of quite different response to volumetric strain could be a potential source of a supplementary freeing of radon (Westerhouse, 1996).

Millich and Neugebauer (1993) report that, in general, two conditions are needed to get directed flow induced by tidal deformations:

– Possibility of fluid circulation. Directed flow occurs if there are spatial differences in the stress dependence of permeability within the flow path.

– Spatial difference in the stress dependence of permeability within the considered region; for instance, variation of permeability with effective stress in one location and constant permeability in another part.

If a volumetric strain is imposed on a composite, fluid-saturated medium with different poroelastic properties, a pore pressure difference will be established at both sides of the contact zone. Some of the radon in the pores and (micro) fractures could move into the mine under a variable pressure gradient.

Furthermore, the emanation coefficient of the clay minerals sandwiched in between the gypsum could be influenced by the tiny tidal strain variations.

6. – Conclusion

With a good accuracy, we have shown that the radon gas concentrations in the former gypsum mine of Walferdange are sensible to earth tides M_2 and O_1. Further measurements will be performed; new experimental methods will be tested.

We will try to observe if the spectral amplitudes of M_2 and O_1 are constant over time. If not, are there any reasons? At earthquake observation sites, where radon tidal effects are recorded, investigations on the temporal evolution of radon tides could indicate changes in the stress/strain situation. We believe that a study of tidal-induced variations of radon concentrations will be another useful step in the effort to use radon as a tracer gas in geophysics.

Until now, no precise static strain-radon coupling mechanism could be proposed, in order to choose the most probable explanation of tidal radon exhalation modifications.

REFERENCES

HOLUB R. F. and BRADY B. T., *The effect of stress on radon emanation from rock, J. Geophys. Res.*, **86** (1981) 1776-1784.

KIES A., MASSEN F. and FEIDER M., *Measuring Radon in Underground Locations, 3rd ICRGG International Colloquium on Rare Gas Geochemistry*, Amritsar, India 1995, in *Rare Gas Geochemistry*, edited by H. S. VIRK (Guru Nanak Dev University, Amritsar) 1997.

LENZEN M., *Ein automatischer Radonsensor für Bohrlochmessungen; Messergebnisse aus der Gipsmine Walferdingen,* Publications du Service Géologique du Luxembourg, Bulletin No. 17/95 (1995).

LENZEN M. and NEUGEBAUER H. J., *An automatic radon sensor for borehole measurements, Rev. Sci. Instrum.,* **68** (7) (1997) 2898-2903.

MILLICH E. and NEUGEBAUER H. J., *Pumping of fluids in porous media induced by earth tides,* KTB-Report 93/2, edited by R. EMMERMANN (Deep Drilling Programme in the Geological Survey of Lower Saxony), 1993, pp. 313-316.

REINSCH C. H., *Smoothing by spline functions I. Numerische Mathematik*, **10** (1967) 177-183.

REINSCH C. H., *Smoothing by spline functions II. Numerische Mathematik*, **16** (1970) 451-454.

SCHERY S. D., GAEDDERT D. H. and WILKENING M. H., *Factors affecting Exhalation of Radon from Gravelly Sandy Loam, J. Geophys. Res.*, **89** (1984) 7299-7309.

SHIRATOI K., *The variation of radon activity of hot springs, Science reports of the Tohoku Imperial University, Sendai Ser. 3*, **16** (1927) 614-621.

STRETTON I. C. and SCHOFIELD P. F., *The static compressibility of gypsum, Geophys. Res. Lett.*, **29** (1997) 1267-1270.

WESTERHAUS M., *Tilt and well level tides along an active fault*, Scientific technical report STR96/05, Geoforschungszentrum Potsdam, 1996.

WOITH H., MILKEREIT C., ZSCHAU J., MAIWALD U. and PEKDEGER, A., *Monitoring of thermal and mineral waters in the frame of READINESS, Proceedings of the 9th International Symposium on Water-Rock-Interaction* (*WRI*), *Rotterdam, 1998*, edited by AREHART and HULSTON (Balkema, Rotterdam) 1998, pp. 809-912.