

Radon behaviour in mineral spring water of Bad Brambach (Vogtland, Germany) in the temporal vicinity of the 1992 Roermond earthquake, the Netherlands

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Abstract

The correlation between the Vogtland micro earthquakes and the fluctuations of ²²²Rn concentration in spring water and soil air has been investigated in the Saxon State Spa of Bad Brambach for more than three years. In contrast to the soil air, spring water shows significant radon anomalies at the time of about 50% of the earthquakes or earthquake swarms ($M_L < 4$, epicentral distance < 50 km). The 1992 Roermond earthquake has confirmed for the first time that it is possible to register an anomaly effect in Bad Brambach related to seismic wave motion from earthquakes with larger epicentral distances. Recent investigations have shown that CO₂ outgassing due to tectonic processes may act as a gas lift for radon. A model conception and first results of continuous radon and CO₂ measurements in spring water are presented.

Introduction

The search for possibilities of earthquake prediction by means of seismological, geophysical, and geochemical parameters has continued for more than five decades. Neunhöfer & Güth (1988) demonstrated a statistical approach in a wide temporal range for the earthquake swarm of 1985/86 based upon seismological data from our investigation area. Other methods, such as the observation of water level fluctuations (Liu et al. 1989, Hill et al. 1993), showed interesting results only during or after periods of strong seismicity (Pazdera & Procházková 1986, Kämpf et al. 1987). In our case the geochemical properties of the confined mineral springs of Bad Brambach were used for the investigation of phenomena related to earthquakes in this region. Some of the registered anomalies are no precursors. Therefore, we prefer to speak of 'coseismic' observations.

The Saxon State Spa of Bad Brambach is situated in the southwest of Saxony, in the western part of the Erzgebirge. It is well-known because of its mineral waters with high natural ²²²Rn and CO₂ concentrations,

ranging from 1.5 to 25 kBq/l and 1000 to 2800 mg/l, respectively.

Our investigation area is located almost exactly on the boundary between the Fichtelgebirge granite in the south and the Vogtland mica slate and phyllite in the north (Fig. 1). The distance to the Roermond earthquake region is about 450 km (Fig. 2). The region is tectonically disturbed by two active fault zones, the Eger Graben (ENE–WSW) and the Marienbad dislocation (NNW–SSE) (Kuschka 1993). Furthermore, it is characterized by the effects of Tertiary and Quaternary volcanism which is responsible, among other things, for the recent seismic activity in this area (swarm and microearthquakes). Probably, the subrecent volcanism is the cause of the heat flow anomaly in this region (Kämpf et al. 1993).

The typical Vogtland microearthquakes have magnitudes <4, but the frequency of occurrence is sometimes relatively high (so-called swarmquakes). According to Kämpf et al. (1992) a distinction should be made between single earthquakes (focal depth 12–15 km) and swarmquakes (focal depth 6–18 km) within the region. Most likely the single events are tectonic

Table 1. Hydrological, chemical, and isotope data of the mineral springs 'Radonquelle' and 'Eisenquelle', Bad Brambach.

Type Spring parameters	Na-Ca-HCO ₃ -SO ₄ , acidulous with Fe + Rn	
	Radonquelle	Eisenquelle
Discharge (l/h)	150	230
Temperature (° C)	7–9,5	7–9,5
Depth of the spring capture (m)	6	5
Content of radon ²²² Rn (Bq/l)	25000	1600
Content of radium ²²⁶ Ra (Bq/l)	1,8	0,06
Content of uranium ²³⁸ U (ppb)	2,5	1,5
²³⁴ U/ ²³⁸ U activity ratio	3,1	2,8
Content of CO ₂ (mg/l)	2540	2740
δ ¹³ C (‰)	– 3,5	– 3,7
δ ¹⁴ C of the CO ₂ (‰ mod.)	< 1	
Mean residence time (tritium) (a)	16–45	30
Content of 'young' water (%)	~ 20	~ 60

quakes in the vicinity of the steeply dipping Marienbad dislocation. On the other hand, the genesis of the swarms is unclear.

According to Hurtig & Oelsner (1979) and Čermak (1979) a deep source of heat has been identified in the investigated area. Kämpf et al. (1989) demonstrated that this heat source could be related to a low-velocity zone in the crust at about 10–15 km depth. In this zone, strong CO₂ degassing activity may occur, and deep gases escape towards the surface. The macroscopic effect of the degassing activity has been identified in the CO₂ bubbling which strongly affects many springs in the Bohemian Massif and the Vogtland.

The mineral springs of Bad Brambach are very useful for investigating earthquake prediction methods on a geochemical basis, because of their chemical properties (Table 1) as well as their location near the most frequent epicenter of the microearthquakes. This epicentral area is located near Kraslice, Western Bohemia, about 20 km NE of Bad Brambach (Neunhöfer & Güth 1988). Furthermore, the area has been explored thoroughly during the last 15 years by means of several modern techniques such as geophysics and isotope hydrogeology (Fröhlich & Gellermann 1989, Fröhlich et al. 1988, Gellermann & Gast 1983, Koch & Tauchert 1986, Watznauer & Koch 1989, Weinlich et al. 1993).

Measurements

In 1989 we started a continuous measurement program of radon in spring water and soil air, as well as of hydrological and meteorological parameters (Fig. 3). After a three-year measuring period it was concluded that soil air is not very suitable to investigate the connection between radon behaviour in the ground and seismic events. Radon in soil air shows a high sensitivity to atmospheric pressure, temperature, soil moisture, and groundwater level fluctuations. This dependence may be expressed by a linear regression function (Heinicke & Koch 1993, Koch et al. 1992). In a comparison between a synthetic radon curve calculated by the regression parameters and the measured values, no typical earthquake-induced anomalies were observed.

The measurements in the spring water are accomplished in the 'Radonquelle' of Bad Brambach (²²²Rn activity 25 kBq/l). Because of this high activity it is possible to detect the gamma activity of the radon daughter products as a radon equivalent (Flexer et al. 1985). A commercial gamma scintillation probe was installed directly in the well, about 60 cm below the water level.

The spectrum of the measuring values was statistically checked, for example by auto- and cross-correlation methods. Furthermore, only values above the one sigma level are interpreted as anomalies. The periodical fluctuations from week to week are due to anthropogenic effects, e.g. damming and removal of

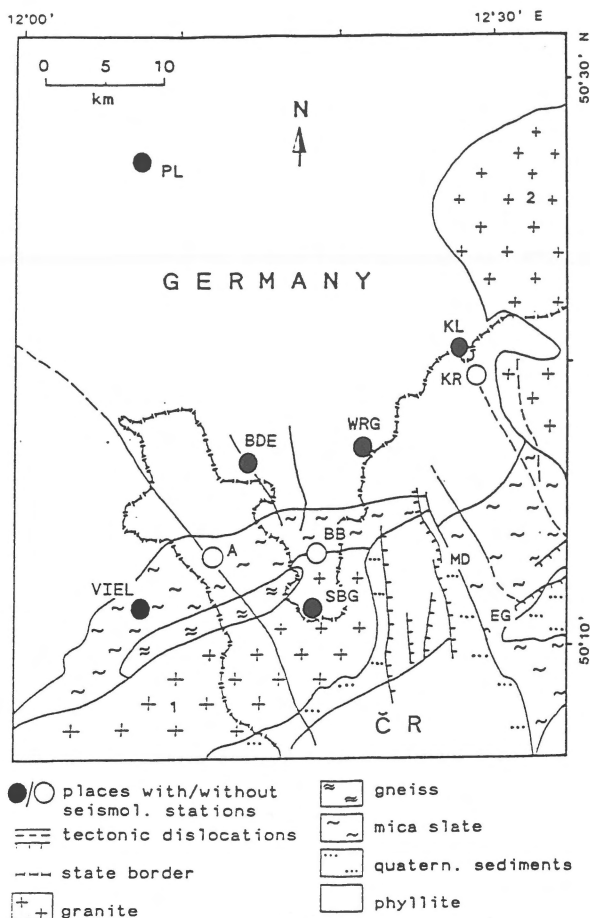


Fig. 1. Measuring area of Bad Brambach (Vogtland) with seismological stations (after Schmedes & Antonini 1991). 1 – Fichtelgebirge granite, 2 – Eibenstock granite, EG – Eger Graben, MD – Marienbad dislocation, A – Aš, BB – Bad Brambach, BDE – Bad Elster, KL – Klingenthal, KR – Kraslice, PL – Plauen, SBG – Schönberg, VIEL – Vielitz, WRG – Wernitzgrün, ČR – Czech Republic.

spring water for therapeutic purposes (2000 l per week, i.e. 8% of the total discharge).

Since April 1992, an improved measuring system has continuously registered the Rn and CO₂ concentrations, electric conductivity, and water flow rate at two mineral springs of Bad Brambach, the 'Radon' and the 'Eisenquelle'. The measuring interval is 15 minutes. The values are recorded by a personal computer.

The results show a good correlation between the parameters CO₂ and radon within an interval measured during a seismically quiet period (Fig. 4). Simultaneous fluctuations are evident in both curves due to anthropogenic influences (pumping effect, water removal).

Results and discussion

Within a measuring period of about three years we found some significant radon anomalies in the spring water before, during, or after microseismic events in the region. Figure 5 shows an example of these data. The used daily averages were calculated from 36 single values per day (measuring interval 40 min). It is assumed that the time of an occurring anomaly depends on the location of the epicenter within the region. No direct dependence on the local magnitude has been observed.

Former Ra-Rn residence time investigations as well as the data interpretation of uranium exploration shafts in the area had shown that the origin of the very high activity of the 'Radonquelle' – a uranium (radium) deposit – is situated at relatively low depth (< 200 m), which is not directly influenced by the tectonic stress field (Fig. 6; Dörr 1991, Fröhlich & Gellermann 1989, Gellerman & Gast 1983, Koch & Tauchert 1986, G. Lange 1992 pers. comm., Watznauer & Koch 1989). It may be assumed that the radon cannot reach the surface by its own force within a relatively short time (some hours to a few days) which enables the formation of a spike-like anomaly. Thus, a different 'transmitter' must be responsible for the signal from the ground due to a seismic event. Such a transmitter could be the CO₂ as a typical component of all the mineral waters in the Bad Brambach area (Table 1). Here, the ratio of the gas volumes V_{CO_2}/V_{Rn-222} plays an important role. It amounts to about $2.2 \cdot 10^{12}$ in the case of the 'Radonquelle'. This means that this spring emits about 1320 m³ CO₂ but only 0.6 mm³ ²²²Rn per year. In particular, tritium and uranium analyses show that the spring waters consists of two main water components, an old mineralized part and a younger one (20–60%, Table 1). The CO₂-enriched old water comes from greater depth because the $\delta^{13}C$ values of the spring gas indicate mantle-derived CO₂ (– 3.5 to – 3.7‰; Dörr 1991, Weinlich et al. 1993).

It may thus be assumed that CO₂-enriched groundwater reflects variations in the strain/stress field built up at seismogenic depths (Shapiro et al. 1982).

Thus, a first model conception may be proposed for the formation of an Rn/CO₂ anomaly generated by a local seismic event (Heinicke et al. 1993). The tectonic strain/stress field triggers the CO₂ generation, and the newly formed gases escape towards the surface acting as a gas carrier for radon. ²²²Rn is subject to natural radioactive decay, but the gas carriers are also continuously supplied with radon originating from relatively

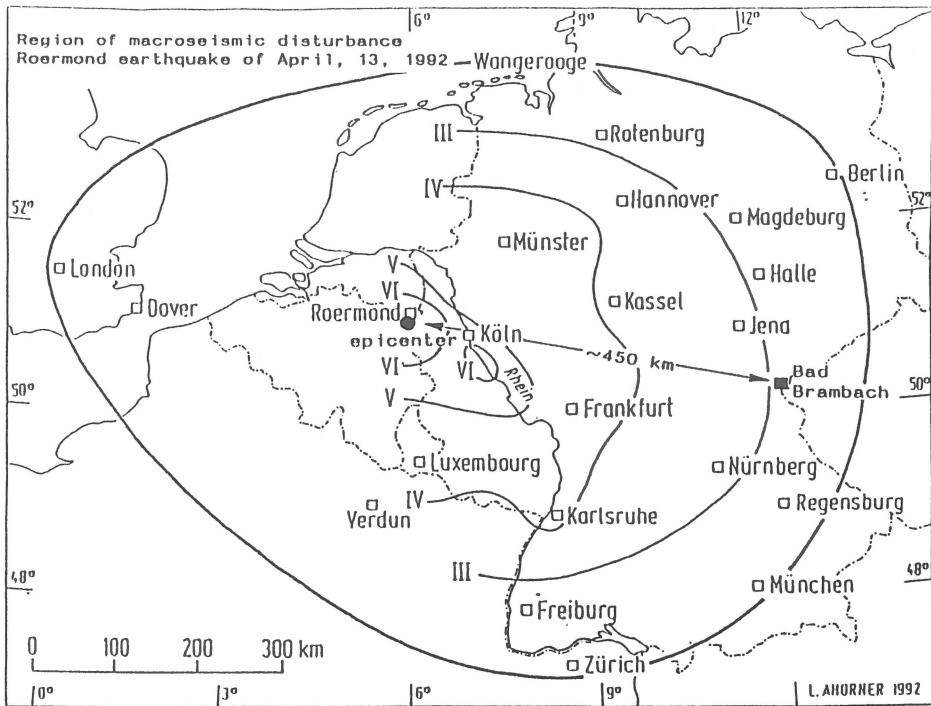


Fig. 2. Location of Bad Brambach in relation to the Roermond earthquake epicenter and the preliminary isoseismal contours (after Ahorner 1992).

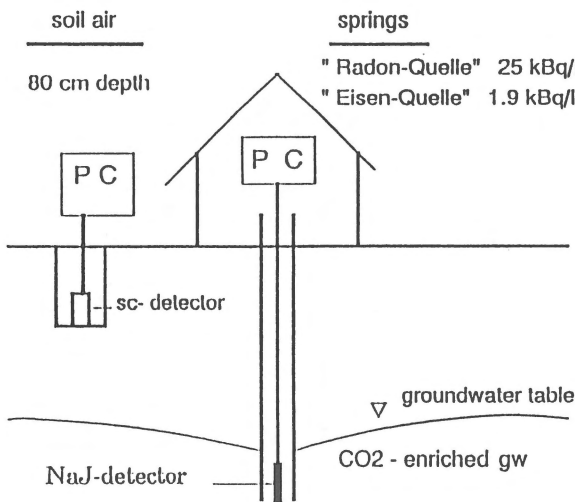


Fig. 3. Schematic overview of the measuring principle at Bad Brambach. Personal computers (PC) record measurements of temperature, air pressure, precipitation (humidity), discharge, conductivity, radon (sc-detector and NaI-detector), CO₂ and groundwater (gw) level.

al. (1992) experimentally confirmed the ability of CO₂ to cause such a process. Toutain et al. (1992) reported about a similar phenomenon after having monitored volcanic gas emanations. The spike-like form of the observed radon anomalies in the 'Radonquelle' of Bad Brambach reflects the above model conception very well. It confirms the assumption of an outburst-like CO₂ release from the depth within a very short interval. This hypothesis is supported by Irwin & Barnes (1980) who found that most of the significant seismic areas of the earth are characterized by many springs with CO₂ of magmatic origin. Thus it should be useful for earthquake prediction research to observe continuously such typical CO₂ sources with regard to variations in CO₂ concentration.

In another case, an anomaly effect was observed in the period of the Roermond earthquake of April 13, 1992 (1.20 h, GMT). Figure 7 first shows a radon anomaly caused by a single microquake in the Vogtland area (February 1992) which is followed by the large 'Roermond anomaly' of April 1992. The microquakes after the Roermond event caused only a small radon peak, whereas a typical swarmquake (end of June

shallow accumulations. This could explain the very close correlation between CO₂ and ²²²Rn. Varhegyi et

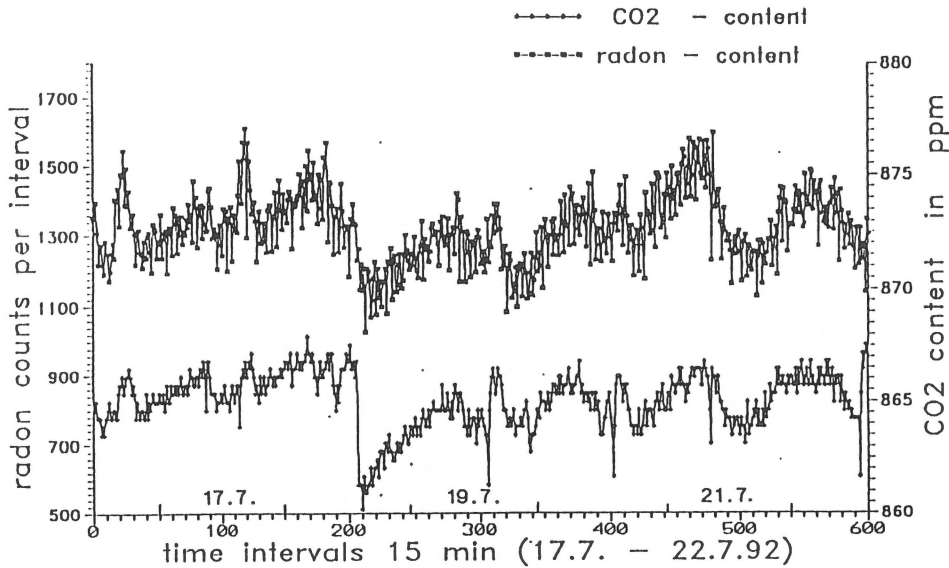


Fig. 4. First results of continuous measurements of ^{222}Rn together with CO_2 at the mineral spring 'Eisenquelle' during a seismically quiet period, showing a good correlation between ^{222}Rn and CO_2 concentration.

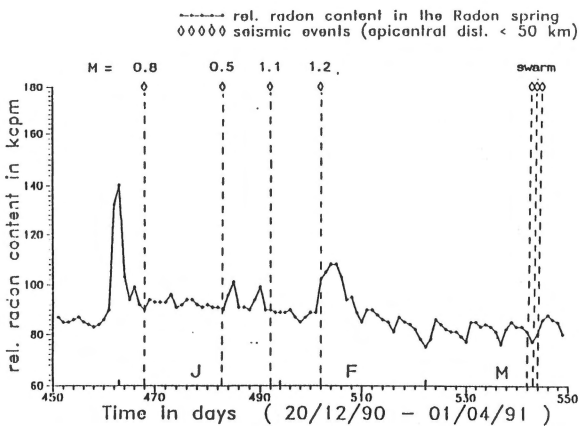


Fig. 5. Example of typical radon anomalies in spring water of the 'Radonquelle' in connection with micro-earthquakes (\diamond). The earthquake swarm of March 1991 is not marked by an anomaly. M = local magnitude.

1992) practically did not influence the radon behaviour in the spring water.

The significant radon increase which followed the Roermond event started about one hour after the quake (maximum about 8 hours after), followed by several peaks some days later. Possibly, the seismic energy of the Rayleigh waves of the Roermond earthquake generated an increased radon emanation in connection

with an enhanced CO_2 degassing. The basis of this effect is the destruction of the binding forces of the gases (Rn , CO_2) in a gas-rock interaction. Thus, the seismic wave motion could act in the same way as a beat on a bottle of mineral water.

It is true that anomaly effects such as in the period of the Roermond event are not suitable for 'earthquake prediction'. However, they are of great interest for the understanding of those processes which generate precursor phenomena within the crust. If seismic waves, depending on their frequency and energy, can cause an anomalous fluctuation of radon and CO_2 , then we may apply this approach with high probability to depth processes as well. Thus, for example, small cracks occurring before an earthquake could show similar effects near the hypocenter (Scholz 1990). This conception would agree with the occurrence of CO_2 anomalies as discussed above.

Conclusions

The results of continuous radon measurements in the Bad Brambach area demonstrated that radon in spring water is a suitable tracer for seismic information from the crust. The Roermond event, most probably, induced a radon anomaly at our measuring site simultaneously with the earthquake. It was followed by smaller anoma-

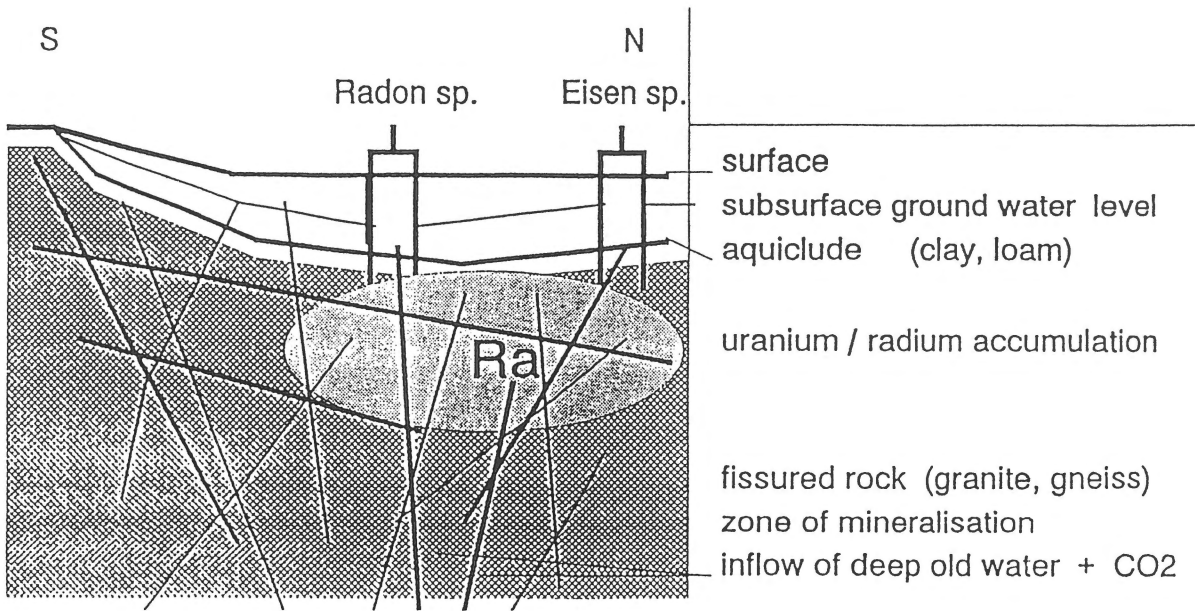


Fig. 6. Koch & Heinicke 'Radon behaviour...'

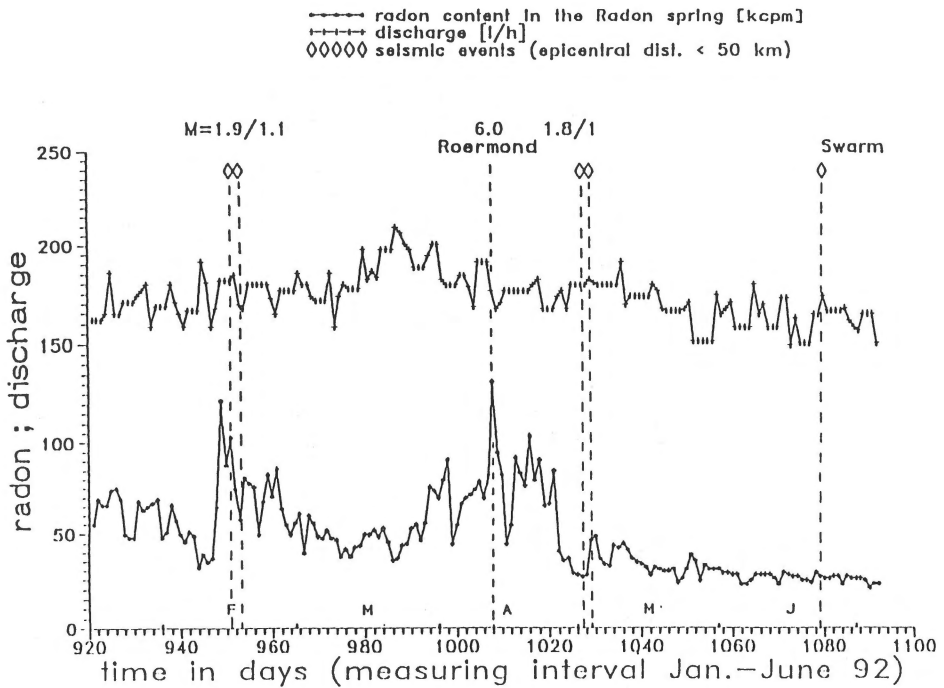


Fig. 7. Example of radon anomalies in spring water of Bad Brambach in the temporal vicinity of microearthquakes with Vogtland origin and of the 1992 Roermond event (epicentral distance about 450 km). In the latter case the radon concentration started to increase about 1 hour after the earthquake and reached its peak 8 hours later.

lies. We suggest that radon alone is not able to penetrate the water up to the surface from depths characterized by geodynamic processes, within the time necessary to induce the observed radon concentration changes. An additional gas carrier should be responsible for such a short-time reaction. This is CO₂ at our measuring site. In rare cases it may be methane or nitrogen as well (Martinelli 1991, Martinelli & Ferrari 1991). Obviously the anomalies did not originate from stress transfer from the hypocenter but from ground vibrations (Rayleigh waves) during the event. This conception could illustrate one of the processes responsible for the appearance of CO₂/Rn anomalies as an earthquake precursor effect.

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References

- Ahorner, L. 1992 Das Erdbeben bei Roermond am 13. April 1992 und die daraus zu ziehenden Lehren für das Erdbebengefährdungspotential im Rheinland – Mitt. Deutsch. Geophysik. Ges. 1–2: 51–57
- Čermak, V. 1979 Review of heat flow measurements in Czechoslovakia. In: V. Čermak & L. Rybach (eds.): *Terrestrial Heat Flow in Europe* – Springer, Berlin, Heidelberg: 152–160
- Dörr, H. 1991 Zusammenfassung der Ergebnisse der Probenahme im Kurpark in Bad Brambach am 24.08.90 – Technical Report, Heidelberg University, 8 pp
- Flexer, S., H.A. Wollenberg & A.R. Smith 1985 Radon in groundwater of the Long Valley Caldera, California – Lawrence Berkeley Lab., Report LBL-23301, Berkeley, 21 pp
- Fröhlich, K. & R. Gellermann 1989 Isotopenphysikalische Untersuchungen zur Hydrogeologie und Genese der Quellwässer Bad Brambachs. In: H. Jordan (ed.): *Radontherapie heute* – Abh. Sächs. Akad. Wiss. Leipzig 57: 58–68
- Fröhlich, K., D. Hebert & R. Gellermann 1988. Umweltnuklid Tritium in Mineralwässern von Bad Brambach – *Isotopenpraxis* 24: 163–164
- Gellermann, R. & H. Gast 1983 Ra-Rn-Datierung der Quellwässer von Bad Brambach – *Z. Physiother.* 35: 129–135
- Heinicke, J. & U. Koch 1993. Dependence of radon anomalies from seismicity and other parameters in Bad Brambach. Proc. 2nd Workshop on Radon Monitoring in Radioprotection, Environmental and/or Earth Sciences, Trieste, Nov.–Dec. 1991: 536–541
- Heinicke, J., U. Koch & G. Martinelli 1993 Investigations of the connection between seismicity and ²²²Rn-CO₂ content in spring waters at the Vogtland area (Germany): First results. In: *Activity Rep. 1990–1992 and Proc. XXIII General Assembly Eur. Seismol. Comm.* – Prague: 317–323
- Hill, D.P., P.A. Reasenbergh, A. Michel et al. 1993 Seismicity remotely triggered by the magnitude 7.3 Landers, California, earthquake – *Science* 260: 1617–1623
- Hurtig, E. & Ch. Oelsner 1979 The heat flow on the territory of the German Democratic Republic. In: V. Čermak & L. Rybach (eds.): *Terrestrial Heat Flow in Europe* – Springer, Berlin, Heidelberg: 186–190
- Irwin, W.P. & I. Barnes 1980 Tectonic relations of carbon dioxide discharges and earthquakes – *J. Geophys. Res.* 85, B6: 3115–3121
- Kämpf, H., P. Bankwitz, P. Vogler, G. Strauch, W. Michler & D. Tauchert 1987 Seismo-hydrochemical investigations on spring of mineral water Marie I (Bad Elster/GDR) before and during the earthquake swarm activity 1985/86. Proc. Workshop on Earthquake Swarm in Western Bohemia Dec. 1985–Feb. 1986 – Mariánské Lázně: 79–88
- Kämpf, H., K. Bräuer, U. Koch, M. Malkovsky, G. Strauch, F.H. Weinlich & S. Weise 1992 Vulkanismus - Mineralwässer - Seismizität im Bereich der Marienbader Störungszone. In: *Exkursionsführer 1. Jahrestagung Ges. f. Geowiss. e.V. – Geoforschungszentrum Potsdam*: 129–155
- Kämpf, H., W. Seifert & M. Ziemann 1993 Mantel-Kruste-Wechselwirkungen im Bereich der Marienbader Störungszone. Teil 1: Neue Ergebnisse zum quartären Vulkanismus in NW-Böhmen – *Z. Geol. Wiss.* 21, 1/2: 117–134
- Kämpf, H., H. Strauch, P. Vogler & W. Michler 1989 Hydrological and hydrochemical changes associated with the December 1985/January 1986 earthquake swarm activity in the Vogtland/NW Bohemia seismic area – *Z. Geol. Wiss.* 17: 685–698
- Koch, U. & D. Tauchert 1986 Ermittlung von Radon-Verweilzeiten an natürlichen Bad Brambacher Wässern – *Z. Physiother.* 38: 387–396
- Koch, U., J. Heinicke, K. Fröhlich & M. Krbetschek 1992 Indikator Radon: Radon im Oberen Vogtland und seine geowissenschaftliche Anwendung – *Wissenschaft und Fortschritt* 42: 253–255
- Kuschka, E. 1993 Das Zentralsächsische Lineament im Südwestvogtland – Tektonik und Mineralisation – *Z. Geol. Wiss.* 21, 1/2: 163–169
- Liu, Lan-Bo, E. Roeloffs & Xiang-Yuan Zheng 1989 Seismically induced water level fluctuations in the Wali Well, Beijing, China – *J. Geoph. Res.* 94, B7: 9453–9462
- Martinelli, G. 1991 Fluidodynamical and chemical features of Radon 222 related to total gases: Implications on earthquake prediction topics – Report, Int. Atomic Energy Agency (IAEA) Vienna, 22 pp
- Martinelli, G. & G. Ferrari 1991 Earthquake forerunners in a selected area of Northern Italy: recent developments in automatic geochemical monitoring – *Tectonophysics* 193: 397–410
- Neunhöfer, H. & D. Güth 1988 Mikrobeben seit 1962 im Vogtland. Seismologische Aspekte und Beziehungen zur lokalen Geologie – *Z. Geol. Wiss.* 16, 2: 135–146
- Pazdera, A. & D. Procházková 1987 Changes in ground water level observed in connection with the earthquake of Dec. 21, 1985. In: *Western Bohemia. Proc. Workshop on Earthquake Swarm in Western Bohemia Dec. 1985–Feb. 1986* – Mariánské Lázně: 76–78
- Schmedes, E. & M. Antonini 1991 Alpen und Böhmisches Masse. In: *Erdbeben in der Bundesrepublik Deutschland 1985* – Bundesanstalt für Geowissenschaften und Rohstoffe, Hannover: 26–45
- Scholz, C.H. 1990 The mechanics of earthquakes and faulting – Cambridge Univ. Press: 439 pp

- Shapiro, M.H., J.D. Melvin, T.A. Tombrello, Jiang Fong-Liang, Li Gui-Ru, M.H. Mendenhall, A. Rice, S. Epstein, V.T. Jones, D. Masdea & M. Kurtz 1982 Correlated radon and CO₂ variations near the San Andreas Fault – *Geoph. Res. Letters* 9: 503–506
- Toutain J.P., J.-C. Baubron, J. LeBronec, P. Allard, P. Briole, B. Marty, G. Miele, D. Tedesco & G. Luongo 1992 Continuous monitoring of distal gas emanations at Volcano, Southern Italy – *Bull. Volcanol.* 54: 147–155
- Varhegyi, A., J. Hakl, M. Monnin, J.P. Morin & J.L. Seidel 1992 Experimental study of radon transport in water as test for a transportation microbubble model – *J. Appl. Geophys.* 29: 37–46
- Watznauer, A. & U. Koch 1989 Die geologischen und hydrogeologischen Verhältnisse im Quellgebiet von Bad Brambach. In: H. Jordan (ed.): *Radontherapie heute*, – *Abh. Sächs. Akad. Wiss. Leipzig* 57: 49–58
- Weinlich, F., K. Bräuer, H. Kämpf, G. Strauch & St. Weise 1993 Mantel-Kruste Wechselwirkung im Bereich der Marienbader Störungszone. Teil 2: Gasgeochemische Untersuchungen an Mineralquellen entlang eines Profils über das Eger-Rift – *Z. Geol. Wiss.* 21, 1/2: 135–142