Gas geochemistry

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Science Reviews

British Library Cataloguing in Publication Data
Gas Geochemistry
A catalogue record of this book is available from the British Library

ISBN 0-905927-79-6

Production editor: Sara Nash
Typeset by: DL Technology, PO Box 9, Buckhurst Hill, Essex, IG9 5BE

Preface

The Second International Colloquium on Gas Geochemistry was organized by the University of Franche-Comté (the Laboratory of Nuclear Microanalysis on the U.F.R.'s Besançon campus, and the Laboratory for Technical Interface Metrology on the Montbéliard campus) and was held between 5 – 9 July, 1993.

This Colloquium, which brought together 135 scientists from 23 different countries, aimed to promote the knowledge of the various applications of gas geochemistry. The Colloquium provided participating scientists with a forum to present and discuss their work, the main results of which are published in the official Proceedings. The issues raised include: the study of rare gases (Rn, He, Ar, *etc.*), their various applications in the earth sciences (geological and hydrogeological prospecting, *etc.*), environmental sciences, and natural and industrial risk analysis. It is worth emphasizing the fact that one of the common threads running through many of papers was radon, a radioactive gas which, although a very effective tracer for applications in the earth sciences, can also represent a danger to the environment in terms of the health of the population.

The richness and diversity of the work presented here demonstrates the interdisciplinary nature of the subject, and the need for just such an event in order to contrast points of view and to permit exchange of knowledge and knowhow.

The smooth running of this Colloquium was due to the excellent cooperation of all the members of the organizing committee and students of the Laboratory of Nuclear Microanalysis, as well as to the support of a number of organizations, public or private, without which this event could not have taken place. In particular, we would like to thank: the General Council of the Doubs ; the Montbéliard Regional Urban District; the University of Franche-Comté ; the French Ministry of Foreign Affairs; the InterUnec Association at Besançon; the International Science Foundation; the Faculty of Law and Economics at Besançon; and the Physical Measurements Department at Montbéliard for providing the facilities.

Moreover, we would like to thank all the firms who, by their very presence, demonstrated their interest in this event, in particular, the Génitron, Algade, Mégarad and Dosirad companies.

This Colloquium began to take a concrete shape with the submission for publication of most of the papers presented. Fifty manuscripts accepted by the reviewers are the subject of this volume, which we encourage you to read.

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Radon and underground climate in the Moestroff Cave

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Resumé

Les variations des concentrations du radon dans la grotte de Moestroff montrent un minimum en été et un minimum moins marqué en hiver. Un modèle de la dynamique de l'air tenant compte de l'existence d'un réseau étroit situé en dessous du labyrinthe principal permet d'expliquer ces variations. Si l'on veut corréler les concentrations de radon aux autres variables climatiques, on est amené à distinguer les mois où la température externe dépasse la température à l'intérieur de ceux où elle est inférieure. Alors que la concentration moyenne du radon diminue avec l'accroissement de la température extérieure pour la station la plus proche de l'entrée, elle passe par un maximum à la station la plus reculée. Les concentrations moyennes du radon sont corrélées positivement au taux du CO_2 *; aucune corrélation n'a pu être trouvée avec les chutes de pluie et la pression atmosphérique moyenne. Les concentrations du radon augmentent avec l'éloignement de l'entrée ; ceci de façon linéaire lors des mois chauds et exponentielle lors des mois froids.*

The Moestroff Cave

The Moestroff maze cave is located at an altitude of 250 m in a plateau separating the valleys of the rivers Our and Sauer in the Grand-Duchy of Luxembourg. The extremely narrow horizontal galleries (mean diameter less than 70 cm) of the cave lie at a depth of about 30 m in the upper Muschelkalk.; the topsoil is a narrow layer of Keuper marls. The entrance to the 3.5 km of accessible galleries lies 12 meters up in a vertical cliff. The bottom of the galleries is covered by heavy clay whereas the ceilings all have longitudinal fissures. There are no flowing waters in the cave, but the atmosphere is extremely humid, approaching practically everywhere 100% relative humidity; the mean yearly temperature is about 10°C.

Many cracks in the bottom of the galleries are leading downwards, but are too narrow and irregular for exploration. In the cave, crawling is the only possible displacement method, which makes progression rather slow and strenuous (Phymoes, 1992).

The extreme confined galleries imposed to build a fully automatically working

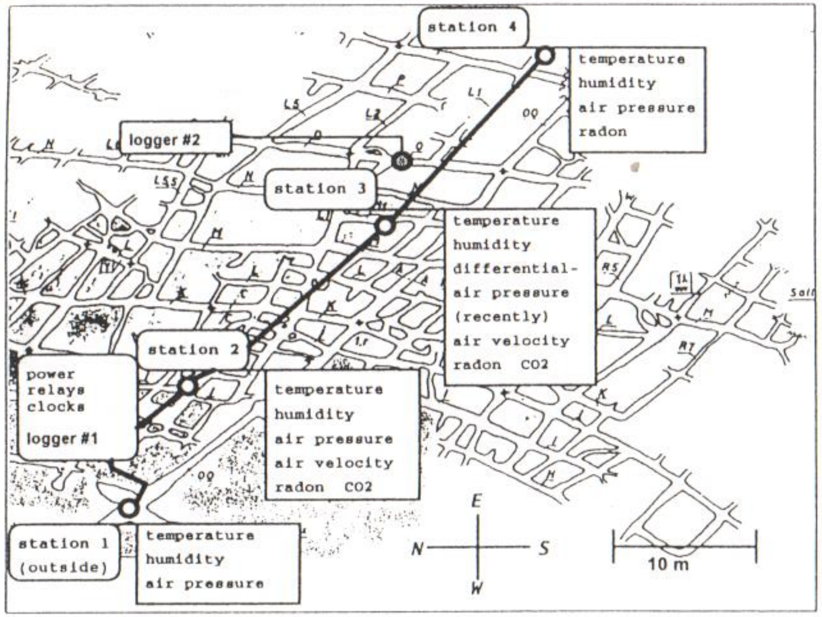

Figure 1 *Sensor stations for measurement of climatic parameters in the Moestroff Cave; the galleries shown represent only a small part of the total cave.*

measurement system, and since July 1992 three measuring stations are functioning in the main gallery of the cave. Figure 1 gives the overall structure of this climatological data net.

The seasonal radon pattern

Some individual measurements showed radon concentrations up to 20 kBq m^{-3} in deep cave locations. These radon concentrations are comparable to those measured in some caves in France and Switzerland (Robe, 1992; Rybach *et al.*,1990). The high radon concentrations are due to the very favourable surface to volume ratio in the Moestroff cave.

To get a clearer picture of the seasonal radon patterns, monitoring began in February 1992. These measurements are done with Karlsruhe Solid State Nuclear Track detectors, usually exposed for 3 weeks. All the radon concentrations discussed in this paper are mean concentrations over that time interval. Working Level spot measurements are made at irregular intervals with a portable working-level meter (Radon-Sniffer, Thomson & Nielsen Electronics Ltd., Canada).

Many authors (Hunyadi, 1991; Middleton, 1991; Wilson *et al.*,1991) who measured radon or radon progeny concentrations in caves report high summer and

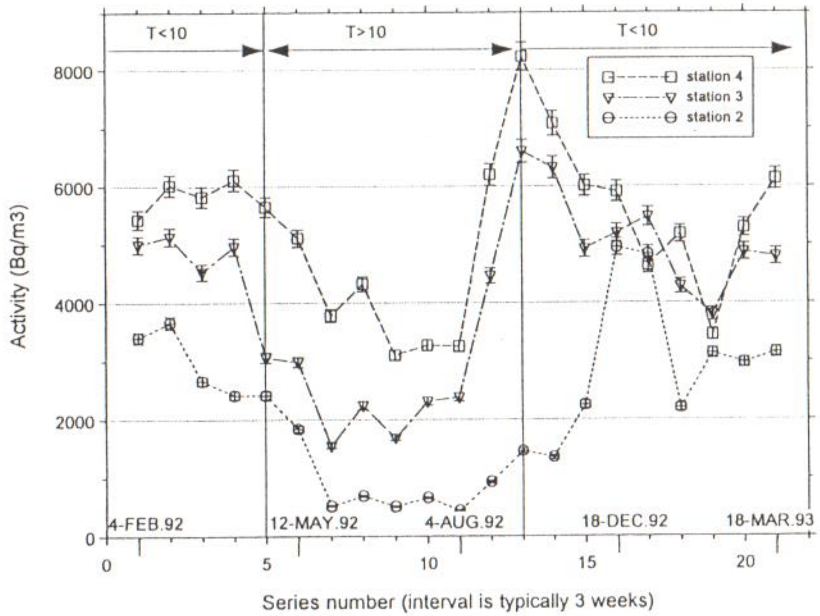

Figure 2 *Seasonal radon activity pattern at 3 stations in the main gallery of the Moestroff Cave.*

low winter concentrations. For a cave system with a predominantly horizontal structure and many fissures and path-ways opening upwards, this has to be expected. In these caves there exist a strong chimney effect during the colder months, flushing the cave with fresh air; during summer time, air movements are restricted or inverted and hence the concentrations higher. As shown by Figure 2, the opposite occurs in the Moestroff cave.

The radon concentration is low in summer when the external temperature exceeds the mean cave temperature (10°C); a less pronounced minimum occurs in winter. An analysis of the measurements distinguishing the periods when the external temperature is lower and when it is higher than the mean cave temperature shows that the three stations do behave quite differently:

- when the external temperature rises, but remains lower than 10°C, the radon concentration falls at station 2, remains practically constant at station 3 and goes up at the farthest situated station 4;
- when the external temperature is higher than 10°C, the concentrations at all stations fall with increasing temperature.

The linear correlations between radon concentration and external temperature are significant at the 0.05 level for station 2 and for the values of station 3 corresponding to temperatures higher than 10°C; at station 4, the concentrations over the global

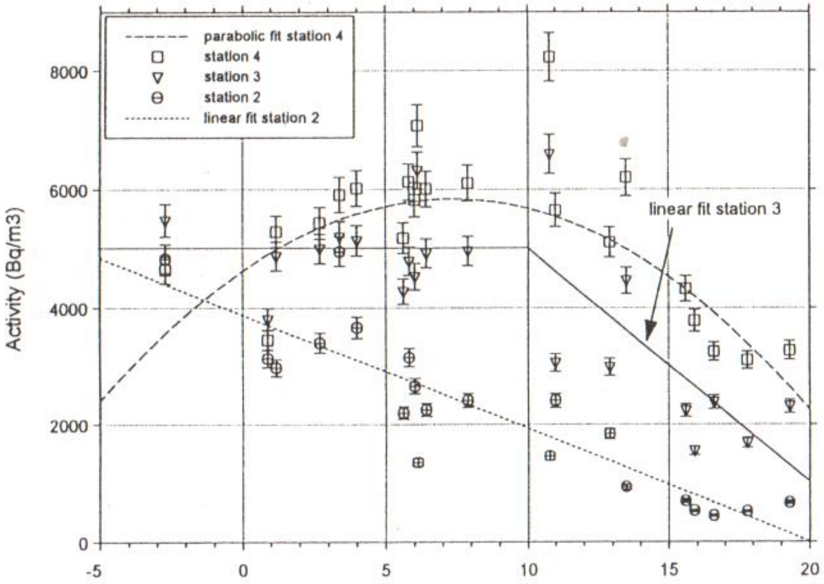

Figure 3 *Radon concentrations at stations 2, 3 and 4 when the outside air temperature is lower or higher than 10°C.*

temperature range are best fitted by a parabola; the maximum radon concentrations correspond to a near zero temperature difference between inside and outside.

Discussion

The seasonal radon changes at the 3 stations can be explained by a possible multistorey structure of the cave: the accessible upper storey may lie about 3m and 5m above two deeper storeys (Figure 4), which are probably inaccessible but communicate with the upper storey through numerous cracks and fissures.

During the summer months, hot air flows in through the main entrance, cools down and flows out through the deeper situated storeys. This pattern reverses during the colder months when warm cave air is replaced by cold air inflowing through the lower openings in the cliff (Figure 4). The existence of this seasonal airflow pattern has been verified by numerous checks with lighted incense sticks.

At station 4, which is best isolated from disturbances coming in through the main entrance or through the deeper storeys, minimum air circulation means maximum radon concentration; this explains the parabolic relationship between radon concentration and external temperature.

Near the entrance (station 2), radon levels decline continuously with rising

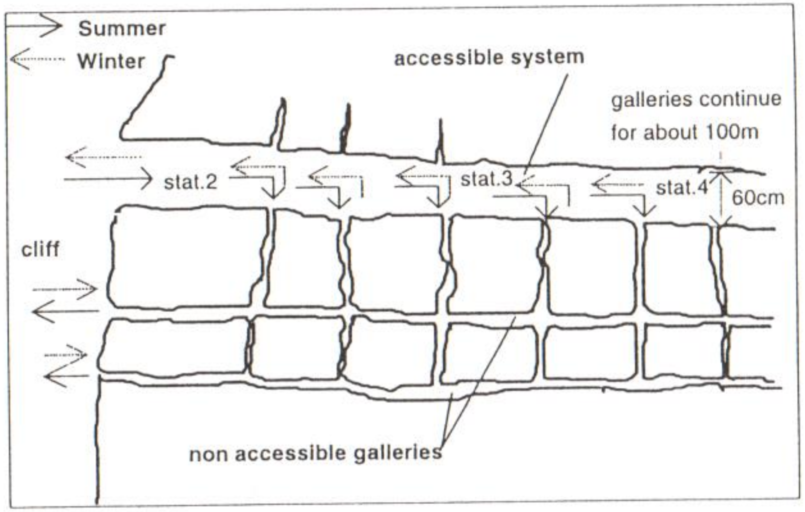

Figure 4 *Summer- and wintertime wind pattern in Moestroff Cave.*

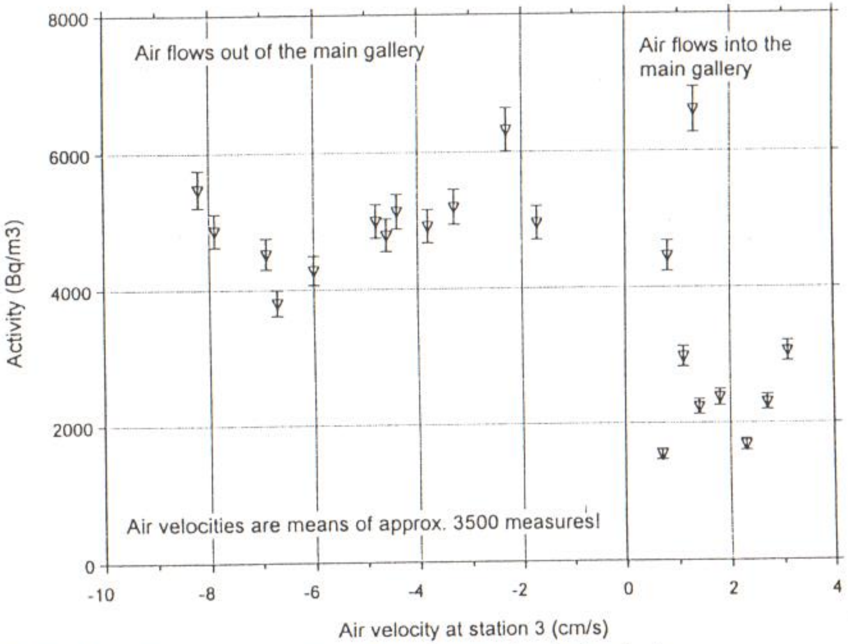

Figure 5 *Radon concentration at station 3 versus local air velocity.*

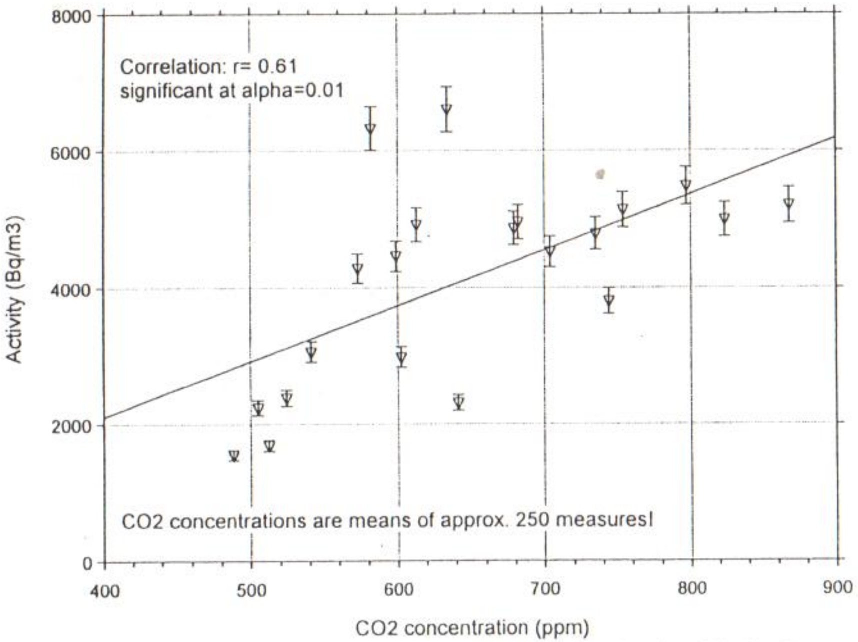

Figure 6 *Radon concentration at station 3 is positively correlated to CO_2 levels.*

external temperature. In winter thermal induced air circulation brings radon enriched air up from the lower storeys; this air flows out to the entrance, so that the concentrations there are highest when this airflow is greatest, i.e. when the external temperatures are lowest. When the external temperature exceeds 10°C, airflow reverses and the fresh air blowing into the cave through the main entrance brings further down the mean radon concentration at station 2.

Figure 5 represents the radon concentration at station 3 versus the local air velocity; for a same velocity, the air inflowing through the main entrance (summer) depresses radon levels much more as does the air uprising from the lower galleries (winter).

Out of the measured parameters, the temperature difference between external air and cave air is the dominant factor governing the wind activity and as a consequence the radon concentrations in the cave; this has been reported by many authors (Hunyadi, 1991; Smart and Atkinson, 1990; Andrews, 1991; Gunn, 1991).

The influence of other parameters

The relative humidity in the cave atmosphere is practically constant at a very high level (95 to 100%), and is not influenced by external rainfall; no correlation between radon concentration and rainfall could be found.

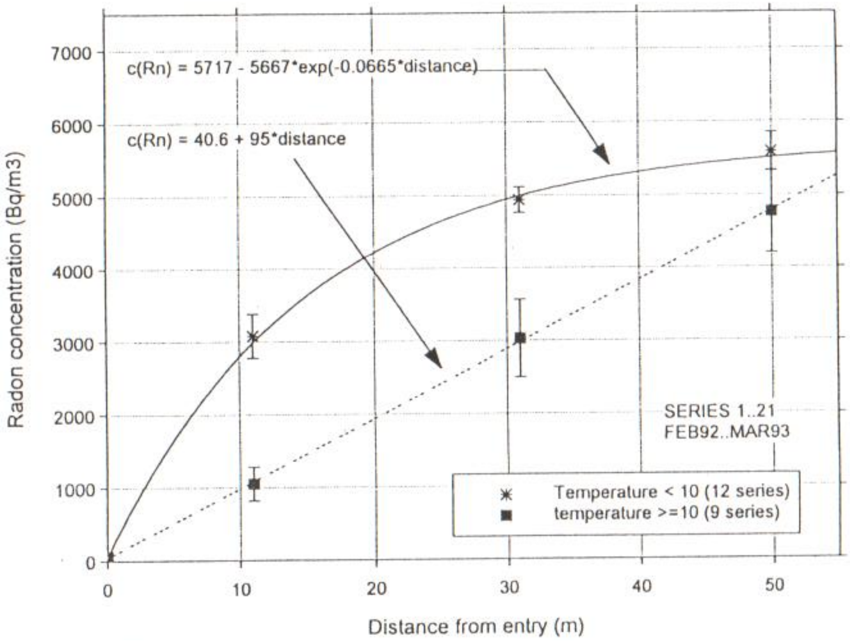

Figure 7 *Mean radon concentrations versus distance from entry.*

Most reports show that instant radon levels rise with decreasing atmospheric pressure. The three week integrating measurements can not control this effect; actually no significant correlation between mean radon levels and the mean air pressure could be found.

The carbon dioxide concentration is measured at station 3 by a very precise (3%) NDIR sensor since March 1992 (one measure every two hours). The positive correlation between the mean CO_2 levels and the radon concentration is significant at the 0.01 level, as shown by Figure 6, where every data-point represents the mean of about 250 CO_2 measurements. Similar results have been reported by Shapiro (Shapiro *et al.*, 1984) at Alandale and Lytle Creek in the USA.

At station 3 the simple linear relationship

$$c(Rn) = -\ 1{,}153 + 8.14 \times c(CO_2)$$

with c(Rn) in Bq m^{-3} and $c(CO_2)$ in ppm means an increase of about 8 Bq m^{-3} of radon per ppm CO_2.

The mean radon levels versus the distance from entry clearly show two different patterns, depending on the external temperature:

if $T_{ext} > T_{int}$, radon levels increase linearly with distance,

if $T_{ext} < T_{int}$ the higher mean values are best fitted by an exponential model.

This latter model has been used by Wigley and Brown (1976) to compute the temperature of the cave air, and also applies to the temperature profile of the Moestroff cave. The relaxation distance, *i.e.* the inverse of the damping parameter, is 15 m for the radon levels, close to the 15.4 m corresponding to the temperature profile in March 1993.

Conclusions

Our long-time study reveals that interactions of meteorological and climatic factors affect radon concentrations on a seasonal time-scale.

The mean radon concentrations are mostly influenced by mean temperature differences. The seasonal radon pattern give a clear hint to the existence of different air movements during the year. A distinction has to be made between the months where the external temperature is higher and those where it is lower than the cave air. The radon levels fall continuously with rising temperature at locations near the entrance, but a parabolic fit suits best for the deepest cave locations where highest radon levels are measured when $T_{ext} = T_{int}$. The radon concentrations are positively correlated to local carbon dioxide levels.

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