Prediction of areas presenting a high radon exhalation potential: A new methodology based on the properties of geological formations and soils

G. Ielsch, C. Ferry, G. Tymen[1] and M.-C. Robe

Institut de Protection et de Sûreté Nucléaire, DPRE/SERGD/LEIRPA, BP. 6, 92265 Fontenay-aux-Roses, France
[1] Laboratoire de Recherches Appliquées Atmosphère Hydrosphère, Université de Bretagne Occidentale, UFR Sciences et Techniques, 6 avenue Le Gorgeu, BP. 809, 29285 Brest cedex, France

Abstract. A research program carried out since 1997 produced a methodology for predicting areas with a strong potential for radon exhalation at the soil surface. This methodology is based on a quantification of the Rn exhalation rate, from a precise characterization of the main local geological and pedological parameters that control the radon source and its transport to the soil/atmosphere interface. It combines a cross mapping analysis of parameters used in a Geographic Information System with a model of the vertical transport of Rn by diffusion trough the soil. This code *(TRACHGEO)* calculates the radon flux density at the surface as a function of the properties of the rock and the soil. This approach is validated in 4 typical areas with different geological contexts, starting from *in situ* measurements of radon fluxes and of radon concentrations in dwellings. A lithogeochemical classification of the geological formations as a function of their U contents and their confrontation to Rn level measurements demonstrate the primordial influence of the U content of the basement on Rn exhalation. This study leads to an initial map of the exhalation potential by assigning a potential class to each lithogeochemistry. Nevertheless, *in situ* radon measurements reveal a high spatial variability on uraniferous lithologies. Tests made by the *TRACHGEO* tool show the need to take account of spatial heterogeneity of soils (in addition of geochemistry) to improve the mapping resolution. The *TRACHGEO* forecasts explain the variability of the Rn exhalation on a larger scale.

1. INTRODUCTION

A research program has been carried out in France since 1997 in order to develop a methodology for predicting areas with a strong potential for exhalation of radon-222 at the surface of the soil [1] [2] [3]. Indeed, the objective is to define areas in which the density of buildings with high radon levels is likely to be highest. Identification of priority areas would then help to target actions taken by the government, and particularly screening measurements in existing buildings.

2. GENERAL METHODOLOGY

Our approach is based on the quantification of the radon-222 exhalation rate at the surface of the soil, from the knowledge of the geological and pedological parameters that influence the radon emanation and its migration until the atmosphere. We use a Geographic Information System, combined with a simplified model of the radon migration through the soil, which calculates the radon flux density at the surface, according to the chemical and physical properties of rocks and soils.

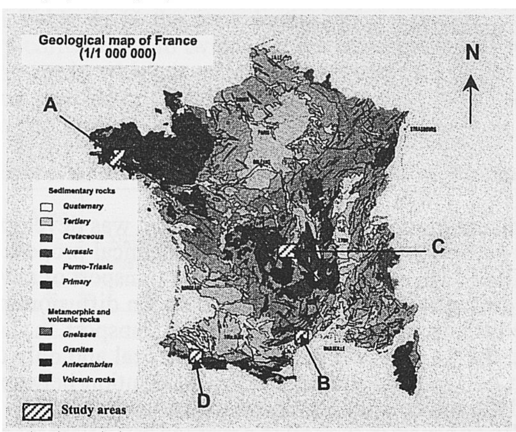

Figure 1. Location of the study areas selected for the methodology validation, on the geological map of France [4].

The main objective was to validate this methodology in 4 typical areas with different geological contexts,

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starting from *in situ* measurements of the radon exhalation rate at the soil surface. Measurements of the indoor radon concentration were also carried out. Each study area was about 30 km in breadth and 50 km in length (Fig. 1). In each transect, measurement paths were chosen in order to run across the different geological formations. The *in situ* radon level measurements and the acquisition of the geological and pedological data were carried out along these paths. The data was characterized and analyzed at a precise scale (1/50 000).

3. DATA ACQUISITION

3.1. In situ radon measurements

Measurement sites were selected along the paths defined in each area, usually at 1- to 3-km intervals. For each geological unit, one point was defined at least. For each site, two measurements of the radon flux density at the soil/atmosphere interface were performed with the « accumulation technique » [5] [6] [7]. The measurement uncertainty is around 20% and the detection threshold is 1 mBq m^{-2} s^{-1}. This data was collected in summer when the weather conditions were fairly stable.
Measurements of the indoor radon concentration were also realized in dwellings, where possible, located near the sites selected for radon exhalation rate measurement. The radon activity was determined by using track-etch detectors (LR115), placed in the main room (ground floor) and exposed for 2 months at least, in winter. The measurement uncertainty varies from 25% for a concentration below 10 Bq m^{-3}, to 4% for a concentration above 400 Bq m^{-3}.

3.2. Radon source term in the basement rocks

The uranium content of the rock corresponds to the radon source term in the basement and thus governs the radon exhalation potential. Therefore, our objective was to determine a range of probable U contents for each geological formation of the transects. The rock uranium content was estimated from the geological maps at scale 1/50 000 and from the chemical information available on these formations. The geological units were first classified according to their lithology. Then, a probable U content range was assigned to each lithological class, using two different approaches: the first one was direct and based on the chemical analysis of rocks sampled in precise geological units (units of the transects completed by analyses of other geological formations corresponding to the same lithology) and the second one (indirect or « lithogeochemical ») used estimations of the variability of U contents for a given lithology, published in the literature.

3.3. Soils characteristics

A soil sample was taken in each radon measurement site (at 50 cm depth) in order to determine its volumetric moisture and porosity (gravimetric method). The soil thickness was estimated from pedological drillings also carried out on each site. Furthermore, each soil sample radium-226 activity was analysed by gamma-ray spectrometry, using a Germanium hyper-pure detector, of N-type (61.9 mm crystal diameter), with a 40% relative efficiency and a resolution value of 1.95 keV at 1.33 MeV. The measurement uncertainty is about 10%.

4. DATA ANALYSIS

4.1. The TRACHGEO model

A simplified model of the radon migration through a soil column, was developed in order to calculate the Rn exhalation rate at the soil surface, from the physical and chemical properties of the rock and the soil. This model, named TRACHGEO (which stands, in French, for transport of radon through a homogeneous layer limited at its base by a geological formation), is based on Rn diffusion in the pore space of the soil. It is a simplified version derived from the non-steady radon transport in the unsaturated zone model, TRACI [8]. We assume that: the Rn transport in the soil is vertical and only due to diffusion in the pore space; there is no adsorption on the solid grains; the Rn distribution between liquid and gas phases is governed by Henry's law and the soil column is homogeneous.
In this case, the steady-state Rn transport in the soil is governed by (z vertical axis, directed upward):

$$D\cdot\frac{d^2C}{dz^2}-\lambda\cdot C+\lambda\cdot\frac{\rho_d\cdot E\cdot C_{Ra}}{p_{eff}}=0 \quad [9];$$

where C is the Rn concentration in the gas phase (Bq m^{-3}); D, the pore average Rn diffusion coefficient (m^2 s^{-1}); E, the Rn emanation factor (-); ρ_d, the dry bulk density of the soil (kg m^{-3}); C_{Ra}, the radium concentration in solid material (Bq kg^{-1}); λ, the radioactive decay constant of Rn (2.1 10^{-6} s^{-1}).
The effective porosity, p_{eff} (-), is given by [9]:

$$p_{eff}=p\cdot(1-m+k_H\cdot m)$$

where p is the total porosity of the soil (-); m, the soil moisture saturation (-) and k_H, the Henry's law constant (0.25).
At the surface of the soil, the radon concentration is zero. Radon concentration in the gas phase of the underlying rock, C^r is supposed not to be affected by transport and is given by:

$$C^r=\frac{\rho_d^r\cdot E^r C_{Ra}^r}{(1-m+k_H\cdot m)\cdot p^r}$$

where r refers to the rock properties (here, it is assumed that the rock moisture saturation is equal to the soil moisture saturation). By continuity, it imposes the Rn concentration in the soil gas phase at the soil – rock interface.
The Rn flux at the soil surface F is obtained by solving the Rn transport equation with the boundary conditions given above:

$$F=p_{eff}\cdot\sqrt{\lambda\cdot D}\cdot\left(\frac{C^s}{th(H/l_d)}+\frac{C^r-C^s}{sh(H/l_d)}\right)$$

where

$$C^s=\frac{\rho_d\cdot E\cdot C_{Ra}}{(1-m+k_H\cdot m)\cdot p}$$

$l_d=\sqrt{\frac{D}{\lambda}}$ is the Rn diffusion length in the soil (m) and H, the total height of the soil column (m).
The Rn pore average diffusion coefficient in the soil used in TRACHGEO, is given by [9]:

$$D=p\cdot D_o\cdot\exp(-6\cdot m\cdot p-6\cdot m^{14p})$$

where D_o is the Rn diffusion coefficient in free air (1.1 10^{-5} m^2 s^{-1}).

4.2. Geographic Information System

A Geographic Information System (G.I.S.) was used to analyze the spatial variations of the different information layers constituted by the geological and pedological data and the radon levels.

5. RESULTS AND DISCUSSION

The results of the radon level measurements (flux density at the soil surface and indoor concentration) are reported for each area in Table 1. In the area A, the radon exhalation rate is highly variable (9-837 mBq m^{-2} s^{-1}) and generally corresponds to twice or three times the world-wide average exhalation rate of radon at the surface of the earth, which is estimated between 16 and 26 mBq m^{-2} s^{-1} [10]. In the other transects, the Rn exhalation rates do not generally exceed 90 mBq m^{-2} s^{-1} and the mean Rn flux is similar to the mean value over the continents.

Table 1. Results of the Rn measurements realized at the soil surface in the 4 study areas.

Area	Rn flux density (mBq m^{-2} s^{-1})			
	Number of measurements	Range	Geometric mean	% of values ≥ 100 mBq m^{-2} s^{-1}
A	85	9 - 837	52	20
B	39	2 - 77	10	0
C	35	6 - 87	18	0
D	31	3 - 300	11	3

The indoor concentrations show strong variations from 8 to 2896 Bq m^{-3}. The proportions of houses presenting Rn concentrations greater than or equal to 1000 Bq m^{-3} (the French intervention limit, as an

annual average) and of houses showing concentrations greater than or equal to 400 Bq m^{-3} (the French precaution limit, as an annual average, and the European reference limit) and below 1000 Bq m^{-3} differ significantly between the 4 areas. The highest levels are essentially observed in the transect A.
The largest variations of radon levels are observed in the transect A, therefore we illustrate our main results by those obtained in this area (Fig. 2).

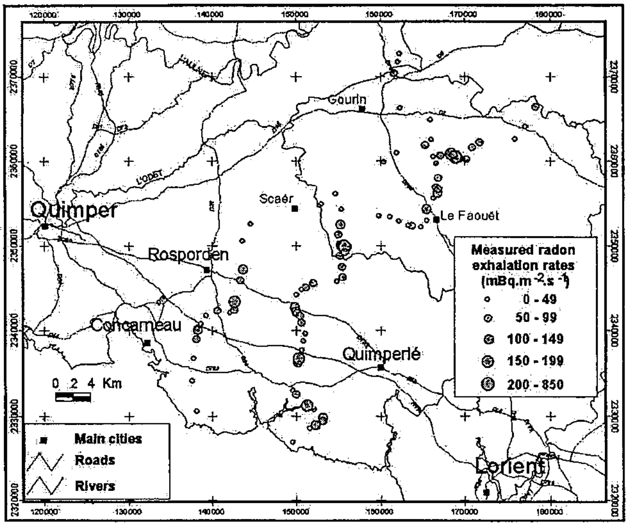

Figure 2. Area A: results of the radon exhalation rate measurements. The measurement sites were selected along a transect oriented NNE-SSW, which runs across the different geological formations observed in the area.

Firstly, the comparison between radon levels measured on the field and lithogeochemistry (U contents) shows that the radon source term of the lithologies, is the primary parameter that controls the spatial variations of radon exhalation rates. It is possible to classify a lithogeochemistry (lithology associated with a range of probable U contents) according to its radon exhalation potential. An example of such a classification is given in Table 2 and is realized by comparing the ranges of the Rn flux densities and of the indoor Rn concentrations measured on each lithogeochemical type. This classification can lead to a map of the radon exhalation potential: an example is given in figure 3 for the area A.

Table 2. Classification of the Rn potential of the lithologies according to their U content and the Rn measurements realized on the 4 study areas. The proportions are relative to the number of measurements performed on each class.

	Class I Very low potential	**Class II Low potential**	**Class III Moderate potential**	**Class IV High potential**
Mean U content of the lithologies (ppm; mean for each geological unit)	≤ 2	≤ 4	≤ 8	≤ 20
Rn exhalation rate at the soil surface ≥ 100 mBq m^{-2} s^{-1}	0%	< 5%	< 5%	30%
Indoor Rn concentration ≥ 400 Bq m^{-3}	< 10%	< 10%	15%	30%
Indoor Rn concentration ≥ 1000 Bq m^{-3}	0%	0%	0%	15%

Nevertheless, we can observe that the radon fluxes are highly variable for a given lithogeochemical type, in particular on uraniferous lithologies corresponding to the high exhalation potential class according to our lithogeochemical classification . Therefore, the exhalation potential mapping that can be realized from the study of the basement geochemistry sometimes over-estimates this potential. The forecasts obtained by TRACHGEO tests show the need to take account of spatial heterogeneity of soils (in addition of geochemistry) to improve the mapping resolution: the theoretical fluxes allow us to explain the spatial variability of the radon exhalation on a larger scale. Therefore, we can estimate a radon exhalation potential for each study site, from the TRACHGEO forecasts. The figure 4 illustrates the theoretical fluxes thus determined in the transect A. We can note that this map allows us to obtain forecasts in good agreement with the measurements (Fig. 2) and which are more precise than on the first map of the Rn exhalation potential only assessed by the lithogeochemical study (Fig. 3). The vast majority of our results

(75% of the sites studied in the 4 areas) are in good agreement with observations in the field. However, our estimations largely over-estimate the Rn exhalation rate for 22% of the sites. We can also note that a very low proportion of sites (3%) presenting a high exhalation potential, cannot be identified by TRACHGEO forecasts: most of these sites are located in faulted areas, suggesting that exhalation variations may be caused by structural anomalies. Indeed, the Rn transport by advection is not taken into account in our approach (Rn migration only by diffusion).

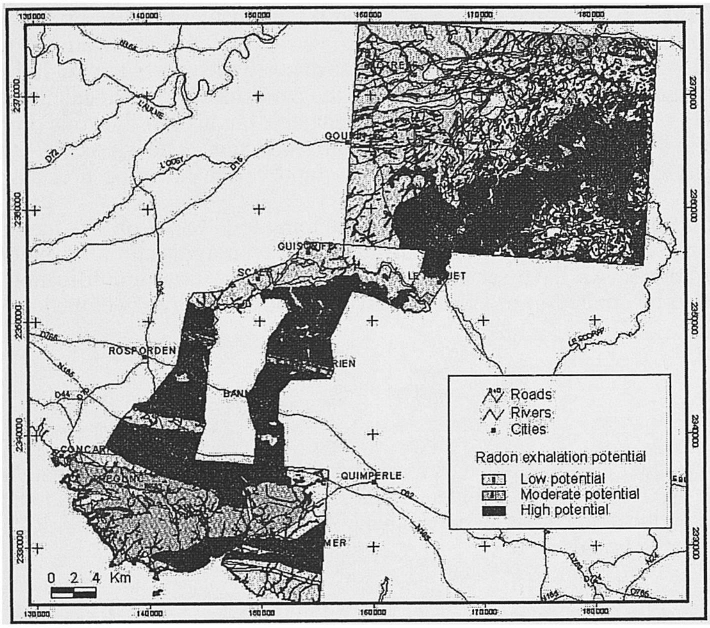

Figure 3. Area A: map of the Rn exhalation potential estimated by comparing the lithogeochemistry of the basement rocks (U contents of the geological formations mapped at the scale 1/50 000) and the range of Rn levels (fluxes at the soil surface and indoor concentrations) measured on each lithogeochemical type (see text for details).

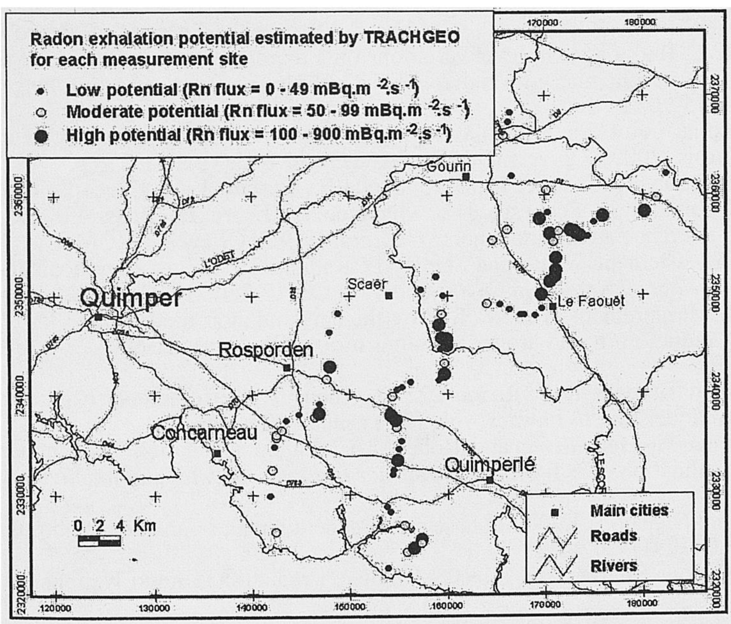

Figure 4. Area A: map of the Rn exhalation potential estimated by TRACHGEO model for each study site. The model takes into account the basement lithogeochemistry and the soils properties.

6. CONCLUSIONS

The proposed methodology is validated on the 4 areas selected for their different geological contexts. The comparison between lithogeochemical maps and the spatial variations of radon levels demonstrates the primordial influence of the U content of the basement on the Rn exhalation, showing that such a geochemical study is a first essential analysis step. It is possible to deduce an initial mapping of the exhalation potential by assigning a potential class to each defined lithogeochemistry. Indeed, the highest radon levels ($\geq$ 100 mBq m^{-2} s^{-1} at the surface of the soil) are mostly observed on lithologies whose mean uranium content can exceed 8 ppm or so. It should also be noted that dwellings presenting high Rn concentrations (values above 1000 Bq m^{-3} and the vast majority of values above 400 Bq m^{-3}) are located on those particular lithogeochemistries. Furthermore, the Rn exhalation rates calculated by TRACHGEO which takes account , not only of the basement geochemistry, but also of the soils properties, are in good agreement with the in situ measurements for 75% of the study sites. The TRACHGEO model, associated with a Geographic Information System, allows us to improve the mapping precision of Rn exhalation potential assessment.
Our final objective is now to develop an operational mapping tool, based on this methodology in order to draw predictive maps of radon exhalation potential that would constitute a guide for the Rn risk management. Nevertheless, we have seen that the exhalation potential amplification factor induced by fracturing remained to be studied in order to improve the methodology developed so far which ignored this phenomenon.

Acknowledgments

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