

# 16ENV10 MetroRADON

# Activity 4.2.1 and 4.2.2

# Relationship between indoor radon concentration and geogenic radon

Lead organisation: BfS Other involved organisations: BFKH, IRSN, JRC, UC, UNSPMF, VINS

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# 1. Motivation

The purpose of the MetroRADON project, funded within the European Metrology Programme for Innovation and Research (EMPIR) is to develop reliable techniques and methodologies to enable SI traceable radon activity concentration measurements and calibrations at low radon concentrations. The need for this project has been largely motivated by the requirements of the implementation of the European Council Directive 2013/59/EURATOM (EU-BSS) (EC, 2013), one aim of which is to reduce the risk of lung cancer for European citizens due to high radon concentrations in indoor air. Furthermore, it is a goal of the project to enable uptake and exploitation of its results and experiences by all stakeholders concerned with radon, from regulators and policy makers, professionals in designing, performing, evaluating and interpreting radon surveys, radon instrument manufacturers to the end-users (e.g. companies providing radon measurement, construction industry) and the scientific community. More details about the MetroRADON project can be found at the project website (MetroRADON, 2020).

Article 103 of the EU–BSS requires that member states identify areas where the radon concentration in a significant number of buildings is expected to exceed the relevant national reference level – referred to as radon priority areas (RPAs) within the MetroRADON project and this report. The definition of RPAs will influence political and technical decisions, which in turn will have economic effects in these countries, such as mandatory radon measurements in workplaces in these areas according to Art. 54 EU-BSS, as well as mandatory preventive measures or priority of awareness programmes. As the definition of RPA in the EU-BSS allows a wide range of interpretation, different concepts and methodologies have been proposed and some already adopted.

Within the MetroRADON project a specific work package (WP 4) is included with the aim to analyse and develop methodologies for the identification of radon priority areas, to investigate the relationships between indoor radon concentrations and quantities including soil exhalation and to develop the concept of a "geogenic radon hazard index" (GRHI) as a tool to help identify radon priority areas. One specific task (WP 4.2) within this workpackage is dedicated to analyse the relationship between indoor radon concentration and geogenic quantities.

# 2. Introduction

#### 2.1 Aim and motivation of Task WP 4.2

The aim of this task is to estimate relationships between indoor Rn or derived quantities such as the probability of exceeding a reference level within an area, and quantities related to geogenic Rn such as the Rn potential or uranium concentration in the ground (see Task WP 3.2, deliverable 3), as some concepts for mapping the geogenic Rn potential and RPA crucially depend on such relationships.

As RPAs are often estimated from quantities other than indoor Rn, such as geogenic Rn, the correlation and a statistical relationship between indoor Rn and the RPA predictor quantity (or quantities) therefore needs to be established, because only indoor Rn is directly linked to reference values according to the EU-BSS. In most cases, the statistical relationships between indoor Rn and geogenic quantities are weak. These relationships have been studied for many years as regression and classification type approaches. The physical and statistical reasons for the weak relationships will be evaluated and explained and their consequences assessed. In this

task, existing models and methodologies will be reviewed and reported, in particular statistical procedures which have sometimes proved a weak point in such analyses in the past.

#### 2.2 Structure of Task 4.2

In activity A4.2.1, the concept of radon potential (RP) is introduced. Various approaches are presented, in particular the "soil radon potential" or geogenic radon potential (GPR) (chapters 3 and 5). The concepts are discussed and a literature review of the statistical relationships and correlation between indoor Rn and geogenic Rn has been performed (chapter 4). Sources of information include journals, reports and conference contributions. Based on the review, physical and statistical reasons for the weak relationships are evaluated and interpreted and a synopsis of the results is produced. Further, possible inconsistencies in the literature and assess their consequences are identified.

Different methods have been developed, particularly in Europe, to assess the GRP that is then sometimes used for radon mapping (chapters 4 and 6) and radon priority area definition (deliverable 5, chapter 3). Those approaches are based on different models (statistical, physical or empirical models) that use different input quantities such as soil-gas radon concentration, radon exhalation rate at soil surface, soil permeability, soil Ra content, radon emanation factor etc. These parameters can be locally measured on the field or calculated. In A4.2.2, several existing approaches to assess a "soil radon potential" are reviewed, both from those identified in A4.2.1 and also others of which we know. In chapter 7, two case studies of GRP estimation are presented.

All results of the activity are discussed and summarised in this report.

The text for the specific activities and the involved partners are listed here in the following table. Additional institutions that joined this task later are VINS (Vinca institute) and UNSPMF (University Novi Sad).

Activity number	Activity description	Partners (Lead in bold)
A4.2.1	BfS, JRC and UC will obtain information about the approaches used to assess a "soil radon potential" and will undertake a literature review of the statistical relationships and correlation between indoor Rn and geogenic Rn. Sources of information will include journals, reports and conference contributions.	BfS, JRC, UC
	Based on this review BfS, JRC and UC will evaluate and explain the physical and statistical reasons for the weak relationships and will produce a synopsis of the results.	
	BfS, JRC and UC will then identify possible inconsistencies in the literature and assess their consequences.	

A4.2.2	Different methods have been developed, particularly in Europe, to assess a "soil radon potential" that is then sometimes used for radon mapping and radon priority area definition. Those approaches are based on different models (statistical, physical or empirical models) that use different parameters such as soil-gas radon concentration, radon exhalation rate at soil surface, soil permeability, soil Ra content, radon emanation factor etc. These parameters can be locally measured on the field or calculated. Certain methods also include a parameter that represents the capacity of soils to favour the radon entry in a house after building. IRSN and BfS will review at least 5 existing approaches to assess a "soil radon potential", both from those identified in A4.2.1 and also others of which they have knowledge.	IRSN, BfS
	IRSN will then compare selected approaches using experimental data from France that IRSN has access to by applying the data to mathematical models and will draw conclusions regarding the applicability of the selected approaches.	
A4.2.3	Based on the results from A4.2.1 and A4.2.2, BfS, IRSN, JRC and UC will write a report outlining the current state of the art with regard to information about the relationship between indoor radon concentration and geogenic radon as well as the new results. These results will also be published in scientific journal articles and presented at conferences. The report will feed into D5 and D6.	BfS, IRSN, JRC, UC

# 3. The radon potential RP

#### 3.1 Introduction: Motivation and purpose of RP

Two factors contribute to indoor radon concentration: 1, radon sources and conditions which control Rn transport in the subsoil, summarized as geogenic factor, and 2, building properties and usage, summarized as anthropogenic factor. Sources include geogenic radon which infiltrates from the ground, outdoor radon, and radon from tap water (Jobbágy et al. 2017 for a recent overview) and natural gas as used for heating and cooking (Neznal et al. 1996). Building materials are another source, which can be included among the anthropogenic factors. However, the most important source is geogenic radon, followed by Rn exhalation from building materials. Outdoor Rn, tap water and natural gas play a minor role in most cases, as far as this has been investigated. Anthropogenic factors, except building materials, include building construction, which determines Rn infiltration from the ground and exchange with the atmosphere, and building usage, i.e. ventilation habits.

For decades, there have been attempts to define a quantity called *radon potential* or RP, intended to "factorize out" the anthropogenic contributions. The RP is supposed to be a standardized quantity in which the anthropogenic factors play no role. It shall measure the availability of radon, originated from natural (geogenic) sources, to exhale from the ground into the atmosphere, or to infiltrate a building. It has, therefore, been said more colloquially, that the RP measures "what Earth delivers in terms of radon".

Basically, two types of approaches have been proposed in the past. They are discussed in the following sections. The particular concept of the geogenic radon potential, GRP, is discussed in more detail in section 4.2.2.

3.2 Concepts of RP in general and GRP in particular

#### a) Standardized indoor concentration

Suppose that at a site, there was a room within a building with both having defined standardized propertiesinstead of the actual room and building. These properties concern those which affect Rn concentration moststrongly: presence of basement, type of interface between ground and building, floor level of the room, type16ENV10 MetroRADONActivity 4.1.1/4.1.25

of windows (which determines the ventilation rate). The ideal building would, for example, be one with basement with concrete slab floor, the room would be on ground floor and have double glass windows. These are called the standard conditions. The deviations between the actual and ideal rooms and buildings are quantified by multipliers. Applying them on the observed Rn concentration would yield the one in an ideal room and house, i.e. according to standard conditions, on the same site. This approach underlies the so-called Friedmann radon potential, developed by H. Friedmann around 1990 duringthe first national Austrian radon survey (1992-2001) (Friedmann 1995, 2005). To some extent, also the European Indoor Radon Map (EIRM) has been guided by this idea, as it maps Rn in ground floor rooms only. The resulting map is shown in Figure 1. A newer version is shown in <a href="https://geogis.ages.at/GEOGIS\_RADON.html">https://geogis.ages.at/GEOGIS\_RADON.html</a>.



Figure 1: Austrian RP map, based on the first Austrian survey (Friedmann 2005).

Among problems of this approach are the following.

(1) The standardization factors or multipliers are found by statistical analysis of actual buildings, in which together with Rn concentration measurements, building, room and usage information has been acquired by questionnaires. This is a common practice with radon surveys. However, usually it is not known, to which degree the information is correct and accurate. Sometime important information is simply not known (e.g. floor of the basement), ambiguous (e.g. building along a slope, in which the same room may be a ground floor room upslope and a first-floor room down slope), insulation capacity of windows (double windows may be poorly insulating if the window frame is degraded), etc. It has turned out, however, that certain relevant information can be summarized in proxy quantities. For example, the age of a building, although itself is not a physical control variable, is a good indicator of its insulation against the ground. As another example, in Germany it has been found that whether a building is located in former East or West Germany is a useful proxy factor, since the building stock in former East Germany is older and physically less radon tight, on average,

than in former West Germany. Put together, selection of appropriate standardization factors requires sophisticated analyses.

(2) The factors, although carefully selected, are inevitably fuzzy factors, to different degree, since they rely on qualitative assessment, as explained above. These factors, as independent variables in a regression or ANOVA logic, are affected by uncertainty which is difficult to quantify. Furthermore, the set of factors can never be complete, because certain factors (e.g., frequency of opening windows) can hardly be quantified. (In the quoted Austrian survey, it has been found that the number of children in a family is a proxy of the ventilation rate.) Therefore, standardization multipliers, or standardization model parameters, are subject to uncertainty, which carries through to the resulting standardized Rn concentration. This means that local variability between Rn concentrations, due to different anthropogenic factors, is partly counterbalanced by model-induced uncertainty of the standardized RP value. If done statistically correctly, one can assume that the errors cancel on regional (e.g., municipality) average, although one cannot assume that a local RP (at one particular location) equals exactly the hypothetical Rn concentration in a standard room in a standard house at that very location. Obviously, it is also impossible to verify if it is.

(3) Certain factors are ignored in this reasoning, namely outdoor Rn, exhalation from building materials or tap water and natural gas contributions.

As a result, the RP based on standardization of measured indoor Rn concentration is only an approximation of the site-specific Rn situation, controlled by geogenic Rn, with anthropogenic factors removed only approximately. Still, it has been found that the Friedmann-RP reflects geological reality (which in turn controls geogenic Rn) rather well (e.g., Bossew et al. 2008). The EIRM, although filtering only for one factor, namely floor level, also leads to a representation of European geology in terms of the regional distribution of Rn concentration.

#### b) Other definitions

There is no unanimous definition of the RP, as this has evolved as a working concept over time, in different contexts. Another definition, quite different from the above, shall be mentioned. In the UK and Ireland, RP denotes the exceedance probability of indoor Rn concentration over a reference level, within an area, RP:=prob(C>RL).

Tanner (1988) proposed the radon availability number (RAN) defined as source times migration distance of Rn in the ground under standard pressure difference. Alonso et al. (2010) proposed using Ra concentration times emanation power, because it can quantify the "potential radiological hazard" of a porous material.

Among schemes based on combined scoring of factors, there is:

The one introduced by the U.S. EPA (Schumann 1993): classes of indoor Rn concentration, eU, geology, soil permeability, prevalent basement type;

The approach proposed in Kemski et al. (2001, 2009) and similarly, the Czech Radon Index (Neznal et al. 2004), are based on joint classification of soil Rn concentration classes and permeability classes;

Wiegand (2001, 2005) suggested a "10-point system" based on scoring categorical variables such as lithology, topography and land cover. Used in Tung et al. (2013).

In Sweden, schemes for regional classifications and for characterisation of building sites based on lithology, permeability, texture, Ra and soil Rn concentration has been introduced (Ek in Long Way 2011, sec. 5.3.9);

Guida et al. (2010): combined scoring of permeability, geology, Ra conc., vegetation cover, morphology, tectonics and karst features;

lelsch et al. (2010): aggregation of classes of Rn source potential, factors which enhance transport, "aggravating" factors.

Details can also be found in the Long Way (2011) document, chapters 5.2 and 5.3.

When using the term radon potential, one must therefore always add to which definition it refers.

For further discussion see section 4.3.4 about the Geogenic Radon Hazard Index and Bossew et al. 2020 (Annex 3).

c) Case study: Spain

The Spanish approach to assess the soil radon potential was born from a specific research project funded by the Nuclear Safety Council (CSN) developed during the years 2015-2017. The work teams were formed by the Autonomous University of Barcelona, University of Cantabria, University of Las Palmas de Gran Canaria and The Polytechnic University of Catalunya. Moreover, the company Geomnia collaborated in the geological part of the project.

The methodology recommended in Spain to determine the radon risk in a terrain on which it is intended to build is based on the Czech method (Neznal et al., 2004). From the measurements of radon concentration in the soil and the intrinsic permeability it is obtained the radon index (RI) or radon potential (RP). According to this approach, the radon index provides a level of risk for a terrain that can be expressed numerically from the radon potential of the terrain. The radon index can be determined from a non-numerical estimate of the air permeability of the soil and the radon concentration measured. Fixed the permeability in three classes (high, medium and low), the radon index can be obtained from Table 1.

Table 1: Radon index classification based on radon activity concentration C (kBq/m<sup>3</sup>) measured in soil and estimated gas permeability.

	Low	<i>C</i> < 30	<i>C</i> < 20	<i>C</i> < 10
Radon index	Medium	30 ≤ <i>C</i> < 100	$20 \leq C < 70$	10 ≤ <i>C</i> < 30
	High	<i>C</i> ≥ 100	<i>C</i> ≥ 70	$C \ge 30$
		Low	Medium	High
			Permeability	

It is established that for terrain of an area equal to or less than 800 m<sup>2</sup>, at least 15 radon measuring points are required to characterize them. For an area of greater extension, 10 m x 10 m grids should be established. In

areas with high radon concentrations a 5 m x 5 m sampling grid is recommended. The classification is based on the evaluation of the measured radon concentration values and their distribution. For each terrain a unique radon value of the soil must be obtained, assigned from the third quartile (75% of the data set).

The soil gas permeability limits established in the Czech method are shown in Table 2.

Table 2: Gas permeability in soil classification (Neznal, 2005).

Class	Permeability k (m²)
High	k > 4.0 10 <sup>-12</sup>
Medium	$4.0\ 10^{-12} \ge k \ge 4.0\ 10^{-13}$
Low	k < 4.0 10 <sup>-13</sup>

If the gas permeability of the soil value is available, it is possible to obtain the RP from the next equation:

 $RP = \frac{C-1}{-\log k - 10}$ 

where C is the radon concentration in soil expressed in kBq/m<sup>3</sup> and k is the permeability (m<sup>2</sup>).

Therefore, the relationship between the Radon index and the radon potential can be obtained from Table 3 or the graphical representation of Figure 2.

Table 3: Radon Potential and Radon Index classification.

Radon Potential(RP)	Radon Index(RI)
< 10	Low
$10 \leq \text{RP} < 35$	Medium
35 ≤ RP	High





The considerations of the use of the Czech method as a standard for the determination of the radon risk in a terrain discussed in the mentioned project are divided in advantages and disadvantages as following:

#### Advantages

1. Radon concentration and permeability of soil are two parameters that should reasonably be good indicators of the potential risk of a terrain. However, the relative importance of diffusion as an entry mechanism in homes will increase as the reference levels decrease.

2. The radon concentration measurement in the soil, following the standard procedure, does not show major problems and it is usually consistent regardless of the type of instrument used.

3. The radon average value in a terrain is usually representative if sufficient measurements are carried out.

#### **Disadvantages**

1. The most important inconvenience is the difficulty to obtain a representative permeability value of the terrain. It is necessary to establish which methodology should be used to obtain an intrinsic permeability value representative of the site.

2. The radon concentration measurements in the soil are affected by the different soil layers. Therefore, it is difficult to apply depth correction and it makes useless the radon measurement at the 80 cm level from the surface. One way to avoid this effect is to make the measurements from the foundation level, but this also indicates that the characterization that has been made of the locations does not have to be correct.

3. The seasonal variations observed in the different locations where the project was developed have not generally affected the risk classification. However, there are studies in the literature that show large fluctuations in radon levels in the soil, especially in fractured soils. No site with large fractures in the project was studied.

4. As far as the group of experts involved in the project knows, this strategy has not been validated experimentally.

5. In Spain there are regions where the Czech method cannot be applied because it is not possible to measure the radon in the soil.

The study of possible alternative methods for estimating the radon concentration in soil did not provide satisfactory results, and this is a further inconvenience to establish a standard method.

Radon levels in houses depend on multiple factors, with entry through the ground by advection being very important, but the other factors cannot be neglected.

The best tool for determining the radon risk in an area is the radon map of dwellings with a good resolution as a result of including a large number of measurements. The radon level maps in homes integrate all the factors: the soil potential, the typical construction characteristics of the area, the habits of the population, and the climate of the place. For this reason, the best way to know if a region should be a priority action is to look at a map of housing levels, what percentage of homes have levels above the established reference level. Only in the case of start an urban development in a large uninhabited region would it make sense to carry out a study of characterization of the land. Nowadays in Spain, the official national radon map, which takes into account data from indoor radon measurements and other information (geological, lithostatrigraphical, etc...), is the Radon Potential Map created by the Spanish Nuclear Safety Council (CSN, 2017a; 2017b; 2019).

#### General Recommendations for new buildings construction

In the opinion of the group of experts involved in the project, the best strategy for radon protection in new buildings includes the following actions:

1. Establish a basic level of protection for the entry of radon in all homes, similar to what has been done in Ireland, and increase it according to the estimated potential risk.

2. Determine the potential risk of each terrain from the potential Spanish radon map and also use the Czech method if it is possible. For risk characterization in large uninhabited areas, use the Czech method.

3. Promote campaigns to measure radon in air in dwellings. The best tool for determining the potential risk in a newly constructed area is to use of a map of radon levels in homes.

4. Establish a mechanism to improve the scale of the potential radon map and to incorporate the new radon level data in houses to the map.

5. Validate the methodology implemented by measuring radon levels in newly built housing, or by designing statistically significant measurement campaigns after a period of a few years to check if radon levels decrease. This validation is the most important task in the coming years from the point of view of the National Radon Protection Plan.

# 4. Estimation and mapping methods

Several steps lead from the data to a map (Figure 3). As a first step, data of input quantities, from Rn concentration in the ground and permeability to other predictors such as geological factors or geochemical concentrations, or also indoor Rn concentration in the case of the 4.2.1.2a (standardized indoor

concentration), must be acquired. This is done by measuring or taking from existing databases, such as geological maps.

Sometimes, relevant target quantities must be calculated from others through models (section 4.2.1.4), which are established and calibrated through special studies.

Target quantities, whether measured of predicted, are aggregated into spatial units which are the basis of the maps, such as grid cells or administrative units. These units constitute the wanted map.



Figure 3: Flow scheme: from data to map

The subject is further discussed in sections 5.2 (measurement of input quantities) and 6 (estimation and mapping).

4.1 Relationships between indoor and geogenic Rn, and predictors and proxies - literature review

(For the terminology of "predictor" and "proxy", see section 4.2a.)

Relationships between geogenic quantities and indoor radon must be known in order to use the former for predicting the latter. In this subsection, literature will be reviewed in this respect, while in the subsequent section, the physical background is discussed.

a) structure of the literature database

For comparability, a set of criteria has been defined along which literature has been evaluated. The result is given as table <Lit- $4_2_1_4$ --all-190424.xls>, to be found as annex.

The columns of the table are:

- 1. identifier;
- 2. motivation, objective, purpose of the study;
- 3. target variable, i.e. the quantity which shall be estimated, modelled or predicted;
- 4. predictor quantities;
- 5. sampling method;
- 6. sampling design;
- 7. temporal aspect, if applicable;
- 8. sample size;

- 9. region in which the study has been performed, and from where the conclusions have been derived;
- 10. physical characteristic of the region;
- 11. size of the region;
- 12. method of statistical analysis;
- 13. main results;
- 14. correlations identified;
- 15. residuals (indication of heterogeneity of population, missing predictors, adequacy of model);
- 16. are the results likely transferable to other regions?, can they be generalized?;
- 17. Comments.

#### b) Summary evaluation

47 papers found in literature were evaluated. The evaluated papers were written between the early 1990s and 2018, mostly from European research institutions.

The papers have the following motivations:

- Predict indoor radon concentration (IRC) and GRP from geogenic quantities;
- Investigate how much of geographical IRC variability can be traced to the ones of its geogenic controls;
- Improve spatial IRC estimation including RPA maps by using covariates as additional predictors.
- Soil gas Rn as tracer of environmental processes.

In most studies, the target variable is IRC; a few are focused on soil gas radon concentration (SRC), outdoor Rn concentration and Rn exhalation from the ground. In some cases, the target variables are derived quantities which are not directly observable, such as IRC exceedance probability, GRP, indices or hazard classes.

The predictors used are of two different types: radiological quantities and non- radiological quantities. Radiological quantities are U, Ra, Th, K, SRC, radon in indoor air, radon emanation coefficient, GDR. The nonradiological quantities are geochemical concentration in soil and rock other than U, Th, K; geological and tectonic variables (geological units, fault density etc.) and soil type. Among anthropogenic predictors are building types and characteristics.

Predictors and response variables can be ordinal (continuous numbers, typically: concentrations or ordered classes: "low, medium, high") or nominal-categorical (geology, i.e. without intrinsic ordering). Different types of variables imply different analytical treatment.

Differences between measurement methods are especially relevant for Rn measurement. IRC was measured with passive integrating monitors in all but one paper (continuous time-resolved monitoring). However, also different passive measurement methods exist, concerning detector material, sensitivity against Tn, exposure time, placement of monitor. In none of the studies evaluated here, measurement method is used as additional

predictor or confounding variable; apparently mostly due to the fact that only one technique was used in a study. Further investigation of possible impact of methodology on the result remains on the agenda.

Data sources of the studies are either surveys or sampling campaigns specifically performed for the study, or existing data which were re-used for the specific objective (or both, if necessary). Using modelled or aggregated data as input involves an additional uncertainty component, namely modelling uncertainty, which is often difficult to assess.

The situation is different with regard to spatial and temporal sampling design:

- Spatial sampling designs follow sophisticated schemes, in many cases. This shows awareness towards the problem of representativeness, which is difficult to solve in particular for IRC surveys. However, if the purpose is a survey which should serve for further decision making in the framework of Rn policy, this is crucial.
- Temporal design: The period of measurements varies among the countries too. This is due to the fact that some countries have strict protocols that indicate when radon measurements must be performed. This is the case of Finland and Norway when measurements have to be done in the winter season (assumed to yield conservative results). But which months are considered to be winter varies between these two countries. In other countries, such as the UK, it can be measured any time of the year and the result is seasonally adjusted. (The problem is minor for 1-year measurements, because annual cycles are averaged out, although the problem of long-term variability between years remains.)

Regarding the method used in the analysis and the results, all papers have used different types of regression analysis, however differently advanced, from conventional simple and multiple regression to machine learning (ML). Traditional geostatistical tools are applied by some researchers if the objective is mapping. Including categorical predictors is traditionally done by ANOVA or by ML in more advanced approaches.

#### c) Transferability of results

An important, but complicated issue is to see to which extent the *results* of the publications in terms e.g. of regression coefficients, can be transferred to other countries or regions rather different from the study areas.

As expounded before and in the next section, Rn quantities (most importantly IRC) result from a complex pathway, or rather network, "from rock to risk". Therefore, results of analyses of response of e.g. the IRC to one or several predictors depend on controls not accounted for in a study; these may indeed be irrelevant in a given study situation, because they can be assumed about constant over the domain of that study. A typical case is climate which may be irrelevant in a regionally confined study. However, the results may not be comparable with ones of a study performed in a region with different climate, in which this factor is different, but also not accounted for. Another example is correlation between IRC and ambient dose rate (ADR): the strength of the association depends on the presence of confounding covariates (e.g. Th concentration in the ground). If they are not accounted for, correlations of the two quantities cannot be reasonably compared between studies.

Even if a number of studies do apply advanced regression models that account, to different degree, for the complex and partly nested interaction structure between predictors, a "large-scale" regression study – or meta-study – which spans the variabilities of all potential controls, is still missing. Its objective would be to model, or "explain" IRC (or other Rn quantities) as response to all predictors that vary across Europe. Notably this concerns climate, building styles and living habits (partly functions of climate) which are variables that exhibit large-scale variability, which may be irrelevant, and therefore (rightly) ignored in regional studies. 16ENV10 MetroRADON Activity 4.1.1/4.1.2 14

At present, it seems impossible to "distil" existing regression and modelling studies, as evaluated in this section, to achieve the objective of a large-scale model in the above sense. This severely limits the more universal usability of most studies, without restricting to the specific situations that underlie them.

On the other hand, the *methods* used by some of the studies seem to be universally applicable. This is to say that the lack of transferability of results is owed to the predictor data, which are regionally specific, but not to methodology.

#### d) Conclusions

(1) As first conclusion, a number of studies prove the association between Rn quantities and geogenic quantities. However, this is not unexpected. But even assuming that the individual studies are correct for their respective (regionally specific) boundary conditions, quantitative results are difficult to generalize beyond the regional conditions, in most cases.

(2) Methodology has become quite developed for the last 20 years or so. Therefore, potential new studies whose objective is to extend applicability, should take advantage from existing ones. In certain regards there is however still space for further development. This concerns more rigid treatment of geochemical quantities as (in a mathematically sense) compositional and closed variables, for which the CODA (compositional data analysis) approach should be further exploited; 2 studies evaluated here have already done so. Further, it seems that the convoluted dependence structure of Rn quantities on different types of environmental controls and proxies limits the applicability of traditional regression models; at current state of knowledge, ML approaches seem to be most suitable to deal with such situation. However, questions, among other, related to interpretation of component effects and uncertainty budgets require further investigation.

4.2 Relationships between indoor and geogenic Rn, and predictors and proxies – physical causes and their statistical manifestations

#### a) General

The physical relationship between quantities which are related to Rn concentrations in different media is very complex, even in a simplified visualization (Figure 4). Even if the nature of the physical processes which generate the relationships is not very complicated – radioactive decay, diffusion, advection, dissolution – the intricate interaction generates the complex behaviour of the system.

Therefore, the statistical relationship between quantities is often weak although they are physically related. This reduces the potential of proxy quantities to Rn concentration to substitute it in cases where no Rn values are available, schematically shown in Figure .

Conceptually, one distinguishes between proxies (or surrogates) and physical predictors, Figure 6. The latter are ones that are in a causal relationship with the target variable, e.g., uranium concentration in the ground as a physical direct predictor of soil Rn concentration. Proxies are ones that are statistically related to the target, but not directly linked by physical causality. An example is terrestrial gamma dose rate component (TGDR) of ADR as  $Z_1$  in the figure, which is statistically related to IRC (= $Z_2$ ) because both share the same predictor, namely the uranium content in the ground ( $Z_0$ ). However, both ADR and IRC are also influenced by other variables, e.g., <sup>137</sup>Cs fallout and Th concentration in soil ( $Z_0$ " and  $Z_0$ ") influencing dose rate and ground permeability ( $Z_0$ '), the IRC; therefore, their correlation is weak. In this example,  $Z_0$ ',  $Z_0$ " and  $Z_0$ "' act as confounders.

For further illustration of the concept, examples of proxies from other fields are shown in a table, taken from the Minitab blog (link not existing any more):

#### **Examples of proxy variables**

Intended variable	Proxy variable
Historical environmental conditions	Widths of tree rings
Quality of life	Per-capita GDP
True body fat percentage	Body Mass Index (BMI)
Cognitive ability	Years of education and/or GPA
Depth that light penetrates into the ocean over large areas	Satellite images of ocean surface color
Hormone levels in blood	Changes in height over a fixed time



*Figure 4: "From rock to risk" – Simplified visualization of the complex relationships between quantities that control Rn concentrations in various media.* 



Figure 5: Correlation between target variable and proxy obscured by interaction of confounders and intermediate influencing factors.



Figure 6: Physical predictors, proxy and target variable.

To sum up, the influences of "confounding" quantities reduces, or even may obscure the statistical association between target quantity (e.g. indoor Rn concentration) and proxies (ADR), predictors (U concentration in the ground) and between Rn quantities (Rn concentration indoors and in the ground).

#### b) Specific: Radon

Specifically, for indoor radon concentration (IRC), the physical phenomena to be considered are Rn generation by radioactive decay of Ra, emanation into pore space and transport in rock, soil and ground water to the surface; subsequently, infiltration into buildings.

<sup>226</sup>Ra concentration (immediate parent nuclide of <sup>222</sup>Rn) may be in equilibrium with <sup>238</sup>U in geological media or not, depending on the chemical environment. Ecological process may separate the two because of their different chemical properties which determine sorption on environmental matrices.

Emanation of Rn from grains into the pore space depends on mineralogy of the grains and water content. Transport in the medium is controlled by porosity (available pore space) and tortuosity (connectedness of pores), both dependent on humidity in rather complicated manner. Diffusive transport depends on temperature, advective transport on pressure gradients and permeability, in turn a function of the above factors.

The transition from the geogenic to the anthropogenic compartment, i.e. the indoor atmosphere, depends on building properties and usage of the building. For the former, the tightness of the building shell against the ground (factors: diffusivity and advection pathways) and advective "suction" (driven by thermal stack effect or wind) of the indoor against geogenic compartment. The latter are usage type (residential, workplace of some type) and usage habits (dependent on cultural and climatic factors – so far little understood!).

The anthropogenic factors translate the geogenic radon potential GRP ("what Earth delivers") into the observed IRC. The ratio between IRC and GRP is very variable on every spatial scale due to the variability of building types and usage patterns. However, as found in many studies (among them some quoted in section 4.1), the spatial pattern of IRC largely reproduces the one of the GRP, while the variability of anthropogenic factors appears as spatial "noise". The reason is that on local and regional scale, the anthropogenic factors

have little spatial trend, as opposed to the geogenic factors (mainly controlled by geology and soil properties whose variabilities are obviously subject to trend).

However, the spatial correlation properties of anthropogenic factors have so far been little investigated. It can be expected that over larger scale climatic trend is present which is a main control of these factors. But to some degree, this is likely also the case in regional scale, subject to geography (lowland / mountains) and degree of urbanisation. Investigation of this subject remains on the agenda.

As a conclusion, in any case, it is evident that the variability of factors blurs the dependency between source (<sup>238</sup>U) and IRC, or even worse, between proxy such as ADR and IRC.

(For thoron, i.e. <sup>220</sup>Rn, the situation is partly more complicated because the decay chain between <sup>232</sup>Th and Tn contains several longer-lived radionuclides, which can render ecological fractionation more complex. On the other hand, the short half life of Tn (56 s) compared to the one of Rn (3.7 d) allows only short migration distances, along which less variability of controlling factors may be expected.)

#### 4.3 Correlation and concordance concepts

The strength of the association between variables can be quantified in different ways. A very common indicator is the *Pearson correlation coefficient* which can be applied for numerical variables and measured their linear association. It is defined as  $r(X,Y) := cov(X,Y)/(\sigma(x) \sigma(Y))$ . The sample r is only asymptotically unbiased with sample size. For small samples, approximate bias correction is available. The Pearson correlation coefficient is not robust against outliers. For X,Y bivariate normal, exact significance tests are available, and approximate ones otherwise.

If X and Y are not linearly, but still monotonically related, often the *Spearman rank correlation coefficient* is used. This is the Pearson coefficient applied to rank-transformed data. It is also less sensitive against outliers. Significance can be tested.

Also the *Kendall correlation coefficient* is a rank coefficient. All pairs of a joint sample of X and Y,  $(x_i, y_i)$ , are screened whether they are concordant, that is, if  $x_i > x_j$  then  $y_i > y_j$  or if  $x_i < x_j$  then  $y_i < y_j$ , otherwise discordant. From the number of concordant and discordant pairs a coefficient is defined. Variants for ties are available. A significant test also exists.

The association between nominal data is investigated via contingency tables. It is measured by statistics derived from  $\chi^2$  statistics, typically the *contingency coefficient* CC. In particular for binary data, 2 × 2 tables are quantified by the *Matthews correlation coefficient* MCC or statistics derived from the odds ratio, such as the *Yule coefficient of colligation*. For details, see the statistics sub-section of section 4.1.1.2, which deals with association of radon priority areas estimated from dwellings and workplaces.

The *intraclass correlation coefficient* (ICC) for grouped data measures the performance of the grouping scheme, or how similar data are which belong to a particular group.

4.4 Synopsis of literature results, assessment of consistency, consequence for RPA estimation and for construction of RHI

Interpretation of literature results has been given in section 4.2, physical interpretation of apparent lack of consistency in section 4.3.

The mostly regional nature of studies and their results, which is the source of apparent inconsistency has consequences for RPA estimation and GRHI construction.

#### a) RPA

RPA estimates that rely on transfer models (mathematically, regression analysis between IRC and controls or proxies) will reflect the regional conditions which underlie the models. For example, if ADR is used as proxy predictor, its classification power as well as the actual classifications will be different in parts of Europe with different environments, because of the large-scale regional cofactors which contribute.

Indeed, this effect is desirable as delineated RPAs should naturally reflect regional conditions. On the other hand, it means that regionally adapted models must be developed for RPA estimation in a particular region, and ones suited for other regions with different border conditions must be used very cautiously, if at all. This may be remediated if "grand" models are available which account for the complete variability of relevant controls over a domain, say Europe (with regard to which regional results are special cases).

#### b) GRHI

Due to lack of experience, the impact of inconsistency between dependence models on the GRHI is so far unclear. In current understanding, potentially most affected seem "local" versions of the GRHI which are based on collations of regional models (see discussion in the GRHI section). However, it seems that the dominant source of inconsistency across borders between regions, along which the GRHI estimates are collated or "sewn", is uncertainty that is due to its estimation from different sets of predictors between regions, but less so owed to predictors that vary between regions – but this would have to be investigated in detail.

For the "global" GRHI version, based on datasets common to entire Europe, the problem does not appear by definition. (In fact, the "global" GRHI version has been developed to avoid inter-regional inconsistency problems, in spite of disadvantages; see the GRHI section.)

# 5. Geogenic radon potential GRP

This section is based on text submitted to the European Atlas of Natural Radiation and a report by BFKH.

## 5.1 Concepts of geogenic RP

While the approach to define the RP by standardizing indoor Rn concentration ("Friedmann RP", see section 3.2) can be called a top-down approach, as the value from which it starts, i.e. indoor Rn concentration, lies very high up in the "rock to risk" scheme. In contrast, the GRP is a bottom-up approach, since it starts from geogenic quantities, which measure geogenic radon source and transport in the ground. Remember that Rn availability on the surface, or its availability for infiltration into a building, depends on both factors.

The main physical mechanism for infiltration into a building is advection through the interface of the building with the ground, possibly to cracks or fissures if there is a slab type foundation. Diffusion may play a role if there is no constructed barrier, as in earth basements of old buildings.

Advective flux is proportional to the pressure gradient across the interface times source strength. Proportionality factors are permeability in the ground and resistance of the interface. Rn availability, normalized against anthropogenic factors (pressure gradient, which depends on air circulation physics of the building, and type of the interface), is therefore proportional to source strength times permeability (k) of the ground, GRP  $\propto$  source  $\times$  k.

The matter is more complicated if diffusive transport is considered too. Diffusive flux depends on concentration gradient and resistance again diffusion, quantified by diffusion coefficients of the ground and of the barrier of which the interface ground-building consists (if any). Diffusive transport is independent of pressure gradients. Therefore, diffusive flux cannot be integrated into the GRP defined as flux normalized by pressure gradient. Diffusive flux through the soil surface  $\propto$  source  $\times \sqrt{D}$ , D – the effective diffusion constant in the soil. For a recent reference on Rn transport in soil, Chakraverty et al. (2018).

Several operational definitions of the GRP have been proposed. Currently most used seems to be version (1), a combination of Rn concentration in soil under equilibrium,  $C_{\infty}$ , de facto in 70 to 100 cm depth (Czech and German definition, respectively). As a formula, the so-called Neznal-GRP is most popular,

#### GRP : = $(C_{\infty}-C_0)/(-10-_{10}\log k)$ (Neznal et al. 2004)

C in kBq/m<sup>3</sup>, k in m<sup>2</sup>. For mean to high permeability (k =  $10^{-11}$  to  $10^{-12}$  m<sup>2</sup>), this is approximately proportional to the advective flux, normalized by pressure gradient. Very low and high permeability result in higher and lower GRP than expected by k C<sub>∞</sub>. The formula has been found semi-empirically, as combination that allows optimal prediction of indoor Rn concentration. C<sub>0</sub> is a very small concentration, set to zero by most users of that approach. In the original publication, C<sub>0</sub>=1 kBq/m<sup>3</sup>. The formula is not applicable for very high permeability, about 5.10<sup>11</sup> m<sup>2</sup>. The relationships of permeability k, the transformed 1/(-10-lg k), migration length and influencing factors is shown in Figure 4.

(2) As alternative, the source term has been proposed to estimate  $C_{\infty}$  as proportional to mean U or Ra concentration down to about 1 m in the ground, times emanation power. (Also porosity and water content has to be considered.) This approach has been developed and used in Estonia, Petersell et al. (2017)(map 6.1 f, theory and references p. 27). U or Ra concentration is usually determined from soil samples in the laboratory, as are emanation power and other soil specific parameters. For Ra concentration, a possible alternative is airborne gamma-ray spectrometry. The European U map of this Atlas could be an input database for the source term required for calculating the GRP.

<b>k</b> [m²]	10 <sup>12</sup> k	1/(-10-lg k	) texture	transport	influence of humidit	migration y length
>>	>>	-	karst		low	some km
10 <sup>9</sup>	1000	-	coarse gravel			some 100 m
1010	100	$\infty$	gravel	advective		some 10 m
$8 \cdot 10^{11}$	80	5				
$5 \cdot 10^{11}$	50	3	coarse sand			up to 10 m
1011	10	1	sand			few m
1012	1	1/2	silt – sand			< 1 m
1013	1/10	1/3	silt			50 cm
1014	1/100	1/4	silt – clay			10 cm
10 <sup>15</sup>	1/1000	1/5	clay	diffusive		cm
<b>10</b> <sup>16</sup>	1/10,000	1/6	fine clay		high	mm



Version (1) requires measuring soil gas radon concentration C<sub>∞</sub>. Soil gas samples are collected using a stainlesssteel probe pounded in the ground with a co-axial hammer to a depth of about 70–100 cm to avoid the influence of atmospheric air. Gas is extracted by purging the probe with a plastic syringe (50 cm<sup>3</sup>), or by an automatic pump connecting the probe directly to the measuring device. This can be done in situ rather quickly and cheaply. The practice has been used, among other, in CZ, DE, AT, BE and IT. It requires careful probing in order to avoid "clean air contamination" of the sample, i.e. intrusion of atmospheric air which dilutes the Rn concentration in the sample.

The main problem is that soil gas Rn concentration, even at 1 m depth, is not constant in time, but subject to seasonal and daily cycles related to temperature and mean soil humidity, and possibly also fluctuating ground water table. Their amplitude depends on soil type and can represent a variability of up to 50% under unfortunate conditions. To minimize the effect, sampling is recommended by practitioners to be performed avoiding certain conditions (frost, saturated soil after rain, untypically dry soil). For these reasons, soil gas survey should be conducted during low-precipitation seasons (typically summer to early fall) to minimise any variations induced by different sampling periods. As such it is believed that all the surveys represent the same populations and that they can be combined for statistical and geo-spatial analysis.

To remediate this problem, it has been proposed to measure soil Rn with passive detectors buried in the ground, such as CR-39 or polycarbonate based monitors (Turek et al. 1997, 2004; Conrady et al. 2011). The method has been used in Israel (Shirav-Schwartz et al. 1997) and in Kosovo (Kikaj et al. 2016) as well as in a soil Rn survey in Rhineland-Palatinate (Germany) (results unpublished). It has been shown that also polycarbonate based material as used for CDs/DVDs can be used for the purpose (Pressyanov et al. 2010, 2014, Mitev et al. 2018). The subject is discussed in detail in section 4.3.2. Integration periods can be between days and possibly years, depending on Rn concentration and detector type. Measuring for days to weeks (apparently the usual period with CR-39) does not solve the problem posed by seasonal variability, though (Kikaj et al. 2016). Polycarbonate is less sensitive and therefore allows longer collecting times, mitigating the problem of temporal variability (Shirav-Schwartz et al. 1997). Pressyanov et al. 2015, 2018 report usability of the CD/DVD 16ENV10 MetroRADON Activity 4.1.1/4.1.2 21

method between exposures of 10 and 10<sup>6</sup> kBq h/m<sup>3</sup>, allowing very long exposure. The method is logistically more complicated than grab sampling, as sites have to be visited twice and the burying and recovery procedure is more labour intensive. Additionally, influence of soil humidity on track-etch monitors is not quite clear, and thoron may interfere substantially. (The latter problem seems to have been solved by installing appropriate diffusion barriers for the buried-TE method and for the CD/DVD method by particular etching procedures, Pressyanov et al. 2003.) A comparative assessment of grab sampling and passive methods has been given in Kemski et al. (2012).

The same problem of temporal dependence applies for point measurement of permeability. A possible alternative is using model based, i.e. calculated, instead of measured permeability. Theoretically, one should be able to estimate permeability from soil parameters like grain size distribution (texture), porosity, humidity and others. Databases of these quantities are available Europe wide. Investigation of whether calculated permeability can or shall substitute the measured one is currently (late 2018) under way in Germany (Petermann et al. 2018); preliminary results are however little encouraging, as it was so far not possible to establish a model that could explain measured permeability values with satisfactory precision. The problem is currently unresolved.

On problems of soil permeability determination in general, Neznal et al. 2005.

In the future, a European GRP map could be based on the U map and soil databases, for example the European LUCAS database (https://esdac.jrc.ec.europa.eu/projects/lucas; Orgiazzi et al. 2018). There is little chance that a GRP map based on method (1) could be accomplished in foreseeable future, because many countries do not plan soil gas Rn surveys (as performed in CZ, DE and IT), and if they do, completion may take long time. (However, it should be said that soil gas Rn and geochemical surveys are logistically much easier than indoor Rn surveys, concerning issues of representativeness and data protection.)

A different way of defining radon potential is based on multivariate cross-tabulation. This method results in an index with a categorical-ordinal quantity, the results are given in classes such as (I, II, III, IV) or (low, medium, high). Classes are assigned based on scores either assigned to a combination of input quantities or calculated as the sum of points delegated to the input quantities. The second type allows for the consideration of multiple factors. Available quantities are soil gas radon, permeability, standardised indoor concentration, equivalent uranium concentration or other geochemical quantities, external terrestrial gamma dose rate, geological categories, quantities related to tectonics, and the presence of 'special features' like mines, caves, water bodies and other extraordinary conditions, which are coded binary (yes, no) (Gruber et al. 2013).

A similar approach has been applied in France, lelsch et al. (2010), Figure 5. In a study in Bourgogne region, the following set of geogenic variables has been used as constituents to create a classified GRP: geology, lithology, U content, fracturing (presence of faults), underground mines, and thermo-mineral sources as quantitative parameters. This was made necessary by the relative sparseness of the soil-gas data in France. The data was provided by previous geological and geochemical surveys, studies and databases, to compile the map they selected a 1.5 km<sup>2</sup> sized minimal object size and calculated the mean U content of the geological units based on the geological map of France (1:1,000,000, digital map). Then the authorsconsidered the various artefacts (mines, geological fractures, etc.) inside the geological units and constructed a map by compiling all considered layers together. For classifying the geogenic radon potential they used two quantitative scales, a more detailed 5 step and a more easily interpretable three step scale. The French approach is discussed in more detail in section 4.2.2.4b.



Figure 5: The geogenic radon potential map of Bourgogne (lelsch et al. 2010).

#### 5.2 Input quantities: Measurement

Physical controls of the GRP are Rn source and Rn transport in the ground. These can be captured in different ways for defining a GRP quantity, e.g. the popular Neznal-GRP. Input quantities are Rn concentration in the ground and permeability.

Measurement techniques:

*Uranium concentration in the ground:* ICP-MS, X-ray fluorescence are most common, gamma spectrometry of <sup>234</sup>Th and <sup>234</sup>Pa lines is possible with care.

*Radium concentration in the ground:* Usually determined by gamma spectrometry of progenies <sup>214</sup>Pb and <sup>214</sup>Bi. The samples have to be sealed for (optimally) one month before measurement in order to have equilibrium between <sup>226</sup>Ra and its progenies.

*Radon concentration in the ground:* Different techniques are available: Grab sampling, continuous sampling and passive exposure. Grab sampling can be taken using different options available in the market such as Neznal probes (www.radon-vos.cz/?lang=en&lmenu=en\_measuring&page=en\_measuring\_rm2) or MARKUS system (<u>https://radoninstrument.com/en/product/markus/</u>). These instruments are ionization chambers (Neznal probes) or silicon detectors (MARKUS). In both cases it is necessary to make a hole in the soil 0.7 to 1 m deep. Then, the measurement system sucks an air sample. After several minutes the instrument gives a reading of the radon concentration in the soil gas where the sample was taken. Although these systems are

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widely used, other methods such as passive detectors can be used too. They are CR39 detectors specially designed to operate under the conditions of 0.7 – 1m deep and the typical exposure time ranges from 1 to 15 days. Finally, continuous devices such as Alphaguard have systems to measure radon in soil gas too. Further information abut this can be found in the ISO standard 11665-11:2016 "Measurement of radioactivity in the environment — Air: radon-222 — Part 11: Test method for soil gas with sampling at depth".

*Permeability:* The measurement of permeability in the soil can be done by active methods such as the one proposed by Neznal "Radon Jok" that allows the measurement of this parameter in situ. The system applies negative pressure to suck air from the soil using the device and calculate the permeability using the known air flow through the probe. The system allows to measure a range of permeabilities from  $k = 10^{-11} \text{ m}^2$  to  $10^{-14} \text{ m}^2$ . Further information can be found in the website <u>www.radon.eu/jok.html</u>.

Permeability can be estimated as the weight percentage of fine fraction (<63  $\mu$ m): high permeable soil have the weight percentage of the fine fraction <15 %, medium permeable soil have weight percentage between 15–65 % and low permeable soil have weight percentage of fine fraction above 65 %.

There is also possibility of getting values of soil permeability in some of the existing databases of soil characteristics. One example is the European map of soil permeability that can be found here: <a href="https://data.europa.eu/euodp/en/data/dataset/jrc-eanr-11\_soil-permeability">https://data.europa.eu/euodp/en/data/dataset/jrc-eanr-11\_soil-permeability</a> .

*Porosity:* The porosity of the soil can be determined experimentally in the laboratory by taking soil samples and applied standard analytical methods.

*Emanation power:* There are two ways to estimate the radon emanation coefficient from the soil: gamma spectrometry and measurements of radon and radium. The gamma spectrometry technique consists on measuring the peaks of <sup>214</sup>Pb and <sup>214</sup>Bi several times in conditions of equilibrium and lack of equilibrium. It is important to guarantee that the soil sample has no radon in the soil pores before the measurement starts. The other way of determining the emanation coefficient is enclosing the soil sample and waiting for secular equilibrium between radium and radon. Then the radon concentration must be measured by active or passive methods as well as the effective volume of the sampling device and the weight. Further information about these methods and the mathematical expressions to calculate the emanation coefficient can be found here: www-pub.iaea.org/MTCD/Publications/PDF/trs474\_webfile.pdf (IAEA Technical report series No. 474 "Measurement and calculation of radon releases from NORM residues, IAEA 2013").

# 6. Geogenic radon potential (GRP): Estimation and mapping

If input quantities, such as C and k values for the Neznal-GRP, are not available, then the radon potential is usually estimated from proxies or surrogates. Such proxies are the standardised indoor radon concentration (measured in defined standard conditions such as ground floor rooms, presence of a basement, etc. to 'factorise out' anthropogenic factors) The standardised indoor radon concentration is correlated to the GRP, with inaccuracies caused by remaining unaccounted for or poorly assessed factors. Other quantities such as equivalent uranium (eU) or dose rate have similarly describable relations to the GRP, however these relations can be locally different, according to the regional predominance of some factors. The controlling factors have to be considered when using substitutes for the soil radon in the formula (Gruber et al. 2013).

Several options exist for compiling maps. The target variable has to be matched to spatial units (area), which will serve as the basis of the map. These spatial units can take various shapes and forms such as administrative or geological units or a grid cell. Geographical units might be a practical choice for the radon potential, and if desired those units can be decompiled into a grid system. The spatial units are then assigned a value derived from the measured target variables inside (arithmetic mean, geometric mean, median, etc.) (Gruber et al. 2013). The technique requires availability of sufficient data. Various estimation or interpolation techniques (local regression methods, different versions of kriging, Bayesian inference or extensive Monte Carlo simulations) can be implemented during the construction of such maps. However, it should be kept in mind that the interpolated concentration is only an estimate, not the true radon concentration, even though it can be useful for the visualisation of the data and in defining areas with higher risk probability (Cafaro et al. 2014). The different spatial units offer different advantages and disadvantages. Administrative boundaries make administrative action easier, but disregard the relation between the radon potential and the geology and soil properties. Grids makes mapping independent from other variables, but ignores variation within the grid cells. Geological boundaries are much more closely related to the radon potential but still there can be variations in the radon potential inside the geological units (lelsch et al. 2010). In case of sufficient data density maps can be made by displaying each point of data, without interpolation for the areas between the data points, which would still give an instinctive grasp of the overall situation (McKinley 2015).

#### Multivariate estimation

Estimation from possibly several predictors additional to, or instead of GRP data, one has to rely on association between target variable (GRP in this case) and predictors. The subject is discussed in detail in section 4.2.1.4. Two examples are quoted here:

In case of the multivariate cross-tabulation, values can be assigned to the various parameters or qualitative categories can be set up. For example, in case of a study on Bourgogne, a five-step qualitative scale was used to define radon source potential based on lithology and uranium content. For the geogenic radon potential map the authors narrowed down the number of categories to three and included the various artefacts such as mines and hot springs into the analyses (lelsch et al. 2010).

In case of the geogenic radon potential formula reliant on soil gas and soil permeability measurements, there are some methods correlating various other parameters if the input is not directly available. Appleton and Miles performed least squares (LS) regression analysis to establish empirical relationships between estimated uranium in the <2mm fraction of topsoils derived from airborne gamma spectrometry data, U measured in the <2mm fraction of topsoil geochemical samples soil gas radon and indoor radon concentrations based on observations in the United Kingdom (Appleton and Miles 2010). The linear relationships were compared to those described for other countries. The described relationships are dependent on the underlying geological units. Similar relationships were described by other authors for Germany, Croatia and the Czech Republic (Appleton and Miles 2010).



*Figure 6: The relationship between indoor radon concentration and soil gas radon by least square regression analysis (Appleton and Miles 2010).* 

Various log - ratio transformation methods (pairwise, additive, isometric, etc.) have been also used for the eliminating the constant sum closure effects caused by the relative nature of geochemical data (McKinley 2015). Yet another method is using correlation coefficient matrices either on the original data or if lognormal distribution is assumed then the logarithms of the data (Pereira et al. 2017). In some cases, (for example the Portugal C2-type granites) the correlation might not be made between the desired parameters due to the high variability of the data. Some other examples are the Global Ordinary Least Squared and the Geographically Weighted Regression, the latter being suggested favourable due to the inclusion of local geographical parameters (De Novelis et al. 2014, Ciotoli et al. 2017).

# 7. Case studies

#### 7.1 Germany

In Germany, several versions of GRP map evolved over time. In the 1990s, Kemski et al. (2001) proposed a map of soil Rn concentration (SRC) in 1 m depth, aggregated into a 3 km × 3 km grid. Interpolation is by inverse square distance weighting within geological units. A simplified litho-stratigraphic geological classification was designed such as to classify geological units according their GRP. The resulting map is published on the home page of the BfS, www.bfs.de/DE/themen/ion/umwelt/radon/karten/boden.html, shown in Figure 7.



Figure 7: Map of Rn concentration in soil, Germany.

In 2012, a map of the Neznal-GRP was proposed, Bossew (2013, 2015). At each measurement point, the GRP was calculated from measured SRC (data mostly the same as above) and permeability. These were normalized to geology by dividing by the GM(GRP) per geological unit. A simplified geological classification scheme as above was applied. The In-transformed normalized values were subjected to sequential Gaussian simulation (SGS). The 100 realizations were back-transformed and statistics computed. The estimation units were 10 km × 10 km grid cells.

In Figure 8, the mean over realizations (so-called E-type map) is shown on the left. The right map shows the relative dispersion between realizations, defined as Qdev90 := (Q95 - Q05)/(Q95 + Q05), Q the quantiles to percentiles 5 and 95. The dispersion essentially reflects density of input data.



*Figure 8: GRP map of Germany based on GSG. Left: Expectation, Right: Relative dispersion between realizations.* 

The latest version is based on machine learning. Random forest turned out the optimal method according to various performance scores. Predictors are related to geology, hydrology, soil properties, meteorology and geochemical concentrations. The result has been published, Petermann et al. (2020).

#### 7.2 France

A map of the radon potential of the geological formations has been established by the IRSN (lelsch et al. 2017b; lelsch et al. 2010) in order to characterize the capacity of the underlying rocks to generate radon at the surface on the French territory. This mapping is based on the characteristics of the geological formations. The main parameters considered are the uranium contents of the underlying rocks and factors that can facilitate the transport of radon to the surface (faults, underground mining works, thermal springs...). This map is based on data from the geological map of France at the scale of 1: 1,000.000.

Moreover, a measurement campaign of the radon in dwellings was carried out over the period 1982-2002 by IRSN and the Ministry of Health (Demoury al. 2013). During this campaign, 12,940 measurements were collected over the French territory. In complement, measurements of indoor radon concentration in public buildings were also used for this study (database of the French Ministry of Heath). They represent 8,253 results acquired between 2014 and 2018. At last, 2,305 measurements acquired from local measurement campaigns in homes were also collected for this study (databases of the French local Health Authorities).

Thus, a totality of 23,499 indoor radon measurement results in 9,967 different municipalities spread over French territory have been used. Table 4 provides the statistics associated calculated from this dataset. Once should note that these measurements overestimate indoor radon concentrations because measurements in public buildings and local measurements campaigns in homes were rather carried out in high radon potential areas.

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All the data could not be located precisely with coordinates. For half of the data, the name of the municipality was available and not the exact location. (A new problem has been caused by the European General Data Protection Directive GDPR (2016), as reporting exact geolocation data is not allowed and aggregation of data (or some similar process) should be used).

*Table 4: Statistics of indoor radon concentrations available for the whole French territory. A.M – arithmetic mean, S.D – standard deviation of the sample* 

	Number			Radon co	ncentration	ו (Bq.m <sup>-3</sup> )		
	of data	Min	Max	Average (A.M)	S.D	1st quartile	Median	3rd quartile
Dataset	23,499	1	28,553	219	571	40	84	202
National campaign in dwellings	12,940	1	4,382	89	159	28	49	93

#### Relation between indoor Radon concentration and geogenic Radon at the national scale

At first, all measurement results were directly compared to the map of the geogenic radon potential in 3 classes. For each municipality, we first calculated the percentage of the surface covered by the "low" category of the geogenic radon potential map. The arithmetic mean of indoor radon concentrations was then calculated for each concerned municipality (Figure 9c). Thus, the mean radon concentration in municipalities with 100% of their surface concerned by a "low" category is 78 Bq/m<sup>3</sup> while that of municipalities with 0% of their surface in a "low" category is 322 Bq/m<sup>3</sup>.

The same method was then applied considering the percentage of "high" radon potential area (Figure 9d).

In both cases, the results obtained show a good consistency between the geogenic radon potential map and the results of indoor radon measurements in dwellings and public buildings.



Figure 9: Comparison between the French geogenic radon potential map (a) and the mean radon concentration by municipality based on 23,498 indoor measurements (b) considering the surface of the municipality (%) covered by a low geogenic radon potential (c) or a high geogenic radon potential (d).

Moreover, from the French geogenic radon potential map, the municipalities have been classified in three categories of radon potential:

- Category 1 municipalities are those located entirely on geological formations with low uranium contents and with no factors that may facilitate the transfer of radon to the surface
- Category 2 municipalities are also located on geological formations with low uranium contents, but a
  part of their surface is concerned by geological factors that can facilitate the transfer of radon to the
  surface
- Category 3 municipalities are the municipalities which present geological formations whose uranium contents are estimated higher compared to the other formations, at least on a part of their surface. For this category, the presence of radon at high concentrations in buildings is most likely.

This classification of municipalities into three categories is currently used in the French regulation with an obligation of radon measurement in certain public buildings in the municipalities of category 3.

Figure 10 shows the relation between the percentage of indoor radon concentrations exceeding of the values of 100, 300, 600 and 1000 Bq/m<sup>3</sup> and the category of the municipality. A good correlation is observed between the municipality categories and the probability of exceeding these values.



Figure 10: Comparison between the French municipalities radon classification (a) and the mean indoor radon concentration by municipalities based on 23,498 indoor measurements (b) considering the percentage of exceeding different values by municipality (c)

#### Relation between indoor Radon concentration and geogenic Radon at the regional scale

The comparison of the means by municipality with the percentage of the surface of each municipality with a "Low" or a "High" geogenic radon potential was also carried out at the scale of a smaller territory. For this, data analysis was restricted to the Haute-Vienne *"département"* (French district), a territory characterized by high radon concentrations in dwellings. In this area, 1,132 measurement results are available and distributed in 112 different municipalities. Table 5 provides the statistics calculated from this data.

Table 5: Statistics of indoor radon concentrations available in Haute-Vienne "département". A.M – arithmetic mean, S.D – standard deviation of the sample.

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Number			Radon co	oncentratio	n (Bq.m <sup>-3</sup> )		
of data	Min	Max	Average (A.M)	S.D	1 <sup>st</sup> quartile	Median	3 <sup>rd</sup> quartile
1,132	7	16,928	746	1,270	177	382	774

Figure 11c shows the relation between the arithmetic mean of indoor radon concentrations (Figure 11b) and the percentage of surface concerned with a "low" geogenic radon potential for the municipalities in Haute-Vienne *"département"*. A very good correlation is observed between these two parameters. Figure 11d shows the same analysis with the surface of municipalities concerned by a "high" geogenic radon potential. A good correlation is also observed.



Figure 11: Comparison between the geogenic radon potential map for Haute-Vienne territory (a) and the average by municipalities based on 1 132 indoor radon measurements over this local territory (b) considering the surface of the municipalities covered by a low radon potential (c) and a high radon potential (d)

#### Relation between indoor Radon concentration and geogenic Radon at a local scale

Finally, an analysis was made on a restricted area of the Haute Vienne *"département"*, with a surface of 50 km x 25 km. This sector, comprising 15 municipalities, is an area with a high radon potential already identified (presence of old uranium mines). During the winter of 2015/16, a measurement campaign was initiated by IRSN to raise awareness of the radon issue and to inform the public on remediation techniques (lelsch et al. 2017a). 706 indoor radon measurements were made in dwellings during this campaign. Table 6 presents the statistical summary of the data acquired. All these data could be geolocated precisely (with coordinates).

Table 6: Statistics of indoor radon concentration data acquired on a small area with a high radon potential. A.M –arithmetic mean, S.D – standard deviation of the sample.

Number			Radon co	oncentratio	n (Bq.m <sup>-3</sup> )		
of data	Min	Max	Average (A.M)	S.D	1 <sup>st</sup> quartile	Median	3 <sup>rd</sup> quartile
707	7	16,928	985	1,474	272	575	1,050

Fehler! Verweisquelle konnte nicht gefunden werden.a shows the precise location of the measurement points on the geogenic radon potential map in 3 classes. Fehler! Verweisquelle konnte nicht gefunden werden.b shows the relation between the geogenic radon potential class on which the dwelling is located and the indoor radon concentration. A very good correlation is observed for these two parameters.



Figure 12: Comparison between the local geogenic radon potential map (a) and the geolocated indoor radon measurements (b)

#### 7.3 European level

Generation of a European map of geogenic radon is an ongoing project of the JRC as part of the European Atlas of Natural Radiation (Cinelli et al. 2018). A first trial version has been proposed by Gruber et al. (2012). Units of the OneGeology map (<u>http://www.onegeology.org/</u>) were assigned values of the GRP and classified according two schemes: frequency of resulting GRP values and correlation with indoor Rn. For the trial maps, the geological units were calibrated with German GRP data (because of otherwise limited availability). While the EGRM presents a unified picture of the collected data, the trial version was calibrated using German geotypes, so for other countries analogies were used. The result is shown in Figure 13.

Further problems are due to some countries missing in OneGeology, partly insufficient classification depth of OneGeology (which was not made for Rn mapping) and inconsistencies in its legend at that time.



Figure 13: Trial versions of the European Geogenic Radon Map (EGRM) with "radiological" (left) and "geological" radon classes (Gruber et al. 2012)

Calibration on German data only appeared problematic because nominally same geologically units can still be different with respect to Rn. Some regions are covered by geology which does not occur in Germany and could therefore not be included. As an example, the mismatch between the European trial maps and the correct Czech geogenic radon maps is shown in Figure 14.



Figure 14: Mismatch between the trial version of the European and the Czech geogenic radon maps.

## 8. Summary, conclusions and open problems

The idea of radon potential as a quantity which "subtracts" individual physical properties of buildings to indicate the natural conditions that control long-term mean indoor Rn concentration (IRC), has been around for more than 30 years. Concepts were reviewed in this task. One may distinguish between "top-down" approaches, whose initial variable is observed indoor Rn concentration, which is normalized with regard to house, room and usage properties, i.e. the anthropogenic factors which control IRC.

An alternative is the "bottom-up" approach, which starts from geogenic control quantities. The GRP is a particular kind of radon potential; it is defined physically from quantities which control Rn generation and transport in the ground. In this action, definitions are discussed as well as the geogenic quantities which are its input, and their measurement. Problems of representativeness of measured values for a measurement location are addressed, which are mainly owed to the temporal variability of some control quantities.

Regarding mapping, the rationale of the RP in general, and the GRP in particular is that the geographic pattern of IRC mainly reflects the one of its geogenic controls. The reason is that the geographic dependence of anthropogenic factors is relatively minor compared to the geogenic ones, at least on regional scale, i.e. anthropogenic ones appear as statistical noise on top of the geogenic pattern. In mathematical terms, the anthropogenic factor appears as a scalar factor relating IRC and its geogenic controls (or predictors) that has to be found by regression-type analysis.

Many regression studies have been performed for many years. In this task of Metro Radon, a literature review of relationships between geogenic quantities which control geogenic and indoor Rn concentration has been performed and the results interpreted. In particular, the correlation between IRC and geogenic quantities, which is poor in many cases, has been discussed. The main problem seems to be that models have been developed regionally, obviously considering only regionally variable controls, about constant ones regarded as fixed and entering regression coefficient. However, on larger scale, e.g. Europe, the latter controls are also

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geographically variable, if over larger distance compared to the regionally variable ones. Therefore, regionally developed models, though correct regionally, may not be applicable beyond the region in which they have been developed. This problem remains a challenge; first European-scale studies have been initiated only recently. Their further development and evaluation remain a task for the future.

The question is closely related to analysis of the spatial statistical properties of the anthropogenic factors, about which so far only very initial studies exist. These have not entered discussion in Metro Radon.

A further open problem, not addressed in Metro Radon, is the one of anthropogenically modified geogenic factors. This is typical for urban and built-up environments where geogenic controls - including geology itself - may not be equal to the one in its surroundings, i.e. open land, where data are usually being acquired in field studies (e.g. due to pavements, landscaping, landfills, historical construction activities etc.). The problem is important because most people live in strongly altered built-up environments. In particular in old European cities, this may challenge correct IRC estimation based on geogenic factors. It is recommended that the topic is addressed thoroughly in future investigations.

Methods to estimate the RP or GRP, respectively, have been addressed in this action. To illustrate it, case studies from two European countries are shown.

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