EURADOS
European Radiation Dosimetry Group e. V.

EURADOS Report 2020-01

Neuherberg, February 2020

12th EURADOS Winter School
Radon: Dosimetry, Metrology and Regulation

Arturo Vargas, Annette Röttger, James W Marsh, Pawel Olko, Martin Dubslaff, Marta García-Talavera, Claudia Grossi, Werner Hofmann, Estelle Rage, Krystian Skubacz and Hannah Wiedner

ISSN 2226-8057

ISBN 978-3-943701-20-3

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ISSN 2226-8057

ISBN 978-3-943701-20-3

Imprint

Issued by:
European Radiation Dosimetry e. V.
Ingolstädter Landstr. 1
85764 Neuherberg
Germany
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www.eurados.org

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Content

Abstract

The COUNCIL DIRECTIVE 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation addresses radon at workplaces and at homes: Radon is directly addressed in the Basic Safety Standards Directive through Article 54 concerning workplace radon levels, Article 74 concerning indoor radon levels, and Article 103 for the national action plan. The new directive brings significant changes, with respect to the former Directive 96/29/Euratom, in terms of protection measures related to exposures due to the inhalation of radon progeny not only in work environments but also in dwellings. The member states had 4 years (deadline 2018) to implement these regulations in their legislation. Consequently, several member states have developed new regulations and have encouraged metrological centers to develop the facilities for a quality measurement system according to the rules of the new directive.

Although protection against radon is primarily based on measurement and optimisation, dose estimates are required for workers if, despite optimisation, radon levels in a workplace remain above the national reference level (ICRP 126). Effective doses arising from unit exposure to radon and its progeny have been calculated using either dosimetric models or using the so-called 'epidemiological approach'. Both approaches give consistent results within their associated uncertainties (ICRP 137). Taking account of both methods, ICRP have recently recommended a single reference dose coefficient to be used, in most circumstances, for workers in buildings and in underground mines. Reference values are also given for specific situations of indoor work involving substantial physical activity, and for workers in tourist caves (ICRP 137). In special cases, where exposure conditions are non-typical, where sufficient, reliable aerosol data are available, and estimated doses are likely to be high, site-specific dose coefficients can be calculated using the dosimetric data provided in ICRP 137. This would require a careful analysis of the European workplaces with a coordinated action with an expert group performing field measurements for dose assessments.

The following papers are based on the presentations given in the EURADOS Winter School 2019 held in the Łódź University of Technology (Poland) on 14th February 2019. According to the topics the papers have been classified in four chapters: i) radon risk: dosimetry and epidemiological studies, ii) Standards and Regulations, iii) Metrology of radon and radon progeny and iv) Radon as a tracer.

1. Radon risk: dosimetry and epidemiological studies

1.1 Effective dose coefficients for inhaled radon and its progeny: ICRP's approach for workers (James W. Marsh and John D. Harrison, Public Health England (PHE), United Kingdom; and Dominique Laurier, Institut de Radioprotection et de Sûreté Nucléaire (IRSN), France)

1.1.1 Introduction

The International Commission on Radiological Protection (ICRP) has recently published dose coefficients for the inhalation of radon, thoron and their airborne progeny as well as recommendations for their use for the protection of workers (ICRP Publication 137). Although protection against radon is primarily based on measurement and optimisation, dose estimates are required for workers if, despite optimisation, radon levels in a workplace remain above the national reference level, or if, from the outset, radon exposure is considered as occupational as in the case of mines. ICRP recommends the use of a single effective dose coefficient of 3 mSv per mJ h m^{-3} (approximately 10 mSv per WLM) to be used in most circumstances, for workers in buildings and in underground mines. This paper discusses risks from radon, the methods used to calculate doses from exposure to radon progeny and the application of the reference effective dose coefficients. It briefly describes the general approach for the management of radon exposure in workplaces based both on ICRP recommendations and the European directive (EC, 2014). It is also shown that the recent UNSCEAR review supports the use of the ICRP reference dose coefficients for radiation protection purposes. The paper is an update of the papers by Marsh et al. (2017) and Harrison and Marsh (2012).

Radon has long been recognised as a cause of lung cancer. The International Commission on Radiological Protection (ICRP) issued recommendations on protection from radon at home and at work in Publication 65 (ICRP, 1991) and in its 2007 recommendations, Publication 103 (ICRP, 2007). Assessments of risks of radon-induced lung cancers have been mainly based on epidemiological studies of underground miners and more recent studies on miners have focussed on lower levels of exposures and lower exposure rates, more similar to those in homes and indoor workplaces. Estimates of lung cancer risk per unit radon exposure derived from these studies have increased. In addition, pooled case-control studies of residential radon exposures have been conducted. Since its 2007 recommendations, ICRP has published three more reports on radon exposure; (i) Publication 115 (ICRP, 2010) on lung cancer risks from radon and radon progeny, (ii) Publication 126 (ICRP, 2014) on radiological protection against radon exposure and (iii) Publication 137 (ICRP, 2017) on Occupational Intakes of Radionuclides (OIR), Part 3. The latter document gives doses coefficients for the inhalation of radon, thoron and their airborne progeny as well as recommendations for their use for the protection of workers.

Recently, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) conducted a review of radon epidemiology and dosimetry (UNSCEAR 2019). It is shown that the review supports the use of the ICRP reference dose coefficients for radiation protection purposes.

Special quantities and units that characterise the concentration and exposure of the radon progeny in air are described in appendix A.

1.1.2 Risk from radon

In ICRP Publication 65, (ICRP, 1991), the lifetime excess absolute risk (LEAR) of lung cancer death from radon and radon progeny was calculated (Table 1). The calculation of lifetime risk requires:

- risk coefficients derived from epidemiological studies, with or without modifying factors such as attained age.
- a projection model, enabling extrapolation of risk outside the range considered by the epidemiological study and transport to other populations.
- background reference rates for all-cause and lung cancer mortality, and
- a scenario of exposure to radon concentrations.

The GSF risk model used in the calculation of LEAR was derived from an epidemiological analysis of uranium miners in eastern Germany (ICRP, 1991). It has modifying factors of attained age and time since exposure but not of exposure rate. The model is a relative risk project model in which it is assumed that the interaction between radiation exposure and the underlying background rates is multiplicative. With this model the LEAR was estimated as 2.8×10^{-4} per WLM following exposure during adulthood for the ICRP Publication 60 reference population (ICRP, 1991).

In the BEIR VI report (NRC, 1999), risk models that take account of the effect of modifying factors of time since exposure, age and exposure rate were derived from the joint analysis of 11 cohorts of miners from China, Czech Republic, United States, Canada, Sweden, Australia and France. More recently, a risk model has been derived from the joint analysis of the French and Czech miner cohorts focusing on periods of work with low levels of exposure and good quality exposure assessment (Tomášek et al., 2008a). Tomášek et al. (2008b) used these risk models to calculate LEAR for the ICRP Publication 103 reference population (ICRP, 2007), and obtained values significantly greater than the previous estimate from Publication 65 (Table 1.1). As a result, ICRP now recommends a detriment-adjusted nominal risk coefficient for a mixed adult population of non-smokers and smokers of 5×10^{4} per WLM or 1.4×10^{-4} per mJ h m^{-3} (ICRP, 2010). This is approximately double the nominal risk coefficient given in ICRP Publication 65 (ICRP, 1993). The increase is mainly due to consideration of low exposures and low exposure rates. It should be noted that LEAR estimates are dependent upon the background lung cancer rates assumed for the reference population and this strongly depends on the prevalence of smoking.

Table 1.1 Estimates of the lifetime excess absolute risk (LEAR) of lung cancer associated to radon exposure, based on studies of underground miners.

Reference	Primary risk model	Background reference rates	Lifetime excess absolute risk of lung cancer (10^{-4} WLM^{-1})[a,b]
ICRP, 1993	ICRP 65 (GSF)[c]	ICRP 60	2.83
Tomášek et al., 2008b	ICRP 65 (GSF)[c]	ICRP 103	2.7
	BEIR VI[d]	ICRP 103	5.3
	Czech-French joint cohort	ICRP 103	4.4

(a) Exposure scenario: 2 WLM y^{-1} from age 18 to 64, lifetime risk at 90 y.
(b) A relative risk project model was assumed in the calculation of LEAR.
(c) The GSF model is described in ICRP Publication 65 (ICRP, 1993).
(d) The BEIR VI exposure-age-concentration model.

In addition to epidemiological studies of underground miners, case-control studies of lung cancer and residential radon exposures have been conducted. In particular, three pooled residential case-control studies have been carried out in Europe, North America and China. These three studies gave results that were statistically compatible and showed that the risk of lung cancer increased by at least 8 % for an increase in radon concentration of 100 Bq m^{-3}. After correcting for random uncertainties in the radon activity concentration measurements, the European pooled residential study gave an excess relative risk of 16 % per 100 Bq m^{-3} increase (Darby et al., 2006). This value was considered by ICRP as a reasonable estimate of the risk associated with relatively low and prolonged radon exposures in homes, considering exposure over a period of 25-30 years (ICRP, 2010).

Although comparisons between residential studies and miner studies are complex, appropriate comparisons of lung cancer risks estimates from recent miner studies and indoor studies show good consistency (Tomášek et al., 2008a; ICRP, 2010; Hunter et al., 2013). For example, based on conversions from WLM to time-weighted averaged radon activity concentration, the excess relative risk (ERR) estimates from the joint European studies on uranium miners were in agreement with those from the joint analysis of the European residential radon studies (Hunter et al., 2013).

1.1.3 Dose conversion convection

In ICRP Publication 65 (ICRP, 1993), the effective dose per unit of radon exposure was obtained using the so-called 'conversion convention method', also referred to as the 'epidemiological approach'. In this method, the estimated detriment associated with unit exposure to radon was divided by the detriment per unit effective dose (Equation 1.1). The former was determined from miner epidemiology and the latter determined mainly from epidemiological studies of Japanese atomic bomb survivors exposed largely to gamma rays. Hence, values of mSv (effective dose) per WLM were obtained and referred to as the dose conversion convention. It was estimated that the detriment coefficient for radon exposure is approximately equal to the lung cancer fatality coefficient because (i) the poor prognosis of lung cancer and (ii) that most of the dose is delivered to the lungs (ICRP, 1993).

$$\frac{Lifetime\ lung\ cancer\ risk\ \ per\ WLM}{Lifetime\ risk\ (detriment)\ per\ Sv} = Sv\ per\ WLM \qquad (1.1)$$

Table 1.2 Dose conversion convention coefficients obtained by equating ICRP values for total radiation detriment (all cancers and hereditary effects) with estimates of lifetime lung cancer risk from radon exposure (Marsh et al., 2010).

Exposed population	Lifetime lung cancer risk (WLM^{-1})	Total detriment (Sv^{-1})	Effective dose (mSv WLM^{-1})
Whole	2.8×10^{-4} (ICRP, 1993)	7.3×10^{-2} (ICRP, 1991)	4
	5.0×10^{-4} (ICRP, 2010)	5.7×10^{-2} (ICRP, 2007)	9
Workers	2.8×10^{-4} (ICRP, 1993)	5.6×10^{-2} (ICRP, 1991)	5
	5.0×10^{-4} (ICRP, 2010)	4.2×10^{-2} (ICRP, 2007)	12

With the nominal probability coefficient (fatality) of 2.83×10^{-3} WLM^{-1} for radon induced lung cancer given in ICRP Publication 65 (ICRP, 1993) and the total detriments per unit Sv of ICRP Publication 60 (ICRP, 1991), rounded values of the dose conversion convention were obtained: 5 mSv WLM^{-1} for

workers and 4 mSv WLM^{-1} for members of the public (Table 1.2). Marsh et al. (2010) updated these values to give 12 mSv WLM^{-1} for workers and 9 mSv WLM^{-1} for members of the public using the revised lifetime risk coefficient for radon exposure of 5.0×10^{-4} WLM^{-1} (ICRP, 2010) and the updated values of the detriment per Sv of ICRP Publication 103 (ICRP, 2007). These revised dose conversion convention values are about a factor of 2 greater than the previous estimates, mainly due to the doubling of the lifetime risk coefficient for radon exposure.

The dose conversion value for members of the public is derived assuming the risk per unit exposure to radon progeny is the same for adults and children. However, because the impact of radon exposure at young ages is unclear, the value for adults is more reliable than those for all ages.

1.1.4 Dosimetric approach

The organ that receives the highest dose from the inhalation of radon gas and its progeny is the lung. Radon (^{222}Rn) gas decays into a series of solid short-lived radioisotopes (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) creating an aerosol of solid particles suspended in air. Nearly the entire lung dose arises from the inhalation of the radon progeny aerosol and not from the gas itself. Because radon (^{222}Rn) is an inert gas, nearly all the radon that is inhaled is subsequently exhaled. However, a large proportion of the inhaled radon progeny deposits in the airways of the lungs. Due to their short half-lives, dose is delivered to the lung tissues before clearance can take place, either by absorption into blood or by particle transport to the gastro-intestinal tract. Two of these short-lived progenies, ^{218}Po and ^{214}Po, emit alpha particles, the deposited energy from which dominates the dose to the lung and the associated risk of lung cancer.

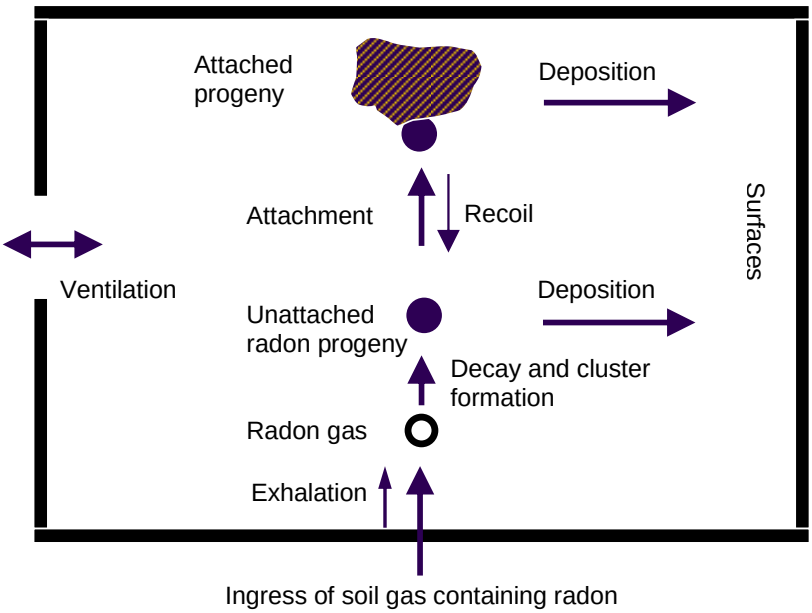

Figure 1.1 Schematic representation of the behaviour of radon progeny in an enclosed space (NRC, 1991; Porstendörfer, 1994)

The ground underneath buildings is usually the main source of indoor radon (^{222}Rn): soil gas carrying radon enters the building mainly due to pressure driven convection flow (Figure 1.1). Radon exhalation from the walls may also occur. The radon progeny aerosol in the atmosphere is created in two steps. After decay of the radon gas, the freshly formed radionuclides react rapidly (< 1 s) with trace gases and vapours and grow by cluster formation to form particles with diameters around 0.5

to 5 nm in size. These are referred to as unattached progeny. The unattached radionuclides may attach to existing aerosol particles in the atmosphere within 1 to 100 s forming the attached progeny. Most of the activity in the attached progeny is found in the accumulation mode with activity median diameters (AMD) between 100 and 450 nm.

Because radon progeny in the air can be removed by plate-out (i.e. by deposition on surfaces), the activity concentrations of the short-lived radon progeny in the air are less than that of the radon gas. This is quantified by the equilibrium factor, *F,* which is a measure of the degree of disequilibrium between the radon gas and its progeny. *F* is decreased further (i.e. there is greater disequilibrium) as a result of ventilation because as the ventilation rate increases, there is less time for the radon progeny to grow-in. If the exposure is characterised by radon gas measurements, a value for the equilibrium factor, *F*, is required to estimate the radon progeny concentration in air for the calculation of doses to the lung.

1.1.4.1 The ICRP Publication 66 Human Respiratory Tract Model

The ICRP Publication 66 Human Respiratory Tract Model (HRTM; ICRP, 1994) can be used to calculate the equivalent dose to the lung. The model considers both the extrathoracic airways and the thoracic airways (lung). The lung is divided into three regions: bronchial region (BB, airway generations 0-8), bronchiolar region (bb, airway generations 9-15), and the alveolar interstitial region (AI) where the gas exchange occurs.

For the purposes of the HRTM, radiosensitive cells in the central airways, have been identified and are assumed to be restricted to tissue layers of given depths and thicknesses. These are basal (BB_{bas}) and secretory (BB_{sec}) cells in the bronchial epithelium and clara cells (a type of secretory cell) in the bronchiolar epithelium. The central airways of the lung are modelled using simple geometric structures, sets of concentric cylinders, which represent the mucus gel layer, sol layer containing cilia, and the target cell layers. The radon progeny deposit in the mucus gel or sol layer and the alpha particles emitted from ^{218}Po and ^{214}Po have enough initial energy to reach the target cell layers. It is the dose the target cell layers in the central airways that are important for radon progeny whereas in comparison the dose to the AI region is significantly lower consistent with observed regional lung cancer distributions.

The equivalent dose to the lung is given by: $H_{lung} = w_R \times [A_{BB}(D_{bas} + D_{sec})/2 + A_{bb} D_{bb} + A_{AI} D_{AI}]$, where w_R is the radiation weighting factor for alpha particles (20), D_x is the absorbed dose in each target region, and A_i are the apportionment factors representing the regional's estimate sensitivity relative to that of the lung. In the absence of adequate quantitative information about the relative sensitivities of the different regions of the lung, ICRP assigned equal apportionment of the lung risk for the three regions of the lung: $A_{BB} = ⅓$, $A_{bb} = ⅓$, $A_{AI} = ⅓$ (ICRP, 1994).

For radon progeny dosimetry, the effective dose can be approximated by multiplying the equivalent lung dose by the ICRP tissue weighting factor for lung (0.12). That is, doses to body tissues from radon and progeny absorbed to blood are very low in comparison with the dose to the lungs. It is important to recognise that the effective dose is used for radiation protection purposes and not for risk assessment, and that simplifying assumptions were made in the development of the HRTM.

The effective dose per unit exposure to radon progeny calculated with the HRTM mainly depend on the breathing rate and the aerosol characteristics of the inhaled radon progeny such as the activity size distribution and the unattached fraction. For radiological protection purposes, most of the parameters in the dosimetric models, such as breathing rate, correspond to values for reference worker or reference person.

1.1.4.2 Effective dose for radon progeny

ICRP Publication 137 on Occupational Intakes of Radionuclides (OIR), Part 3 (ICRP, 2017) gives effective doses per unit exposure to radon progeny for indoor workplaces, mines, and tourist caves (Table 1.3). Reference aerosol parameter values were chosen for the difference exposure scenarios based on measurement data published in the open literature.

Table 1.3. Calculated values of effective doses per unit exposure to radon progeny for homes, indoor workplaces, mines and tourist caves (ICRP, 2017) (a). Dose from inhaling ^{222}Rn gas is excluded.

Place	Unattached fraction[b], f_p	F[c]	Effective dose per unit exposure[d]		
			mSv per WLM	mSv per mJ h m^{-3}	mSv per Bq h m^{-3}
Indoor workplace	0.08	0.4	20	5.6	1.2 x 10^{-5}
			(14)[e]	(3.9)[e]	(8.6 x 10^{-6})[e]
Mine	0.01	0.2	11	3.1	-
Tourist cave	0.15	0.4	23	6.6	1.5 x 10^{-5}
Home	0.08	0.4	13	3.7	8.2 x 10^{-6}

a) Effective doses coefficients for indoor workplaces, mines and tourist caves were calculated by the ICRP Task Group (ICRP, 2017) whereas the value for home was calculated by Marsh and Bailey (2013).
b) f_p = unattached fraction in terms of the potential alpha energy concentration (PAEC).
c) F = equilibrium factor.
d) 1 WLM = 3.54 mJ h m^{-3} and 1 WLM = (6.37 x 10^5/F) Bq h m^{-3} of ^{222}Rn exposure.
e) Approximate values calculated for a sedentary worker with a lower breathing rate of 0.86 m^3 h^{-1} compared with the reference breathing rate of 1.2 m^3 h^{-1}.

The dose calculated for an indoor workplace (5.6 mSv per mJ h m^{-3}) is higher than that for homes (3.7 mSv per mJ h m^{-3}) mainly because of the higher breathing rate assumed for a reference worker (1.2 m^3 h^{-1}) compared with the averaged breathing rate assumed for an adult while at home (0.78 m^3 h^{-1}). The reference breathing rate for a worker (one-third of time spent sitting, two-thirds of time spent undertaking light exercise) may be considered as an over-estimate for sedentary occupations such as officer workers (Harrison and Marsh, 2012). Assuming a lower breathing rate of 0.86 m^3 h^{-1} (⅔ sitting, ⅓ light exercise) would reduce the dose coefficient from 5.6 mSv per mJ h m^{-3} to approximately 3.9 mSv per mJ h m^{-3} (20 mSv WLM^{-1} to approximately 14 mSv WLM^{-1}) for an indoor workplace.

It is acknowledged that there is large variability in the measured aerosol parameter values depending upon the actual exposure conditions. For this reason, ICRP Publication 137 also gives dosimetric data as a function of aerosol size so that 'site-specific' dose coefficients can be calculated in cases where reliable aerosol data are available.

1.1.5 Applications of dose coefficients

The updated dose conversion convention value for workers derived from epidemiology is 3.4 mSv per mJ h m^{-3} (12 mSv WLM^{-1}). In comparison, effective dose coefficients calculated with ICRP

reference biokinetic and dosimetric models gave values of 3 to 4 mSv per mJ h m^{-3} (11 to 14 mSv WLM^{-1}) for workers in mines, sedentary indoor workers and exposures in homes. Thus, there is good consistency between the dose coefficients obtained by dosimetric calculations and by epidemiological comparisons. However, this does not indicate accuracy or precision in the estimates as there are uncertainties associated with both approaches. Taking account of both approaches, ICRP recommends a single rounded value of 3 mSv per mJ h m^{-3} (approximately 10 mSv WLM^{-1}) for workers in buildings and in underground mines to be used in most circumstances without adjustment for aerosol characteristics. ICRP also recommends a reference value of 6 mSv per mJ h m^{-3} (approximately 20 mSv WLM^{-1}) for specific situations of indoor work involving substantial physical activity, and for workers in tourist caves.

In cases where aerosol characteristics are significantly different from typical conditions, where sufficient, reliable aerosol data are available, and estimated doses warrant more detailed consideration, it is possible to calculate site-specific dose coefficients using the dosimetric data provided in ICRP Publication 137 and the associated electronic annex (ICRP, 2017).

1.1.5.1 Comparison with UNSCEAR report

Recently, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) conducted a review of radon epidemiology and dosimetry. A summary of its finding has been published in its report to the UN General Assembly (UNSCEAR, 2019) and the full scientific report is to be published in 2020. It included a review of published dosimetry assessments for exposures in homes, indoor workplaces and mines. For homes, the dose coefficients ranged from 1.1 to 5.9 mSv per mJ h m^{-3} with an arithmetic mean of 3.1 mSv per mJ h m^{-3}. Thus, the ICRP reference dose coefficients are consistent with these values.

UNSCEAR reviewed epidemiological studies of lung cancer in underground miners and estimated lifetime risks using similar methodology to that applied in ICRP Publication 115. Estimates of LEAR were calculated by applying the BEIR VI risk model individually to updated miner cohort studies (Wismut (Kreuzer et al., 2018), Czech Republic (Tomášek, 2012); Eldorado (Lane et al., 2010)) as well as to the combined 11 studies used in the BEIR VI report (NRC, 1999). The LEAR estimates ranged from 2.4 to 7.5 x 10^{-4} per WLM for the Wismut and Eldorado studies respectively (UNSCEAR, 2019). These values are consistent with a LEAR value of 5 x 10^{-4} per WLM proposed by the ICRP for radiation protection purposes.

The ranges in the LEAR estimates derived by UNSCEAR correspond to dose conversion convention values for workers of 1.6 to 5.1 mSv per mJ h m^{-3} (5.7–18 mSv per WLM) calculated here using the ICRP Publication 103 detriment per Sv value for adults. Again, the ICRP reference dose coefficients are consistent with these values.

1.1.6 Protection against radon.

ICRP's protection policy against radon is based on setting reference levels and applying the principle of optimisation to maintain or reduce exposures to as low as reasonably achievable (ALARA). For indoor radon, the reference level is expressed as an annual average radon activity concentration and represents a level where action would almost certainly be warranted to reduce exposure. The ICRP statement on radon included in Publication 115 (ICRP, 2010) lowered the upper reference level for homes from 600 Bq m^{-3} to 300 Bq m^{-3} in line with the change in the nominal risk coefficient for radon. In its most recent publication on protection against radon exposure (ICRP, 2014), the Commission retains the upper reference level value of 300 Bq m^{-3} for dwellings and recommends the same value

of 300 Bq m^{-3} for all other buildings and workplaces. However, it is the responsibility of national authorities to establish their own national reference level (NRL), taking the prevailing economic and societal circumstances into account.

1.1.6.1 Specific graded approach for control of radon exposure

For some workplaces, such as thermal spas, caves, mines and other underground workplaces, national authorities may consider from the outset that workers' exposure to radon is occupational. However, in most workplaces, radon exposures are not considered as occupational because the exposures are more related to the location than the work activity. Workplaces in this category include most mixed-use buildings, such as schools, hospitals, offices, post offices, jails, shops, restaurants, cinemas and general workshops. In these workplaces, a specific graded approach is recommended in which the first step is radon measurement and to reduce the activity concentration of radon ALARA (Figure 1.2). If difficulties are met in the first step, a more realistic approach is recommended as a second step, consisting of optimising protection using the actual parameters of the exposure situation, such as occupancy, together with a reference level of 10 mSv annual effective dose. If despite all reasonable efforts, doses remain above 10 mSv y^{-1}, then the workers are considered as occupationally exposed and the relevant requirements for occupational exposure would apply. The effective dose of 10 mSv y^{-1} corresponds to an average radon concentration of 650 Bq m^{-3} using an effective dose coefficient of 3.0 mSv per mJ h m^{-3}, an occupancy of 2000 h y^{-1} and $F = 0.4$. However, for specific situations of indoor work involving substantial physical activity, and for workers in tourist caves, ICRP recommends a reference value of 6 mSv per mJ h m^{-3} (approximately 20 mSv WLM^{-1}).

1.1.6.2 European directive

The European directive has a similar approach to that of the ICRP for the management of radon in the workplaces (EC, 2014). Radon measurement should be carried out in workplaces as specified in the directive to demonstrate compliance with NRLs. The NRL for indoor workplaces should not be higher than 300 Bq m^{-3}, unless it is warranted by national circumstances. If following optimisation, the annual average radon concentration in a workplace continues to exceed the NRL, then the relevant regulator must be notified. A dose assessment is required and if this exceeds an effective dose of 6 mSv per year then this should be managed as a planned exposure situation whereas if below or equal to 6 mSv per year they must be kept under review [Article 35 (EC, 2014)]. The meaning of the annual average radon concentration is an estimate of the average radon activity concentration over a year for 100% occupancy. The actual occupancy is considered in the dose assessment according to Article 35 (EC, 2014). The effective dose of 6 mSv y^{-1} corresponds to an average radon concentration of 450 Bq m^{-3} using an effective dose coefficient of 3.0 mSv per mJ h m^{-3}, an occupancy of 2000 h y^{-1} and $F = 0.4$.

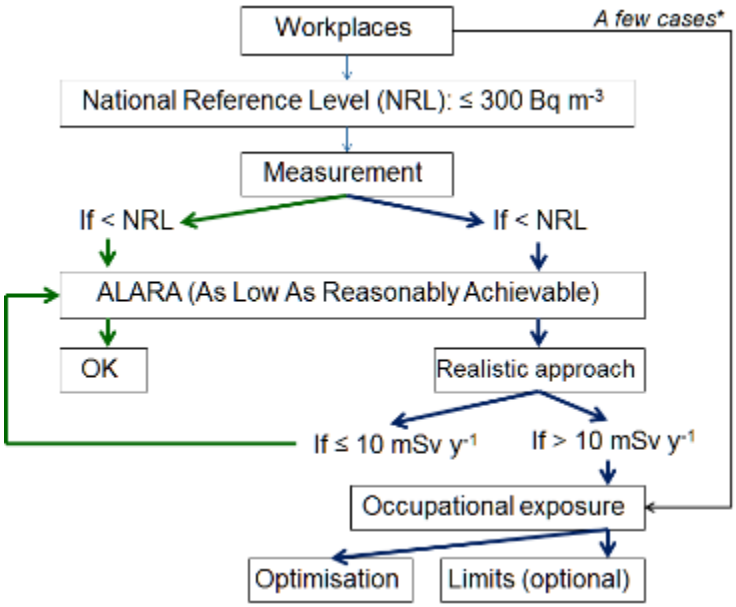

Figure 1.2 Specific graded approach for the management of radon exposure in workplaces. Adapted from ICRP Publication 126 (ICRP, 2014). *Workplaces where, from the outset, radon exposure is considered as occupational by national authorities.

1.1.7 Summary and conclusions

There is strong evidence from cohort studies of underground miners and case-control studies of residential radon exposures that exposures to radon and its progeny can cause lung cancer. Lung cancer risk estimates from both miner studies and indoor studies show good consistency. Radon exposure is considered to be the second leading cause of lung cancer after smoking. Because of the synergistic effect of tobacco smoking and radon, radon is much more likely to cause lung cancer in people who smoke or have smoked in the past. However, radon is the primary cause of lung cancer among people who have never smoked.

Although protection against radon is based primarily on measurement and optimisation, dose estimates are required for workers in certain situations. Based on both dosimetry and epidemiological data, ICRP recommends the use of a single effective dose coefficient of 3 mSv per mJ h m^{-3} (approximately 10 mSv per WLM) to be used in most circumstances, for workers in buildings and in underground mines (ICRP, 2017). In terms of measurements of ^{222}Rn gas exposure, this corresponds to 6.7×10^{-6} mSv per Bq h m^{-3} assuming an equilibrium factor, F of 0.4.

UNSCEAR's recent review of radon epidemiology and dosimetry provides support for the use of ICRP's recommended effective dose coefficient of 3 mSv per mJ h m^{-3} for radiation protection purposes. It must be emphasised that the UNSCEAR's established dose conversion factor of 1.6 mSv per mJ h m^{-3} should not be used for dose assessment for workers, instead the ICRP dose coefficients should be used for this purpose. UNSCEAR uses its established value when comparing radon exposure levels with other sources of radiation exposures.

Protection against radon in homes and indoor workplaces is based on measurement of radon gas and the application of reference levels and optimisation. In line with its revised nominal risk coefficient for radon, the Commission now recommends an upper derived reference level of 300 Bq m^{-3} for dwellings. The same value is recommended for all other buildings and workplaces. A

specific graded approach for the control of radon in workplaces is recommended. However, for some workplaces, such as underground mines and thermal spas, national authorities may consider from the outset that workers' exposure to radon is occupational.

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APPENDIX A: SPECIAL QUANTITIES AND UNITS

Special quantities and units used to characterise the concentration of the radon and radon progeny in air, and the resulting inhalation exposure has been described in ICRP Publications 65 and 137, (ICRP, 1993; 2017) as well as in the ICRU Report 88 (ICRU, 2012). A brief summary is given here.

A.1 Concentration

The *radon activity concentration* is the activity per unit volume of the gas, expressed in units of Bq m^{-3}.

The dose to the lung mainly arises from the inhalation of airborne radon progeny and the alpha particles emitted during their decay and that of their short-lived progeny. Consequently, the quantity "*potential alpha energy concentration* (PAEC)" of the radon progeny mixture was historically used as a measure of concentration that was an indicator of dose and risk. The PAEC is the total alpha energy ultimately emitted by the decay of the short-lived progeny in unit volume of air. The historical unit of PAEC is the working level (WL). One WL was originally defined as the concentration of the potential alpha energy associated with radon progeny in equilibrium with 100 pCi per litre (3,700 Bq per litre). However, in the UNSCEAR 1982 report (UNSCEAR, 1982) and in ICRP Publication 65 (ICRP, 1993), it was redefined as any combination of the short-lived radon progeny in 1 m^3 of air that will result in the emission of 1.300×10^8 MeV of alpha energy (i.e. a PAEC of 1.300×10^8 MeV m^{-3} or 2.08×10^{-5} J m^{-3}).

The *equilibrium equivalent concentration* (EEC) is defined as the activity concentration of radon gas, in equilibrium with its short-lived progeny, which would have the same potential alpha energy concentration as the existing non-equilibrium mixture. One WL equals approximately 3,750 Bq m^{-3} of EEC of ^{222}Rn. The EEC is therefore a measure of the radon progeny concentration or more precisely the PAEC.

A.2 Equilibrium Factor, *F*

The *equilibrium factor*, *F* is defined as the ratio of the EEC to the radon gas concentration. In other words, it is the ratio of the PAEC for the actual mixture of radon decay products to that which would apply at radioactive equilibrium.

A.3 Exposure

Exposure is the time integral of the concentration. The potential alpha energy (PAE) exposure is the time integral of the PAEC in air and has the historical unit of the *working level month* (WLM). The WLM is defined as the cumulative exposure from breathing an atmosphere at a concentration of 1 WL for a working month of 170 hours. One 1 WLM = 3.54 mJ h m^{-3}. For ^{222}Rn, if the exposure is expressed in terms of the radon gas concentration then the two units are related via the equilibrium factor: 1 WLM = (6.37×10^5 / *F*) Bq h m^{-3}.

A.4 Unattached Fraction

The *unattached fraction*, f_p is defined as the fraction of the potential alpha energy concentration (PAEC) of the short-lived progeny that is not attached to the ambient aerosol. If there is a need for a

more precise definition of unattached progeny, then (ICRU, 2012) proposes 5 nm diameter as an upper limit for the unattached progeny (i.e. clusters carrying progeny).

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1.2 Lung cancer risk form radon and radon progeny - Epidemiological studies (Estelle Rage, Institut de Radioprotection et de Sûreté Nucléaire (IRSN), France)

1.2.1 Introduction

Radon is a radioactive gas of natural origin, emitter of alpha particles and coming from the disintegration of uranium from the soil. It is colorless, odorless and it is present everywhere at the surface of the earth and concentrates in confined places, such as mines or home. Consequently, population of workers such as miners, but also the general population is chronically exposed to radon and its short-lived progeny over the lifespan (ICRP, 2010). After inhalation of radon and its progeny, the radionuclides distribute in the whole body, mainly in the lungs leading to an irradiation of the bronchial epithelium. The dosimetric models confirmed that the received dose is delivered mainly to the lung, for more than 90 %, and the dose delivered to the other organs is 100-fold lower (Marsh et al., 2012).

Historically, an unusual mortality from respiratory disease was observed among young miners as soon as the 16th century. At the end of the 19th century, these diseases were identified as bronchial cancer and at the same time, in 1898, Pierre and Marie curie discovered the radium. At the beginning of the 20th century, in 1924, a first mention of occupational disease was made, and several years later, the inhalation of radon was presented as a possible cause for these diseases (by Planck). Just after the Second World War, the intensive extraction of uranium began. In France, this extraction began in 1946, and the first measures of radioprotection were set up ten years later. The first epidemiological studies conducted among miners were launched in the 1960's and the first epidemiological studies conducted among the general population were launched in the 1990's. 1988 is a key date where the International Agency for Research on Cancer classified radon as a known pulmonary carcinogen in humans (IARC, 1988).

1.2.2 Epidemiological studies among uranium miners

Radon concentrates in uranium mines, but also in iron, tin or fluorspar mines. About tens countries around the world have set up cohorts of miners to study the effect of radon exposure on health risks and especially on lung cancer risk. Indeed, miners constitute a relevant population for epidemiological studies due to their chronical exposure to ionising radiation (IR), and especially to radon. They are exposed to different levels of doses and dose rates, depending on the mine location and the exposure period, providing contrasting levels of exposure. Cohorts of miners are generally well followed-up in terms of dosimetric, administrative or mortality information. The international pooled 11 underground miner cohort study including 60,606 miners highlighted a significant association between the increased risk of death from lung cancer and cumulative radon exposure within each cohort and in the combined cohort[4]. The analysis of time-dependent modifying factors showed a decrease of this risk with the age at exposure and with the time since exposure (NRC, 1999). An interaction between radon exposure and smoking habits was suggested as sub-multiplicative. No evidence of an association for other health effect was observed with radon exposure (NRC, 1999).

To give some elements on cohort building and to illustrate main known results, the following mainly deals with the French cohort of uranium miners.

The French cohort has been set up in 1982 and included subjects according to defined selection criteria (males, employment at least one year, between 1946 and 1990 in the French CEA and COGEMA companies) (Laurier et al., 2004). Miners were then followed-up over the time, from 1946 to the end of 2007 (current end point), for 35.4 (range 0.1-61.0) years in average. At this time, vital

status was supplied by a national registry, and for deceased miners, causes of death were collected from the specific national database and for former miners, from the occupational medical service of the COGEMA company. Additionally, occupational history and annual occupational IR exposures were respectively collected from administrative databases of the company and from a specialised dosimetric department. The French cohort is characterised by three periods of radon exposure assessment. From 1956, the date where radiation protection measures were set up, an individual recording of radon exposure was performed from ambient measurements. From 1983, individual measurement was done from an integrated system of individual dosimeter (ISID). Before 1956, only few measures were done and a reconstitution of the exposure was estimated by an expert group. Uranium ore dust exposure was reconstructed between 1956 and 1958, assessed from ambient measurements between 1959 and 1982, and then measured by ISID. External gamma rays exposure was individually measured from 1956. The cumulative radon exposure was 36.6 Working Level Month (WLM) in average in the whole cohort (1946-2007) for a mean duration of 11.8 years. The cumulative radon exposure was 17.8 WLM in average from 1956 (1956-2007). Indeed, the forced ventilation set up in 1956 clearly marked a sharp decrease of the annual exposure (annual mean = 21.3 WLM before 1956 and 1.7 WLM from 1956) (Rage et al., 2015).

Most of the main results observed in the French cohort have been confirmed in other cohorts.

Mortality from lung cancer has been assessed by the Standardised Mortality Ratio (SMR) comparing, according to categories of age and calendar period, the risk of mortality from lung cancer observed in the cohort with the risk of expected cases if the cohort came from the general male French population, used as a reference population. Among the 5,086 French miners, 211 of them died from lung cancer. A significant SMR of 1.34 [95 % Confidence Interval (CI):1.16-1.53] expressed a significant increase of the risk of death from lung cancer (Rage et al., 2015).

The exposure-risk relationship was assessed with a linear excess relative risk (ERR) model, fitted by a Poisson regression. A 5-years lag was applied to take into account a minimum time between the radon exposure and the time to develop a lung cancer potentially related to radon exposure. An increase of the risk of lung cancer death was significantly associated with cumulative radon exposure (ERR / 100 WLM = 0.71 [95 % CI: 0.31-1.30]).

At low level, the ERR for lung cancer death remained significant, as observed in the French +1955 sub-cohort including the 3,377 miners hired from 1956 only, i.e. at lower radon exposure. A total of 94 cases of lung cancer were observed and the ERR remained significant (ERR = 2.42 [0.09-5.14] per 100 WLM), with a value higher than in the whole cohort (Rage et al., 2015). As another example, an increase of the lung cancer risk was observed at lower radon exposure in the Wismut cohort, including a total of 26,766 miners hired from 1960 in the + 1960 sub-cohort (N cases = 334; ERR = 1.3 [0.7-2.1] per 100 WLM) (Kreuzer et al., 2015) compared to the whole cohort including 58,987 miners exposed to a higher level of radon (N cases = 3,016; ERR = 0.19 [0.16-0.22] per 100 WLM) (Walsh et al., 2010). The increase of the risk observed at low exposure is probably due to the higher accuracy of the exposure assessment in the recent period compared to the large uncertainties related to the exposure assessment in the older period (Hoffmann et al., 2017).

Multiple chronic IR exposure. In addition to radon, uranium miners were also exposed to long-lived radionuclides (LLR) coming from uranium ore dust and to external gamma rays. In the French + 1955 sub-cohort, the risk of lung cancer death was found to be significantly associated with each of these IR exposures. The question of the own effect of each IR exposures arose. But due to the multi-collinearity between these exposures, the limited size of the cohort and the lack of statistical power that ensues from it, the models applied could not be fitted and didn't allow the estimation of the

own risks (Vacquier et al., 2011). In the other side, the large German Wismut cohort conducted similar analyses on the + 1960 sub-cohort and found that the ERR associated with radon exposure (ERR / WLM = 0.013 [0.007-0.021]) was only slightly modified and remained significant after adjustment for external gamma rays (ERR / WLM = 0.011 [0.004-0.019]) or LLR (ERR / WLM = 0.014 [0.007-0.022]) (Kreuzer et al., 2015).

To consider **multiple chronic radiation exposure**, a **dosimetric approach** was also investigated in the frame of the European collaborative Alpha-Risk project conducted between 2005 and 2009 (Tirmarche et al., 2010). Absorbed doses were calculated for different target organs (such as red bone marrow, kidney, or liver). For the lungs, the calculation of the absorbed doses was based on the Human Respiratory Tract Model (HRTM) as described in ICRP Publication 66 (ICRP, 1994). Different aerosol parameter values were assumed for different exposure situations, according to the type of drilling (dry/wet), type of mine (underground/open pit), type of ventilation (poor/medium/good), and the use of diesel engines. Different breathing rates were also assumed, depending upon the type of job (hewer/other) (Marsh et al., 2010; 2012; Rage et al., 2012). The mean cumulative total absorbed lung dose was 134 mGy among the 3,271 exposed French miners, 428 mGy among the 9,979 exposed Czech miners and 312 mGy among the 29,086 exposed German miners. Alpha emitters (i.e. radon gas, radon progeny, and LLR) contributed to 58 % of the total lung dose among the French miners and to 87 % among the Czech and German miners. Among alpha contributors, radon progeny represented more than 97 % of the alpha lung dose. The analysis of the dose-risk relationship in the joint European cohort showed a significant association between the risk of lung cancer death (N = 1,444 cases) and the total lung dose (ERR/Sv = 0.07 [90 % CI: 0.06-0.08]), but also with the alpha and non-alpha lung doses in separate models. Significant associations were also observed for each of the alpha component (ERR/Sv = 0.07 [90 % CI: 0.06-0.08]), despite large uncertainties on estimates for LLR (ERR/Sv = 9.38 [90 % CI: 7.19-12.17]). In multivariate analyses where all IR exposures were considered together, the risk remained significant for radon gas + radon progeny (ERR/Sv = 0.05 [90 % CI: 0.03-0.06] and for LLR (ERR/Sv = 2.92 [90 % CI: 1.13-5.17]), whereas the association with non-alpha lung dose did not remain (Laurier et al., 2009).

The analysis of **time-dependent modifying factors** showed a decrease of the lung cancer risk with the time since exposure and the attained age (Vacquier et al., 2009) as observed in the international 11 country miner cohort analysis (NRC, 1999) (BEIR VI). An analysis conducted among the combined Czech-French cohort confirmed their modifying effect (Tomasek et al., 2008). **Tobacco** is the first risk factor for lung cancer. Unfortunately, it has not been systematically collected at the setting up some uranium miner cohorts that is why nested case-control studies have been conducted. On the basis of the Czech (Tomasek, 2011), French (Leuraud et al., 2007) and German (Schnelzer et al., 2010) nested case-control studies, the Alpha-Risk project investigated a combined nested case-control analysis including a total of 1,236 cases of lung cancer death and 2,678 controls for which information on smoking habits have been collected (non-smokers/former smokers/current smokers). The association remained significant after taking into account for **smoking status**, and the increased risk remained among each category of smokers. As suggested in the international 11 country miner cohort analysis (BEIR VI), the analysis of the relation between lung cancer risk and radon exposure and smoking is in favor of a sub-multiplicative interaction between radon and smoking (Leuraud et al., 2011).

1.2.3 Epidemiological studies among general population

Lung cancer risk **from residential radon** exposure has been estimated from about twenty case-control studies conducted from the 1990's. Individual studies had limited statistical power, like the

French study which included 486 cases of lung cancer death and 984 controls. The estimated relative risk presented a borderline increase (RR/100 Bq m^{-3} = 1.04 [CI95 %:0.99-1.11]) (Baysson et al., 2004). Only few of these studies were able to detect a significant increase of lung cancer risk associated with radon exposure. Gathering several case-control studies allows an increase of the statistical power. Three combined analyses conducted among the Chinese, North American and European studies observed an increase of lung cancer risk associated with residential radon exposure. The Chinese study, gathered two studies, included 1,050 cases and 1,995 controls, and found a significant relative risk (RR/100 Bq m^{-3} = 1.13 [CI95 %:1.01-1.36]) (Lubin et al., 2004). The North-American study, gathered seven studies, included 3,662 cases and 4,966 controls, and found a borderline association (RR/100 Bq m^{-3} = 1.10 [CI95 %:0.99-1.26]) (Krewski et al., 2006). The European cohort, the largest one, gathered 13 studies from nine countries (Darby et al., 2005). In this last study, a standardised protocol was used for the 13 studies, specifically based on identical inclusion criteria, common questionnaire, and reconstruction of indoor radon exposure for 30 years. A total of 7,148 cases and 14,208 controls were included. The indoor mean radon concentration was 104 Bq m^{-3} among cases and 97 Bq m^{-3} among controls. The risk of lung cancer increased by 8.4 % (95 % confidence interval 3.0 % to 15.8 %) per 100 Bq m^{-3} increase in measured radon (RR/100 Bq m^{-3} = 1.08 [CI95 %: 1.03-1.16]). After consideration of uncertainties related to estimations of radon concentration, this risk increased by 16 % (95 % confidence interval 5 % to 31 %) per 100 Bq m^{-3} increase in measured radon (RR/100 Bq m^{-3} = 1.16 [CI95 %:1.05-1.31]). At low exposure, for homes where radon concentrations were lower than 200 Bq m^{-3} the exposure-risk relationship remained significant (Darby et al., 2005). Smoking history has been reconstructed for all included subjects. The risk of lung cancer death was significantly increased among each category of smokers (current smokers, ex-smokers < 10 years, ex-smokers ≥ 10 years, and nonsmokers), with a higher risk among smokers than nonsmokers (Darby et al., 2007).

Another way to quantify the risk of lung cancer associated with radon exposure is the **attributable fraction**. A recent analysis assessed the national residential radon exposure mean for 66 countries, ranging from 5 Bq m^{-3} in Cuba to 133 Bq m^{-3} in Poland, with an average among the pooled countries at 38 Bq m^{-3}. The population attributable risk (PAR) has been assessed from several risk models. Similar results were provided by different models used among miner studies, with a PAR ranging from 13.6 % to 16.5 %, meaning that 13.6 % to 16.5 % of lung cancer deaths could be attributable to residential radon (Gaskin et al., 2018).

In summary, epidemiological results coming from uranium miner studies and residential studies present a good consistence and confirmed the significant increased risk of lung cancer death associated with cumulative radon exposure. Both observed a persistence of the risk at low level of radon. Taking into account for smoking habits did not modify the significance of the exposure-risk relationship and studies are in favor of a sub-multiplicative interaction between smoking habits and radon exposure. To date, lung cancer is the only highlighted risk associated with radon exposure. Clearly, isolated increased risks have been observed, such as leukemia, skin cancer, brain cancer or stomach cancer in different studies, but the estimation of low risks needs to be refined, particularly by large combined studies. Currently, a large international project, the Pooled Uranium Miners Analysis (PUMA), is setting up to include more than 120,000 uranium miners arising from seven cohorts (from Canada, Czech Republic, France, Germany, and USA, Richardson et al., 2017). This project will particularly allow to better address questions concerning low exposure and low exposure rate effects on lung cancer risk, to have greater ability to describe temporal modifier factors, and to better quantify the impact of radon progeny versus other radiological exposures present in uranium mines. Risks for cancers other than lung and non-cancer diseases will be also

investigated. Future findings from the PUMA study should provide additional information to strengthen our understanding of radon and radon progeny-related to lung cancer risks, and also to other cancers and non-cancer risks.

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1.3 Alpha particle microdosimetry in the lungs: Variability of energy deposition at the cellular level (Werner Hofmann, University of Salzburg, Austria)

1.3.1 Introduction

The variability of energy deposition at the cellular level results from random variations of stochastic macroscopic factors (source variability) and microscopic factors (microdosimetry). Internal microdosimetry comprises both sources of variability. Stochastic macrosocopic factors are the variability of radon progeny deposition in a stochastic lung, the microdistribution of radon progeny surface activities on bronchial airway surfaces, the variability of mucus clearance velocities, and the diameter-related depth distribution of basal and secretory cell nuclei in bronchial epithelium. Microscopic factors causing fluctuations of energy deposition in cell nuclei are the hit probability and the related multiplicity of hits, the random track lengths of alpha particles in spherical target cell nuclei, and the random energy deposition along the track of an alpha particle as a result of the range distribution of alpha particles (Bragg curve). Two microdosimetric parameters have been proposed to describe the variability of energy deposition in microscopic targets, the hit probability and the frequency of cellular hits and specific energy distributions for single and multiple hits. Several approaches have been proposed to establish a relationship between these microdosimetric parameters and cellular radiobiological effects, such as hit-related models, effect-specific track length models, effect-related interpretations of specific energy distributions, and finally track structure models (nanodosimetry).

Current radon progeny lung dosimetry models provide information on average absorbed doses in basal and secretory cell nuclei, either in individual airway generations, as in the case of airway generation models, or in bronchial (BB) and bronchiolar (bb) regions (Winkler-Heil et al., 2007). However, cellular radiobiological effects relevant for lung cancer induction depend on the energy actually deposited in individual cell nuclei and not on a computed average value for all target cells in bronchial epithelium. Thus, in contrast to the average dose approach, the microdosimetric approach considers the variability of energy deposition in individual cell nuclei, which is particularly significant at low doses and dose rates, where only a small number of cells in epithelial tissue is actually hit, receiving a wide range of cellular doses.

In radon progeny lung dosimetry, steady-state ^{218}Po and ^{214}Po surface activities on cylindrical airway surfaces represent the alpha particle source and basal and secretory cell nuclei at a given depth in bronchial epithelium represent the target (Figure 1.3). The variability of energy deposition at the cellular level is the result of random variations of macroscopic factors (source variability) and microscopic factors (target variability). While the source variability represents an internal dosimetry problem, microdosimetry considers only the target variability, thus internal microdosimetry comprises both source and target variability,

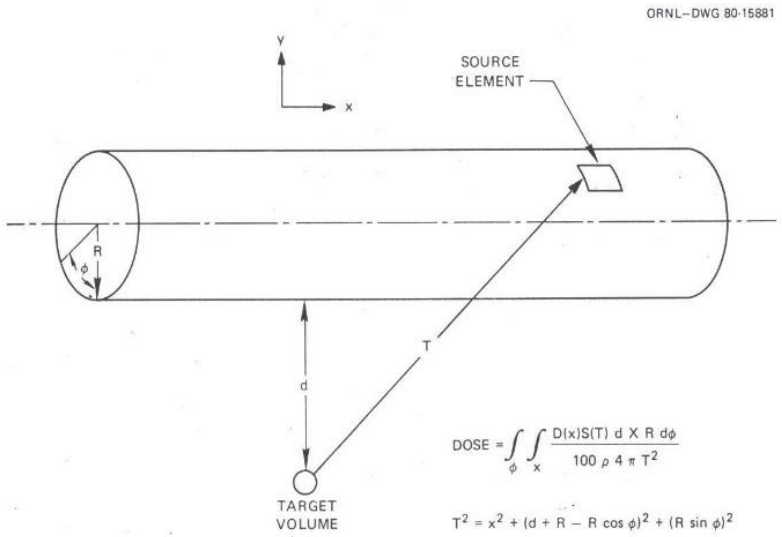

Figure 1.3 Irradiation geometry of sensitive target cells in bronchial epithelium by radon progeny alpha particles emitted from bronchial airway surfaces.

1.3.2 Macropscopic factors – source variability

Stochastic macroscopic factors are (1) the inter- and intrasubject variability of radon progeny deposition in an asymmetric, stochastic airway structure, (2) the microdistribution of radon progeny surface activities on bronchial airway surfaces, such as accumulations at airway bifurcations, (3) the variability of mucus clearance velocities as a result of the morphometric variability, (4) the distribution of basal and secretory cell nuclei across the bronchial epithelium, and (5), the diameter-related thickness of the bronchial epithelium and the resulting depth distribution of basal and secretory cell nuclei. These macroscopic factors can be described by statistical distributions and thus require the application of stochastic modelling techniques, e.g. Monte Carlo methods.

The asymmetric, stochastic lung model of Koblinger and Hofmann (1985; 1990), derived from the morphometric measurements of Raabe et al. (1976), provides the morphometric basis for stochastic dose calculations. This model describes the inherent asymmetry and variability of the human airway structure, describing airway diameters and lengths in each airway generation by lognormal probability density functions. Upon inhalation, the random walk of an inhaled particle through the airway system is randomly selected by Monte Carlo methods from individual airway parameter distributions and their statistical correlations. As a result of the asymmetry and variability of the airway geometry and related flow rates, deposition fractions of inhaled radon progeny in each airway generation exhibit a wide range of values, which can be approximated by lognormal distributions (Hofmann et al., 2010).

Experimental (Kinsara et al., 1995) and CFD simulation studies (Balásházy and Hofmann, 2000) for inhaled attached and unattached radon progeny in bronchial airway bifurcation models have demonstrated that inhaled nuclides are preferentially deposited within airway bifurcation zones, thereby producing activity hot spots in the vicinity of the carinal ridge. This local deposition enhancement is caused by secondary motions which are functions of the local flow rate. Since flow rates significantly decrease with penetration into the lung, enhanced deposition at bifurcations is only effective in large bronchial airways. This localised deposition is further enhanced by the reduced mucociliary clearance, or even stasis, at airway branching sites (Hofmann et al., 1990). While these

radon progeny accumulations increase only moderately average bronchial cellular doses relative to uniform distributions, related microdosimetric specific energy distributions reveal significant differences (Fakir et al., 2005).

Since the mucus velocity in a given bronchial airway is related to the diameter and length of that airway, the variability of linear airway dimensions in the stochastic lung model leads to corresponding distributions of mucus velocities in bronchial airway generations (Hofmann and Sturm, 2004).

Morphometric measurements of Mercer et al. (1991) have provided information about the volumetric densities of basal and secretory cell nuclei across the bronchial epithelium. Basal and secretory cell nuclei differ not only in their depth distributions, i.e. deeper lying basal cells vs. shallow lying secretory cells, but also in their relative magnitudes, i.e. higher frequency of basal cell nuclei in large bronchial airways.

The depths of basal and secretory cell nuclei in a given airway depend on the diameter of that airway. Based on the morphometric measurements of Mercer et al. (1991), the correlation of basal and secretory cell depths with corresponding airway diameter can be expressed by a polynomial function (ICRU, 2012). As a result of the asymmetric stochastic airway structure, where airway diameters in a given bronchial airway generation are described by lognormal distributions, the depths of basal and secretory cell nuclei are also lognormally distributed.

Steady-state ^{218}Po and ^{214}Po surface activities on cylindrical airway surfaces, resulting from the combined action of deposition, clearance and radioactive decay, finally represent the alpha particle source for subsequent cellular dose calculations. Thus, the variability of deposition fractions, mucus clearance velocities and bronchial airway surfaces leads to corresponding distributions of the resulting surface activities for both attached and unattached radon progeny.

The effect of macroscopic factors on cellular doses illustrating the source variability can be demonstrated by the calculation of average absorbed doses to basal and secretory cell nuclei, i.e. variations in energy deposition are not considered (Figure 1.4). Three sources contribute to the variability of average cellular doses: (1) the variability of the ^{218}Po and ^{214}Po surface activities, (2) the variability of target cell depths, and (3) the variability of source-target distances, affecting the range of alpha particles and the related stopping power (Bragg curve). While these macroscopic factor lead to significant variations of absorbed doses in individual cell nuclei, sometimes referred to as microdosimetry, this approach is still based on the macroscopic dose concept as it does not consider the variability of energy deposition within cell nuclei. Since the simulation of the effect of macroscopic factors on average absorbed doses to basal and secretory cell nuclei requires the application of stochastic modelling techniques, in this case Monte Carlo methods, the most appropriate terminology might be stochastic macroscopic dosimetry.

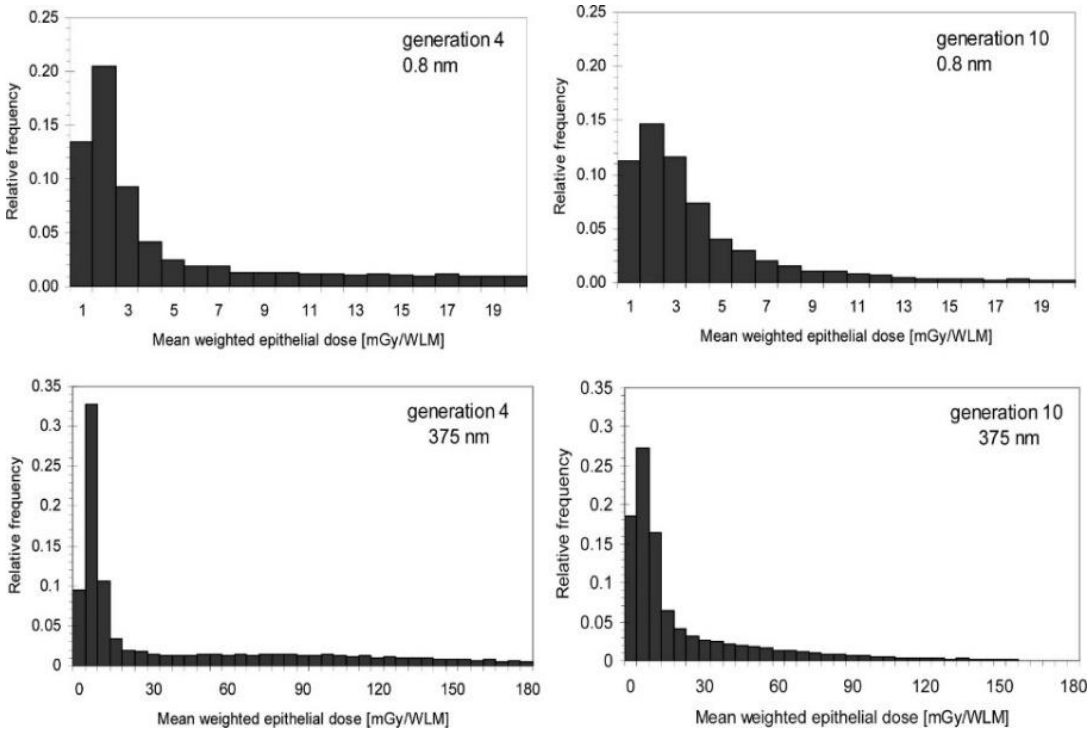

Figure 1.4 Probability distributions of basal and secretory cell doses, produced by ^{218}Po and ^{214}Po alpha particles, for 0.8 nm (unattached fraction) and 375 nm (attached fraction) radon progeny aerosols in bronchial airway generation 4 and bronchiolar airway generation 10 for defined exposure conditions.

1.3.3 Microscopic factors – target variability

Microdosimetry, initially developed for external radiations (Rossi, 1968), describes the variability of energy deposition in microscopic targets, thus illustrating the target variability. While fluctuations of energy deposition in macroscopic targets. e.g. organs or tissues, can be neglected, and thus are properly represented by average absorbed doses, they become increasingly important for microscopic targets, such a cell nuclei or DNA components, where they can vary over a wide range of values. For high LET radiations, such as alpha particles, these random fluctuations of energy deposition are smaller than for low LET radiations, such as gamma rays.

Microscopic factors causing fluctuations of energy deposition in individual cell nuclei are: (1) the hit probability, i.e. the probability of hitting a cell nucleus and the related frequency of single and multiple hits (Poisson distribution), and, in the case of a hit, (2) the random track lengths of alpha particles in spherical target cell nuclei (crossers) or incomplete traversal (stoppers), (3) the random energy deposition along the track of an alpha particle as the result of the range distribution of alpha particles from source to target and the corresponding LET distribution of the intersecting tracks (Bragg curve), and (4) energy straggling (Vavilov distribution) of traversing alpha particles and the contribution from grazing alpha particles ejecting electrons into the target volume (this effect is small compared to that of intersecting tracks and thus may be neglected for microdosimetric calculations.

In classical microdosimetry, energy deposition ε in microscopic targets is represented by the specific energy $z = \varepsilon/m$, the stochastic equivalent of absorbed dose D (ICRU, 1983). The distribution of specific

energies can be described by the probability density f(z), which contains a δ-function at z = 0 for the probability of no energy deposition. For alpha particle irradiation, energy deposition can be expressed either by the specific energy for single hits, $f_1(z)$, or by the specific energy distribution for single and multiple hits at a given absorbed dose D, f(z;D). For comparison, the mean specific energy is a non-stochastic quantity, conceptually equivalent to the average absorbed dose D in micrometer-sized target volumes, commonly used in cellular stochastic macroscopic dosimetry.

The stochastic equivalent of LET in microdosimetry is the lineal energy $y = \varepsilon/L$ (ICRU, 1983), where L is the average chord length of intersecting tracks (for spherical targets with radius R, L = 4/3 R). The distribution of lineal energies can be described by the probability density f(y) and the mean lineal energy is a non-stochastic quantity conceptually equivalent to LET. Note that linear energy is defined only for single energy deposition events.

In the case of alpha particles, f(z) can be approximated by the track length distribution for a given LET and f(y) by the LET distribution (Hofmann et al., 2000).

Initially formulated for external irradiation, Roesch (1977) extended the fundamental concepts of microdosimetry to internally-deposited alpha-emitting radionuclide sources: The alpha activity is represented by a distribution of point sources, where $f_1(r;z)$ is the single event density for a point source as a function of distance r from the target. If the target receives exactly n single energy deposition events from several point sources, $f_n(z)$ can be calculated as the n-fold convolution of $f_1(r;z)$ using the Fourier transform technique. Since the probability of exactly n energy deposition events at dose D follows the Poisson distribution, f(z;D) is finally given by the compound Poisson process.

In the case of radon progeny alpha particles emitted from cylindrical bronchial airway surfaces, Aubineau-Laniece et al. (2002) and Fakir et al. (2005) developed a Monte Carlo code for the calculation of specific energy spectra in bronchial target cells based on the analytic RADONA code of Caswell et al. (1994). Compared to the analytic approach of Roesch (1977), single event distributions are calculated by Monte Carlo methods, while the convolution of the single event densities is based again on the analytic Fourier convolution method.

In the case of high LET radiations, such as alpha particles, two microdosimetric parameters have been proposed to describe the variability of energy deposition in microscopic targets (cell nuclei): (i) the hit probability and the frequency of cellular hits following a Poisson distribution, and, (ii) specific energy distributions f(z) for single hits, $f_1(z)$, or for multiple hits at a given absorbed dose D, f(z;D). The hit probability concept considers the variability of cellular hits, while energy deposition is expressed by the mean specific energy, i.e. the variability of energy deposition within cell nuclei is not considered. For comparison, the specific energy distribution concept includes both the variability of cellular hits and the variability of energy deposition in cell nuclei.

The microdosimetric approach is especially relevant for low level radon exposures, where low doses of alpha particles are characterised by a small number of cells affected, i.e. small hit probabilities. However, the cells hit may receive relatively high doses, while the majority of cells are not hit at all. For example, in the low dose region, defined by the action of single cellular hits, an increase in tissue dose increases only the number of cells hit but not the average dose received by individual cells. With the onset of multiple hits, when most cells have already received at least one hit, average cellular doses rise in proportion to the tissue dose. Thus, in the case of alpha particles, the low dose region is characterised by a low number of cells hit, but not by low cellular doses.

At low radon exposures, specific energy distributions in basal and secretory cell nuclei result from single energy deposition events (Hui et al., 1990). While the shapes of the single-event specific energy distributions for both target cells are practically identical, the slightly smaller height of the $f_1(z)$ distribution for the basal cells is caused by their deeper location in bronchial epithelium and hence smaller hit probabilities. With increasing tissue dose, distinct peaks of specific energy distributions produced by multiple hits start to appear, moving the f(z;D) distributions stepwise to higher specific energies with increasing number of hits, although decreasing in height in line with the Poisson distribution of hits (Sedlák, 1996).

In the case of local radon progeny deposition hots spots in bronchial airway bifurcations and resulting high local tissue doses, cells located directly at the carinal ridge receive multiple hits even at low exposure levels (Fakir et al., 2005). This is illustrated in Figure 1.5 for three selected cellular sites, along the cylindrical airway (R1), at the transition zone of the bifurcation (R2) and directly at the carinal ridge (T). Although the specific energy distributions in R1 and R2 differ in the number of alpha particle hits, they are still produced by single hits, For comparison, the two specific energy distributions in T for two different depths in tissue differ also in the number of hits received, but here the differences in multiple hits lead to distinctly different distributions along the z-axis, The higher the specific energy peak, the higher is the probability of alpha particle hits.

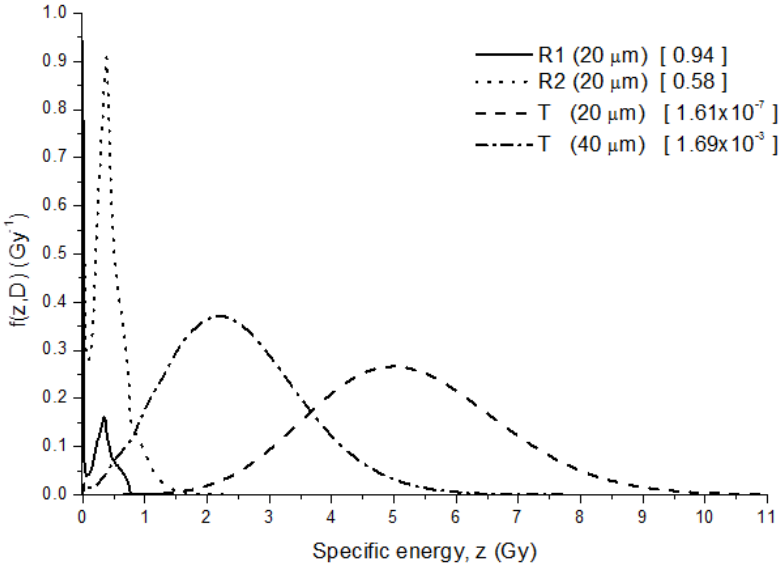

Figure 1.5 Dose-dependent specific energy spectra f(z;D) in three different target locations R1, R2 and T of a bronchial airway bifurcation for non-uniform radon progeny surface activities, normalised to a cumulative exposure of 20 WLM (residential radon exposures). The number in parenthesis are the probabilities of zero events, indicating the fraction of cells not hit at all.

1.3.4 Relationship between microdosimetry and radiobiological effects

Since the capabilities to calculate hit probabilities and specific energy distributions are currently available, the question arises how to relate these microdosimetric parameters to experimentally observable cellular biological effects relevant for lung cancer induction. At present, no unique

relationship between hit probabilities and specific energy distributions and cellular radiobiological effects has been established. However, several approaches have been proposed to establish such an approximate relationship for practical purposes. These models comprise (i) hit-related models, (ii) effect-specific track length models, (iii) effect-related interpretation of specific energy distributions, and, finally, (iv) track structure models (nanodosimetry).

The basic hypothesis of hit-related models is that biological effects are related to the fraction of cells hit by alpha particles and, in the case of a hit, to the number of alpha particle hits (Truta-Popa et al., 2011). At low radon levels, only a very small number of cells will be affected by single hits, thus leading to a linear dose response.

In effect-specific track length models, the random intersection of cell nuclei and the multiplicity of cellular traversal are related to effect-specific probabilities per unit track length (PPUTL) as functions of LET (Hofmann et al., 2000). Since the PPUTLs are derived from experimental in vitro data, they are specific to a given radiobiological effect, such as cell killing or oncogenic transformation. In the case of alpha particles, $f_1(z)$ is approximated by the track length distribution and f(y) by the LET distribution,

Examples of effect-related interpretations of specific energy distributions are the dual radiation action model (classical microdosimetry), effect-specific threshold or boundary energy models, and the hit-size-effectiveness function.

The theory of dual radiation action is based on the assumption that the biological response to an irradiated cell results from the interaction of two sublesions in a sensitive volume, leading to a linear-quadratic dose-effect relationship (Kellerer and Rossi, 1972). At low doses of alpha particles, the two sublesions can be produced by a single alpha particle, thus exhibiting a linear response.

The concept of an effect-specific threshold or boundary energy is based on the assumption that specific energies below and above a defined threshold energy z_o can be related to specific radiobiological effects, such as cell killing (above z_o) or oncogenic transformation and carcinogenesis (below z_o) (Sedlák, 1996).

The hit-size-effectiveness function concept provides a formal relationship between microdosimetry and radiobiology without any assumptions on cellular mechanisms by combining the hit concept with the specific energy concept (Bond et al., 1985). The basic assumption of this concept is that both energy deposition in subcellular sites and the subsequent cellular response are random processes. The hit-size-effectiveness function E(z) for different biological endpoints derived from measurements with various types of radiation, with LETs ranging from 1 to 350 keV μm^{-1}, is plotted in Figure 1.6 (Sondhaus et al., 1990). Values of E(z) represent the probability of a quantal response to a hit of size z. The similarity of the hit-size-effectiveness functions for a wide range of radiobiological effects and types of radiation suggest a fundamental response of a cell to ionising radiation.

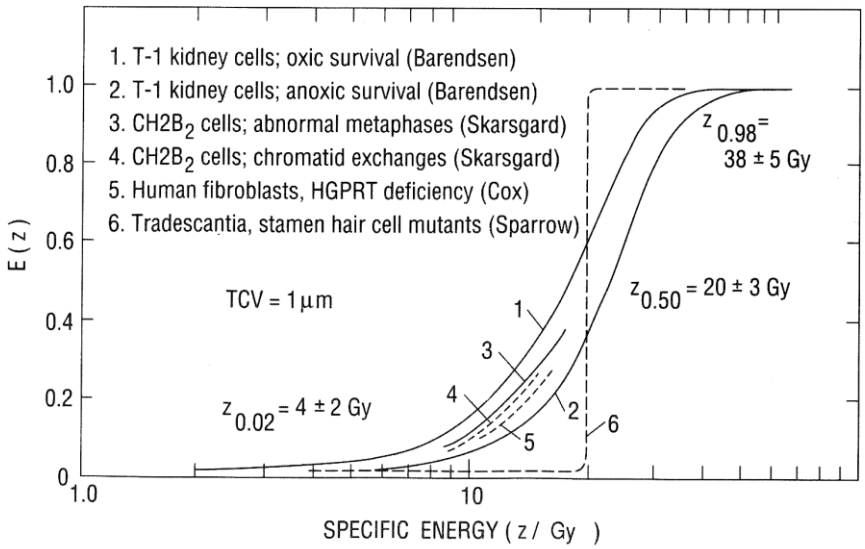

Figure 1.6 Hit-size-effectiveness function E(z) for different biological endpoints and types of radiation.

In the track structure models, subcellular radiobiological effects are related to the structure of charged particle tracks, either to the radial dose distribution around a charged particle track as in the amorphous track structure model (Katz, 1987) or to detailed track structure simulations at the DNA level, i.e. in nanometer sites (nanodosimetry) (Friedland and Kundrát, 2013).

1.3.5 Summary and conclusions

The variability of energy deposition at the cellular level results from random variations of stochastic macroscopic factors (source variability) and microscopic factors (target variability). Internal microdosimetry comprises both source and target variability.

The source variability can be described by distributions of average cellular doses (mean specific energy) in individual cell nuclei (stochastic macrosocopic dosimetry). The target variability can be characterised by hit probabilities, which consider the variability of cellular hits but not the variability of energy deposition within cell nuclei, and specific energy distributions which include both the variability of cellular hits and the variability of energy deposition within cell nuclei (microdosimetry), The microdosimetric approach is especially relevant for low radon level exposures, where low doses of alpha particles are characterised by a small number of cells affected while the majority of cells is not hit at all.

At present cellular radiobiological effects cannot directly be predicted on the basis of computed specific energy distributions. However, several substitute microdosimetric approaches have been proposed to establish such a relationship, such as hit-related concepts, effect-specific track length models, and effect-specific interpretations of specific energy distributions. Since microdosimetry refers to energy deposition at a given point in time, the application of microdosimetry to the prediction of lung cancer risk is limited by two factors: bystander or non-targeted effects and biological mechanisms modifying the initial response after irradiation.

Acknowledegements

The contributions of R. Winkler-Heil, L. Koblinger†, I. Balásházy, D.J. Crawford-Brown, H. Fakir, R.S. Caswell†, P. Pihet and I. Laniece-Aubineau are gratefully acknowledged.

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2. Standards and Regulations

2.1 Radon regulation in Spain: Overview (Marta García-Talavera, Nuclear Safety Council, CSN, Spain)

2.1.1 Introduction

While radon at the workplace has been regulated in Spain for several years, the transposition of Directive 2013/59/Euratom (BSSD) entails a much broader approach to reducing radon exposure for the population as a whole, including the development of a National Radon Action Plan. This directive has also propelled the completion of two long-term projects: the Radon Potential Map of Spain, which identifies radon priority areas; and the new Basic Document on radon protection included in the Technical Building Code, which sets the standards that buildings must meet in Spain. In addition, guidance on the assessment of radon exposure and doses at the workplace (CSN, 2012) is under revision in order to adopt recommendation from ICRP Publication 137, Part 3 and to account for recent developments in measurement instrumentation. These regulatory developments, together with the gaps to be yet addressed, are outlined below.

2.1.2 Radon Regulation at the Workplace

Since 2001, radon exposure at the workplace has been broadly regulated in Spain under Title VII of the 1996 Basic Safety Standards directive. In 2011, CSN Instruction IS-33 developed the title in greater detail, thus fostering regulatory compliance (CSN, 2011). This Instruction specifies:

- the types of workplaces where employers are obliged to take radon measurements (i.e. underground workplaces and those containing a groundwater source);
- the reference level above which a radiation protection programme must be applied; and
- an obligation to retest for radon every five years.

The transposition of the new BSSD entails several regulatory changes that will be enacted by a new regulation, currently undergoing legislative process: reduction of the reference level from 600 Bq m^{-3} to 300 Bq m^{-3}; and a dose-based criterion, namely 6 mSv y^{-1}, above which workers' exposure needs to be managed as a planned exposure situation. The number of workplaces under regulation will also increase considerably, not only because of the lower reference level but also because all workplaces at ground floor within radon priority areas will be required to take radon measurements. Application of the optimisation principle referred to in article 54.3 of the BSSD is one of the most challenging regulatory requirements to put into practice. Lack of clear guidance and qualified experts leaves employers with legal uncertainty regarding to what level radon concentrations or exposures need to be reduced in order to attain optimised values. Consequently, regulatory efforts should be made in this area in the short term.

CSN (2012) specifies the methodology to assess radon concentrations or exposures. It requires dividing the workplace into homogenous radon concentration zones (HZs), and then placing a given minimum number of detectors per HZ, depending on its area. Detectors must be exposed for at least three months during the heating season, except for underground workplaces, where exposures must cover either one full year or the entire open season. Short-term variations (day/night or workday/holiday effect) can be accounted for by measuring with a continuous radon monitor during at least five non-consecutive cycles. The emergence on the market of more affordable radon monitors calls for a revision of the latter guideline as well as for setting a requirement on the minimum accuracy and precision of the devices employed.

At workplaces where areas with radon concentration of over 300 Bq m^{-3} have been identified, worker doses will need to be assessed by applying the dose coefficients in ICRP Publication 137, Part 3. When doses are likely to exceed 6 mSv y^{-1}, Member States are given the flexibility to determine which of the relevant requirements for planned exposure situations are to be applied. In Spain, these will include those pertaining to:

i) information to workers on job-specific risk and protection measures;
ii) assignment of individual doses;
iii) dose limitation;
iv) dose record keeping and recording; and
v) signage of areas where doses over 6 mSv y^{-1} might be received.

In regard to service provision, accreditation by ISO 17025 will be required for radon measuring laboratories. Moreover, where radon concentrations exceed the reference level, consultation with a technical radiation protection unit (TRPU) will be mandatory, in order to assess worker doses and to get advice on the relevant radiation protection measures. TPRUs are entities authorised and inspected by CSN regarding work procedure, technical means and human expertise. A new authorisation scheme for TRPUs on radon was set by CSN in 2018. When applicable, the monitoring of radon personal doses needs to be carried out by a radon personal dosimetry service.

2.1.3 Radon Regulation in the Building code

First published in 2006, the Spanish Technical Building Code is a national-level regulation made up of a collection of Basic Documents (BD) that set out the rules and minimum requirements for new buildings and refurbishments. The new BD on protection against radon (which will enter into force later in 2019) requires different levels of preventive measures in new buildings, depending on the risk category assigned to the building site: '0' requires no specific protection; 'I' implies installing either a passive crawl space or soil depressurisation system or a radon barrier (membrane) between the soil and the indoors; 'II' involves a combination of both techniques, also providing for active depressurisation if the preventive measures should fail to achieve radon concentrations below the reference level.

The risk category of the building site is assigned based on municipality. Municipalities, in turn, are classified using the Radon Potential Map of Spain (CSN, 2017). 'Radon potential' is defined for each geologic cartographic unit as 'the 90th percentile of radon concentrations in dwellings (referred to ground floor)', according to the methodology defined in García-Talavera et al. (2013). To classify municipalities, the following steps were completed using a custom ArcGIS Pro geoprocessing tool (García-Talavera and López, 2019):

i) In the Radon Potential Map of Spain (see Figure 2.1), units with P90 > 300 Bq m^{-3} (coloured in orange and pink) and P90 ϵ (200, 300) Bq m^{-3} (coloured in light brown) were selected.
ii) For each municipality overlaying those radon potential units (RPUs), the building area was obtained from Cartociudad©, a collaborative project led and coordinated by the National Geographic Institute of Spain.
iii) The overlap between the municipality building area (A_i) and the RPUs of interest was calculated as a percentage of A_i.

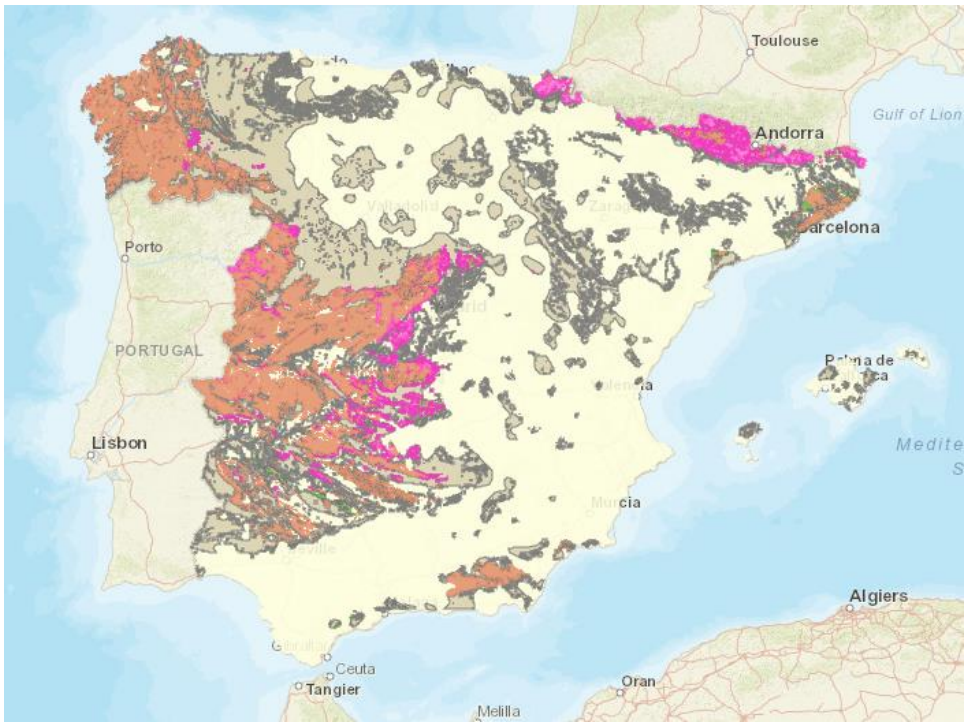

Figure 2.1 Radon Potential Map of Spain. P90 > 300 Bq m^{-3} (coloured in orange and pink) and P90 ϵ (200, 300) Bq m^{-3} (coloured in light brown)

The criteria for classification are given in Table 2.1 and the resulting map is shown in Figure 2.2.

Table 2.1 Criteria for municipalities classification

	Building area overlap with RPU
Class II municipalities	> 5% with > 300 Bq m^{-3} RPU
Class I municipalities	up to 5% with > 300 Bq m^{-3} RPU; or > 5% with 200-300 Bq m^{-3} RPU

2.1.4 National Radon Action Plan

Under the BSSD, Member States are required to establish national action plans addressing long-term risks from radon exposures in dwellings, buildings with public access and workplaces (Article 103). The first National Radon Action Plan in Spain will cover the period 2020–2024 and will be renewed as five-year programmes. It will represent the collaborative effort among five national organisations, autonomous regions and local entities to eliminate avoidable radon-induced lung cancers. The Plan will be structured in five key thematic areas:

- Core knowledge and infrastructure
- Building
- Occupational exposure
- Radon priority areas (instruments supporting the realisation of local action)
- Public communication

Figure 2.2 Map of Radon Zones by Municipality (established in the Technical Building Code). Class II municipalities coloured in brown.

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3. Metrology of radon and radon progeny

3.1 Uncertainties and Traceability in radon measurements (Annette Röttger, Physikalisch-Technische Bundesanstalt (PTB), Germany)

3.1.1 Introduction

The formal definition of the term "uncertainty of measurement" is given by "Evaluation of measurement data – Guide to the expression of uncertainty in measurement" (GUM or JCGM 100: 2008) and in the International Vocabulary of Metrology (VIM) as follows: "Uncertainty (of measurement): a parameter, associated with the result of a measurement, that characterises the dispersion of the values that could reasonably be attributed to the measurand."

These documents were issued and regularly updated in succession to the "Arrangement on the mutual recognition of the equivalence of national standards and of calibration certificates issued by national metrology institutes (MRA)" in 1999. Since then a comparison has had to deal with a single quantity, the result of each participant has had to be given in the form of a value with an assigned uncertainty. Furthermore, the uncertainty budget for the calculation of the result has had to be included, as well as the information about the traceability of each input quantity to national or international standards.

This paper gives an overview of how uncertainties associated with calibration and field measurements can be dealt with. The question of how to calculate a dose with assigned uncertainties is addressed in an example.

3.1.2 Uncertainties in Metrology

Determination of uncertainties is one important key issue for scientific developments. It provides the possibility to compare and to benchmark results, it is fundamental to accomplish decisions and from the metrological point of view, it is the counter stone to develop our metrological infrastructure further.

Every quality system must include traceability and uncertainty evaluation. Therefore, the ISO standard "General requirements for the competence of testing and calibration laboratories" (ISO, 2017) is used as the fundament for the implementation of a quality system and this standard is supported by the Guide to the expression of uncertainty in measurement (JCGM, 2008a; b), which is due to its importance available for free downloadable at BIPM website.

Providing traceability is one central task of National Metrology Institutes (NMI). NMIs have to realise, to maintain and to disseminate the legal units in compliance with the International System of Units (SI). Thus, a NMI thus is at the top of the metrological hierarchy in its country. The calibration certificates issued by the NMI document a calibration traceable to national measurement standards.

Radon is a dominating part of the natural radiation exposure of humans. Therefore, measurements of the radon activity concentration are performed in houses and at workplaces worldwide. To assure the quality of the obtained data and to provide a database for epidemiologic studies, the establishment of traceability and uncertainty assessment must be done with uttermost priority.

An important point is, to clarify the aim of the measurement and corresponding to that have the following questions answered:

1. What is the measurand? E.g. exposure $P(C,t)$, activity concentration $C(^{222}Rn)$, $C(^{220}Rn)$, equilibrium factor F, …

2. What range of the measured value is to be expected? Do I have a traceable calibration in that range for the respective quantity?
3. Which range of uncertainty has to be achieved at the required range of measurement?

Taking these questions into the planning of a measurement campaign will increase the impact of the research work drastically.

3.1.3 Summary of the GUM concept

The GUM provides concepts and basic principles of how to deal with uncertainties in measurement. It provides procedures for the stages of uncertainty evaluation (formulation and calculation stage).

Summarising the most important ideas from the GUM (JCGM, 2008a), yields: The purpose of measurement is to provide information about a quantity of interest: a measurand. No measurement is exact. When a quantity is measured, the outcome depends on the measuring system, the measurement procedure and other effects. The result is an indication value. The dispersion of the indication values would relate to how well the measurement is made. Its average would provide an estimate of the true quantity value that generally would be more reliable than an individual indication value. The measuring system may provide indication values that are not dispersed about the true quantity value, but about some value offset from it. The difference between the offset value and the true quantity value is sometimes called the systematic error value. There are two types of measurement error quantity: systematic and random.

A systematic error (an estimate of which is known as a measurement bias) is associated with the fact that a measured quantity value contains an offset. A random error is associated with the fact that when a measurement is repeated it will generally provide a measured quantity value that is different from the previous value. It is random in that the next measured quantity value cannot be predicted exactly from previous such values. There are two types of measurement error quantity, systematic and random.

All input quantities and the measurand are a sample from a probability distribution. Taking measurements is obtaining random numbers from a distribution.

Note: The purpose of the type A and type B classification is to indicate the two different ways of evaluating uncertainty components and is for convenience of discussion only; the classification is not meant to indicate that there is any difference in the nature of components resulting from the two types of evaluation.

Both types of evaluation are based on probability distributions, and the uncertainty components resulting from either type are quantified by variances or standard deviations.

- Type A evaluation is calculated from series of repeated observations, (example: frequent reading of a device)
- Type B evaluation means using available knowledge, (example: calibration factor of a device)

In other words: type A standard uncertainty is obtained from a probability density function derived from an observed frequency distribution, while a type B standard uncertainty is obtained from an assumed probability density function based on the degree of belief that an event will occur.

Application of the GUM is not the only way to access the value of the assigned uncertainty of a measurement. But it is the way we internationally agreed to do it so far. The GUM is a toolbox, and there is ongoing development in it.

3.1.4 Traceability of measurements: The role of the national metrology institutes

On 20 May 2019, the new specifications for the SI units will come into force. The states of the Metre Convention have adopted a fundamental reform of the SI units. The kilogram, ampere, mole and Kelvin are redefined. In future, they will be defined by natural constants. The last artefact, the original kilogram is now obsolete. The kilogram, ampere, kelvin, and mole will then be defined by setting exact numerical values for the Planck constant, the elementary electric charge, the Boltzmann constant, and the Avogadro constant, respectively. The metre and candela are already defined by physical constants, subject to correction to their present definitions. Though these are in fact world shaking changes, we scarcely notice them because the new definitions aim to improve the SI without changing the size of any units, thus ensuring continuity with existing measurements. Nevertheless, the origin of traceability for each unit is shifted. An example of the traceability chain the radon activity concentration is given in Figure 3.1 in the new SI system with the concept of fundamental constants.

The so-called Calibration and Measurement Capabilities (CMCs) of an NMI can be found in the BIPM key comparison database, which is a public website containing all information relating to the International Committee for Weights and Measures (CIPM) Mutual Recognition Arrangement (MRA), an arrangement establishing the equivalence of measurements made by, and certificates issued by, all the participating institutes (signatory National Metrology Institutes and other designated institutes). The CIPM MRA is the framework through which National Metrology Institutes demonstrate the international equivalence of their measurement standards and the calibration and measurement certificates they issue. The outcomes of the Arrangement are the internationally recognised (peer-reviewed and approved) Calibration and Measurement Capabilities (CMCs) of the participating institutes. In an ideal case, before a field measurement is performed, the device intended to be used has been calibrated. In principle, a calibration after a field measurement is also possible, but the time between calibration and measurement should not be too long. In the case of radon the calibration has to be done in the region of activity concentration that is expected to occur. A calibration at 10 kBq m^{-3} is not suitable for a measurement at 100 Bq m^{-3}.

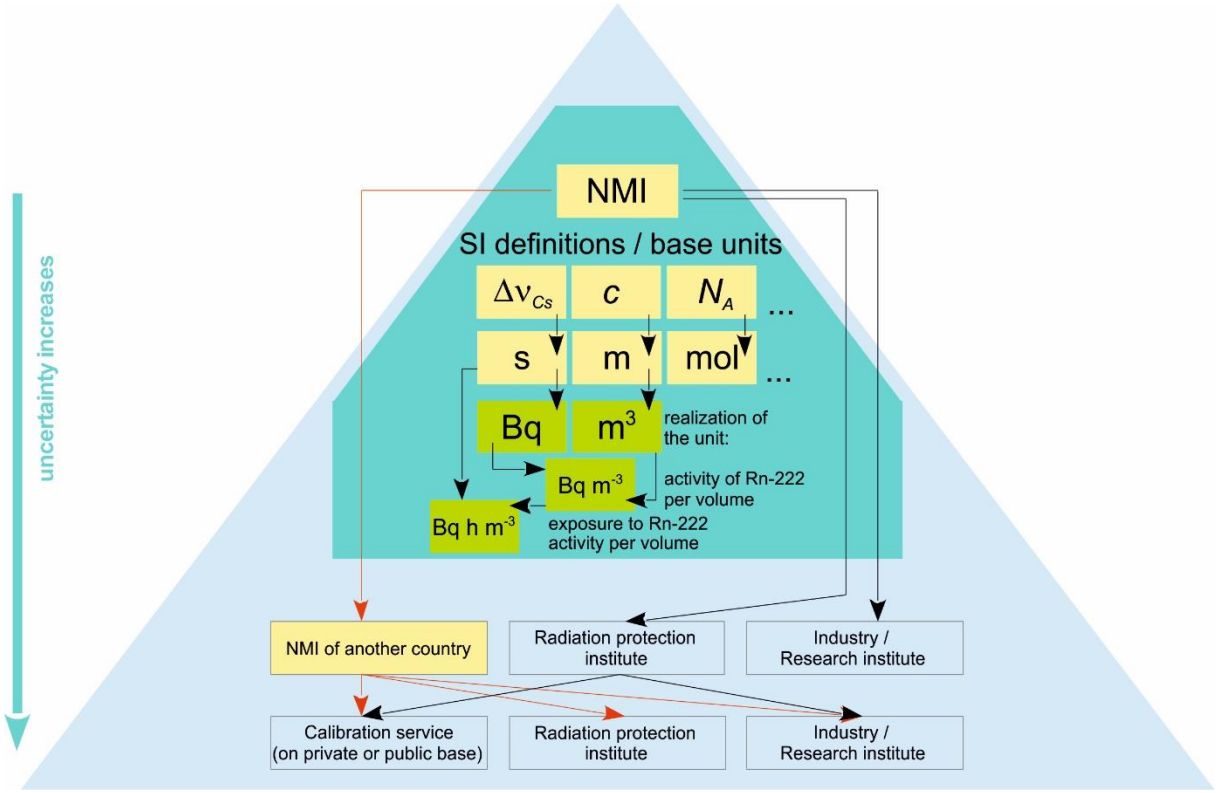

Figure 3.1 Traceability of Bq m^{-3} and Bq h m^{-3} in the concept of the new SI system. The basic units in yellow (seconds, metres and moles are defined by fundamental constants) are realised by an NMI. The green ones may be realised by an NMI but have to be at least traceable to an NMI. The length of the traceability chain will influence the total uncertainty.

Though this seems to be obvious, it is done rather often, partly lack of knowledge but sometimes even due to the idea that a calibration at higher radon levels gives a better statistical significance and thus a smaller uncertainty for the calibration factor. In most cases, this is not feasible, since this approach ignores the question of linearity, cross influence of ^{222}Rn, the respective background reading and the internal method used by the instrument that could change depending on the radon concentration level.

In practice, a scientist carrying out a specific radon measurement might not think about the calibration of the measuring device as fundamental as it really is. However, the devices on the market are not all reliable and their reading of the value is seldom correct within the uncertainty stated by the manufacturer. A certification produced by a manufacturer is only valid metrologically if the manufacturer is accredited for issuing such a certificate.

To realise the unit, a calibration has to be performed to establish traceability. The uncertainty of the calibration will be an intrinsic part of each later measurement in field. Therefore, the calibration should be chosen with the same care as the device itself. The following physical characteristics of the measuring device are required before starting a measurement campaign: traceability, uncertainty, detection limit and range of application.

Three typical procedures for the calibration of monitors for the concentration of ^{222}Rn are used worldwide:

1. a primary method based on a reference activity concentration realised by a primary radon gas standard and a calibration volume (both values are traceable to national standards),
2. a secondary method based on calibration via a reference monitor enclosed in the same atmosphere as the system under test, and
3. a primary / secondary calibration in a constant atmosphere based on a radium emanation source. This method is primary or secondary with respect to used components.

All facilities operating one or more of these methods have to be traceable according to the list given in the Calibration and Measurement Capabilities of the BIPM. All methods provide valid calibration factors. In case of methods 1 and 3, achievable relative combined uncertainties are 2 % to 4 % for k = 2. In case of method 2 the relative combined uncertainties can vary largely. Realistic ones are 4 % to 10 % for k = 2.

The obtained calibration factors differ in the assigned uncertainty but all of them involve the determination of background activity concentration. This can be a main source of uncertainty in lower level measurements. The ignorance of the background reading yields to measurement errors.

A simple example for the calibration of nuclear track detectors is presented in the following uncertainty budget. The calibration conditions are well known in terms of the exposure period, $\Delta t = (t_2 - t_1)$ and radon activity concentration $\boldsymbol{C}$. This calibration is performed in a radon reference chamber, starting at time t_2 and ending at t_1. The exposure is determined with a secondary standard (i.e. with reference instrument) that has been calibrated using a primary standard.

The exposure is given by

$$\boldsymbol{P} = \boldsymbol{C} \cdot (\boldsymbol{t_2} - \boldsymbol{t_1}) \qquad \text{with} \qquad \boldsymbol{C} = \boldsymbol{k_r} \cdot (\boldsymbol{C_r} - \boldsymbol{C_{rbg}}) \tag{3.1}$$

Where $\boldsymbol{C_r}$ is the indicated activity concentration and $\boldsymbol{C_{rbg}}$ is the specific background value obtained with the reference instrument and $\boldsymbol{k_r}$, is the calibration factor for the instrument. The uncertainty budget for exposure P is given in Table 3.1.

Table 3.1 Uncertainty budget for the exposure P according to GUM based on Equation 3.1. The index gives the amount of influence of a single uncertainty to the combined uncertainty.

Quantity	Value	Standard uncertainty	Index
C	30.7 kBq m^{-3}	0.5 kBq m^{-3}	
k_r	1.031	0.014	89.7 %
C_r	29.8 kBq m^{-3}	0.13 Bq m^{-3}	9.9 %
C_{rbg}	59 Bq m^{-3}	3 Bq m^{-3}	0.0 %
t_2	49.00 h	0.04 h	0.2 %
t_1	0.0 h	0.04 h	0.2 %
P	1,500 kBq h m^{-3}	22 kBq h m^{-3}	

During this exposure a number of m nuclear track detectors are exposed. The track density n that is obtained will have a variation. This variation can be used to assign an uncertainty to the track density $u(\bar{n})$ itself (this is a simplified example, for a special batch of etch track detectors only).

If a group of *m* nuclear detectors is exposed to different radon levels, a linearisation can be performed and if the linear model passes the consistency check, the calibration coefficient, κ can be calculated as follows:

$$\kappa = \frac{P}{(\bar{n} - \bar{n}_{bg})} . \quad (3.2)$$

The calculated value of κ and its associated uncertainty is given in Table 3.2 for this example. Further details can be found in (ICRU, 2012; Röttger et al., 2016).

Table 3.2 Example for the analysis of an exposure of the nuclear track detectors at 1,500 kBq h m^{-3} and 12 detectors without exposure (background detectors) to obtain a calibration coefficient κ according to Equation 3.2.

Quantity	Value	Standard uncertainty	Index
P	1,500 kBq h m^{-3}	22 kBq h m^{-3}	3.2 %
$\bar{n}$	2,030 cm^{-2}	160 cm^{-2}	96.5 %
$\bar{n}_{bg}$	69 cm^{-2}	6 cm^{-2}	0.3 %
κ	0.77 kBq h m^{-3} cm^2	0.06 kBq h m^{-3} cm^2	

3.1.5 Application: Radon measurements at homes and workplaces

A calibrated device is required to make indoor measurements at homes and work places, from the metrology point of view this is seen as a field measurement. This device has assigned uncertainties from its use in field measurement and from its former calibration (realisation of the unit), which is a measurement as well. In other words, the measured average radon activity concentration over the detector exposure period has a combined uncertainty according to GUM covering both, the calibration measurement and the field measurement. Obviously, it is not possible to measure with a smaller uncertainty in field than the uncertainty of the calibration. It is important to keep in mind that the background track density is determined by the transit detectors in calibration and in application.

Following the example given in section 3.1.4, the uncertainty of a single number of tracks is given by $u(n_i) = \sqrt{m} \cdot u(\bar{n})$. An unknown exposure to a radon activity concentration (mean value $\bar{C}$ determined on $\bar{n}$ nuclear tracks) can be obtained:

$$P = (\bar{n} - \bar{n}_{bg}) \cdot \kappa \quad (3.3)$$

$$\bar{C} = \frac{(\bar{n} - \bar{n}_{bg}) \cdot \kappa}{\Delta t} = \frac{P}{\Delta t} \quad (3.4)$$

Table 3.3 Example for the analysis of nuclear track detectors to obtain an exposure according to Equations 3.3 and 3.4.

Quantity	Value	Standard uncertainty	Index
κ	0.77 kBq h m^{-3} cm^2	0.06 kBq h m^{-3} cm^2	56.8 %
$\bar{n}$	3,290 cm^{-2}	220 cm^{-2}	43.2 %
$\bar{n}_{bg}$	50 cm^{-2}	5 cm^{-2}	0.0 %
P	2,500 kBq h m^{-3}	260 kBq h m^{-3}	
Δt	2,000 h	14 h	0.4 %
$\bar{C}$	1.25 kBq m^{-3}	0.13 kBq m^{-3}	

The final determined exposure in this field measurement is expressed in the formula $P = (2.5 \pm 0.6) \cdot 10^3$ kBq h m^{-3} (with a coverage factor k = 2) and the mean value for the radon activity concentration during the time of measurement (2,000 h) is given by $\bar{C}$ = (1.25 ± 0.26) kBq m^{-3} (with a coverage factor k = 2).

However, this method is only valid strictly if the conditions and constraints set up by all equations are valid in the field measurement as well. A good example for that is the presence of ^{222}Rn. Its activity concentration for the given example is assumed to be negligible in the calibration procedure and in the field measurement, or the nuclear track detectors must be free of this influence parameter as e.g. temperature, humidity, pressure, ambient dose rate, as recommended in ICRU (2012) and ISO (2012).

The decision threshold and detection limit can be calculated according to ISO 11929 (ISO, 2010a). The decision threshold and detection limit are defined by the respective uncertainties for special exposure conditions. With the probability of the error of first and second order typically (a = b = 5 %) the results can be expressed by

$$\text{decision threshold:} \quad \bar{C}^* = k_{1-\alpha} \cdot \tilde{u}(\tilde{\bar{C}} = 0) \tag{3.5}$$

$$\text{detection limit:} \quad \bar{C}^\# = \bar{C}^* + k_{1-\beta} \cdot \tilde{u}(\tilde{\bar{C}} = \bar{C}^\#) \tag{3.6}$$

In the given example $\bar{C}^*$ = 6 Bq m^{-3} and $\bar{C}^\#$ =15 Bq m^{-3} is determined. Detailed and simple examples for the determination of the uncertainties for different detector types, their decision threshold and detection limit can be found in ISO (2010b; 2012). Please note: The measurement of the ^{222}Rn exposure will in most cases be influenced by the presence of ^{222}Rn. This is not an uncertainty to be considered but an error to be corrected – but the correction has an associated uncertainty influencing the final result (IEC, 2015).

3.1.6 Dosimetry based on radon activity concentration measurements

The effective dose can be calculated from the ^{222}Rn exposure measurement, from a ^{222}Rn activity concentration measurement, from a progeny exposure or progeny activity concentration measurement in a given time of exposure.

In the wording of the GUM, the measurand is the effective dose, while the input quantities can differ: ^{222}Rn exposure P from a mean ^{222}Rn activity concentration $\bar{C}$ with a measured or assumed equilibrium factor F, a progeny exposure P_{RnP}or equilibrium equivalent activity concentration C_{eq}:

$$H = P_{RnP} \cdot k_{ICRP} = C_p \cdot t \cdot k_{ICRP} \quad \text{with} \quad C_p = C_{eq} \cdot k_u = \bar{C} \cdot F \cdot k_u \tag{3.7}$$

The factor k_u is a unit conversion (^{222}Rn: 5.57(10) · 10^{-6} mJ Bq^{-1} and ^{220}Rn: 7.565 (8) · 10^{-5} mJ Bq^{-1}) due to the decay chain, while the factor k_{ICRP} is the respective ICRP dose conversion, see Table 3.4 and Table 3.5 with uncertainties assigned according to number of digits in the ICRP publications. Here we consider a measurement over the full exposure time of 2,000 h with a mean ^{222}Rn activity concentration $\bar{C}$.

Table 3.4 An example for an uncertainty budget with the "old" conversion from ICRP 65 (ICRP, 1993), with uncertainty assigned according to number of digits in the ICRP publications. The uncertainty of the equilibrium factor is based on assumption only.

Quantity	Value	Standard uncertainty	Index
$\bar{C}$	300 Bq m^{-3}	50 Bq m^{-3}	53.6 %
t	2,000 h	11.5 h	0.0 %
F	0.4	0.0577	40.2 %
C_{eq}	120 Bq m^{-3}	26.5 Bq m^{-3}	
C_p	668·10^{-6} mJ m^{-3}	147·10^{-6} mJ m^{-3}	
k_u	5.5682·10^{-6} mJ JBq^{-1}	1·10^{-9} mJ JBq^{-1}	0.0 %
P_{RnP}	1.34 mJ h m^{-3}	0.30 mJ h m^{-3}	
k_{ICRP}	1.43 mSv m^3 (mJ h)$^{-1}$	0.081 mSv m^3 (mJ h)$^{-1}$	6.2 %
H	1.911 mSv	0.435 mSv	

Table 3.5 With the new dose conversion from ICRP 137 (ICRP, 2017) with uncertainty assigned according to number of digits in the ICRP publications. All other input quantities are chosen from Table 3.4. The estimate of the effective dose here is higher, and the uncertainty seems to be more reasonable from the state of the art.

Quantity	Value	Standard uncertainty	Index
k_{ICRP}	3 mSv m^3 (mJ h)$^{-1}$	0.5 mSv m^3 (mJ h)$^{-1}$	16.0 %
H	4.0 mSv	1.0 mSv	

Dose assessment based on the mean measured ^{222}Rn activity concentration $\bar{C}$ with a measured or assumed equilibrium factor F, a measured progeny exposure P_{RnP} or a measured equilibrium equivalent activity concentration C_{eq} during the time of exposure t does result in quite different quality of result when it comes to the range of uncertainty. It is therefore important to define the aim of the measurement before a campaign as good as possible and choose devices, calibration and measurand according to that aim.

The results of the uncertainty analysis here are only valid in the restrictions of the cited ICRU and ICRP publications.

This paper was prepared in the scope of the EURADOS Winter School 2019. It is evident, that it can provide a start in uncertainty analysis only, but it does include knowledge of the metrology of radon

of more than 20 years. The author wants to thank therefore all people from PTB, ICRM, MetroRadon and EURADOS who took part in discussions on uncertainties and traceability.

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3.2 Quality of radon measurements - Radon and radon progeny chambers for calibration (Martin Dubslaff, Federal Office for Radiation Protection in Germany (BfS), Germany)

3.2.1 Introduction

As a consequence of the new European Council Directive 2013/59/EURATOM, Germany introduced a new Radiation Protection Act and a new Radiation Protection Ordinance wherein radon measurements play a considerable role to reduce the exposure of people to radon. Thereby, the quality assurance of radon measurements at workplaces has been assigned to the Federal Office for Radiation Protection (BfS). One essential requirement for reliable and accurate measurements are calibrated measurement systems with traceability to national standards and reasonable low uncertainties. This holds true especially for measurements of the radon-222 activity concentration close to the new reference value of 300 Bq m^{-3}. For dose determinations, also the Potential Alpha Energy Concentration (PAEC) of the short-lived radon-222 decay products may be measured and thus, systems for this measurand have to be calibrated. Quality management systems and intercomparisons between laboratories are necessary to ensure and prove the quality of measurements and calibrations.

In this report calibration methods and radon calibration chambers are presented. As one example the ISO 17025 accredited BfS Radon Calibration Laboratory in Berlin is presented in more detail, which offers radon calibration services, intercomparisons and proficiency testing.

3.2.2 Measurement and measurands

High quality of radon measurements can only be achieved if the measurement results are reliable and if the measurands are traceable to the national standard with reasonable uncertainties.

As shown in Table 3.6, common measurands for radon measurements are

- Radon-222 activity concentration [Bq m^{-3}]
- Radon-222 exposure [Bq h m^{-3}]

and for radon decay product measurements

- Potential alpha-energy concentration (PAEC) of the short-lived Radon-222 decay products [µJ m^{-3}]
- Equilibrium-equivalent (Radon-222 activity) concentration [Bq m^{-3}]

Table 3.6 Measurands of radon and radon decay product calibrations

	Radon-222 gas		Radon-222 decay products	
Measurand	C_{Rn}	P_{Rn}	*PAEC*	*EEC*
	^{222}Rn activity concentration in air	^{222}Rn exposure	Potential alpha-energy concentration of the short-lived radon-222 decay products	Equilibrium equivalent (radon-222 activity) concentration
Unit	Bq m^{-3}	Bq h m^{-3}	µJ m^{-3}	Bq m^{-3}
Description	^{222}Rn activity per volume	Mean ^{222}Rn activity concentration multiplied by exposure time	Sum over all alpha energies of ^{222}Rn decay products per volume that are potentially emitted during decays from ^{218}Po to ^{210}Pb (a definition can be found in ICRU Report 88 [5])	An assumed ^{222}Rn activity concentration, in equilibrium with its short-lived decay products, which would have the same *PAEC* as the actual existing mixture of nuclides
Examples of measuring techniques	Electronic measuring devices with, e.g., scintillation cells, ionisation chambers or alpha spectrometers	Exposimeters: Integrating measuring devices, e.g., solid-state nuclear track detectors (SSNTD) or electrets	Electronic measuring devices with grab sampling, deposition of decay products on filters and/or wire-screens and analysis by alpha spectrometry	

Using the total *PAEC* per Bq in equilibrium (see ICRP publication 65 [4]), *PAEC* and *EEC* can be converted via the following Equation 3.8:

$$PAEC = EEC \cdot \frac{5.56\ \text{nJ}}{\text{Bq}} \tag{3.8}$$

From IRCP publication 65 Equation 3.9 to calculate *EEC* from activity concentrations C_i of nuclide i can be derived:

$$EEC = 0.105 \cdot C_{Po218} + 0.516 \cdot C_{Pb214} + 0.379 \cdot C_{Bi214} + 5 \cdot 10^{-8} \cdot C_{Po218} \tag{3.9}$$

3.2.3 Calibration equipment

To find a relation for a correction of indication values of a measuring instrument a comparison between its indication values and a measurement standard is performed under specified conditions (compare definition 2.39 in JCGM, 2012). Independently from the actual radon measurand, an equipment with following three units is necessary to run radon calibrations: a radon emanating source, an exposure chamber and a measurement standard traceable to the national metrology institute.

3.2.3.1 Sources

Prevailing kind of sources in calibration laboratories is the flow-through source consisting of dry ^{226}Ra salt that can be purged with air. For a constant radon emanation and exhalation of the source, the humidity of the purging air has to be constantly low. To adjust the actually required activity and use only a fraction of the maximal transferred activity of the source, controlled flow dividers in bifurcation can be installed at the output of the source.

Another source often used to create a reference atmosphere is a ^{222}Rn gas standard (see 3.2.3.3).

3.2.3.2 Chambers

Small calibration containers like stainless steel barrels with a volume of some 100 l are cheap, need less space and ask small sources to create a considerable high radon activity concentration. For small number of items and long-term exposures it can be also beneficial to use small containers without blocking a whole walk-in chamber or to use many small containers in parallel with different activity concentrations each.

Big walk-in calibration chambers have practical advantages, for example, more and larger devices can be exposed. In walk-in chambers a monitoring and adjustment of environmental parameters, e.g., temperature, humidity or aerosol concentration can be installed. A bigger volume also causes less disturbance of radon activity concentration while inserting objects into the atmosphere and leads to a lower plate-out influence during ^{222}Rn decay product calibrations. However, to keep the atmosphere homogenous can be challenging especially for short-lived decay products and in case of ^{222}Rn calibrations so that only a part of the whole chamber might be used for the calibration.

In principal there are two possibilities to run a radon calibration chamber, in decay mode and in constant mode.

- In decay mode, a certain radon-222 activity is injected, e.g., a gas standard.The activity exponentially decreases during the calibration due to radioactive decay. The reference activity concentration can be calculated for each point in time if chamber volume and initial activity is known. Alternatively, the activity concentration can be measured by means of a reference device if the delay and sampling behaviour of the calibration object is considered properly.
- In constant mode, a continuous dosing is applied to compensate the radioactive decay and the activity concentration is measured by an instrument as measuring standard. To realise the constant mode, often flow-through sources are used. The constant mode is essentially needed for long term exposures, e.g., for Rn decay product calibrations (a longer time period is required to achieve constant calibration conditions oder so) or for exposures of exposimeters such as track detectors.

3.2.3.3 Measurement standards

A measurement standard can be, for instance, a measuring instrument that has a lower measurement uncertainty than the calibration object to be calibrated. Several metrology institutes also offer ^{222}Rn gas standards with a certified activity that can be used to create a ^{222}Rn reference atmosphere. A certified ^{222}Rn activity is flushed into a certified volume to create a known reference atmosphere.

The calibration hierarchy includes different levels with increasing uncertainty from the highest to the lowest level: national standards, reference standards and working standards (see 5.3, 5.6 and 5.7 of JCGM, 2012).

3.2.4 Calibration procedures at the BfS Radon calibration laboratory

In Europe, several laboratories offer calibration services related to radon-222. Some of them are National Metrology Institutes or Designated Institutes (NMI or DI) or are accredited according to ISO 17025 for the measurand ^{222}Rn activity concentration in air. Very few are accredited for the measurand ^{222}Rn PAEC or EEC.

In Germany, the BfS represents the recognised national reference laboratory and is responsible for quality assurance of radon measurements. A comprehensive program for quality assurance is performed comprising calibration services, intercomparisons and proficiency tests. The BfS Radon Calibration Laboratory is accredited according to ISO 17025 since 1999 for the measurands ^{222}Rn activity concentration and PAEC of short-lived ^{222}Rn decay products. Non-accredited exposures to ^{220}Rn (thoron) and mixed atmospheres are realised on a level of a factory calibration. The technical capabilities include two walk-in chambers and six stainless steel containers that can be operated in a constant or decay mode and a vacuum chamber for a realisation of a ^{222}Rn reference atmosphere.

One of the two chambers is dedicated to PAEC calibrations. In this chamber aerosol size distribution and particle concentration can be controlled. The following table 3.7 summarises the capabilities and the adjustable or monitored parameters at the BfS Radon Calibration Laboratory for each calibration container. The dots (•) indicate the availability to adjust the parameters given in the left column.

Table 3.7 Calibration chambers and capabilities of the BfS Radon Calibration Laboratory.

Parameter	Range	30 m³ *PAEC* chamber	11 m³ radon chamber	0.4 m³ containers	0.17 m³ vacuum chamber
Rn-222 activity concentration	50 ... 12,000 Bq m^{-3} (accredited range)	•	•	•	•
Air temperature	-2 ... 40 °C	•	•	monitoring only	monitoring only*
Relative humidity	10 ... 90 %	•	•	monitoring only	•
Air pressure	800 hPa ... ca. 1,000 hPa (atmospheric pressure)	monitoring only	monitoring only	monitoring only	•
Potential alpha-energy concentration of the short-lived Rn-222 decay products	0.32 … 640 µJ m^{-3} (accredited range)	•			
Aerosol particle concentration	200 ... 50,000 particles/cm^3	•			
Equilibrium factor	0.1 ... 0.9	•			
Unattached fraction	< 1 ... 60 %	•			

* temperature is adjustable in a secondary chamber which can be connected to the chamber

3.2.4.1 Blank indication determination

Every calibration of electronic radon measuring instruments generally is started with a blank measurement in a radon-free atmosphere. The calibration object is placed in a calibration container which is continuously purged with sufficiently aged dry gas (air) and a mean indication value C_0 of long-term measurement is determined. Finally, this blank indication is used to correct the indication values (see following chapter).

Blank indication determination is of growing importance since measurements at ^{222}Rn activity concentrations below the reference value of 300 Bq m^{-3} are requested (EURATOM, 2013).

3.2.4.2 Calibration of radon-222 activity concentration measuring devices

The standard procedure for a calibration of electronic measuring instruments comprises a determination of calibration factors at three different ^{222}Rn activity concentration levels.

In Figure 3.2, the ^{222}Rn activity concentrations of the standard and of the calibration object versus time is given. The calibration chamber is operated in stable mode with a continuous dosing by purging a ^{226}Ra flow-through source.

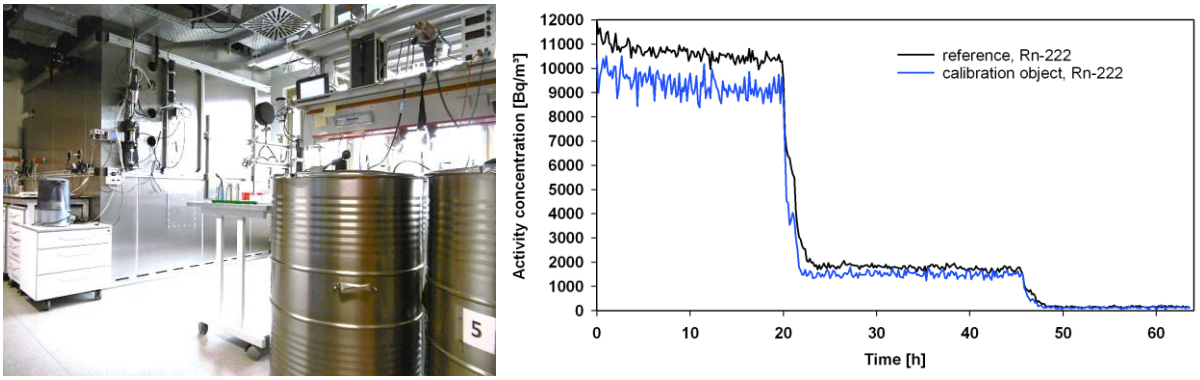

Figure 3.2 Left: BfS Radon calibration laboratory with calibration chamber and calibration containers. Right: Calibration measurements at three different ^{222}Rn activity concentration levels (11 kBq m^{-3}, 1.8 kBq m^{-3}, 0.2 kBq m^{-3}).

During the calibration, environmental conditions, e.g., temperature, relative humidity and atmospheric pressure are monitored.

Indication of the calibration object and value of the standard measuring device are compared after subtracting the blank indication. The selected time period for the comparison of both measurements depends on the sensitivity of the measuring instruments and activity concentration level during the calibration.

For each calibration level a calibration factor is calculated:

$$k_M = \frac{C_{Ref}}{C_M - C_0} \tag{3.10}$$

C_{Ref}: mean ^{222}Rn activity concentration within a selected time period

C_M: mean ^{222}Rn activity concentration measured by the calibration object within this time period

C_0: blank indication of the calibration object determined in radon-free air

The calibration factor k_M for the calibration object and its uncertainty $U(k_M)$ together with measurement results and the specified environmental conditions are provided in a calibration certificate.

The relative uncertainty of the calibration factor $U(k_M)$ is calculated from the uncertainty of the reference or working standard $U(C_{Ref})$ and the uncertainty of the calibration object $U(C_M)$ and the uncertainty of the blank indication $U(C_0)$ as follows:

$$\frac{U(k_M)}{k_M} = \sqrt{\frac{U^2(C_{Ref})}{C_{Ref}^2} + \frac{U^2(C_M) + U^2(C_0)}{(C_M - C_0)^2}} \tag{3.11}$$

The calibration and measurement capability, i.e,, the best $U(C_{Ref})/C_{Ref}$ with $k = 2$ at BfS is shown in Table 3.8 for different levels of radon concentration.

Table 3.8 Calibration and measurement capability (CMC) $U(C_{Ref})$ according to accreditation certificate D-K-15063-01-00 (BfS)

^{222}Rn activity concentration	CMC (best $U(C_{Ref})/C_{Ref}$ with k = 2)
50 ... < 80 Bq m^{-3}	17 %
80 ... < 300 Bq m^{-3}	12 %
300 ... < 1,000 Bq m^{-3}	8 %
1,000 ... < 12,000 Bq m^{-3}	5 %

In calibration certificates, always the expanded uncertainty with a coverage factor of k = 2 for a coverage probability ≈95 % is stated. Factor k has to be corrected according to GUM Table G.2 (JCGM, 2008) if the measurement comprises a small number of values only.

In Germany, the measurand ^{222}Rn activity concentration in air is realised and disseminated by BfS only. A ^{222}Rn reference activity is transferred into a reference volume of 166.2 dm^3 which is certified by national metrology institute PTB. The resulting ^{222}Rn activity concentration reference atmosphere is subsequently used to calibrate a measuring instrument that serves as measurement standard of the laboratory. During this decay-mode calibration, the temperature and the pressure are monitored. Afterwards, with this measurement standard the working measurement standards for routine calibration services are periodically calibrated.

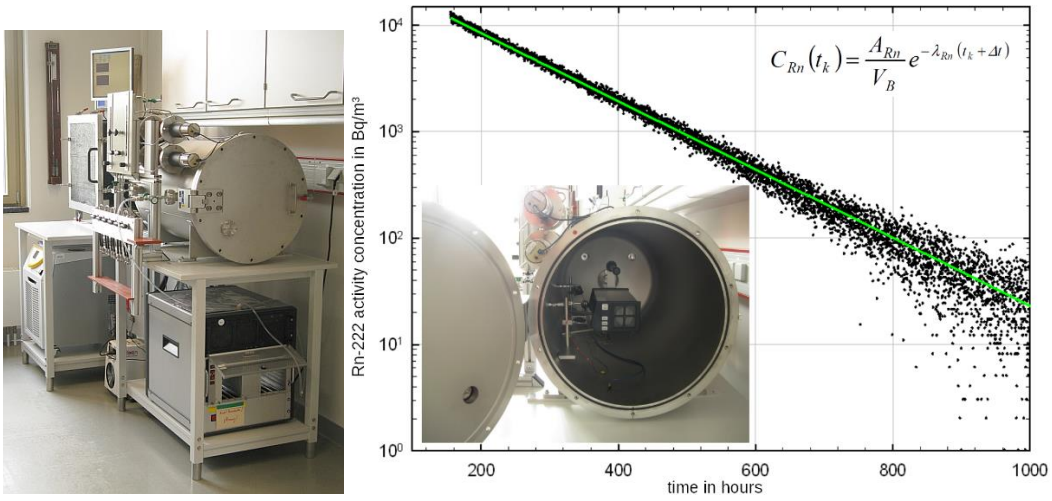

Figure 3.3 Vacuum chamber as volume standard certified by PTB and calibration measurement within the ^{222}Rn activity concentration reference atmosphere.

The tightness of the vacuum chamber can be double checked by residual radon measurement, pressure monitoring and additionally by comparison of the decay constant of ^{222}Rn λ_{Rn} with the effective decay constant from the exponential curve fit of measurement. The effective decay constant $\lambda_{eff} = \lambda_{Rn} + \lambda_{leak}$ should be equal to the known λ_{Rn} if leakage rate λ_{leak} is zero. For the measurement in Figure 3.3 a one-sided statistical test shows a probability of 99.998 % that λ_{leak} is not larger than 0.

3.2.4.3 Calibration of radon-222 *PAEC* measuring devices

At BfS, the typical PAEC calibration procedure includes two calibration points. One between 10 and 20 µJ m^{-3} with low unattached fraction (< 5 %) and one between 5 and 10 µJ m^{-3} and higher unattached fraction (between 10 and 30 %). Aerosols are created by a NaCl atomiser and the number and size distribution of aerosols can be measured using a condensation particle counter (CPC) and a scanning mobility particle size spectrometer (SMPS). Typically, the mean particle size amounts to 150 nm. The number of aerosols is adjusted according to the preferred unattached fraction by continuous dosing of aerosols. Fans with adjustable tilt and power ensure a sufficiently homogeneous atmosphere. In Figure 3.4, the PAEC calibration chamber at the BfS Radon calibration laboratory is shown.

The reference measurement is performed hourly by volume-controlled deposition of the decay products on a glass fiber filter of high efficiency (> 99.9 %) and on an overlying mesh wire screen for the determination of the unattached fraction (Reineking and Porstendörfer, 1990). At least five PAEC reference measurements are done accompanied by measurements of the environmental parameters (temperature, pressure, relative humidity, aerosol concentration, aerosol size distribution, air velocity).

Figure 3.4 PAEC calibration chamber at the BfS Radon calibration laboratory.

In order to determine the activity concentrations of the decay products, filter and mesh wire screen are measured in parallel using alpha spectrometers. With the activity concentrations of the radionuclides, EEC and PAEC can be calculated (compare 3.2.2) separately for the attached and the unattached fraction.

The volume flow rate for the sampling is traceable to the national standard and radiometric reference measuring system is calibrated via a combined or sequential alpha and gamma measurement (alpha from ^{214}Po and gamma from ^{214}Bi) with ^{214}Po and ^{214}Bi always in radioactive equilibrium due to the short half-life of ^{214}Po. The measurement is traceable via gamma measurement of a sealed ^{226}Ra planar source with an activity certified by the national metrology institute (PTB Germany).

The calibration and measurement capability, i.e,, the best $U(C_{Ref})\,/C_{Ref}$ with $k = 2$ at BfS is shown in Table 3.9 for different levels of PAEC of the short-lived ^{222}Rn decay products.

Table 3.9 Calibration and measurement capability (CMC) $U(C_{Ref})$ according to accreditation certificate D-K-15063-01-00 (BfS).

PAEC of the short-lived ^{222}Rn decay products	CMC (best $U(C_{Ref})/C_{Ref}$ with k=2)
0.32 ... < 6.4 µJ m^{-3}	10 %
6.4 ... < 64 µJ m^{-3}	6 %
64 ... < 640 µJ m^{-3}	6 %

3.2.5 Interlaboratory comparisons and proficiency testing

Intercomparisons between independent laboratories is essential to ensure the quality of radon-222 measurements and calibrations. As a consequence, different institutions offer international intercomparisons. Some examples of institutions that performed intercomparisons in the past few years are the Public Health England (PHE/Great Britain in 2016), the National Institute for Insurance against Accidents at Work (INAIL/Italy in 2018), Central Laboratory for Radiological Protection (CLOR/Poland in 2018) and the Federal Office for Radiation Protection (BfS/Germany in 2017, 2018 and 2019).

Based on experiences from more than a decade of doing intercomparison excersises, BfS gained a profound insight into the performance of passive radon detector systems. A proficiency testing has been designed to evaluate the individual results of participants. To pass this test is one requirement for institutions that want to provide exposimeters for radon measurements according to the new Radiation Protection Ordinance in Germany.

For the intercomparison and proficiency testing each participant sends in a set of exposimeters (currently 35 SSNTD or 24 electrets). The sets are then split into exposure groups (4 for SSNTD or 3 for electrets) and one group that will be not exposed to ^{222}Rn to determine the exposure and other influences during the transportation, e.g., due to storage in a post office during shipment at a certain background radiation or high temperature.

The exposure groups are exposed to different levels of ^{222}Rn activity concentration where at least 2 exposure values are close together. After exposure, the exposimeters are sent back to the participants for evaluation. The intercomparison is performed as blind test. Each participant reports its results to BfS before the reference exposure values are disclosed. Participants get a personal report showing their individual performance and a final report is published with anonymised results (see Foerster et al., 2019).

For the proficiency testing, a lower limit $UG = 0{,}7 - \frac{30}{P_{Ref}}$ and an upper limit $OG = 1{,}3 + \frac{30}{P_{Ref}}$ is defined for each different exposure value P_{Ref} which delivers a trumpet curve to allow higher deviation at lower exposure points. If the exposure value of an exposimeter is not within the limits $UG \leq \frac{P_{mess}}{P_{Rn,Ref}} \leq OG$ it is considered as outlier.

To pass the test an accepted number of outliers must not be exceeded which is derived from Beck et al. (2014) and shown in Table 3.10.

Table 3.10 Accepted outliers at BfS proficiency testing for exposimeters.

number of detectors in the test	accepted number of outliers
8	0
18	1
28	2
38	3

Example: If a participant sends 35 SSNTD that include 7 for transit background determination, 28 are exposed during the intercomparison and 2 outliers are allowed according to the table shown above.

Metrological institutions and calibration laboratories itself also compare each other. Laboratories accredited to ISO 17025 are moreover constrained to perform intercomparisons with other calibration laboratories or metrological institutions. As part of the EMPIR MetroRadon project, BfS conducts an interlaboratory comparison in 2018 and 2019 where an electronic radon measuring instrument is calibrated at different European institutions at three calibration points. The results will be published when all calibrations and data evaluation has been completed.

3.2.6 Outlook

To improve the measurement quality, BfS steps up efforts to promote the development and use of measurement instruments that comply with IEC 61577 series (IEC, 2006). The quality and traceability of calibrations at low activity concentrations will be improved. BfS will intensify its efforts in the field of thoron (radon-220) calibrations to support studies that investigate the presence of thoron and the influence of thoron and its progenies on radon measurements. In Germany measurements of radon gas concentration will be supported by BfS to identify workplaces exceeding the reference level of 300 Bq m^{-3}. More detailed investigation on PAEC, equilibrium factor and aerosol characteristics will spotlight the importance of radon progeny measurements especially for the dose assessment at work places.

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3.3 An Introduction to the MetroRadon Project – Metrology for Radon Monitoring (Hannah Wiedner, Physikalisch-Technischer Prüfdienst des Bundesamts für Eich- und Vermessungswesen (BEV), Austria)

3.3.1 Introduction

The ongoing MetroRADON project is a 3-year Joint Research Project in which 17 European metrology and research institutes aim to provide metrology for radon monitoring in the scope of the environmental Call of the EMPIR Programme. The European Metrology Programme for Innovation and Research (EMPIR) has been developed as an integrated part of Horizon 2020, the EU Framework Programme for Research and Innovation. It is supported from the European Commission using Article 185 of the European Treaty.

The purpose of MetroRadon is to develop reliable techniques and methodologies to enable SI traceable radon activity concentration measurements and calibrations at low radon concentrations and will help to establish a basic European metrological infrastructure for radon measurements, enabling sound monitoring of radon and radon protection in Europe. The need for this project is mostly motivated by the requirements of the implementation of the European Council Directive 2013/59/EURATOM (EU-BSS), aiming to reduce the risk of lung cancer for European citizens due to high radon concentrations in indoor air. Another goal of the project is 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 construction industry and scientific community.

The methods developed in the project will assist EU member states in the establishment of their national radon action plan, which is required under the EU-BSS. The novel development of a European unified index of geogenic Rn hazards, which can be defined flexibly independent of the data available, will provide a consistent picture of susceptibility to geogenic Rn across Europe. The definition of this Radon Hazard Index (RHI) will be an important tool for the harmonised implementation and performance of national radon action plans of EU member states according to the EU-BSS requirements.

Novel calibration methods and traceability validation at low radon activity concentrations will be devised, and new and stable radioactive reference sources developed to enable these calibrations and achieve sufficiently low uncertainties.

For the first time, the distortion of the radon measurement results due to the presence of thoron will be considered and corrected at low radon activity concentrations. Traceability to a primary thoron standard will be ensured and refined, enabling the thoron influence to be reliably investigated.

Guidelines and recommendations on the new calibration and measurement procedures will be published. Traceability of European radon calibration facilities using the new procedures and novel reference sources will be evaluated and the project partners will ensure that the results of this project will be taken up by end users, standards organisations, regulators and international bodies and associations.

3.3.2 Motivation

Radon is estimated to cause between 3 % and 14 % of all lung cancer cases depending on the average radon level in the country (WHO, 2009). For Europe, this corresponds to between 15,000 to 20,000 people per year dying of lung cancer caused by radon exposure. Accurate and reliable radon

measurement data are necessary in order to optimise counter measures to reduce the public's exposure to radon and related follow-up costs.

Radon has already been studied peripherally in a few other Joint Research Projects but never before as the main focus. Within ENV57 MetroERM (Metrology for radiological early warning networks in Europe, 2018) the focus was on environmental monitoring of man-made radionuclides (e.g. ^{137}Cs, $^{239/240}$Pu, etc.), and only the influence of radon and its progeny on these monitoring measurements was studied, not the measurement of radon itself (e.g. the influence of radon progeny concentrations on dose rate detectors (Task 1.6 MetroERM)). In the scope of IND57 MetroNORM (Metrology for processing materials with high natural radioactivity, 2018) only very specific questions regarding radon exposure of workers in waterworks and the emanation of radon from building materials were addressed (Task 3.3 MetroNORM). In MetroRADON (2018) the focus lies on the measurement of radon itself and all relevant exposure situations due to radon in dwellings and workplaces are considered.

The need for this project is mostly motivated by the requirements of the implementation of the European Council Directive 2013/59/EURATOM (EU-BSS, 2014), aiming to reduce the risk of lung cancer for European citizens due to high radon concentrations in indoor air. The directive lays down basic safety standards for protection against the dangers arising from exposure to ionising radiation, evoking new challenges for metrology, and radon measurements and calibrations in Europe. EU Member States are required to ensure that levels of relevant radon activity concentrations as laid down in the EU-BSS do not exceed 300 Bq m^{-3} and are obligated to transpose the EU-BSS into national legislation by 2018 for immediate implementation.

According to the EU-BSS, European Member States are obligated to identify and evaluate existing exposure situations and to determine the corresponding occupational and public exposures. With this in mind, national radon action plans addressing long-term risks from radon exposures in dwellings, buildings with public access and workplaces for any source of radon ingress (Article 100(1) and Article 103) have to be prepared. This requires reliable calibration and measurement methods for low radon activity concentrations between about 100 Bq m^{-3} and 300 Bq m^{-3}. A significant improvement in the metrological infrastructure in Europe in the field of radon calibrations at low activity concentrations is a prerequisite in order to be able to fulfil the requirements of the EU-BSS.

The EU-BSS will impact a range of stakeholders including those responsible for the transposition of the directive into national law and its implementation: from regulators and policy makers, professionals designing, performing, evaluating and interpreting radon surveys, radon instrument manufacturers to the construction industry.

3.3.3 Project objectives, current state of the art and progress beyond

MetroRADON is a 3-year Joint Research Project funded under the European Metrology Programme for Innovation and Research (EMPIR, 2018) in which 17 European institutions aim to provide metrology for radon monitoring. The project consortium consists of National Metrology Institutes, research institutes, bodies charged with the implementation of the EU-BSS into national law and universities. The following will highlight the aims and objectives of MetroRADON and give an overview over and how the project results will go beyond the current state of the art.

Figure 3.5 shows the main objectives of MetroRADON by work package.

The results and findings of MetroRADON will be published in peer-reviewed open access publications in scientific journals, as well as reports, guidelines and recommendations that will be available via the MetroRADON website www.metroradon.eu.

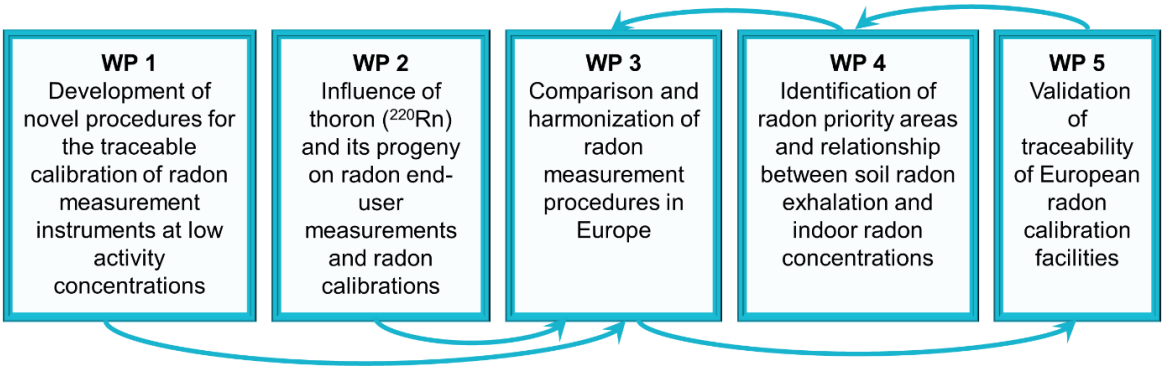

Figure 3.5 Graphical depiction of the aims of MetroRADON.

3.3.3.1 Traceable calibration of radon (^{222}Rn) measurement instruments at low activity concentrations and radioactive reference sources with stable and known radon emanation rates

Under the EU-BSS, EU Member States are required to ensure that levels of relevant radon activity concentrations do not exceed 300 Bq m^{-3}, hence effective implementation of the EU-BSS will require accurate and reliable measurement of low radon activity concentrations. Currently, traceable radon measurements are only conducted at activity concentrations > 500 Bq m^{-3}. According to the BIPM key comparison database of calibration and measurement capabilities (BIPM, 2018), there are currently only a few European facilities that offer radon activity calibrations, and all of them relate to very high radon activity concentrations (of the order of MBq m^{-3}), which are not relevant in the context of the EU-BSS. Although an intercomparison of calibrations for high radon activity concentrations has been conducted in the past, this was in the early 2000s. Two new two CCRI(II) comparisons of existing radon gas primary standards at different European National Metrology Institutes/Designated Laboratories (NMI/DI) for ^{222}Rn and ^{220}Rn in the range of a few kBq will be undertaken, thus providing up-to-date information on the existing radon gas primary standards for higher radon activity concentrations and closing a metrological gap in Europe by realising traceable radon reference atmospheres in the activity concentration range from 100 Bq m^{-3} to 300 Bq m^{-3}, while minimising the uncertainties.

The established metrological procedure (primary standard) is to use a decaying radon gas standard in a defined volume for calibrations of radon activity concentrations above 1 kBq m^{-3}. Using this method for low activity concentration calibrations is not practical, as it is time consuming and expensive due to the need for a radon gas standard for each calibration and specific, very leak-tight chambers. Better, more (long-term) stable and reliable sources and methods that are easier to use and could be used at more calibration facilities have been developed in order for traceable calibrations and measurements to be performed below 300 Bq m^{-3} with reasonable uncertainties. In the framework of the MetroRADON project new radioactive reference sources for ^{222}Rn (radon) and ^{220}Rn (thoron) with stable and known radon emanation rates for the realisation of reference fields for radon activity concentration in air have been produced using and testing various approaches. Emanation sources are less complex, cheaper and less sophisticated than conventional radon standards and will enable more laboratories to establish radon calibration capabilities. To maximise

the benefit for practical applications, these new sources are being used to develop new calibration methods and procedures for the realisation of reference atmospheres. These novel procedures will be used to calibrate radon measurement instruments traceable to primary standards in a range at low activity concentrations (100 Bq m^{-3} to 300 Bq m^{-3}) with relative uncertainties ≤ 5 % (k = 1). Existing established procedures for the calibration of active and passive radon monitors detailed in ICRU Report 88 (ICRU, 2012) will provide a solid base for a quality assured evaluation of the performance of the new procedures. Open access to these new procedures and guidelines will provide end-users with the possibility to calibrate radon measurement instruments at low activity concentrations with adequate measurement uncertainties necessary for the EU-BSS. Figure 3.6 shows radon/thoron chambers used in the project. These chambers are used to establish radon/thoron atmospheres and test the newly developed methods and sources.

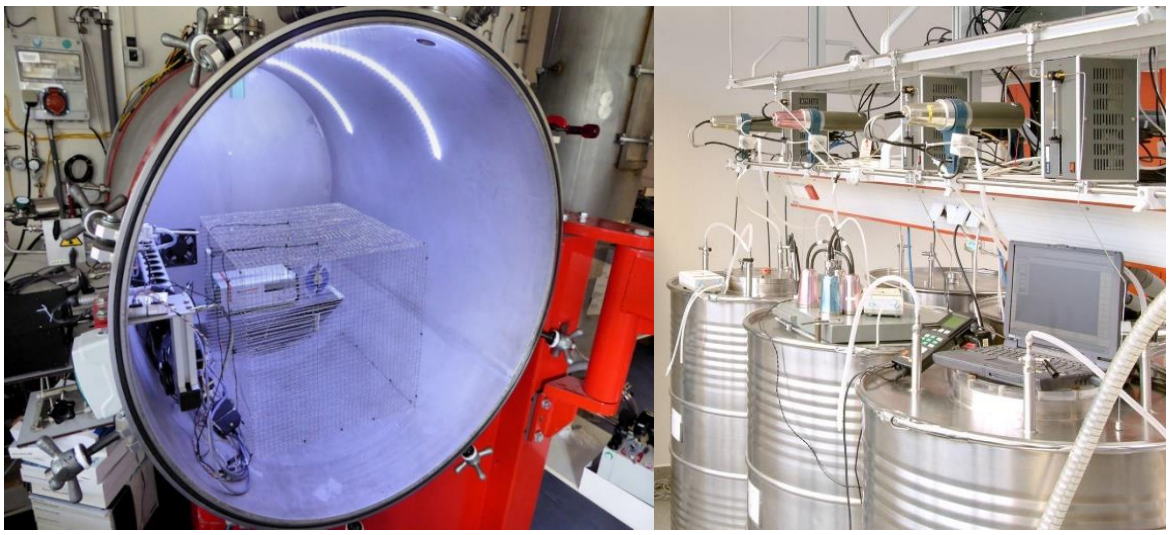

Figure 3.6 Left: inside view of the IRSN (France) radon/thoron chamber, right: stainless steel containers for the calibration of radon instruments at BfS (Germany). Source: IRSN/BfS.

3.3.3.2 Influence of thoron and its progeny on radon end-user measurements and radon calibrations

It has been observed that the presence of the radon-isotope thoron (^{220}Rn) and its progeny (^{212}Pb, ^{212}Bi + ^{212}Po/^{208}Tl) can have an influence on ^{222}Rn activity concentration measurements. This influence, if not properly corrected, can introduce bias in the radon risk estimates or can generate false alarms if these detectors are used to identify dwellings with radon concentrations that exceed reference/action levels. Both thoron and its progeny need to be taken into account, as the generated thoron progeny can remain within the detector volume long after the decay of the parent thoron atoms. Although several scientific studies on the influence of thoron on radon measurements are available in the literature, information is generally not harmonised and, therefore, not usable by end users and decision makers. Detailed knowledge of the influence of thoron on radon measurements is limited and techniques to correct for or to reduce thoron's influence on radon activity concentration measurements do not currently exist.

One aim of MetroRADON is to investigate and reduce the influence of thoron (^{220}Rn) and its progeny on radon (^{222}Rn) end-user measurements and radon calibrations. For this purpose, the sensitivity of radon monitors and detectors to thoron with traceability to a primary thoron system is currently being investigated and evaluated. Reference thoron atmospheres have been established and

evaluated and the secondary thoron reference instruments to be used in the experimental research have been calibrated using the primary thoron system at Institut de Radioprotection et de Sûreté Nucléaire (IRSN, France). Additionally, the sensitivity of radon (thoron) monitors and detectors to radon and thoron under mixed radon and thoron atmospheres and under temperatures in the interval typical for the real environment (e.g. -15 °C to +60 °C) will be investigated. Conclusions about the dependence of the signal on the specific environmental conditions (radon to thoron ratio, temperature, time variations of radon/thoron concentrations and temperature) will be drawn and the consequences for the design of radon surveys under real conditions (e.g. work places, soil gas, etc.) will be considered and analysed. Figure 3.7 shows the application of the techniques for soil gas measurements. Separately, technical concepts and solutions will be proposed to firstly correct the thoron-related bias to the radon signal in radon monitors and secondly to reduce the thoron-related bias to the radon signal in radon monitors through the use of membranes that act as a barrier to thoron. The properties of different filters, foils and membranes that might serve as efficient barriers for thoron, whilst not reducing radon permeability significantly, have been investigated in order to propose methods for reducing the influence of thoron on radon measurements. The findings of the influence on thoron on radon monitors and recommendations on the construction of radon monitors that are not sensitive to thoron will be made available to end-users in reports, guidelines and recommendations via the MetroRADON website.

Figure 3.7 Taking a sample for soil gas radon measurements.

3.3.3.3 Existing radon measurement procedures and approaches to optimise the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe

Measurements of radon concentrations have been conducted in Europe for years. While they have been subject to quality assurance by comparison exercises in the past (at least for relatively high radon activity concentrations), research needs to be undertaken in order to harmonise the different radon measurement techniques and calibrations. Radon surveys (both indoors and outdoors) and radon measurements are carried out differently in European countries, dependent on political decisions, the aim of the survey, availability of resources and infrastructure. Therefore, strong heterogeneities of radon data still exist in Europe. National or regional approaches chosen to identify

areas with observed or suspected high probability of radon concentrations in buildings above the reference level (EU-BSS Article 103) can vary. One strategy relies on directly measured indoor radon data, others on indirect concepts based on the geogenic radon potential (e.g. based on soil radon exhalation rates). There are also different methods used to define the geogenic radon potential of an area and these different methodologies and procedures may lead to inconsistencies, i.e. different values of the nominally same quantity. Intercomparisons on surface soil radon exhalation rates and radon concentrations in soil gas are rare and there are few laboratories whose results have been tested under in-situ conditions, and hence comparability of data cannot be not guaranteed. In order to ensure that radon data and decisions on the identification of radon priority areas (RPAs) are comparable regardless of the approach used, these methods need to be compared and standardised in order to provide comparable results with moderate uncertainties. This use of different strategies and inconsistencies may lead to communication problems between stakeholders and impair credibility, and as a further consequence reduce the efficiency of measures undertaken as part of Radon Action Plans (RAPs) as required by the EU-BSS. New standards, guidelines and protocols can help to steer political and technical decisions with respect to the design of new radon surveys and measurements in harmonised directions (via ISO standards, EC guidelines etc.). Since the EU-BSS are currently being transposed into national law the timing is opportune to provide assistance and to discuss on a scientifically high-level international platform how this is best done in a quality assured and harmonised way.

During MetroRADON existing indoor and geogenic radon survey data is being analysed and evaluated in order (i) to identify the rationale and methodologies used, (ii) to identify the extent and possible sources of inconsistencies in the results of indoor radon surveys and (iii) to propose approaches to reduce inconsistencies and improve harmonisation of indoor and geogenic radon data. It will be evaluated which data are useful for different purposes such as the evaluation of workplaces, preventive measures, estimation of population dose caused by radon, etc. The collection of radon survey methodologies and evaluation of their comparability will provide guidance for technical and political decisions in implementing the EU-BSS in Member States (especially in those who are at the beginning of their radon protection programme) as well as on a European level (e.g. European Atlas of Natural Radiation). One of the partners in the MetroRADON project, the European Commission Joint Research Centre in Ispra, Italy, is responsible for the creation of European radon maps (as part of the European Atlas of Natural Radiation). The data harmonisation, aimed at by this project, will provide the possibility to combine radon measurements at a European level and to develop a consistent European radon map.

Two intercomparisons (indoor and outdoor) have been performed under field conditions to provide a direct comparison between different methodologies and to identify physical reasons for possible inconsistencies, particularly related to sampling and measurement techniques. It is neither realistic nor reasonable that European harmonisation of radon data and surveys is carried out from scratch, nor that all countries are obliged to initiate new surveys or measurements or change their mapping strategies according to a guideline or a defined standard. By comparing existing radon measurement procedures in different European countries, the results will be used to optimise the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe. Information about indoor radon and geogenic radon surveys in Europe regarding strategy, methodologies and their potential for use as a basis for implementation for the EU-BSS will be provided to the relevant stakeholders, including results of the on-site inter-comparison exercise. Methodologies to harmonise indoor data (i.e. seasonal correction, short-term and long-term

measurements) will be published and will assist national radiation protection authorities to develop more appropriate approaches to radon protection to ensure public health.

3.3.3.4 Methodologies for the identification of radon priority areas, the development of the concept of a Radon Hazard Index (RHI), and the relationship between soil radon exhalation rates and indoor radon concentrations

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 (RPAs). 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 EU-BSS Article 54, as well as mandatory preventive measures or priority of awareness programmes. Given its possibly important economic, logistic and political consequences, RPA definition has become a highly politicised issue in some countries. One step in the quality-assured implementation of EU-BSS requirements should, therefore, be proper discussion and involvement of stakeholder interests, since this provides the necessary condition for a radon protection policy that satisfies the needs of society.

As the definition of RPAs in the EU-BSS allows a wide range of interpretation, different concepts and methodologies have been proposed and some already adopted. Currently there are many approaches used to define geogenic radon risk areas which usually form the basis for the definition of RPAs, and this leads to most data being incomparable as the models and concepts used are vastly different. Since RPAs were introduced, their identification has been an important topic in all EU Member and Candidate States (and beyond, as some non-Member States have also decided to adopt the regulation or parts of it). Quality assurance of all steps in the radon protection chain is necessary, in particular given they are methodically different but have important potential economic and political impact.

In MetroRADON methodologies for the identification of RPAs are analysed and developed. Relationships between indoor radon concentration and quantities related to geogenic radon, including soil exhalation, are being investigated. Information is being collected and the methods for radon mapping and delineation of RPAs which are already being used in different countries or regions compared. It will be evaluated what purpose they can be used for (e.g. in workplaces, preventive measures, public radon exposure) and if and how certain methods, developed in one country for a specific purpose, could be used or adapted for other purposes or used in other countries or regions. Strategies to deal with RPAs which have been defined inconsistently across borders will be developed. In addition, approaches for Europe-wide mapping methods will be evaluated and further developed.

The use of CDs and DVDs for retrospective radon measurements and their potential to define radon priority areas will be evaluated. These methods employ CDs or DVDs that are available in almost all public and private buildings in Europe as "detectors" and allow the average radon concentration to be assessed in retrospect, as well as systematic changes due to constructive (including energy-efficiency) interventions. New techniques for measurement of radon exhalation from soil, based on liquid scintillation counting of polymers or track-etching of CDs, are being developed and evaluated. The aim is to analyse and develop methodologies for the identification of radon priority areas (i.e. areas with high radon concentrations in soil, as defined in the EU-BSS).

Finally, a methodology for a harmonised geogenic "radon hazard index" (RHI) will be proposed which could be used as a tool to help identify RPAs and to investigate the relationship between soil

radon exhalation rates and indoor radon concentrations. The methodology for a harmonised RHI will reduce the effort required by Member States to implement and operate national radon action plans. European radiation protection authorities will be able to use the concepts of RHI and the identification of RPAs to make improved and more robust decisions leading to increased public health.

3.3.3.5 Validation of the traceability of European radon calibration facilities, and guidelines and recommendations on calibration and measurement procedures for the determination of radon concentration in air

The desire to improve and harmonise radon measurements in air has increased over the last decade as radon activity concentrations in air measured with different radon monitors were found to be inconsistent with each other when the monitors were placed in the same environment. At present, secondary radon standards are calibrated at relatively high activity concentrations, however calibrations and measurements at low activity concentrations with sufficiently low uncertainties, as required in the context of the EU-BSS, are not available.

During MetroRADON a validation of the traceability of existing European radon calibration facilities at NMIs/DIs, accredited laboratories, other calibration laboratories and universities is undertaken. The results of the exercise will provide an overview of the radon calibration capabilities and will enable radon calibration facilities to improve their methods and expertise regarding radon calibrations. Available information about the validation of traceability of European calibration facilities for radon concentration measurement in air will help end-users to choose and develop calibration facilities tailored to their needs. Information about the validation of traceability of European calibration facilities for radon concentration measurement in air will be provided to all relevant stakeholders and guidelines and recommendations on calibration and measurement procedures for the determination of radon concentration in air will be published.

3.3.4 Conclusions

The largest fraction of the average annual effective dose of each European inhabitant from natural sources of ionising radiation is caused by the inhalation of radon (or more precisely by radon progeny). In large European countries, thousands of lung cancer cases are attributed to indoor radon exposure. The need for this project is mostly motivated by the requirements of the implementation of the European Council Directive 2013/59/EURATOM (EU-BSS) which has the objective to provide a sound and fair basis of radon protection for European citizens and aiming to reduce the risk of lung cancer for European citizens due to high radon concentrations in indoor air. The EU-BSS, which is currently in the process of transposition into national legislation by 2018 for immediate implementation, requires the monitoring of radon levels and the implementation of appropriate precautions against exposure to radon for the protection of the European population in the field of natural radiation. Effective implementation of the EU-BSS will require accurate and reliable measurement of low radon activity concentrations. MetroRADON will help both to establish a basic infrastructure so that metrologically sound measurements can be made to support the implementation of the EU-BSS and sound decision making for radiological protection. The results of the project will effectively support the metrological infrastructure for traceable radon measurements in Europe. These aims agree and complement the plans of the new EURADOS e.V. subgroup WG3.3 Radon. Therefore, a letter of agreement was signed concerning the collaboration in the field of radon monitoring in July 2018.

Acknowledgements

This work is supported by the European Metrology Programme for Innovation and Research (EMPIR), JRP-Contract 16ENV10 MetroRADON (www.euramet.org). The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States.

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3.4 Occupational exposure to radon progenies in underground coal mines (Krystian Skubacz, Główny Instytut Górnictwa, Śląskie Centrum Radiometrii Środowiskowej (GIG), Poland)

3.4.1 Introduction

The aim of the paper is to present possible sources of hazards related to the enhanced level of natural ionising radiation in Polish underground mines, the reasons for their formation, mitigation and monitoring methods, and the result of evaluation of doses to miners.

The sources of enhanced natural ionising radiation like radium bearing waters flowing out of the rock, radioactive sediments that precipitate out of these waters and radioactive aerosols generated after decaying of the radon, occur in Polish underground mines. The higher concentration of short-lived radon progeny can be therefore observed close to locations of sediment accumulation. However, the radon exhaled from cracked rock near the mining areas is much more important source of exposure to miners. Additionally, the radium bearing waters and sediments occur only in some places like water galleries designated for collection of mine waters before pumping to the surface and water pumping stations, which as restricted areas are only available to the authorised staff, and as a result the number of exposed miners is limited.

The hazard related to short lived radon progeny strongly depends on the mining and ventilation system, and the equilibrium factor between radon and its progeny can change in a very wide range from 5 % up to 95 %. That's why the potential alpha energy concentration is measured in Polish underground mines instead of radon concentration. Obligatory measurements of the potential alpha energy concentration of short lived radon progeny have been performing in the Polish underground mines since 1989. The highest value, equal 15 $\mu J\ m^{-3}$, has been measured in the stream of used air close to the place where hard coal was mined. In such places the rocks are crushed and often ventilation is rather poor which results in increasing of the radon exhalation rate and equilibrium factor between radon and its short-lived decay products.

The measuring units must comply the requirements of the State Mining Authority to apply it in underground mines. The applied devices are mainly based on thermoluminescence detectors that are able to register alpha energy released by short lived radon progeny collected on a filter.

3.4.2 NORMs in underground working of Polish hard coal mines

Currently, there are 23 underground mines in Poland, including 18 hard coal mines, 3 copper mines, 1 zinc and lead mine, and a salt mine. In addition, there are also closed mines available to tourists, where silver, gold, uranium, salt and hard coal were mined in the past. The presented research results apply only to hard coal mines. There are radioactive underground waters containing elevated concentrations of radium-226 and radium-228 (Table 3.11). Radioactive sediments are precipitated from these waters when they contain barium and are mixed with waters containing sulfates. Such deposits are the source of external gamma radiation, and air kerma rate at a distance of 1 m can reach 100 $\mu Gy\ h^{-1}$. Moreover, the additional dose can be caused by the ingestion and inhalation intake of nuclides which are present in these sediments. In places where occur radioactive sediments and waters the potential alpha energy concentration of short-lived radon progeny increases. However, the highest concentrations of these isotopes were measured close to the mining areas, where there are no radioactive deposits and the concentration of radium in the rocks is of about 20 $Bq\ kg^{-1}$ (Chałupnik et al., 2001; Skubacz et al., 2011). The reason is the poor ventilation and increased radon exhalation due to the crushing of rocks by mining machines. The doses

corresponding to these natural hazards evaluated for ten-year period are illustrated in Figure 3.8 (Wysocka et al., 2018).

Table 3.11 Natural radionuclides in environment and measured values in Polish underground mines.

Source	Surface	Polish underground mines
Short-lived radon progeny	Open air, 1 m above the ground: 0.05 μJ m^{-3} Dwellings: 0.10 μJ m^{-3}	up to 15 μJ m^{-3} (60 μJ m^{-3})*
Waters	Rivers, lakes: 0.004 kBq m^{-3}	^{226}Ra: up to 390 kBq m^{-3} ^{228}Ra: up to 200 kBq m^{-3}
Sediments	Soil, ^{226}Ra: 0.025 kBq kg^{-1}, ^{228}Ra: 0.025 kBq kg^{-1}	up to 500 kBq kg^{-1} (^{226}Ra+^{228}Ra)
Gamma radiation	Open air: 0.069 μGy h^{-1}	up to 100 μGy h^{-1}

*) the increase in the potential alpha energy concentration to 60 μJ m^{-3} was not a result of usual mining operation but by the uranium deposit accidentally exposed in one of hard coal mines.

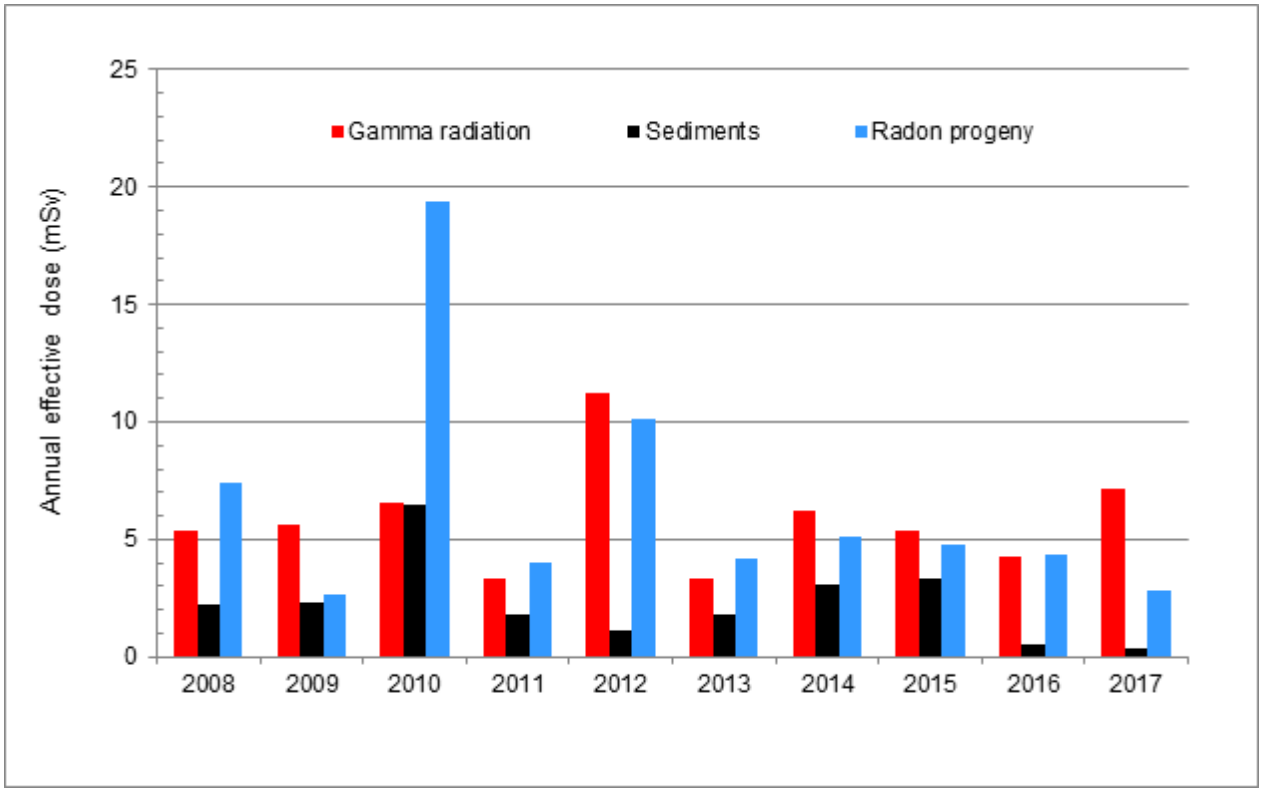

Figure 3.8 Maximum annual effective doses related to natural sources of radiation hazards in Polish underground hard coal mines.

3.4.3 Short-lived radon progeny

According to calculations based on the Jacobi-Eisfeld lung dosimetry model (Jacobi and Eisfeld, 1980) for aerosols of 0.25 μm in size which are deposited in the airways with the least efficiency, radon contributes only 2 % to the effective dose, while its short-lived progeny 98 % assuming that all these isotopes are in radioactive equilibrium (Figure 3.9). On the other side, due to the ventilation and mining system the evaluated equilibrium between radon and short-lived radon progeny changes in the range from below 5 % up to 95 %. That's why according to the Polish legal regulations, the potential alpha energy concentration is measured in underground mines instead of the radon concentration.

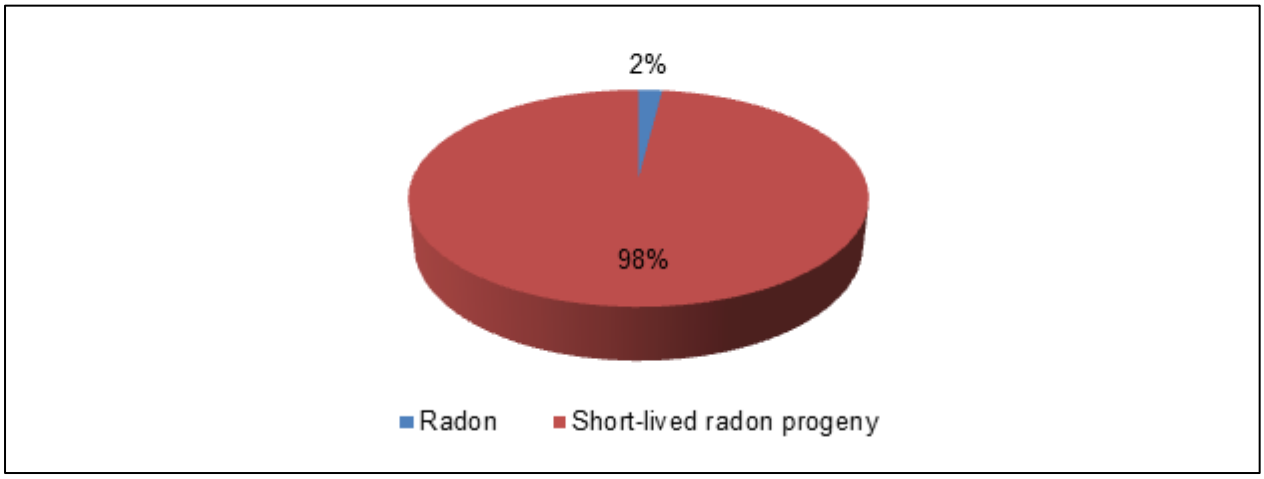

Figure 3.9 Contribution of radon and radon progeny to the total dose.

Obligatory measurement locations of the potential alpha energy concentration are the followings:

- Workplaces close to the air outlet from the longwalls,
- Workplaces close to the air outlet from the road head with the separate ventilation,
- Stationary workplaces such as workshops, special mine rooms, and switching stations,
- Temporary or stationary workplaces where air kerma rate is greater than 0.6 μGy h^{-1}.

The measuring devices are equipped with the cyclones to separate the respirable fraction from particles that are not able to reach the lower respiratory tracks (Figure 3.10). Inside the cyclones are so-called alpha probes with thermoluminescent detectors (TLDs) with $CaSO_4$:Dy or $CaSO_4$:Tm as a luminophore. They are located above the filter that intercepts the respirable particles passing through the cyclones. The TLDs are placed in three sockets to take into account the inhomogeneity of the dust layer on the filter and reduce the limit of detection. These detectors record the radiation emitted by the short-lived radon progeny. The limit of detection for such measuring devices is of about 0.02 μJ m^{-3} at 5 % significance level taking into account 7-hours measurement. Such devices have to meet the safety standards related to the underground mines.

Aspirator AP-2000 EX

Aspirator SKC 224-PCEX8

Dust sampler Barbara 3a

Figure 3.10 Integrating measuring devices for measurement of the potential alpha energy concentration.

In Table 3.12 are collected results from 10-years period of measurements of the potential alpha energy concentration in Polish underground hard coal mines (Wysocka et al., 2018). There always were places where the annual effective dose was greater than 1 mSv or even 6 mSv taking into account the presently recommended conversion factor of 1.4 Sv $(J\ m^{-3}\ h)^{-1}$. Such value is still valid in the legal requirements. However, according to the new ICRP report no. 107 the dose conversion factor should be increased to 3 Sv $(J\ m^{-3}\ h)^{-1}$.

Table 3.12 Results from 10-year period of measurements performed in Polish underground mines.

Year	Number of results			Range	Average	Median	Standard deviation
	$C_\alpha \leq 0.5$ μJ m^{-3}	$0.5<C_\alpha\leq 2.5$ μJ m^{-3}	$C_\alpha>2.5$ μJ m^{-3}	μJ m^{-3}			
2008	2871	36	1	0.01 - 2.8	0.08	0.05	0.13
2009	2954	17	-	0.01 - 1.1	0.07	0.04	0.08
2010	2973	45	5	0.01 - 7.3	0.09	0.05	0.25
2011	3039	25	-	<0.01 - 1.5	0.06	0.04	0.09
2012	2944	20	1	<0.01 - 3.7	0.08	0.05	0.11
2013	3127	39	-	<0.01 - 1.6	0.07	0.04	0.11
2014	3172	50	-	0.01- 1.9	0.12	0.09	0.13
2015	3140	24	-	0.01- 1.8	0.10	0.08	0.10
2016	2992	26	-	0.01 - 1.7	0.11	0.08	0.10
2017	3143	41	-	0.01 -1.1	0.15	0.12	0.11

3.4.4 Mitigation methods

There are some mitigation methods to reduce the radon risk. Covering of side-wall with sealing compound like phenol-formaldehyde resin + catalyst reduces the potential alpha concentration of short-lived radon progeny. Such countermeasure was applied in one of the hard coal underground mine. As a result, the concentration was decreased nearly eight times (Figure 3.11). However, such activities are usually carried out to reduce the underground fire hazards.

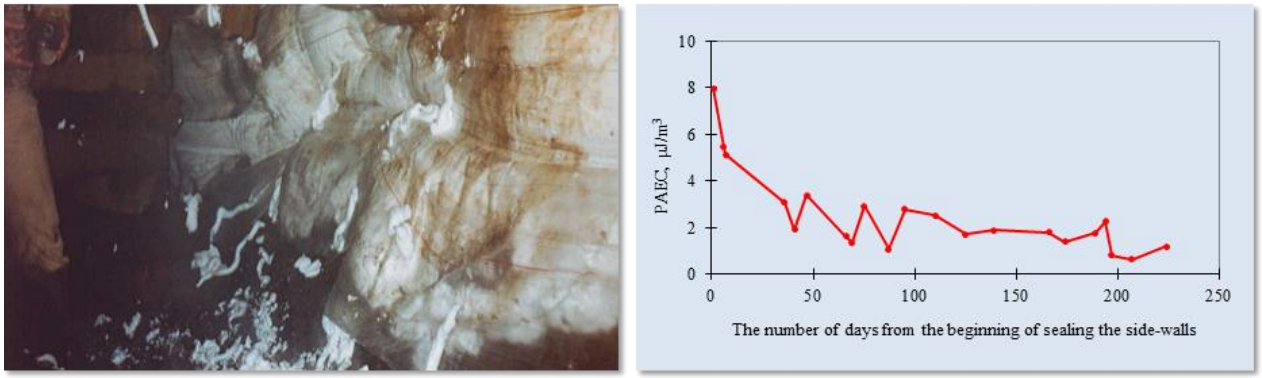

Figure 3.11 Side-wall sealing.

The second method which was successfully used was air filtration as presented in Figure 3.12. In this case the potential alpha energy concentration decreased from about 4 µJ m^{-3} up to 0.5 µJ m^{-3}. The flow rate trough the filter was ranged from 270 to 360 m^3 min^{-1}. The disadvantage of this method was the relatively fast clogging of the filters where there was high dust concentration.

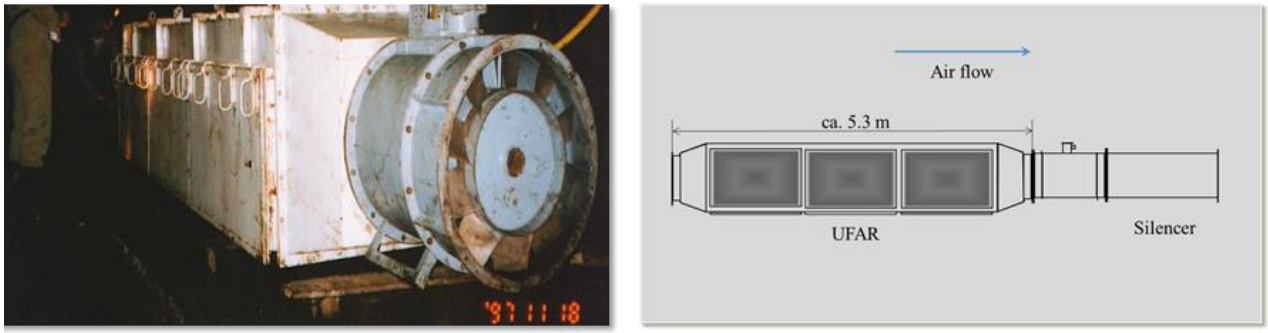

Figure 3.12 Air filtration as a method of reduction of the potential alpha energy concentration.

3.4.5 Size distributions of ambient and radioactive aerosols

The dose conversion factors depend on breathing mode and particle size distribution of radioactive aerosols. For practical reasons, the routine evaluation of doses is based on the recommended value of the dose conversion factor, which for occupational exposure is equal to $k_F = 1.4$ Sv (J m^{-3} h)$^{-1}$. The evaluation of the dose conversion factors corresponding to the measured ambient particle size distribution and the developed transformation method to the size distribution of radioactive aerosols was made for all key working places in an underground hard coal mine. An example of that transformation is illustrated in Figure 3.13.

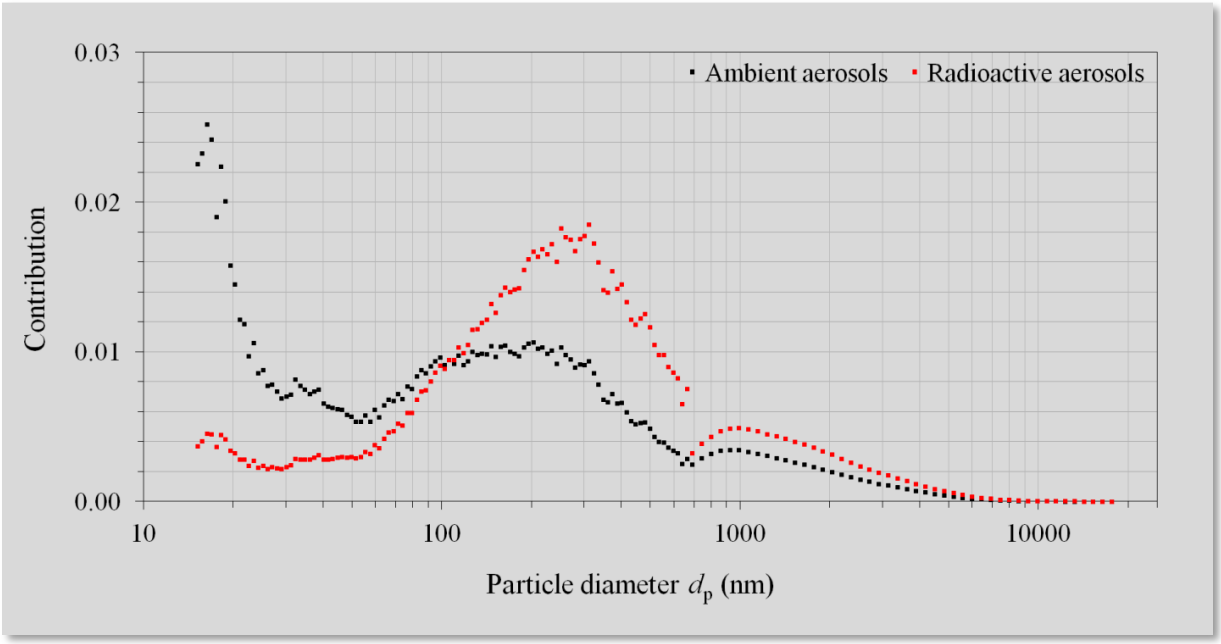

Figure 3.13 The count and activity contributions of ambient and radioactive aerosols measured close to the road head during heading machine operation.

Such investigations were made in the place where the fresh air stream from the surface flows into the mine to ventilate underground excavations (downcast shaft bottom), mine excavation made for the purpose of making available the coal bed and transportation of miners and materials (cross cut), the place where the mine water is collected before pumping it to the surface (water gallery), the area where a new mine excavation is drilled (road head), the place where the hard coal is mined out (coal face or longwall), and a location where used air (dead air) is discharged from the mine to the surface (upcast shaft bottom). The estimated dose conversion factors were much higher than the recommended value of 1.4 Sv $(J\ m^{-3}\ h)^{-1}$. These values are reported in Table 3.13 for breathing rate of 1.2 $m^3\ h^{-1}$ via mouth and 0.75 $m^3\ h^{-1}$ via nose, and were close to the results obtained in German underground mines (Skubacz et al., 2016; Zock et al., 1996).

Table 3.13 Dose conversion factor evaluated for underground workings of a hard coal mine based on measurement of the ambient aerosol size distributions.

Measurement point	Unattached fraction (%)	Dose conversion factor k_F $(Sv\ (J\ m^{-3}\ h)^{-1}$	
		Mouth Breathing rate: 1.2 $m^3\ h^{-1}$	Nose Breathing rate: 0.75 $m^3\ h^{-1}$
Downcast shaft bottom	0.7	6.0	4.7
Cross-cut	0.7	4.4	3.4
Water gallery	1.7	4.9	3.7
Road head	2.0	4.3	2.8
Coal face (30 m away)	1.6	4.6	3.1
Coal face (80 m away)	0.4	7.1	5.5
Upcast shaft bottom	1.1	5.3	4.1

3.4.6 Summary

- The main sources of the natural radioactivity in Polish underground mines are radioactive waters, sediments and short-lived radon progeny.
- The concentration of natural radioisotopes in rocks is usually similar to their natural level in soil and the increase of the potential alpha energy concentration strongly depends on the mining and ventilation system. There is mostly applied the longwall system that leads to the crushing of rocks and increased exhalation of radon from grinded materials.
- The equilibrium factor between radon and its progeny can change in a very wide range from 5 % up to 95 %. That's why the potential alpha energy concentration is measured in Polish underground mines instead of radon concentration.
- The evaluated dose conversion factors for radon hazards were much higher than recommended value of 1.4 Sv $(J\ m^{-3}\ h)^{-1}$ for the occupational exposure.

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4. Radon as a tracer

4.1 ^{222}Rn as tracer for climate studies: from the improvement of the metrology to the scientific applications. (Claudia Grossi and Arturo Vargas, Universitat Politècnica de Catalunya (UPC), Spain)

4.1.1 Introduction

The atmospheric increase of greenhouse gases (GHGs) has a well-known impact on climate change. GHGs emissions from natural as well as anthropogenic sources are being estimated and reported by each national agency. Understanding the processes causing these emissions is important for the implementation of future emission reduction strategies (IPCC, 2013).

The radioactive noble gas radon (^{222}Rn), due to its chemical and physical characteristic (e.g. Nazaroff and Nero, 1988), is being applied in different studies for investigating, among others, soil-atmosphere exchanges. Both atmospheric ^{222}Rn and GHGs concentrations are used within the Radon Tracer Method (RTM) to estimate local/regional nocturnal GHGs fluxes (e.g. Szegvary et al., 2009; Levin et al. 2011; Vogel et al., 2012; Grossi et al., 2016; Grossi et al., 2018).

For the RTM being applied, the knowledge of the environmental atmospheric radon concentration and its soil flux are key parameters. European GHGs monitoring infrastructures are including atmospheric ^{222}Rn monitors in their stations (e.g. Grossi et al., 2016; Grossi et al., 2018). Atmospheric ^{222}Rn measurements currently carried out are mainly based on three different measurement principles. These different measurements are not yet harmonised between them and no traceability to international standards has been developed so far. On the other hand, radon flux data from direct measurements and models are still to an early stage. Reliable measurements techniques and protocols have not been approved and radon flux inventories for the atmospheric and radiation protection scientific communities are not yet validated (Szegvary et al., 2009; IAEA, 2012; López-Coto et al., 2013; Karstens et al., 2015; Schmithüsen et al., 2016).

Due to the present situation, nowadays the research in this field is mainly focused on: i) the building of robust and user friendly atmospheric radon monitors; ii) the enlargement of the atmospheric ^{222}Rn measurements in Europe and their harmonisation; iii) the improvement of continuous radon flux measurements; iv) the standardisation of the Radon Tracer Method for the retrieval of GHGs fluxes in rural and urban areas. The state of the art of the research done by the Institut de Tècniques Energètiques (INTE) of the Universitat Politècnica de Catalunya (UPC), in collaboration with other researchers, is here summarised and presented.

4.1.2 The building of robust and user friendly atmospheric radon monitors

A portable monitor for continuous measurements in outdoor air was designed and build at the INTE-UPC (Grossi et al., 2012). This Atmospheric Radon MONitor (ARMON) performs a direct measurement of ^{222}Rn and ^{220}Rn concentrations based on the α spectrometry of ^{218}Po and ^{216}Po, respectively, on a passivated implanted planar silicon detector surface and using a high electrostatic field. Sample air is previously filtered to remove ambient aerosols and progeny. So fresh progeny is generated within the 20L detector volume. The positive ions of polonium, resulting exclusively from the α-decay of ^{222}Rn and ^{220}Rn, arrive on the surface of the detector because of the electrostatic field. The ARMON can perform hourly measurements of atmospheric ^{222}Rn concentrations with a minimum detectable concentration of around 250 mBq m^{-3}. Each monitor is calibrated at the INTE-UPC radon chamber (Vargas, 2004) considered as the reference chamber in Spain by the Spanish Nuclear Safety Council

(CSN). The INTE-UPC radon chamber was previously traceable to the German National Metrology Institute Physikalisch-Technische Bundesanstalt (PTB) and now to the Swedish radiation safety authority, also if this traceability only valid above 100 Bq m^{-3}, which is by 2 to 3 orders of magnitude bigger than atmospheric radon concentrations. The instrument Atmos 12 DPX is used as second standard.

4.1.3 The enlargement of the atmospheric ^{222}Rn measurements in Europe and their harmonisation

The black labels in Figure 4.1 show the locations of the European long-term atmospheric GHGs or aerosol stations where atmospheric ^{222}Rn concentrations are also measured. In order to address the lack of stations over the Mediterranean basin the Institut Català de Ciències del Clima (IC3), in collaboration with the INTE-UPC and the Universidad de Huelva (UHU), installed three ARMONs in coastal and mountainous GHGs or aerosol stations in Spain (Grossi et al., 2016).

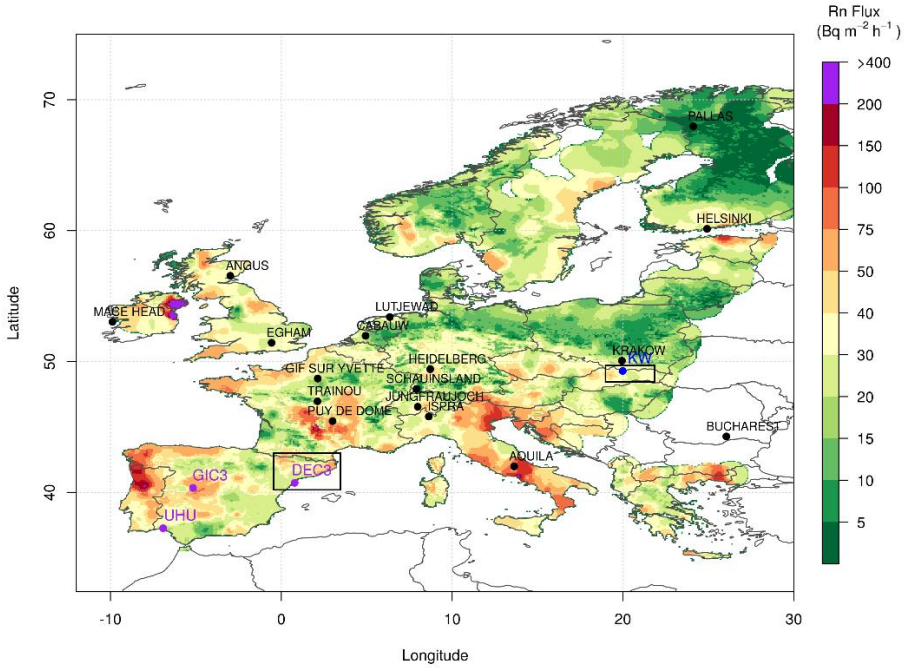

Figure 4.1 Location of operative atmospheric ^{222}Rn stations in Europe (black labels) and recent Spanish stations (violet labels) where co-located measurements of GHGs or aerosol are carried out in agreement with the literature (Grossi et al., 2016).

At European level, the two systems mainly operating are a dual-flow-loop two-filter monitor (Zahorowski et al., 2004), which samples and measures radon directly, and a one-filter monitor (Levin et al., 2002), which samples and measures radon progeny. A third method is being applied at three Spanish ^{222}Rn stations (Grossi et al., 2016) which performs a direct measurement of ^{222}Rn and ^{220}Rn (thoron) concentrations using electrostatic deposition of ^{218}Po and ^{216}Po, respectively.

Due to the diversity of the aforementioned techniques, biases and compatibility issues could limit the comparability of the results obtained by independent local studies and their application for regional-or-global applications. The International Atomic Energy Agency suggested a harmonisation of datasets obtained by the different techniques (IAEA, 2012).

Xia et al. (2010), Griffiths et al. (2016); Grossi et al. (2016); Levin et al. (2017) and Schmithüsen et al. (2017) have carried out studies to improve the accurate monitors responses and to harmonise their results. However, so far, these studies have been performed using only two individual instruments in parallel. Grossi et al. (2017) carried out a 3-months inter-comparison campaign in fall 2016 in Gif Sur Yvette (France) where, for the first time, co-located measurements from monitors based on the three measurement principles were included. Specifically, two single-filter ^{222}Rn progeny monitors (a Heidelberg Radon Monitor (HEIDELBERG) and one by the Laboratoire des Sciences du Climat et de l'Environnement (LSCE)); two two-filters ^{222}Rn monitors by the Australian Nuclear Science and Technology Organisation (ANSTO) and an electrodeposition Atmospheric Radon MONitor (ARMON) by the INTE-UPC, were simultaneously running and sampling air at 2 m above ground level (a.g.l.) and at 100 m a.g.l. under different climatic and aerosol conditions (see Figure 4.2).

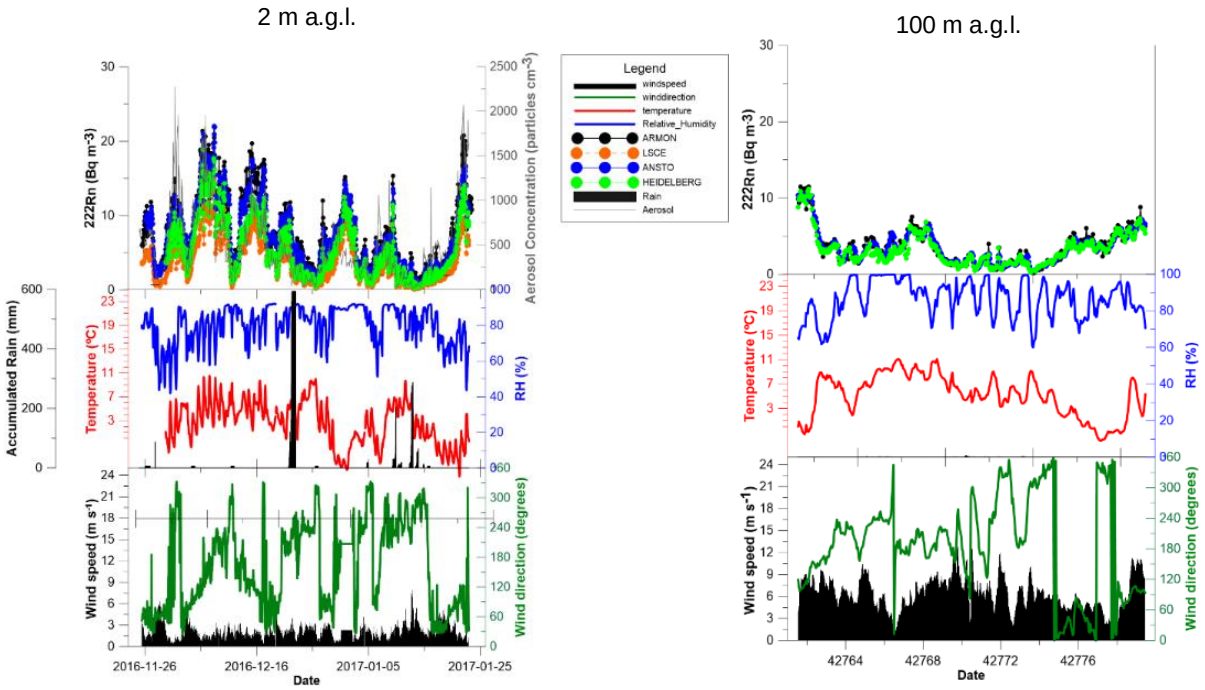

Figure 4.2 Time series of hourly ^{222}Rn concentrations of different types of radon monitors and the respective environmental parameters measured at 2 m above ground level (a.g.l.) (left panel) and at 100 m a.g.l. (right panel) at Gif Sur Yvette (France) (Grossi et al., 2017).

Results of this study show a general agreement in the response of the radon and radon progeny monitors. A decrease in the ^{222}Rn concentration measured by progeny monitors was observed during rain episodes or when the inlet was close to the soil surface. However, due to the really dry conditions of the inter-comparison campaign, there is still the need of a statistical analysis of the rain influence. It is clear the importance of building in the next future a robust metrology for atmospheric radon measurements and of designing a portable atmospheric radon monitor, calibrated with a traceable method, which should be used to calibrate all European monitors in the field.

4.1.4 The improvement of continuous radon flux measurements

The ^{222}Rn source needs to be known on a local as well as global scale. Obtaining experimental radon flux data on global scale is not feasible. ^{222}Rn flux inventories based on indirect methods or models can help but their reliability has to be carefully checked. The exhalation of ^{222}Rn from the soil surface is influenced by environmental factors. These previous influenced need to be investigated to

correctly validate ^{222}Rn flux inventories. However, random sampling and temporal ^{222}Rn flux measurements are insufficient to clarify the relationship between exhalation and environmental factors (Karstens et al., 2015; Yang et al., 2017). In addition, ^{222}Rn flux observations can also be affected by the measurement technique itself and inter-comparison studies need to be performed (Grossi et al., 2011). Yang et al. (2017) show that the use of an accumulation chamber prevents soil surface from precipitation, convection or wind induced effects, and thereby affects the conditions of the soil surface. ^{222}Rn flux metrology and long-term continuous measurement data is needed.

A COntinuous ^{222}Rn Flux (CORAF) monitor was designed and build at the INTE-UPC in order to perform long-term measurement over selected sites. The CORAF is based on the accumulation method (Morawska and Phillips, 1980). The ^{222}Rn concentration in the chamber is measured by a Doseman instrument (www.sarad-radonshop.com). A system made of a small pump and two valves allows the ventilation of the accumulation chamber every set time. Figure 4.3 shows the CORAF installed directly in the soil. Early experiments have been carried out in order to investigate the influence that water content has on the ^{222}Rn exhalation from a selected soil (Figure 4.4). Here the radon concentrations within the accumulated chamber are presented with different colours in agreement with the date of the experiment (between the 3rd of April 2014 and the 3rd of June 2014).

Figure 4.3 COntinuous RAdon Flux (CORAF) monitor designed and build at the INTE-UPC.

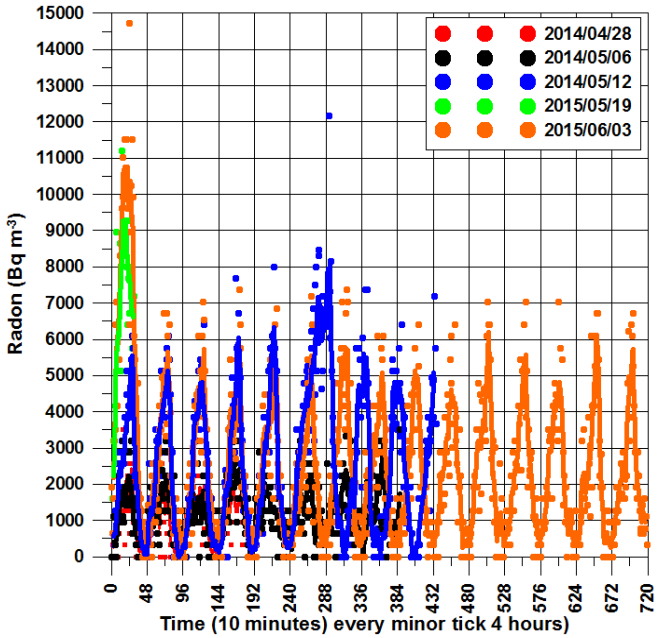

Figure 4.4 Time series of hourly ^{222}Rn concentrations measured within the accumulation chamber of the CORAF under different water content conditions of the soil sample (right side).

4.1.5 The standardisation of the Radon Tracer Method (RTM) for the retrieval of GHGs fluxes in rural and urban areas

The ClimaDat station at Gredos (GIC3), belonged to the IC3, has been continuously measuring atmospheric concentrations of methane (CH_4) and ^{222}Rn, as well as meteorological parameters, since November 2012. To understand the variability of CH_4 emissions, nocturnal fluxes of CH_4 were estimated using the Radon Tracer Method (RTM) and the emission inventory (Grossi et al., 2018). Both flux estimation methods have been applied using the same source region as modelled by the atmospheric transport model FLEXPARTv9.0.2. Figure 4.5 shows the monthly means of the obtained results. The RTM was applied using the radon flux data obtained with the model of Lopez-Coto et al. (2013) and rescale factor obtained by Karstens et al. (2015). The ratio between the CH_4 fluxes obtained using the EDGAR inventory and the RTM is reduced from 3 to 1 rescaled RTM results are used. This fact underlines the importance of using a valid radon flux inventory.

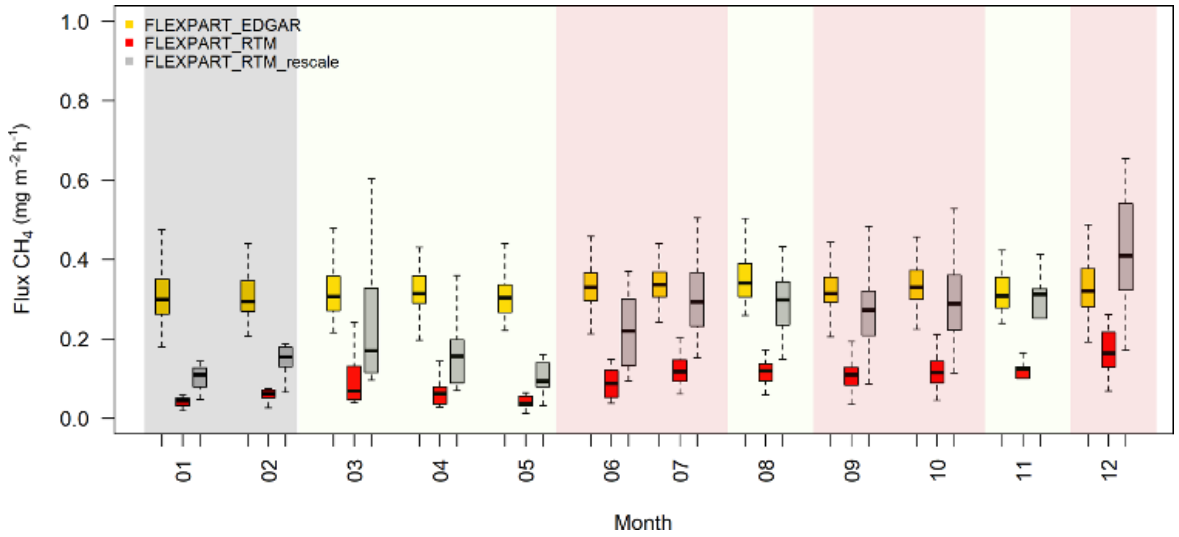

Figure 4.5 Boxplots of monthly CH_4 fluxes (mg CH_4 m^{-2} h^{-1}) calculated for the GIC3 area using the RTM technique (red), the EDGAR inventory (yellow) and RTM technique using the ^{222}Rn flux comparison factor found by Karstens et al. (2015) (grey) (Grossi et al., 2018).

4.1.6 Conclusions and Future steps

The utility of the radon tracer method in helping to the improvement of greenhouse gases emission inventories implies the importance of standardise this method, Particularly, the use of the RTM has been shown, while also highlighting the need to improve this method, especially in regard to: i) validation of the ^{222}Rn flux maps applied within the RTM; ii) harmonisation of atmospheric radon concentrations in Europe; iii) standardisation of the footprint calculation. Atmospheric ^{222}Rn measurements are being carried out within European networks, because of the large radon tracer applications and it will be important to have as many stations as possible. In addition, the harmonisation of all data-sets and a robust metrology to calibrate the European atmospheric radon monitors is - needed. ^{222}Rn flux inventories currently available have to be validated and this leads to the need of continuous ^{222}Rn flux data on local scale and a radon flux metrology chain.

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