4th European IRPA Congress Geneva June 23-27, 2014

Radiation Measurement and Protection in Radon-Balneotherapy – a Review

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1. Introduction

Radon (Rn) and its decay products (DPs) are natural, ubiquitous, multifaceted, *pharmacologically active* substances. Rn and DPs are alpha-, beta- and gamma-emitters and hence offer wonderful opportunities to both old and young explorers for license-free, interesting, multi-method, low-cost samples and measurements for research, practice, teaching, and radiation protection and radiometric preparedness for nuclear events.

The author has been radiation protection practitioner in a radon-spa since 1990, conducting measurements with specially designed methods and instruments which proved to be of value above and beyond their original purpose. This may serve as an example for young researchers how an outside request for help with a practical problem develops unexpectedly into a useful and rewarding field of applied science and public health.

Spas offering radon-balneotherapy are subject to radiation protection ordinances in Germany, Austria, the Czech Republic and other countries. Though the number of such workplaces, workers and patients is small, the special radiometric conditions in the air, in the water, on surfaces, on and in the human body prevailing in radon-spas are a real challenge to anyone dedicated to unusual measurements. Radiation measurements in radon-balneotherapy should be made, and have been made regularly in the radon-spa Sibyllenbad (140 km north-east of Regensburg), for four reasons: 1. legal (radiation protection); 2. economic (quality assurance); 3. psychological (for personnel and patients); 4. scientific (developing methods to measure low-level radiation in view of their biological effects).

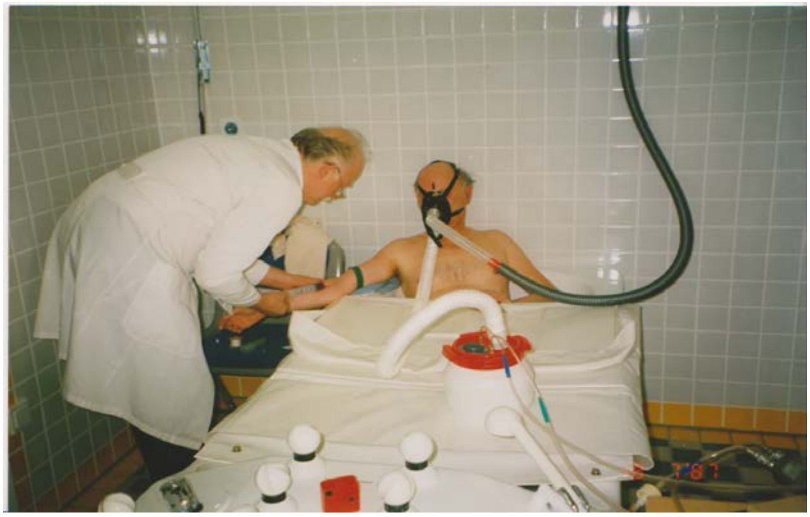

Fig. 1: The author breathing outdoor air, expiratory air measured, blood taken

Radon cures in air or in water are prescribed by specialized physicians for a number of medical indications. Neither the indications nor their observed or claimed medical effects will be discussed in this paper. The primary function of a radiation protection practitioner is to protect the personnel from undue exposure and to verify the radon concentrations in the air or in the water promised to the patients. Rn emanation rates from water and room air ventilation rates determine both the concentrations of Rn and DPs, unattached or attached to aerosol particles, as well as the factor of equilibrium and the aerosol particle size distribution, all essential for the lung dose conversion factor.

Environmental conditions in radon-spas are quite different from those in other workplaces such as mines or waterworks or in homes. They differ significantly from spa to spa, in particular between those offering water in bath tubs and those offering air in caves or rock galleries. Measurements should have a high time resolution and be easily performed on the spot with results immediately available for demonstration to personnel and patients.

In Sibyllenbad, patients are exposed within 2 or 3 weeks in 10 sessions, 20 min each, to radon water with about 1200 to 1500 Bq L^{-1} under high rates of ventilation of 6-8 h^{-1} and a factor of equilibrium of 0.05. Hence, the lung is not a critical organ. Therapeutic exposure comes from DPs deposited on the skin, from Rn taken up from the water through the skin into the blood, and from DPs born in the body. Exposures of the personnel for actual working hours in well ventilated therapy rooms are below 1 mSv a^{-1}, the legal limit being 6 mSv a^{-1}.

Four of the novel instruments/methods are: 1. The Living Level Monitor. 2. Measuring Rn-water with glass fibre filters. 3. Calibration of Rn in air, water and solids. 4. Portable large-area detectors for gross alpha and for soft betas and gammas. Additional methods were employed to study and evaluate the uptake of radon by the human body.

2. The Living Level Monitor LLMS 500

It is well known that the physiologically active agent for the lung is not Rn-222 but the DPs or, more correctly their potential-alpha-energy concentration PAEC. Because of the respective half-lives of 3, 27, and 20 min of the first three DPs, only 10 % of the PAEC come from the first alpha-emitter Po-218, and 90 % come from the beta/gamma emitters Pb-214 and Bi-214 converting to the ultimate alpha-emitter Po-214. Hence, the prime physical quantity to be measured is the concentration not of Rn but of the DPs. There is a simple reason why Rn and not the DPs has been made the reference agent in radiation protection ordinances. It is easier and cheaper to measure Rn by passive methods, such as nuclear track detectors or formerly charcoal canisters. With these methods concentrations are averaged over several days or months, quite suitable for homes and ordinary workplaces, but not for radon-spas. DPs require active methods.

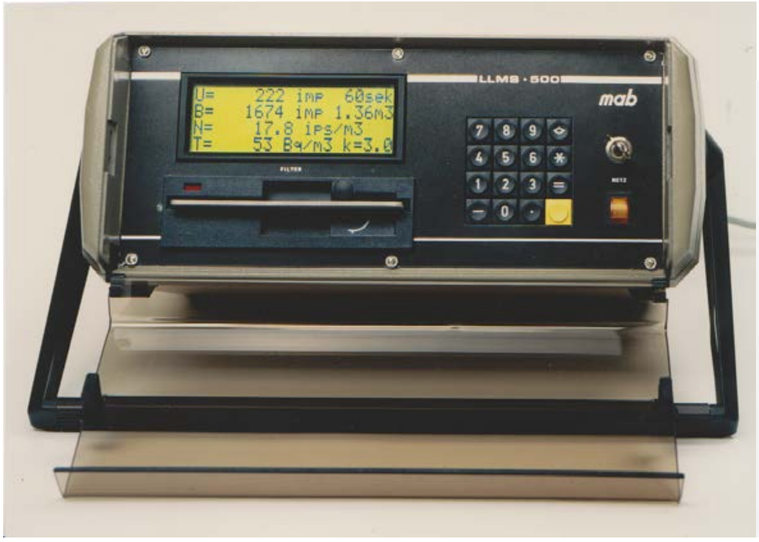

Fig. 2: The Living Level Monitor LLMS 500

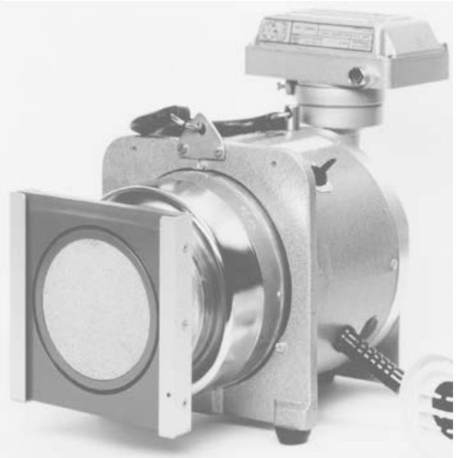

Fig. 3: Staplex sampler

The LLMS 500 [1, 2] consists of two portable units: the sampler and the Monitor. The improved sampler and the novel Monitor were developed in cooperation with the former Münchener Apparatebau mab. The sampler is a powerful Staplex high-volume sampler provided with a mechanical flowmeter, a calibrated Quantometer with continuous reading in

XXXXXX.xx m^3. Glass fibre filters of type MN 85/90 Macherey-Nagel are used. They have an effective diameter of 10 cm, are 0.35 mm thick, filter class S DIN 24184, with quantitative retention for ≥ 0.5 µm aerosol particles. With such filters, a flow rate of 800 L min^{-1} is achieved. With two filters, the lower one collects an activity of less than 1 % of the upper one. For convenience when changing filters, the curling nut and meshed grid support on the Staplex mouth have been replaced by a special gadget. This gadget consists of a 10 cm Ø sintered metal support for the filter and a holder for the diskette. The diskette consists of two pieces of glued paperboard, 13.5 cm by 19.5 cm, and each 1 mm thick, between which the filter is mounted in an opening 10 cm in diameter. The diskette provides a fixed geometry also for powders between transparent adhesive tapes.

The Monitor contains the detector, a pulse counter, electronic hardware and software. The detector is an 11x11 cm^2 sealed beta/gamma-sensitive proportional detector with 90% argon 10% methane counting gas, and a 4.5 mg cm^{-2} aluminium window. Normal environmental radiation generates about 200 cpm. When the diskette is inserted into the panel opening, the sample comes to rest between the detector and a 0.1 mm tungsten sheet, which is an effective beta reflector and increases the measuring efficiency by a factor of 1.25. The efficiency for DPs is e = 33 %, the calibration factor $k = e^{-1}$ = 3 Bq cps^{-1}. A sample of 1.9 g KCl with e = 16 % for K-40 serves as control sample.

The front panel of the LLMS 500 has a floppy disk opening for the sample diskette and a 4x4 key array with numbers or signs for choosing different modes of measurement (environment, air, water, solids), and the parameters m^3, L, g, k, the counting time in min and the number of automatic repetitions up to 999 in steps of one. At the end of a measurement cycle the chosen parameters and the results in respective units are displayed on a four-line 20-digit LC-display. Results are printed online or stored for later printing. In effect, in 1993 the LLMS 500 was the first portable commercial radiation measuring instrument offering a menu and a data logger.

In addition to measuring DPs from air and radionuclides in solids, the LLMS 500 was instrumental in discovering the quantitative adsorption of DPs from water on the same type of glass fibre filters as used for aerosol particles. The LLMS 500 was used in 1993 and later in demonstrations to measure air filters and a table cloth exposed in 1986 in Regensburg to Chernobyl fallout. With the repetitive mode of measurement, DPs with a first effective half-life of 45 min are easily distinguished from Cs-137 or possibly I-131. After Fukushima, two samplers were used to collect about 1000 m^3 of air in 12 h on 6 filters with a total of about one Bq I-131, which were measured by gamma spectrometry. This is just one example for radiometric preparedness provided by special equipment developed for radon-spas.

3. Measuring Rn with glass fibre filters

There is a need for a simple, reliable, sensitive and fast method of measuring radon in water at a source, in radon-spas, in the field, or in lecture rooms. Such a method has been found by playing around with the LLMS 500 and radon water from Sibyllenbad. The new method has been certified by several international intercalibration exercises with the established methods of gamma spectrometry and emanometry.

Glass fibre filters were found [3] to be very efficient for adsorption of the short lived DPs from water when filtered in a Büchner funnel without suction. The retention is more than 95 %. The funnel should be made of porcelain, not of plastic, otherwise the interfacial tension retains several mm of water on the filter. Filtration of 0.5 L in a funnel of 11 cm in diameter, takes about 5 min. The filter is quickly dried on a hot plate with 400 W. The dry filter is conveniently measured with the LLMS 500. The effective half-lives of the DPs collected, as measured with the LLMS 500 or gamma-spectrometry, are in sequence 44, 34, 30, 30, 27 min. The detection limit depends on the filter, the detector and its background, and the volume filtered. The detection limit is as low as 1 Bq L^{-1} when using a pure alpha detector.

In water, radioactive equilibrium between Rn and its DPs is obtained within 3 h, if the water is kept in a completely filled and closed PET bottle, verified experimentally to be gastight. Hence the DPs activity concentration measured is equal to the Rn activity concentration wanted. If the results for fresh water and aged water from the same source are compared, a deviation from radioactive equilibrium may be quite meaningful. Adsorption of the DPs is complete for all neutral drinking water. A slight acidity may be compensated by NaOH, but heavy mineral content is disturbing adsorption. Fresh rain after several dry days contains up to 200 Bq L^{-1} DPs only.

4. Calibration of Rn in air, water, and from solids

The volatile and transient nature of Rn-222 causes a lack of user-friendly, certified standards for radon. Up to now, instruments for measuring radon in air or from solids could not be tested by the individual user for their proper performance and calibration. Commercial radium standards emanating radon are expensive, time consuming and require both a license and delicate handling. A recalibration by the manufacturer or an official calibration service is expensive and means absence of the instrument for a considerable time. Hence there was an urgent need for a simple, fast and reliable procedure for quality assurance and in-laboratory calibration of instruments of the types mentioned.

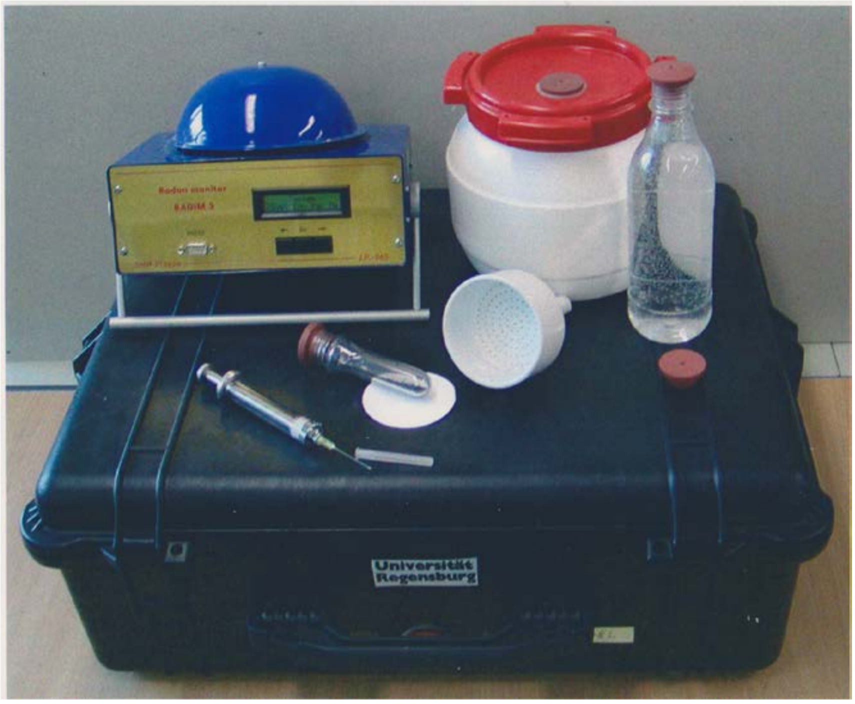

Fig. 4: The parts for in-laboratory calibration of Rn monitors

The calibration procedure [4] can be performed anywhere with a minimum of means, time and effort. The harmless and constant source of radon consists of a gastight plastic container of 3.8 L volume which is filled with 4.5 kg of natural soil from along a road 50 km away from Regensburg. The locality is known for slightly elevated concentrations of uranium and radium due to weathered and fine-dispersed uranium minerals. Hence, the dried soil has a favourable factor of emanation of 0.5. If no appropriate soil is available, crushed uraniferous granite, sandstone or black shale or some uranium minerals may do, though their factor of emanation is much lower, requiring a higher solid activity. The source is in radioactive equilibrium three weeks after the initial filling.

The container is fitted with a septum of chlorbutyl rubber as used in 250 or 500 mL glass bottles for electrolyte-infusion solutions. Empty bottles may be obtained from medical suppliers. The activity concentration of the air in the free volume of the container is left constant when drawing 10 or 20 mL several times from the container with a syringe. Withdrawn air is replaced through a syringe needle. The source is ready for use at any time.

Small and equal volumes of air containing equal activities of Rn are transferred from the source to any combination of two or to all three types of gastight containers, each provided with a septum plug:

1. a 48 L case (Peli Products, Torrance, CA) with one or two instruments of any type for measuring Rn in air,
2. a 0.5 L PET bottle with water. After 3 h the DPs are measured on a glass fibre filter.
3. a 22 mL PET tube with 0.5 g of activated charcoal, grain size about 1 mm, which adsorbs the Rn completely. After 3 h the DPs in the charcoal are measured with the LLMS 500.

Since the volumes of all containers are L or mL, the injection of a low, harmless activity of about 100 Bq gives rise to high activity concentrations in Bq m^{-3}. If the Rn water produced in this manner is measured by gamma-spectrometry, a link to a certified solid reference activity is established.

5. Portable large-area detectors for gross alpha and for soft betas

DPs deposited on various parts of a human body were measured after therapeutic exposure to Rn water. For this purpose, handheld scintillation detectors of 75 mm diameter, 44 cm^2, were developed: with ZnS for gross alpha and with a thin plastic scintillator for soft betas and gammas, both with a convenient data logger. Results were presented at the 4th Biophysics Workshop. [5] Typical body surface activities were 0.4 to 1 Bq cm^{-2} after 20 min in Rn water.

Fig. 5: The portable large-area alpha detector in the author's laboratory

The alpha scintillation probe is sensitive enough for fast retrospective determination of radon exposure by measuring the alpha radiation of Po-210 which is in equilibrium with the Pb-210 implanted on glass samples of windows, mirrors or pictures from rooms with long-term Rn exposure. [6] In 1988 C. Samuelson used a large pulse ionisation chamber with a spectrometric detector, samples with an area of about 200 cm^2, and a counting time of 12 h. Nuclear track detectors are more convenient for on the spot handling, but require up to 60 days of exposure and subsequent in-laboratory etching and counting.

The background of the alpha scintillation probe was reduced from 50 cph to 4 cph and the efficiency was increased from 28 % to 34 %. A detection limit of 5 Bq m^{-2} is calculated for a counting time of 30 min, equivalent to a 20-year exposure of about 250 Bq m^{-3}.

6. Additional methods to study the uptake of Rn

For a correct quantification of the exposure of the personnel in Sibyllenbad, the activity concentration was measured as a function of the aerosol particle size distribution with a special aerosol spectrometer. [7] Though it was shown that the “dose conversion convention” of ICRP 65 is of limited suitability for estimating the effective dose under the unusual environmental conditions prevailing, the correct amount is only 1 mSv a^{-1} for a maximum of 800 working hours in actual fact.

The transfer of Rn from the water, through the skin into blood and into expiratory air was measured with appropriate methods for a person resting 20-30 min in radon water. [8] For waterborne Rn concentrations of (1500 ± 100) Bq L^{-1}, measurements gave (4 ± 1) Bq L^{-1} in the blood and (3.4 ± 0.5) kBq m^{-3} = Bq L^{-1} in the expiratory air, breathing outdoor air (Fig. 1).

The intracorporal deposition of DPs was determined on four persons after 40 and 30 min in Rn water of about 1500 Bq L^{-1} by whole-body gamma spectrometry. [9] About 3000 Bq were found by extrapolation from measurements starting 2.5 h after leaving the water, the time to drive from Sibyllenbad to the University of Leipzig.

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