Evaluation of alpha monitors for detection of Rn-220 in air

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2 Introduction

2.1 Instrument description

The RTM 1688-2 (SARAD GmbH, Dresden, Germany) is a versatile radon and thoron monitor covering the complete field of radon measuring. To perform an alpha spectroscopic analysis, the immediate Radon/Thoron daughter products Po-218/Po-216 are ionized during the decay of the gas. The generated ions are collected on a planar semiconductor detector by an applied electrostatic field. After a short time, the activity of the daughter products on the detector is in equilibrium with the Radon/Thoron concentration within the sampled air. To separate the different daughter products from each other, the method of alpha spectroscopy is used [1].

2.2 Instrument purpose

The purpose of the instrument is to measure Rn-220 concentrations in the outlet air duct in a certain range for Oncoinvent to be able to determine if the emission is within the permitted level.

2.2.1 Hot exhaust

Oncoinvent applies for a permit of 76 GBq of Rn-220 emission per year. However, this amount corresponds to the maximum allowed emission. From the knowledge and experience we have regarding emission and humidity, and despite improved humidity controls, we must expect a mayor part of the emission to take place during wet season (summer/autumn). The duration of the dry season, however, entails an adequate monitoring of Rn-220 concentration also in this season.

In 2021 the highest concentration measured in a single one-hour peak, was 225 000 Bq/m^3 (on 09 Sep 2021).

The instrument therefore needs to be suitable for Rn-220 monitoring in the range of 500 – 225 000 Bq/m^3.

2.2.2 General exhaust

Of the annual Rn-220 emission permit only about 8% is expected through the general exhaust. The majority of this emission is expected to take place during direct handling of radioactivity. In between the level of exhaust in these ducts are expected to be from background level to ten times as much.

3 Information about instruments from the manufacturer

3.1 Calibration of the instrument at the manufacturer site

At the moment Oncoinvent holds seven RTM1688-2 instruments. Instruments with serial numbers RTM6-00331, RTM6-00332, RTM6-00333, RTM6-00392 are currently in use for quantification of Rn-220 in the exhaust air (see table 2). All the instruments under evaluation are regularly calibrated by the manufacturer (e. g., see calibration certificate for instrument RTM6-00392 in attachment [A1]). As a result of a calibration, instrument sensitivity is reported.

The sensitivity of each instrument is determined by comparison to a reference instrument within a sealed calibration chamber under the ambient conditions stated in the certificate (usually, absolute humidity is about 10 g/m^3, which corresponds to 50 % relative humidity at 22.5 ºC). The reference instrument undergoes as annual re-calibration by the German federal office for radiation protection “Bundesamt für Strahlenschutz”. The accredited measurands (Rn-222) are with standards directly based on the primary standards of Physikalisch-Technische Bundesanstalt (Germany). The duration of the comparison measurement is set as to ensure a maximum statistical error (3σ confidence interval) of ±5 % for the instrument under calibration. The statistical error (3σ confidence interval) of the reference measurement is below ±1%, the systematic deviation related to the calibration standard does not exceed ±8 % (also see attachment [A1]).

The instruments are calibrated in this way according to ISO 17025, and the calibration is performed using a Rn-222 source. The calibration coefficient for Rn-220 is thereafter derived from the calibration coefficient for Rn-222 (fast mode). This is possible since the difference in calibration factors is only dependent on the difference in decay constant between the two isotopes. The RTM instruments measure decay events from the primary daughter in the radon decay chain: Po-218 for Rn-222 and Po-216 for Rn-220. These nuclei become charged ions for a short period after the alpha particle emission and these ions are collected onto the detector surface by an electrostatic force. Then the detector measures the energy from the alpha particles emitted in the polonium decay.

The probability for an alpha particle to hit the semiconductor detector is 50% since the alpha will either be emitted into the detector crystal or out from the detector crystal. The efficiency of the ions collection is the same for both Po-218 and Po-216. The energies emitted by the two alphas (6.00 MeV for Po-218, 6.79 MeV for Po-216), are well separated in the spectrum, but their counting efficiencies are approximately the same. This is why calibration performed using Rn-222 can be used to determine sensitivity towards Rn-220. The detector chamber of the RTM1688-2 actually consists of four following chambers, each equipped with its own detector. In the transport from the inlet to the last chamber (flow time from the inlet to the outlet is approx. 50-60 seconds) the Rn-220 concentration will decrease due to decay. This is not the case for Rn-222. The counting efficiency constant for Rn-220 is therefore in practice the calibration constant for Rn-222 times the reduction of Rn-220 as passing the detectors. According to vendor, this relation between both calibration factors was verified years ago when the chamber at PTB Braunschweig still was in operation and will be true if there is exact knowledge on chamber size and the air flow is constant at all times.

3.2 Sensitivity and precision

The precision of the value measured by the instrument is partly defined by the counting statistics. For radioactivity measurements the uncertainty for a number of counts, N, is equal to $\sqrt{N}$ (1σ). To measure the activity with a relative uncertainty of 20%, $\frac{\sqrt{N}}{N} = 0.2$ should be true, which is the case for N = 25

The correlation between counted numbers and radon concentration is given by the equation, see SARAD application note [3].

$$C = \frac{1000 \times N}{T \times K_{eff}} \qquad 1.$$

N: counted numbers

K_{eff}: Counting efficiency from the certificate of the instrument

T: counting time in minutes

According to calibration certificates issued by SARAD for the instruments operating in Oncoinvent, their sensitivity towards Rn-220 ranges from 1.56(±0.7%) to 2.31(±0.7%) cts/(min*kBq/m^3). This means that, 25 counts will result in a reported Rn-220 concentration of 180 Bq/m^3 for the instrument with the highest counting efficiency and 267 kBq/m3 for the instrument with the lowest.

The relative uncertainty will in both cases be 20 %, which means that higher Rn-220 concentrations is needed to achieve the same degree of certainty with the instrument with the lowest efficiency compared to the instrument with the highest. Much lower Rn-220 concentrations can be detected and reported with a lower precision.

In addition to the counting statistics, other uncertainties will also influence the overall uncertainty. The calibration factor itself and the fact that the counting efficiency depends on air humidity also make an impact. In the email correspondence with the manufacturer representative [Veikko Oeser] has reviled their best approach to correct detection efficiency for the humidity conditions different from those used for the calibration (see section 3.1) is a linear correlation. The correction is done so that if humidity is larger than that at calibration conditions the corrected activity value is larger than the original (calculated using the number of counts alone) and vice versa. The exact correction is performed by the following equations (see attachment [A6]):

$$C(Rn)_{corrected} = K(aH) \times C(Rn)_{measured}, \qquad 2.$$

Where $C(Rn)_{corrected}$ is humidity corrected radon concentration,

$K(aH)$ is absolute humidity correction factor,

$C(Rn)_{measured}$ is measured radon concentration.

The absolute humidity correction factor is determined as follows:

$$K(aH) = 1 + m \times (aH - aH_0), \qquad 3.$$

Where $m = 0.025$ m³/g,

aH is absolute humidity during a measurement,

$aH_0 \sim 10$ g/m^3 is absolute humidity during the calibration,

The measured counts are also corrected for tailing from alphas with higher energies, but this has shown to have little to no impact the measured Rn-220 concentration in absence of Rn-222 (see attachment [A5]). The overall uncertainty at about 180-267 Bq/m^3 must therefore be expected to be at least 20% (1σ).

4 Evaluation of the instruments in Oncoinvent

4.1 Evaluation of data with regard to precision and range

SARAD claims to have humidity effects under control. They also claim their instruments are able to operate in a range from [0;10] MBq/m^3 [4] despite the fact that the maximum range of the calibration is [500;25000] Bq/m^3 and quite often much narrower than that. To examine the effect of humidity and measurement range, instrument RTM6-00392 will be used as an example. For all the other instruments the parameters determined by SARAD, as well as the parameters observed based on the instrument's performance during the operation in Oncoinvent are virtually the same (1σ confidence interval).

According to the instrument's latest certificate [A1], ambient conditions used for the calibration were 50 % rel. humidity and 21.5-22.0 ºC. The measurement is performed using Rn-222 chamber with the concentration ranging from 1941 to 3545 Bq/m^3. Sensitivity towards Rn-220 determined in this way is specified then as 1.930 ± 0.013 cts/(min*kBq/m^3). For a 60 minutes measurement this gives the sensitivity (counting efficiency) can be calculated as follows:

$S = 1.930 \cdot 60/1000 \approx 0.116$ cts/(Bq/m^3).

This number should be equal to the slope of the regression curve for the measurements performed under those conditions. To examine the sensitivity, from all the data collected by the instrument RTM6-00392, only those measured in the same conditions of humidity and temperature were extracted:

- relative humidity of exactly 50 %,
- temperature from 21.5 to 22 ºC,
- Rn-220 concentration from 1236 to 10722 Bq/m^3.

This resulted in 30 time points summarized in figure 1. The number of counts in the region of interest 2 used in accordance with [1]. As figure 1 shows, the difference between the slope of the experimental curve (0.117) and the expected value (0.116) is less than 1%.

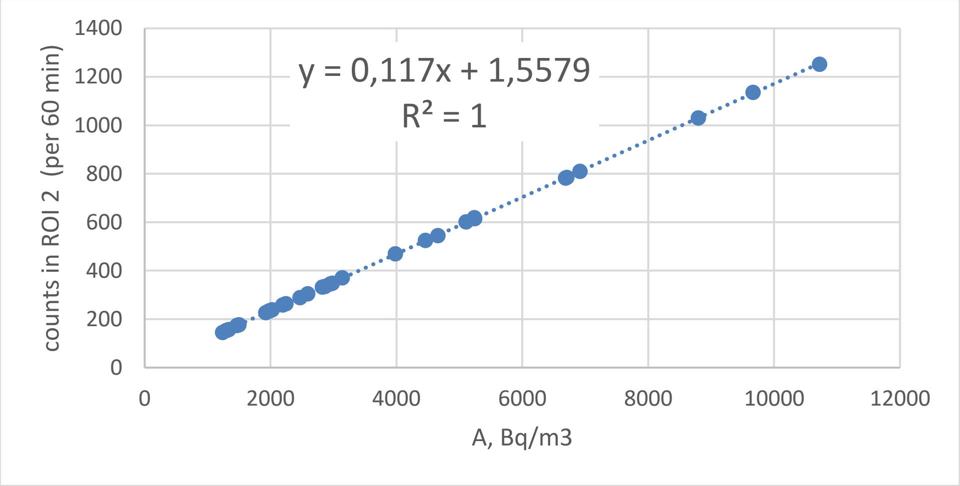

Figure 1 – Correlation of selected raw data (attachment [A2]) from the instrument RTM6-00392

To see to what extent this correction affects the correlation between the counts and Rn-200 concentration in the next test, the whole set of data collected by this instrument from 04 Aug 2021 to 29 Oct 2021 was analyzed (figure 2). Over the time of data collection, variation of the parameters was as follows:

- relative humidity from 17 to 64 %,
- temperature from 20 to 24 ºC,
- Rn-220 concentration from 0 to 225145 Bq/m^3.

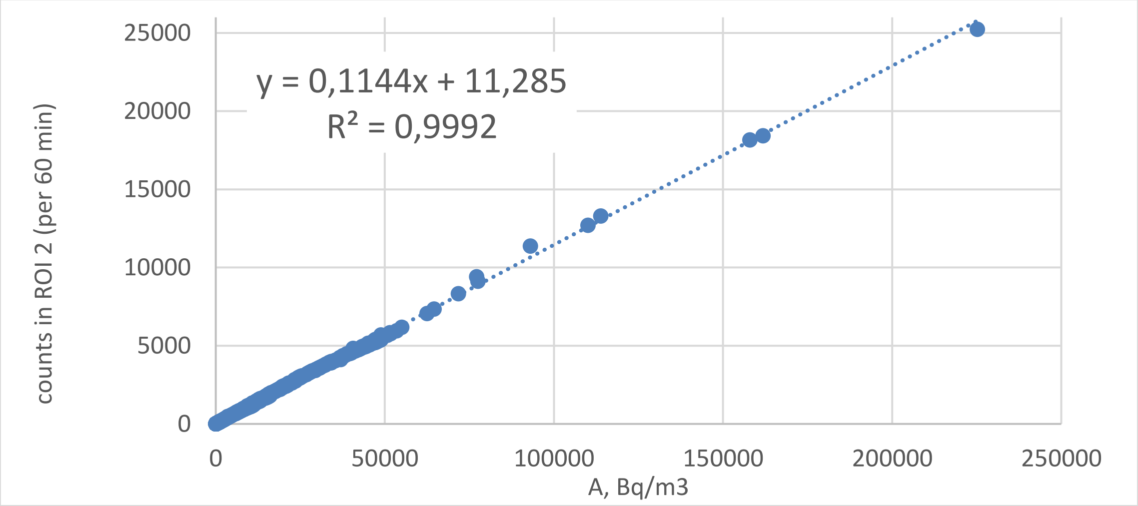

Figure 2 – Correlation of the raw data (attachment [A3]) from the instrument RTM6-00392

As figure 2 shows, even with this wide range of conditions, the slope of the regression curve does not differ from the expected value more than 2 %, and the data points with higher count rates are no different from those with lower count rates. It is therefore expected that the precision in measured Rn-220 concentrations is determined by counting statistics and not affected by the other factors to any significant extent. This is true for the measurements in the whole range, including those performed in the most humid conditions that can be present in Oncoinvent facility.

It is both reported in the literature [5-7] and were observed by us in the modelling experiments that extremely high humidity (over 80 %) can affect the measurements of radon concentration. But the correlation above confirms that the humidity that can be present in the ventilation of Oncoinvent does not affect the measurements of Rn-220 significantly.

4.2 Detection limit

Several approaches for determining the detection limit are possible.

4.2.1 Currie approach

When measuring radioactivity, the formulas of L.A. Currie [8] have become the consented approach. These formulas are also the fundament for the ISO standard 11929:2010 [9] for determination of decision threshold and detection limits. The formulas of Currie are all based on the variation in a zero signal. It is also conventional that detection limit should always be stated together with a level of confidence. At Oncoinvent both in the laboratories and outdoor, there will always be a background level of both Rn-220 and Rn-222. It is therefore hard to determine what a true variation of a zero signal from the instrument will be.

From 22. of December to 4. of January we used one of our RTM instruments; RTM6-0455, to measure the Rn-220 concentration in the inlet air. As a best approached to achieve a true blank we have therefore used this measurement. The dataset contains 77 datapoints all measured for 4 hours.

The average counts in the region for Rn-220 counts; ROI2, is 2.8 counts with a 1 σ variation of 1.9 count. The square root of 2.8 is 1.8, which means that either the uncertainty is larger than what just origin in counting statistics or the number of datapoints are too few for the variation value to converge with the theoretical. Dividing the average number of counts by four hours to get the counts in a one-hour measurement results in 0.7.

The lowest non-zero number of counts in one measuring period is 1 count. Curries formula states that the decision level or also called critical level (CL) is determined by the equation:

$$CL = k \times \ \sigma_0, \qquad 4.$$

where σ_0 is the variation in the zero signal,

k is a constant to determine the confidence level.

The value of *k* when choosing level of confidence is related to the standard normal distribution. If a confidence level is chosen so that only 5% (one in 20) zero signals will be regarded as a true value, *k* = NORMSINV(0.95) = 1.645. If k = 2, then the confidence level is 97.72 %, and only 2.28% of the zero signals will be falsely interpretated as a true value. By putting k equal to either 1.645 or 2, and σ_0 of $\sqrt{1}$ into equation 3 and rounding up to a whole number of counts *CL* = 2 is obtained.

Up to now, we did not subtract any background level from our Rn-220 measurements in the outgoing air. If we were to do that, the equation for critical limit would be translated to the following equation [10]:

$$CL = k \times \sqrt{R_b\left(\frac{1}{t_b} + \frac{1}{t_g}\right)}, \qquad 5.$$

where R_b is count rate of the background,

t_b is time of background measurement,

t_g is time of sample measurement.

Using background count rate R_b = (2.8/240), t_b = 240 minutes, t_g = 60 minutes, and k = 2, results into critical limit of 2.2. Rounding this result down tells us is that, with this set of data, the likelihood for 2 counts in a 60-minute measurement period to be a false positive is less than 2.3 %. Examining data sets from instruments RTM6-0033, RTM6-00332 and RTM6-00392, with low counting number we see that 2 counts in more than 98 % of the cases is regarded as a true value and denoted with a corresponding concentration [Bq/m^3]. In the residual 2 % cases, 2 counts are denoted with a less than value sign (< #) equivalent to a “below the detection limit” reading. The correlation between counts and Rn-220 concentration is determined as sensitivity (counting efficiency). Out of the seven instruments Oncoinvent holds, the lowest counting efficiency for Rn-220 is 1.56 cts/(min*kBq/m^3) and the best is 2.31 cts/(min*kBq/m^3). Corresponding critical levels are shown in table 1. The critical level must be interpreted as follows: if a measurement result below this limit (14-21 Bq/m^3) will occur, there is a higher probability for them to origin form false positive signals. The uncertainty at this concentration level is then 70% (1σ).

Table 1 – Background level evaluation (1 h, 97 % confidence level)

Measured background level	Counting efficiency / sensitivity	Critical level range
6 ± 3 Bq/m^3	1.56 cts/(min*kBq/m^3)	21 kBq/m^3
	2.31 cts/(min*kBq/m^3)	14 kBq/m^3

According to [8], detection limit (DL) is the true signal that may end up below the CL and be therefore mistakenly regarded as a zero signal. The likelihood for this to happen is determined by the following Currie equations:

$$DL[\text{counts}] = 3.29 \times \sigma_0, \text{ (95\% confidence)} \qquad 6.$$

$$DL[\text{counts}] = 4 \times \sigma_0, \text{ (97.72\% confidence)} \qquad 7.$$

Using the square root term in equation 4. This gives either 3.09 or 3.75 counts in 60 minutes. The transformation from counts to Rn-220 concentration than can be made dividing number of counts by measurement time and counting efficiency.

4.2.2 ICH approach

According to [11], detection limit is determined as follows:

$$DL[Bq/m^3] = \frac{3.3\times \sigma_0}{\mathrm{t}_g \times S}, \qquad 8.$$

where S is counting efficiency (see section 4.1)

t_g is time of sample measurement.

In [11] the equation comes without a certainty statement but is unmistakably similar to the equation from Currie with 95% confidence level.

4.2.3 SARAD approach

In the application note AN-002 [3]. SARAD gives a third way to calculate the detector’s detection limit based on measurement time, counting efficiency of an instrument and choosing the number of counts that will give the desired confidence level:

$$DL[kBq/m^3] = \frac{N}{T \cdot S}, \qquad 9.$$

where N is number of counts required to reach the deaired level of confidence, T is time of integration per measurement,

S is counting efficiency reported in the calibration certificate for each instrument.

Then for the measurement settings currently used in Oncoinvent T = 60 min and choosing N = 3 will give a 95 % confidence interval.

4.2.4 Detection limit calculations for Oncoinvent instruments

In table 2 the sensitivity (counting efficiency) values for Oncoinvent instruments are specified. The three approaches are then used to derive the detection limits.

Table 2 – Calculation of detection limit

Instrument Serial no.	Counting efficiency, [cts/(min*kBq/m³]	Detection limit, Bq/m³			
		SARAD, N = 3; 95%	ICH	Currie	
				N = 3; 95%	N = 4; 97.7%
RTM6-00331	1.780(±0,8%)	28	31	28	37
RTM6-00332	1.87(±0,7%)	27	29	27	36
RTM6-00333	1.56(±0,7%)	32	35	32	43
RTM6-00392	1.930(±0,7%)	26	28	26	35
RTM6-00470	2.31(±0,7%)	22	22	22	29

Table 2 shows that the approximate detection limit for the instruments used is 30 Bq/m^3.

These levels must be understood as true Rn-220 concentrations that only in one out of twenty (5%) cases will end up with less than two counts and therefore will be faultily determined as a zero signal, when measuring for 60 minutes. The uncertainty in concentration for reported concentrations of these amounts will be close to $\frac{\sqrt{3}}{3}$ which corresponds to 58% confidence at 1σ.

5 Conclusion regarding instrument fitness for the purpose

Requirement:	Delivery:
An instrument with a range for measuring Rn-220 from just above the background level to a few hundred kBq/m^3	The instrument is able to measure concentrations down to about 14-21 Bq/m^3 and has linear response up to 225 kBq/m^3.
The detection limit must be determined	The detection limit is determined for 1 h measurement as approx. 30 Bq/m^3 (95 % confidence level)

Requirement:	Delivery:
An instrument for continues measurement of Rn-220 concentration in the air of the ventilation duct.	The detection mode described in application note AN-004 [1] shows a selective detection of Rn-220 with fast response.

The overall conclusion is that the RTM-1688-2 instruments are well fit for the purpose.

6 References

[1] SARAD Application note 004 for use of RTM 1688-2 monitors: https://www.sarad.de/cms/media/docs/applikation/AN-004_Thoronmeasurements_EN_11-03-08.pdf
[2] Oncoinvent report: The effect of air humidity on Radon-220 release, corrective and preventative actions
[3] SARAD Application note 002 for use of RTM 1688-2 monitors: https://www.sarad.de/cms/media/docs/applikation/AN-002_MeasurementPrincipals-Statistics-TestPlanning_EN_09-05-12.pdf
[4] SARAD Technical Data on RTM 1688-2 monitors: https://www.sarad.de/cms/media/docs/datenblatt/Datasheet_RTM1688_2_EN_17-12-13.pdf
[5] Iimoto, T., et al. Journal of environmental radioactivity, 2005, 78.1: 69-76
[6] Dubčáková, R., et al. Radiation protection dosimetry, 2011, 145.2-3: 295-299.
[7] Sorimachi, A, et al. Radiation measurements, 2012, 47.6: 438-442.
[8] Currie, Lloyd A., Detection in Analytical Chemistry; Importance Theory, and Practice, ACS symposium series 361, 1988
[9] ISO 11929:2010: Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation — Fundamentals and application
[10] Ström, Daniel J., False Alarms, true Alarms and Statistics: Correct usage of Decision Level and Minimum Detectable Amount, Counting Education Lecture, Health Physics Society, Minneapolis, Minnesota, 1998
[11] ICH Expert Working Group. "Q2 (R1): validation of analytical procedures: text and methodology." Federal Register 60 (1995): 11260

7 Attachments

[A1] RTM6-00392 SARAD calibration certificate
[A2] RTM6-00392: raw data fragment
[A3] RTM6-00392: raw data from in-line measurements from 04 Aug 2021 to 29 Oct 2021
[A4] RTM6-00470: raw data from the inlet air measurements from 22 Dec 2021 to 04 Jan 2022
[A5] email correspondence SARAD: Christian Bartzsch 30. March 2021
[A6] email correspondence SARAD: Veikko Oeser, email 26.Jan.2022