Hellenic Nuclear
Physics Society

HNPS
Advances in
Nuclear Physics

Proceedings of the
30th Hellenic
Symposium on
Nuclear Physics
and Applications

University of Ioannina
October 7-8, 2022

Editors
N. Nicolis
C. Papachristodoulou
N. Patronis
C. Stamoulis
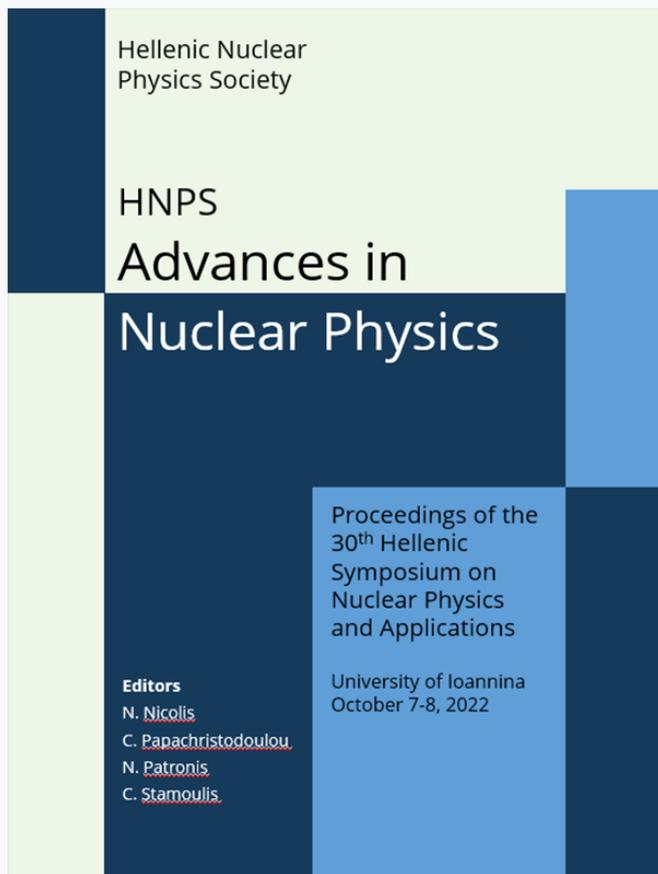
Measuring radon concentrations in air at the progeny non-secular equilibrium time frame

Nikolaos Petropoulos, Sotia Argyrou

Measuring radon concentrations in air at the progeny non-secular equilibrium time frame

S. Argyrou, N.P. Petropoulos[*]

Nuclear Engineering Laboratory, School of Mechanical Engineering, National Technical University of Athens, 15780 Athens, GREECE

Abstract The Nuclear Engineering Laboratory of NTUA (NEL-NTUA) regularly tests radon-in-air concentration measuring instruments. Most common instruments involve open ionization chambers, radon absorbing devices and grab sampling devices. Most types of such instruments do not provide a calibrated radon concentration as soon as the air sampling has ended, since there should exist an adequate time gap, within which sampled radon would reach secular equilibrium with some or most of its immediate progeny. This gap should be around 10-20 min for radon to equilibrate with the Po-218 nucleus, or it could be more than about 3 hours for radon to equilibrate with the Bi-214 / Po-214 progeny. However, if there could be organized calibration experiments at the progeny non-secular equilibrium state, there are some instrument cases, for which this time gap difficulty may be better understood and overcome. To this end, in this work, the Bateman differential equations for all gamma and alpha emitting radon daughters are solved for radon daughters activity at any given time between 0 min and 3 hours using a spreadsheet. This would allow for obtaining a calibration correction factor to be applied at any given time before equilibrium, resulting for radon concentration estimations without the limitations of waiting. Following this solution, the results were theoretically assessed in terms of applicability. Calibration experiments performed for ionization chambers, radon absorbents and Lucas Cells at various radon concentrations in air at the progeny non-secular equilibrium time frame corroborate the theoretical approach.

Keywords radon, radon progeny, radon measurement, secular equilibrium

INTRODUCTION

Radon in air concentration measurements using active instrumentation present peculiarities and could result in serious error, if conducted before acceptable secular equilibrium between radon and its progeny without a proper calibration. That is why most active instruments give credible results only after radon within their active volume (or chamber) gets into equilibrium with some of the progeny produced by radon decay. To better explain why, let us assume a radon measurement instrument with an active volume is 0.25 L. Further, let us also assume that the minimum detectable radon concentration in air should be 200 Bqm^{-3}, a value accepted in EU as the lowest radon action (i.e. warning) level. Air with such a radon concentration diffused or absorbed or even grabbed within this active volume is hardly measurable, since its disintegration rate within 0.25 L becomes only $(200/1000) \times 0.25 = 0.05$ Bq or 3 DPM. In addition, these miniscule DPMs are due to, difficult to detect, $-\alpha$ particles since radon is solely an $-\alpha$ emitter of 5.6 MeV particles. Moreover, usually the radon in the volume is blurred by an unknown quantity of its progeny, possibly infiltrating. Having these into mind most instruments try to block the passage of progeny into their volume or, if this is not possible allow time for this progeny to decay and measure radon in the volume after some reasonable time, thus allowing progeny from this radon decay to develop and add detectable Becquerels to the Becquerels of radon already in the instrument's chamber. Calculations show that unwanted progeny entering from the atmosphere decay to a minimum after a maximum of ~2.5 hours, while desirable progeny coming from the radon decay reach their maximum activity in 3 hours. Therefore, radon measurements conducted in less than three hours after radon entry in the

instrument's chamber are usually subject to questions, depending of course on the technology involved.

RADON PROGENY DECAY CONSIDERATIONS

Radon decays producing progeny following the decay chain below:

$$Rn-222 \rightarrow Po-218 \rightarrow Pb-214 \rightarrow Bi-214 \rightarrow Po-214$$
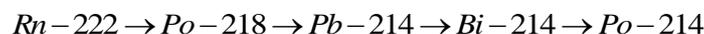
Other and more progeny could be available further down the chain, however, their half life is significantly greater, hence, they develop much later, significantly outside any reasonable time window adequate for timely measurements. The following Table 1 summarizes the properties of the progeny presented in the chain mentioned above.

Table 1. *Some properties of radon decay products*

Isotope	Half-life	Emitter mainly of:	Emitted energy	Time for approximately full decay	Time for acceptable equilibrium (*)
Po-218	~3.1 min	-α	~6 MeV	~20 min	~10 min
Pb-214	~27 min	-γ	352 keV et al.	~160 min	~120 min
Bi-214	~20 min	-γ	609 keV et al.	~120 min	~180 min
Po-214	~20 min	-α	~7.7 MeV	~120 min	~180 min

(*) after which the isotope concentration becomes greater than 0.90 the respective radon concentration

Assuming that each radon decay product of Table 1 infiltrates into the active volume of the assumed measuring instrument, not being in equilibrium with the incoming radon, at an unknown, yet same concentration of $C <= 200$ Bqm^{-3}, its decay is necessary, so that the 200 Bqm^{-3} of pure radon could be detected and measured without noise. Figure 1 depicts the time needed for their acceptable full decay both individually and as a whole. The matter becomes more complicated, if these products enter in the volume with different and still unknown concentrations. Yet, as a rule, following Fig. 1, the infiltrating products noise is considered negligible after the full decay of Pb-214, or after about 2.5 hours. At this time the sum of all progeny has effectively decayed no matter what their initial activity was. During the 2.5 hours of noisy progeny decay the radon activity is hardly affected.
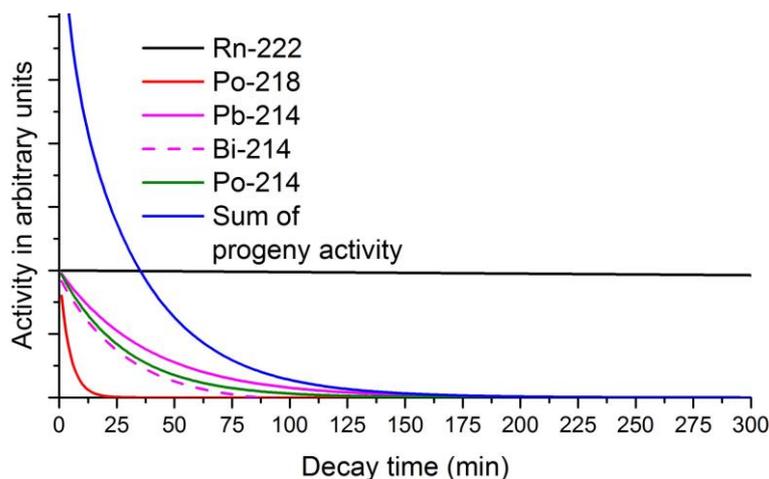

Figure 1. *Decay times of radon progeny with arbitrary initial activity*

RADON DETECTION MODES

Opposite to the infiltrating radon progeny, the progeny growth following the decay of radon in the active volume is well known and well desired. Especially if this progeny reaches secular

equilibrium certain radon measurement modes could be accomplished. These modes could be mainly four, as it follows:

MODE (1): Measurement using the –α of Rn-222 and Po-218. This mode aims to double the DPM available from the initial radon concentration and is feasibly applied after a 10 min growth of new progeny and a concurrent 20 min decay of existing progeny. Such an instrument operates correctly after about 15 – 20 min.

MODE (2): Measurement using the photons of Pb-214 and Bi-214. This scenario doubles the DPM available from the initial radon (since radon would not emit photons) and is feasibly applied after ~3 hours of new progeny growth. Its performance in less than 3 hours (out of equilibrium) depends on corrected smaller calibration factors.

MODE (3): Measurement using the –α of Rn-222, Po-218 and Po-214. This scenario triples the DPM available from the initial radon and is feasibly applied after a 120 min decay of existing progeny and a concurrent 180 min growth of new progeny. Such an instrument operates correctly after ~3 hours. Its operation in less than 3 hours (out of equilibrium) depends on corrected smaller calibration factors.

MODE (4): All modes, when applied, get significantly simpler, if the instruments active volume is protected against entry of unknown progeny using special filters; and this is the usual case.

As well known, the instruments following MODE (1) + (4) are mostly those employing an open ionization chamber, a prominent example is the *AlphaGuard* by Bertin GmbH. Most adsorption methods, like those using charcoal follow MODE (2) + (4). Any commercially available grab sampling *Lucas Cell* follows MODE (3) + (4).

BATEMAN EQUATIONS SOLUTION

The Bateman radon progeny growth differential equations, as approximately solved using any spreadsheet as in [4], may result in Figs. 2 and 3 graphically indicating time to equilibrium for –α emitting and photon emitting decay products respectively for any initial radon concentration. This spreadsheet solution presents a maximum deviation of about 4% from the expected progeny activity. Both figures corroborate the application of all MODE (1) + (4), MODE (2) + (4) and MODE (3) + (4) measurement scenarios as described in the previous section.
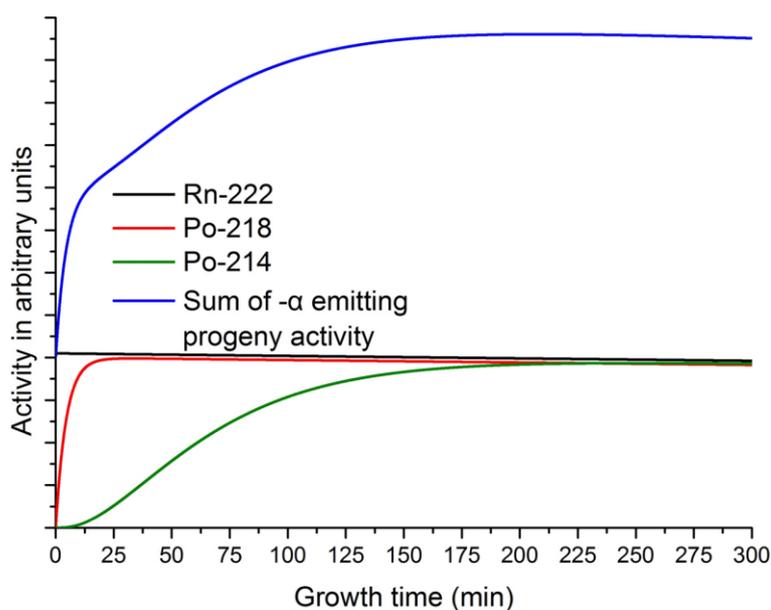

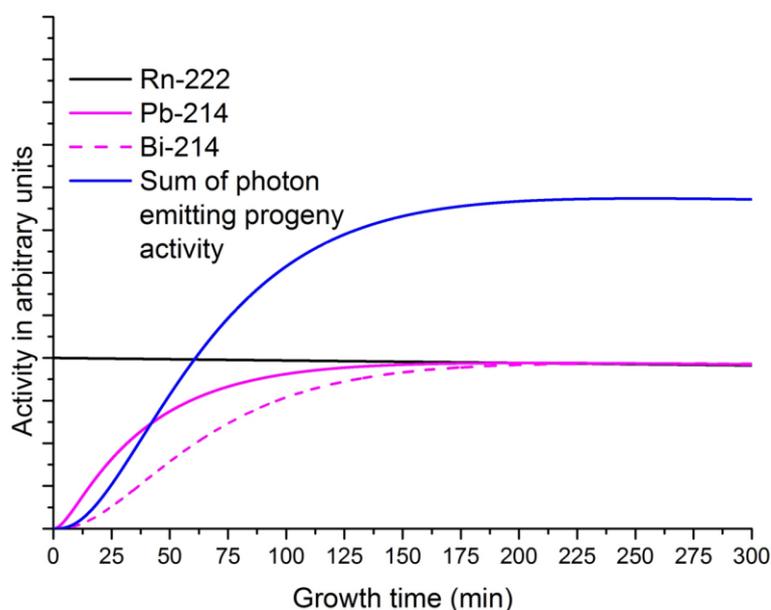**Figure 2**. *Growth times of –α emitting radon progeny with arbitrary initial activity*

Figure 3. *Growth times of photon emitting radon progeny with arbitrary initial activity*

DISCUSSION AND OUTCOMES

Instruments following MODE (1) + (4) measure radon correctly within reasonably small time after air sampling in their active volume. Therefore, no actual calibration factor correction is necessary in case of non-secular equilibrium. Instruments following MODE (2) + (4), see [1], will measure erroneously before equilibrium is reached, unless some calibration factor correction is applied to the calibration factor of measurements under equilibrium. The calibration correction in this case is close to be exponential in terms of time, however the exponential time growth is conveniently slow. Similar applies for instruments operating in MODE (3) + (4), e.g. the Lucas Cell as in [2]. Nevertheless, the calibration correction in this case is not easily applicable, given that correction factor does not depend smoothly on progeny growth time. Figure 4 presents the estimated correction factors for the calibration factor under equilibrium vs time for all considered modes. The results of own studies concerning absorbents and found in [3] and recently concerning Lucas Cells in [4] connect well to the theoretical assessment within this work.
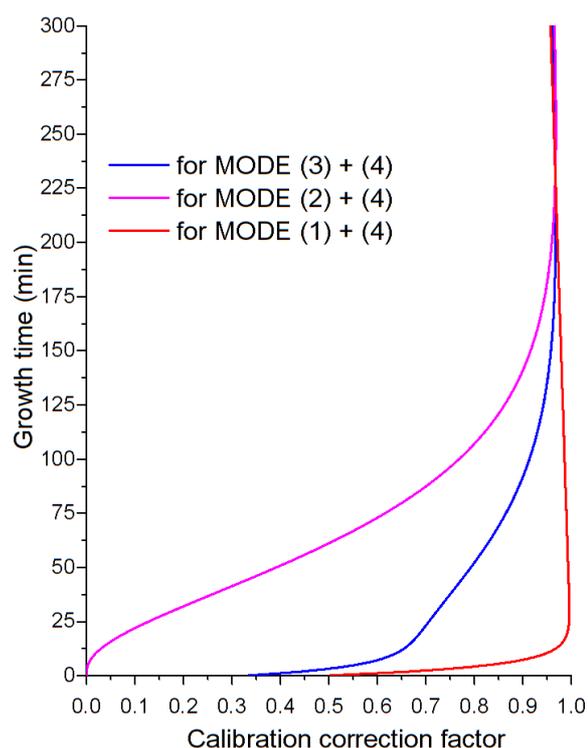

Figure 4. *Calibration correction factor within the non-secular equilibrium time frame*

References

[1] A. George, Health Physics 46(4), p. 867 (1984).
[2] H. Lucas, The Review of Scientific Instruments, 28(9), p. 680 (1957)
[3] J. Paschalides et al., Applied Radiation Isotopes, 68, p. 155 (2010)
     https://doi.org/10.1016/j.apradiso.2009.08.017
[4] S. Argyrou, Diploma Dissertation, NEL-NTUA (2022) http://dx.doi.org/10.26240/heal.ntua.23973