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National Institute for Occupational Safety and Health

SC&A's Evaluation of ORAUT-RPRT-0084, "Two-Count Filter Method for Measurement of Thoron Progeny in Air"

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Abbreviations and Acronyms

ABRWH, Board	Advisory Board on Radiation and Worker Health
Bi	bismuth
hr	hour
L	liter
min	minute
NIOSH	National Institute for Occupational Safety and Health
nsec	nanosecond
ORAUT	Oak Ridge Associated Universities Team
Pb	lead
pCi	picocurie
Po	polonium
Rn	radon
sec	second
Th	thorium
thoron	^{220}Rn
Tl	thallium
WLM	working level months

1 Introduction and Background

The Advisory Board on Radiation and Worker Health tasked SC&A with a technical review of ORAUT-RPRT-0084, revision 00, "Two-Count Filter Method for Measurement of Thoron Progeny in Air," issued March 27, 2017 (ORAUT, 2017; "RPRT-0084"). In RPRT-0084, the National Institute for Occupational Safety and Health (NIOSH) evaluates a methodology to calculate the internal dose to lungs from inhalation of thoron (^{220}Rn) and its progeny using bismuth-212 (^{212}Bi) and lead-212 (^{212}Pb). Thoron is a member of the thorium-232 (^{232}Th) decay chain, whereas ^{212}Bi and ^{212}Pb are members of the thoron decay chain. ^{212}Bi and ^{212}Pb are the primary radionuclides of interest when calculating the internal dose from inhalation of thoron and its progeny due to their relatively long half-lives when compared with thoron reaching equilibrium.

This report presents SC&A's evaluation of the technical approach NIOSH used in RPRT-0084 to estimate ^{212}Pb concentration in the ambient air using the two-count filter method as follows: (1) pull air through a filter for 6 hours and measure the total alpha activity on the filter after the pump stops, and (2) pull air through the same filter for an additional 18 hours and measure the total alpha activity on the filter after the pump stops. These two measured alpha activities are then used to calculate the concentration of ^{212}Pb in the air when long-lived alpha emitters and ^{222}Rn progeny are also present in the air.

Section two of this report outlines the forward and reverse problem approaches NIOSH used in RPRT-0084 to calculate ^{212}Pb concentration in air.

2 Overview of ORAUT-RPRT-0084

This section briefly summarizes RPRT-0084 to develop an understanding of the two-fold approach NIOSH presented to calculate the concentration of ^{212}Pb in air to indirectly calculate the internal dose from inhalation of thoron and its progeny.

2.1 Introduction (RPRT-0084 section 1.0)

As noted in section 1, thoron (^{220}Rn) is a member of the ^{232}Th decay chain, whereas ^{212}Bi and ^{212}Pb are members of the ^{220}Rn decay chain. Members of the ^{220}Rn decay chain are as follows (NIOSH, 2018):

1. ^{220}Rn: 55.6 sec, alpha decay
2. ^{216}Po: 0.15 sec, alpha decay
3. ^{212}Pb: 10.64 hrs, beta decay
4. ^{212}Bi: 60.6 min, alpha decay (36 percent), beta decay (64 percent)
5. ^{212}Po: 304 nsec, alpha decay
6. ^{208}Tl: 3 min, beta decay

The half-lives of polonium-216 (^{216}Po) and polonium-212 (^{212}Po) alpha emitters are very short and as such do not contribute to the working level calculation. Additionally, these alpha emitters represent an insignificant dose due to only a small number of atoms inhaled. Thallium-208

(^{208}Tl) is a beta emitter and has a relatively short half-life. Thus, ^{212}Bi and ^{212}Pb are the primary radionuclides of interest when calculating the internal dose from inhalation of thoron and its progeny.

An equation is derived for the two-count filter method in RPRT-0084 using the following two-fold approach (ORAUT, 2017, p. 6):

1. **Forward problem approach:** "Calculate the total alpha activity expected to be on the filter paper when it is counted at 6 and 24 hours after the air sampler pump is turned off" using "known concentrations of ^{212}Pb, ^{212}Bi, and a long-lived alpha emitter in the air."

2. **Reverse problem approach:** Derive an equation to calculate the concentration of ^{212}Pb in ambient air using the total alpha activity determined in the forward problem approach on the filter paper at 6- and 24-hour intervals after the air sampler is turned off.

If the concentration of ^{212}Pb in air calculated using the equation (13) (equation 3-7 in RPRT-0084), shown in section 2.3 of the review, derived in the reverse problem approach gives the same ^{212}Pb air concentration determined using the forward problem approach, then the equation is deemed to be correct.

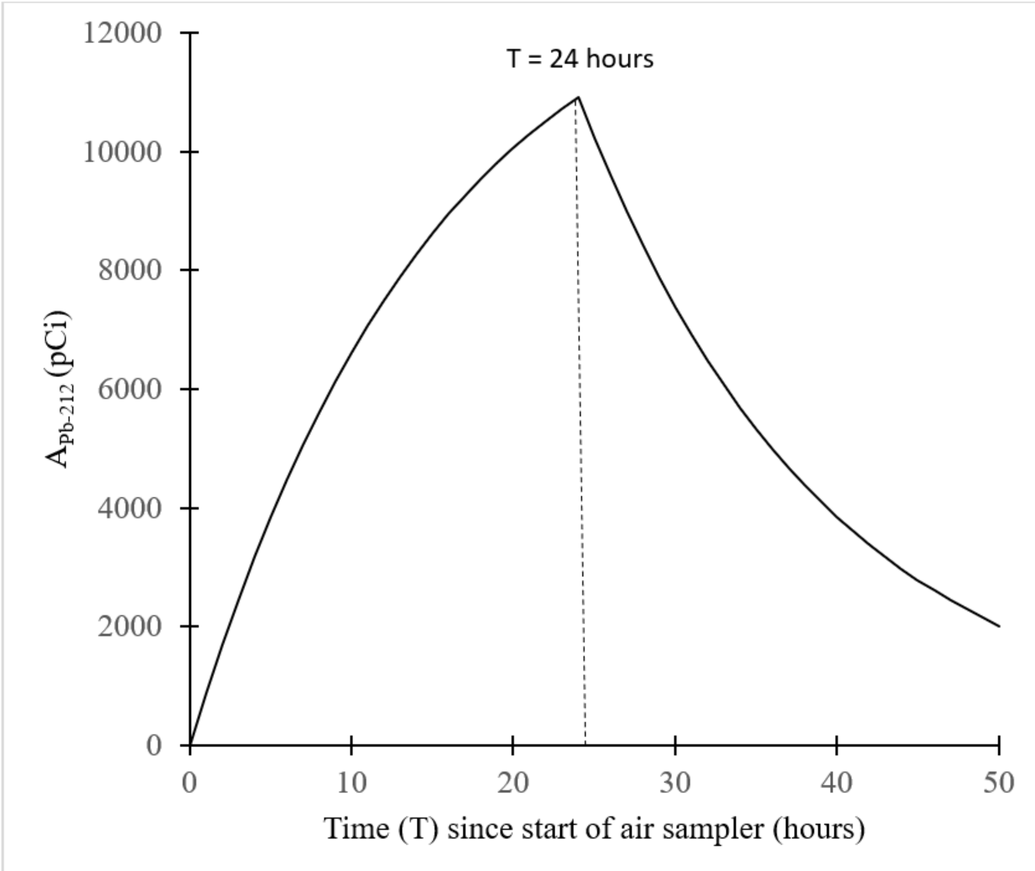
2.2 The forward problem approach (RPRT-0084 section 2.0, pp. 6–9)

The concentration of the source term is assumed to be a constant stream of ^{212}Pb and ^{212}Bi released into the air sampler. The forward problem approach calculates the total alpha activity on the filter at 6 and 24 hours after the sampler pump is turned off, respectively. If A_{Pb0} is the initial ^{212}Pb activity on the filter at the time T when the sampler pump is turned off, then per equation (1) (equation 2-1 in RPRT-0084),

$$A_{\mathrm{Pb0}} = \frac{\mathrm{C_{Pb}F}}{\lambda_{\mathrm{Pb}}}\left(1 - \mathrm{e}^{-\lambda_{\mathrm{Pb}}\mathrm{T}}\right) \tag{1}$$

where C_{Pb} is the ^{212}Pb concentration in air going into the sampler, which NIOSH assumed to be 1 picocurie per liter (pCi/L); *F* is the flow rate of the sampler, which is 15 L/minute (min) = 900 L/hour (hr); λ_{Pb} is the decay constant for ^{212}Pb, which is 0.06513/hr; and T is the length of time the pump is on, which is 24 hours. Figure 1 shows ^{212}Pb activity on a filter paper as a function of time (T). The horizontal axis runs from 0 to 50 hours, and the vertical axis runs from 0 to 12,000 pCi of ^{212}Pb. As expected, the ^{212}Pb activity builds exponentially from the time the air pump is turned on to the time it is turned off at 24 hours, and then decays exponentially.

Figure 1. Initial deposition of ^{212}PB (A_{Pb0}) on the filter paper at T = 24 hours



Substituting the parameters in the last paragraph into equation (1) (equation 2-1 in RPRT-0084) gives the initial deposition of ^{212}Pb on the filter paper, $A_{Pb0} = 1.0924 \times 10^4$ pCi. The decayed ^{212}Pb activities on the filter after the pump stops are then calculated at 6 and 24 hours (A_{Pb6} and A_{Pb24}) using the following exponential decay equations (2) and (3) (master equations 2-2 and 2-3 in RPRT-0084):

$$A_{Pb6} = A_{Pb0}e^{-\lambda pb\times 6} = 7390.4 \text{ pCi} \tag{2}$$

$$A_{Pb24} = A_{Pb0}e^{-\lambda pb\times 24} = 2288.4 \text{ pCi} \tag{3}$$

A_{Pb6} and A_{Pb24} correspond to ^{212}Pb activities on the filter at T = 30 hours and T = 48 hours, respectively, in figure 1. ^{212}Bi activities are assumed to be in transient equilibrium with the ^{212}Pb activities after the pump stops and when ^{212}Pb activities were determined at 6 and 24 hours. That is, the two-filter method with counts at 6 and 24 hours after the pump stops is only dependent on the concentration of that thoron progeny and insensitive to the concentration of ^{212}Bi in the air. Thus, the activities of ^{212}Bi at 6 and 24 hours after the pump stops can be determined from A_{Pb6} and A_{Pb24} activities, respectively, by multiplying them with ^{212}Bi to ^{212}Pb ratio using the following equation (4) (equation 2-5 in RPRT-0084):

$$r = \frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \tag{4}$$

where r is the ^{212}Bi to ^{212}Pb ratio, λ_{Bi} is the decay constant for ^{212}Bi, which is 0.6869/hr. Substituting λ_{Pb} and λ_{Bi} into equation (4) yields r = 1.105. Therefore, the ^{212}Bi activities on the filter at 6 and 24 hours (A_{Bi6} and A_{Bi24}) are as shown in equations (5) and (6) (master equations 2-6 and 2-7 in RPRT-0084):

$$A_{Bi6} = rA_{Pb6} = 1.105 \times 7390.4 \text{ pCi} = 8166.4 \text{ pCi} \tag{5}$$

$$A_{Bi24} = rA_{Pb24} = 1.105 \times 2288.4 \text{ pCi} = 2528.7 \text{ pCi} \tag{6}$$

Only A_{Bi6} and A_{Bi24} are used in determining total alpha activities because ^{212}Bi emits alpha particles, as noted in section 2.1. The total alpha activities on the filter at 6 and 24 hours (A_6 and A_{24}) are as shown in equations (7) and (8) (equations 2-8 and 2-9 in RPRT-0084):

$$A_6 = A_{Bi6} + A_\alpha \tag{7}$$

$$A_{24} = A_{Bi24} + A_\alpha \tag{8}$$

where A_α in equations (7) and (8) (equations 2-8 and 2-9 in RPRT-0084) is the activity from a long-lived alpha emitter on the filter. NIOSH emphasized that A_α is not a parameter of interest in this report. However, for the benchmark problem, NIOSH assumed that A_α accumulated into the filter in 24 hours is per equation (9) (equation 2-10 in RPRT-0084):

$$A_\alpha = \left(0.05\,\frac{\text{pCi}}{\text{L}}\right)\left(15\,\frac{\text{L}}{\text{min}}\right)(24\text{ hr})\left(\frac{60\text{ min}}{\text{hr}}\right) = 1080\text{ pCi} \tag{9}$$

Hence, the total alpha activities on the filter at 6 and 24 hours (A_6 and A_{24}) after the pump is stopped are determined by substituting parameters A_{Bi6}, A_{Bi24}, and A_α into equations (7) and (8) (equations 2-8 and 2-9 in RPRT-0084) as shown in equations (10) and (11):

$$A_6 = 8166.4 \text{ pCi} + 1080 \text{ pCi} = 9246.4 \text{ pCi} \tag{10}$$

$$A_{24} = 2528.7 \text{ pCi} + 1080 \text{ pCi} = 3608.7 \text{ pCi} \tag{11}$$

The alpha activities determined in equations (10) and (11) will be used as input parameters into an equation (13) (equation 3-7 in RPRT-0084) derived by NIOSH in the next section that returns the concentration of ^{212}Pb in the air. The equation derived is deemed to be correct if the ^{212}Pb concentration of 1 pCi/L is returned.

2.3 The reverse problem approach (RPRT-0084 section 2.0)

The total alpha activities determined on the filter at 6 and 24 hours (A_6 and A_{24}) after the pump is turned off are used in the reverse problem approach to calculate the concentration of ^{212}Pb in the air. To begin with deriving the final equation for the reverse problem approach, equations (7) and (8) (equations 2-8 and 2-9 in RPRT-0084) are first subtracted from each other. After substituting

A_{Bi6}, A_{Bi24}, A_{Pb6}, and A_{Pb24} values and rearranging the subtracted equation, it can be concluded as shown in equation (12) (equation 3-4 in RPRT-0084):

$$A_{Pb0} = \frac{A_{24} - A_6}{r\left(e^{-\lambda_{Pb} \times 24} - e^{-\lambda_{Pb} \times 6}\right)} \tag{12}$$

Equating the right-hand sides of equations (1) (equation 2-1 in RPRT-0084) and (12) (equation 3-4 in RPRT-0084), the final equation can be solved to calculate the ^{212}Pb concentration (C_{Pb}) in the sampled air as shown in equation (13) (equation 3-7 in RPRT-0084):

$$C_{Pb} = \left[\frac{\lambda_{Bi} - \lambda_{Pb}}{\lambda_{Bi}}\right]\left[\frac{A_{24} - A_6}{\left(e^{-\lambda_{Pb} 24} - e^{-\lambda_{Pb} \times 6}\right)}\right]\left[\frac{\lambda_{Pb}}{F\left(1 - e^{-\lambda_{Pb} \times T}\right)}\right] \tag{13}$$

After plugging parameters into equation (13) (equation 3-7 in RPRT-0084) when the pump is turned off (i.e., T = 24 hours), the result is C_{Pb} = 1 pCi/L, which confirms that the derived equation is applicable to calculating the ^{212}Pb concentration in the sampled air.

3 SC&A's Evaluation of ORAUT-RPRT-0084

This section summarizes SC&A's evaluation of the approach used by NIOSH in developing RPRT-0084.

3.1 Evaluation of NIOSH's approach to the two-count filter method

SC&A did not identify any issues with the general NIOSH approach in RPRT-0084 to evaluate the two-count filter method for measurement of thoron progeny in air.

3.2 Evaluation of NIOSH's analytical methods

SC&A evaluated the two-count filter method employed by NIOSH in RPRT-0084 to analytically estimate the ^{212}Pb concentration in the ambient air. ^{212}Bi and ^{212}Pb are the primary radionuclides of interest when calculating the internal dose from inhalation of thoron and its progeny. In the two-counter filter method NIOSH employed, SC&A evaluated both the forward and reverse problem approaches to estimate ^{212}Pb concentration in the ambient air.

Even though NIOSH determined a constant long-lived alpha emitter A_α = 1,080 pCi on the filter in 24 hours in equation (9) (equation 2-10 in RPRT-0084) to estimate the total alpha activities on the filter at 6 and 24 hours (A_6 and A_{24}, respectively) after the pump is stopped, A_α is not a parameter of interest in this report. That is, A_α is an independent variable when calculating ^{212}Pb concentration in the ambient air. SC&A verified this by substituting A_{Bi6} and A_{Bi24} determined from equations (5) and (6) (master equations 2-6 and 2-7 in RPRT-0084), respectively, and A_α = 0 into equations (7) and (8) (equations 2-8 and 2-9 in RPRT-0084), as shown in equations (14) and (15):

$$A_6 = 8166.4 \text{ pCi} + 0 \text{ pCi} = 8166.4 \text{ pCi} \tag{14}$$

$$A_{24} = 2528.7 \text{ pCi} + 0 \text{ pCi} = 2528.7 \text{ pCi} \tag{15}$$

After plugging equations (14) and (15) parameters (A_6 and A_{24}, respectively) into equation (13) (equation 3-7 in RPRT-0084) when the pump is turned off (i.e., T = 24 hours), the result still remained C_{Pb} = 1 pCi/L. This additionally confirmed that equation (13) (equation 3-7 in RPRT-0084) is applicable to calculating the ^{212}Pb concentration in the sampled air.

4 Summary and Conclusions

SC&A found the approach used to develop a sampling plan to be reasonable and technically correct.

SC&A found the analytical methods used in the forward and reverse problem approaches to be acceptable. In section 3.2, SC&A provided some expanded discussion concerning the effects that changes in variable parameters could have on the results.

SC&A did not identify any documentation issues that would affect the readability or application of the two-count filter method.

5 References

National Institute for Occupational Safety and Health. (2018). *Dose conversion factors for Radon WLM* (DCAS-TIB-011, rev. 05). https://www.cdc.gov/niosh/ocas/pdfs/tibs/dc-t11-r5-508.pdf

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