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Two-Count Filter Method for Measurement of Thoron Progeny in Air	ORAUT-RPRT-0084	Rev. 00
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03/27/2017	00	New document initiated to describe a two-count filter method for measurement of thoron progeny in air. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Thomas R. LaBone.

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ACRONYMS AND ABBREVIATIONS

GKE	general kinetics equation
hr	hour
L	liter
min	minute
NIOSH	National Institute for Occupational Safety and Health
ORAU	Oak Ridge Associated Universities
pCi	picocurie
SRDB Ref ID	Site Research Database Reference Identification (number)

1.0 INTRODUCTION

Thoron (^{220}Rn) is an inert radioactive gas that is a member of the thorium (^{232}Th) decay chain as shown in Figure 1-1.

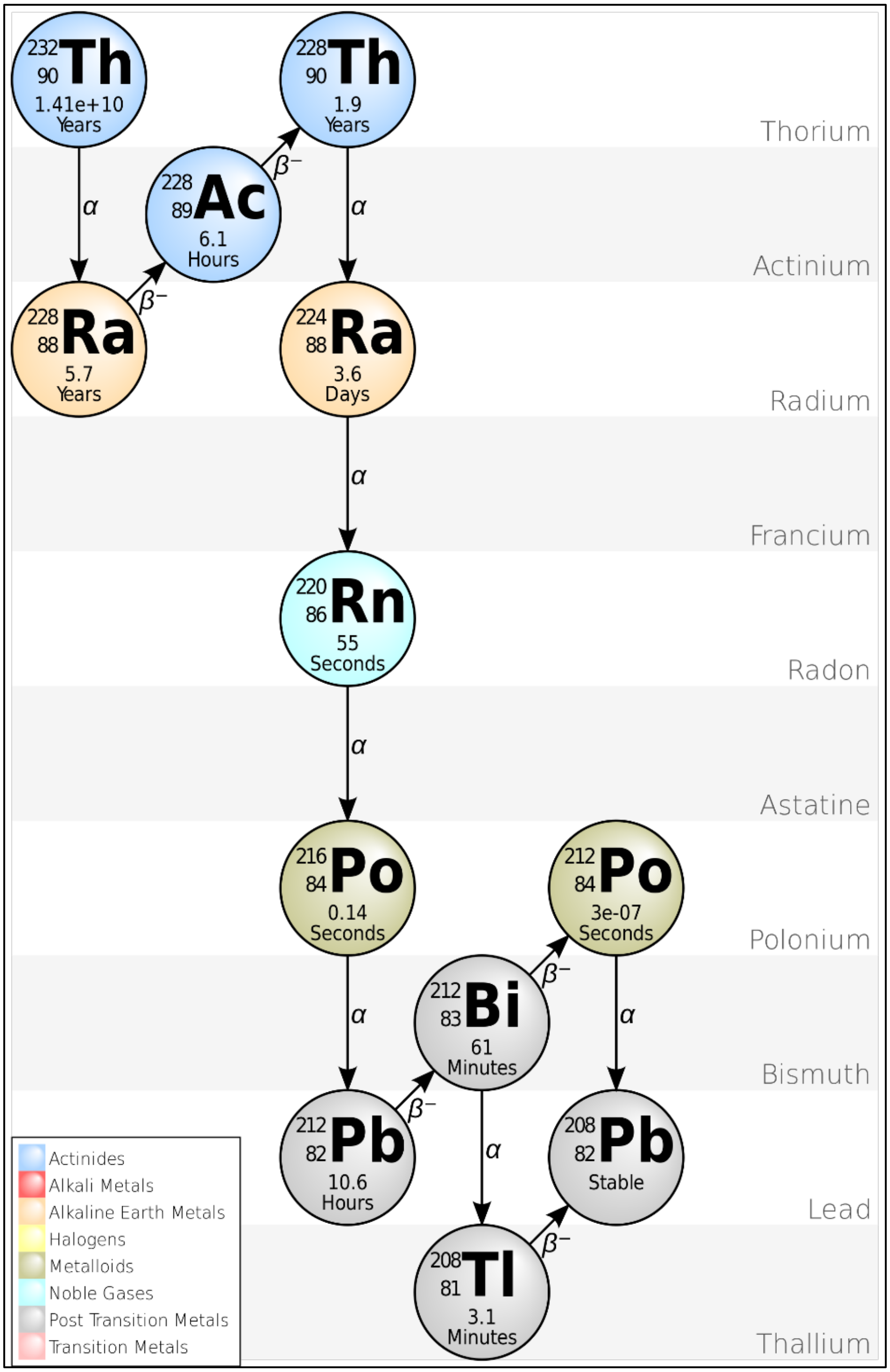

Figure 1-1. ^{232}Th decay chain (Wikipedia 2017).

As discussed in DCAS-TIB-011, *Lung Dose Conversion Factors for Thoron WLM* (NIOSH 2013), ^{212}Bi and ^{212}Pb are the primary radionuclides of interest when calculating the internal dose from inhalation of thoron and its progeny. The two-count filter method can be used to estimate the concentration of

^{212}Pb in ambient air. The method consists of pulling air through a filter for a prescribed length of time and measuring the total alpha activity on the filter at 6 hours after the pump stops and again at 24 hours after the pump stops. Given these two alpha activities, the concentration of ^{212}Pb in the air can be calculated when long-lived alpha emitters and radon (^{222}Rn) progeny[1] are also present in the air.

The goal here is to derive an equation for the two-count filter method. The approach is two-fold:

1. Start with known concentrations of ^{212}Pb, ^{212}Bi, and a long-lived alpha emitter in the air and from this 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. This is referred to as the "forward problem," and its end product is a benchmark solution.

2. Work back from the total alpha activity on the filter at 6 and 24 hours and derive an equation that gives the concentration of ^{212}Pb in the air. This is referred to as the "reverse problem."

If the equation derived in the reverse problem gives the same ^{212}Pb air concentration as used in the forward problem, the equation is deemed to be correct.

In summary, the primary purpose of this report is to provide the technical basis for Equation 3-7, which is used to estimate the concentration of ^{212}Pb in air given alpha counts of the air filter at 6 and 24 hours after the filter pump is stopped.

2.0 THE FORWARD PROBLEM

The source term is assumed to be a constant stream of ^{212}Pb and ^{212}Bi into the air sampler, and the concentration of each radionuclide is assumed to be 1 pCi/L. The goal is to calculate total alpha activity that will be on the filter at 6 and 24 hours after the sampler pump is turned off. Terms used in the calculations are:

F = flow rate of sampler (15 L/min = 900 L/hr)
T = length of time pump is run (24 hr)
λ_{pb} = decay constant for ^{212}Pb (0.06513/hr)
λ_{bi} = decay constant for ^{212}Bi (0.6869/hr)
C_{pb} = ^{212}Pb concentration in the air going into the sampler (1 pCi/L)
t = variable time (hr)
dt = infinitesimally short time increment (hr)

As shown in Figure 2-1, at time t a very small amount of ^{212}Pb (the product of C_{pb}, *F, and dt*) has deposited on the filter. Between times t and T when the pump is turned off, this small amount of ^{212}Pb will have decayed to ^{212}Bi. Equation 2-1 sums the small depositions that take place at all times

1 The radon progeny on the filter are assumed to have completely decayed away by the time the filter is counted for the first time.

between the time the pump is started and the time it is turned off to give the ^{212}Pb activity A_{pb0} on the filter at time T:

$$\begin{aligned} A_{pb0} &= \int_{t=0}^{t=T} \left[C_{pb} F dt \right] \left[e^{-\lambda_{pb}(T-t)} \right] \\ &= C_{pb} F e^{-\lambda_{pb} T} \int_{t=0}^{t=T} e^{\lambda_{pb} t} dt \\ &= \frac{C_{pb} F}{\lambda_{pb}} \left(1 - e^{-\lambda_{pb} T} \right) \end{aligned} \tag{2-1}$$

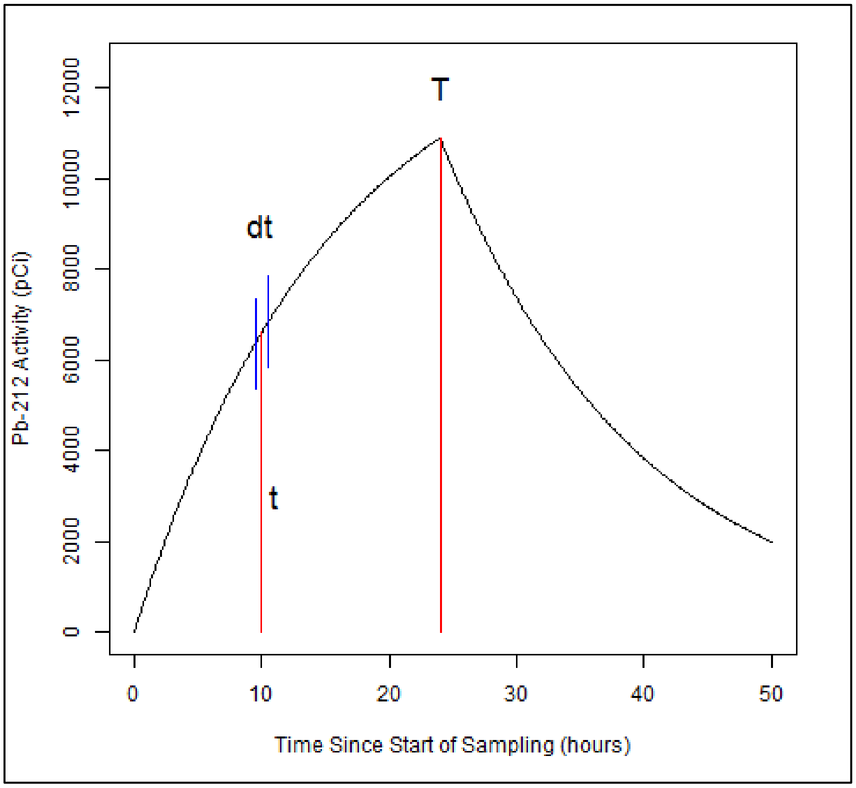

Figure 2-1. Deposition of ^{212}Pb (A_{pb0}) on the air filter.

Substituting the values of the parameters into Equation 2-1[2] gives the activity of ^{212}Pb on the filter when the pump stops:

$$A_{pb0} = 1.092 \times 10^4 \text{ pCi} \tag{2-2}$$

[2] In the numerical examples given in this paper the conversion of units is implicit, i.e., unit conversion constants have been omitted for clarity.

The ^{212}Pb activities on the filter at 6 and 24 hours after the pump stops are:

$$A_{pb6} = A_{pb0} e^{-\lambda_{pb} 6} = \frac{C_{pb}F}{\lambda_{pb}}\left(1 - e^{-\lambda_{pb}T}\right)e^{-\lambda_{pb} 6} = \frac{C_{pb}F}{\lambda_{pb}}\left(e^{-\lambda_{pb} 6} - e^{-\lambda_{pb}(T+6)}\right) \quad (2\text{-}3)$$

$$A_{pb24} = A_{pb0} e^{-\lambda_{pb} 24} = \frac{C_{pb}F}{\lambda_{pb}}\left(1 - e^{-\lambda_{pb}T}\right)e^{-\lambda_{pb} 24} = \frac{C_{pb}F}{\lambda_{pb}}\left(e^{-\lambda_{pb} 24} - e^{-\lambda_{pb}(T+24)}\right) \quad (2\text{-}4)$$

At the times of the 6-hour and 24-hour counts, the ^{212}Bi activities are assumed to be in transient equilibrium with the ^{212}Pb activities, i.e., the unsupported ^{212}Bi on the filter is insignificant in comparison with the activity of supported ^{212}Bi and can be ignored in this application. Another way to state this is that the two-filter method with counts at 6-hours and 24-hours after the pump stops is insensitive to the concentration of ^{212}Bi in the air and cannot provide information on the concentration of that thoron progeny.

Therefore, the ratios of ^{212}Bi to ^{212}Pb activity on the filter at these times is:

$$r = \frac{\lambda_{bi}}{\lambda_{bi} - \lambda_{pb}} = 1.105 \quad (2\text{-}5)$$

The ^{212}Bi activities on the filter at 6 and 24 hours are therefore:

$$A_{bi6} = rA_{pb6} = \left(\frac{\lambda_{bi}}{\lambda_{bi} - \lambda_{pb}}\right)\left(\frac{C_{pb}F}{\lambda_{pb}}\right)\left(e^{-\lambda_{pb} 6} - e^{-\lambda_{pb}(T+6)}\right) \quad (2\text{-}6)$$

$$A_{bi24} = rA_{pb24} = \left(\frac{\lambda_{bi}}{\lambda_{bi} - \lambda_{pb}}\right)\left(\frac{C_{pb}F}{\lambda_{pb}}\right)\left(e^{-\lambda_{pb} 24} - e^{-\lambda_{pb}(T+24)}\right) \quad (2\text{-}7)$$

Because the alpha particles are emitted by ^{212}Bi (not ^{212}Pb), the total alpha activities on the filter at 6 and 24 hours are:

$$A_6 = A_{bi6} + A_\alpha \quad (2\text{-}8)$$

$$A_{24} = A_{bi24} + A_\alpha \quad (2\text{-}9)$$

where A_α is the activity from a long-lived alpha emitter on the filter, which is not a parameter of interest in this application[3].

For the benchmark problem, it is assumed that:

$$A_\alpha = (0.05 \text{ pCi/L})(15 \text{ L/min})(24 \text{ hr})\left(\frac{60 \text{ min}}{\text{hr}}\right) = 1{,}080 \text{ pCi} \tag{2-10}$$

Therefore, the total alpha activities on the filter at 6 and 24 hours after the pump is stopped are:

$$A_6 = \left(\frac{\lambda_{bi}}{\lambda_{bi} - \lambda_{pb}}\right)\left(\frac{C_{pb}F}{\lambda_{pb}}\right)\left(e^{-\lambda_{pb}6} - e^{-\lambda_{pb}(T+6)}\right) + A_\alpha = 9{,}245 \text{ pCi} \tag{2-11}$$

$$A_{24} = \left(\frac{\lambda_{bi}}{\lambda_{bi} - \lambda_{pb}}\right)\left(\frac{C_{pb}F}{\lambda_{pb}}\right)\left(e^{-\lambda_{pb}24} - e^{-\lambda_{pb}(T+24)}\right) + A_\alpha = 3{,}608 \text{ pCi} \tag{2-12}$$

Ultimately, an equation will be derived that takes these alpha activities and returns the concentration of ^{212}Pb in the air. If a concentration of 1 pCi/L is returned, it can be taken as evidence that the equation faithfully implements the model and that the math has been done correctly.

3.0 THE REVERSE PROBLEM

In the reverse problem, the total alpha activity on the filter at 6 and 24 hours after the pump is turned off is known, and the goal is to calculate the concentration of ^{212}Pb in the air. From Equations 2-8 and 2-9, it can be concluded that:

$$A_{24} - A_{bi24} = A_6 - A_{bi6} \tag{3-1}$$

Then

$$A_{24} - rA_{pb24} = A_6 - rA_{pb6} \tag{3-2}$$

Note that the activity of the long-lived alpha emitter drops out of the equations as desired. This relationship can be rearranged to produce:

$$\begin{aligned} A_{24} - A_6 &= r\left(A_{pb24} - A_{pb6}\right) \\ &= r\left(A_{pb0}e^{-\lambda_{pb}24} - A_{pb0}e^{-\lambda_{pb}6}\right) \end{aligned} \tag{3-3}$$

Then

$$A_{pb0} = \frac{A_{24} - A_6}{r\left(e^{-\lambda_{pb}24} - e^{-\lambda_{pb}6}\right)} \tag{3-4}$$

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[3] At some facilities there was interest in the long-lived alpha emitter, and the two-count method was used in the opposite way to strip out thoron background.

From Equation 2-1 it is known that:

$$A_{\mathrm{pb0}} = \frac{C_{pb}F}{\lambda_{\mathrm{pb}}}\left(1 - e^{-\lambda_{\mathrm{pb}}T}\right) \qquad (3\text{-}5)$$

Equating the right-hand sides of Equations 2-1 and 3-4 gives:

$$\frac{A_{24} - A_6}{r\left(e^{-\lambda_{\mathrm{pb}}24} - e^{-\lambda_{\mathrm{pb}}6}\right)} = \frac{C_{pb}F}{\lambda_{\mathrm{pb}}}\left(1 - e^{-\lambda_{\mathrm{pb}}T}\right) \qquad (3\text{-}6)$$

which can be solved for C_{pb}, the ^{212}Pb concentration in the sampled air:

$$C_{pb} = \left[\frac{\lambda_{\mathrm{bi}} - \lambda_{\mathrm{pb}}}{\lambda_{\mathrm{bi}}}\right]\left[\frac{A_{24} - A_6}{e^{-\lambda_{\mathrm{pb}}24} - e^{-\lambda_{\mathrm{pb}}6}}\right]\left[\frac{\lambda_{\mathrm{pb}}}{F\left(1 - e^{-\lambda_{\mathrm{pb}}T}\right)}\right] \qquad (3\text{-}7)$$

When the numbers are put into Equation 3-7 the result is:

$$C_{pb} = 1.0 \text{ pCi/L} \qquad (3\text{-}8)$$

which confirms that the calculation was done correctly.

In the derivation of Equation 3-7, it is assumed that the ^{212}Bi on the filter was in transient equilibrium with the ^{212}Pb when the 6- and 24-hour counts were performed. As discussed before, this means that no unsupported ^{212}Bi was present on the filter when the counts were made. Both assumptions are eminently reasonable considering the half-lives of ^{212}Pb and ^{212}Bi and the times involved.[4] These assumptions should be checked if the filter is counted less than 6 hours after the pump is stopped.

4 The validity of both assumptions was also confirmed with more detailed calculations that are given in Attachment A.

REFERENCES

NIOSH (National Institute for Occupational Safety and Health), 2013, *Lung Dose Conversion Factor for Thoron WLM*, DCAS-TIB-011, Rev. 4, Division of Compensation Analysis and Support, Cincinnati, Ohio, May 1. [SRDB Ref ID: 126391]

Skrable, K. W., C. French, G. Chabot, and A. Major, 1974, "General Equation for the Kinetics of Linear First Order Phenomena and Suggested Applications," *Health Physics*, volume 27, pp. 155–157. [SRDB Ref ID: 165844]

Wikipedia, 2017, "Thorium," Wikimedia Foundation, San Francisco, California, February 17. [SRDB Ref ID: 165836]

ATTACHMENT A
EXACT SOLUTION TO THE FORWARD PROBLEM

The calculations given above use the assumption that ^{212}Bi and ^{212}Pb are in transient equilibrium at the times of interest (i.e., the 6- and 24-hour counts). This greatly simplifies the calculations because it is not necessary to explicitly model the decay of the ^{212}Bi. Here, Skrable's general kinetics equation (GKE) (Skrable et al. 1974) is used to derive the 6- and 24-hour filter activities without assuming transient equilibrium.

The GKE shown below is generally applicable to first-order processes such as radioactive decay and the biokinetics of assimilated material, so the notation is somewhat generic (and perhaps imposing). Here, the GKE is applied to members (species) of a radioactive decay chain, so the notation and the equations will simplify significantly.

$$N_n = \sum_{i=1}^{n}\left[\left(\prod_{j=i}^{n-1} k_{j,j+1}\right)\sum_{j=i}^{n}\left(\frac{N_i^0 e^{-k_j t}}{\prod_{\substack{p=i \\ p\neq j}}^{n}\left(k_p - k_j\right)} + \frac{P_i\left(1-e^{-k_j t}\right)}{k_j\prod_{\substack{p=i \\ p\neq j}}^{n}\left(k_p - k_j\right)}\right)\right] \quad \text{(A-1)}$$

where

P_i = atom production rate for the *i*th species. For the problem, this is the rate at which atoms of ^{212}Pb and ^{212}Bi are deposited on the filter.

N_i^0 = the number of atoms of the *i*th species present at time 0.

N_i = the number of atoms of the *i*th species present at time *t*.

$k_{j,j+1}$ = transfer rate constant from the *j*th species to the *j*+1 species/compartment. Here these are the radioactive decay constants.

k_j = total removal rate constant from the *j*th species. Here these are the decay constants and are the same as the transfer rate constants (i.e., $k_j = k_{j,j+1}$).

Note that the GKE deals with atoms, and activity is calculated once the final solution in terms of atoms is obtained.

The calculation is broken down into two parts: the first is over the time from when the pump is turned on to the time when it is stopped, and the second is at times after the pump has stopped.

When the pump is started there are no atoms of ^{212}Pb or ^{212}Bi on the filter, so $N_i^0 = 0$ for all members of the decay chain. The GKE in this case reduces to:

$$N_n = \sum_{i=1}^{n}\left[\left(\prod_{j=i}^{n-1} k_{j,j+1}\right)\sum_{j=i}^{n}\left(\frac{P_i\left(1-e^{-k_j t}\right)}{k_j\prod_{\substack{p=i \\ p\neq j}}^{n}\left(k_p - k_j\right)}\right)\right] \quad \text{(A-2)}$$

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For ^{212}Pb, set $k_1 = \lambda_{pb}$, $k_2 = \lambda_{bi}$, $t = T$ (i.e., the pump has just been shut off):

$$P_1 = P_{pb} = \frac{C_{pb}F}{\lambda_{pb}} = 1.841 \times 10^6 / \text{hr} \qquad \text{(A-3)}$$

P_1 is the number of atoms per hour deposited on filter, and $N_1 = N_{pb0}$. After working through all the summations and *pi* products the result is:

$$N_{pb0} = \frac{P_{pb}}{\lambda_{pb}}\left(1 - e^{-\lambda_{pb}T}\right) = 2.234 \times 10^7 \qquad \text{(A-4)}$$

Therefore:

$$A_{pb0} = \lambda_{pb}N_{pb0} = 1.092 \times 10^4 \text{ pCi} \qquad \text{(A-5)}$$

which is the same result for the ^{212}Pb activity on the filter given previously in Section 2.0. Similarly, for ^{212}Bi:

$$P_2 = P_{bi} = \frac{C_{bi}F}{\lambda_{bi}} = 1.745 \times 10^5 /\text{hr} \qquad \text{(A-6)}$$

and $N_2 = N_{bi0}$. In this case the GKE gives:

$$N_{bi0} = \frac{P_{bi}}{\lambda_{bi}}\left(1 - e^{-\lambda_{bi}T}\right) + \lambda_{pb}\left[\frac{P_{pb}\left(1 - e^{-\lambda_{pb}T}\right)}{\lambda_{pb}\left(\lambda_{bi} - \lambda_{pb}\right)} + \frac{P_{pb}\left(1 - e^{-\lambda_{bi}T}\right)}{\lambda_{bi}\left(\lambda_{pb} - \lambda_{bi}\right)}\right] = 2.314 \times 10^6 \qquad \text{(A-7)}$$

A_{bi0} was not calculated before because it was not needed, but for completeness it is shown here:

$$A_{bi0} = \lambda_{bi}N_{bi0} = 1.193 \times 10^4 \text{ pCi} \qquad \text{(A-8)}$$

After the pump is stopped the production rates for ^{212}Pb and ^{212}Bi are equal to zero. The GKE with $P = 0$ for all members of the decay chain is:

$$N_n = \sum_{i=1}^{n}\left[\left(\prod_{j=i}^{n-1} k_{j,j+1}\right)\sum_{j=i}^{n}\left(\frac{N_i^0 e^{-k_j t}}{\prod_{\substack{p=i \\ p \neq j}}^{n}\left(k_p - k_j\right)}\right)\right] \qquad \text{(A-9)}$$

Now all times are relative to the time the pump was stopped. That is, N^0 for the first species = N_{pb0} and N^0 for the second species = N_{bi0}. The GKE for ^{212}Pb reduces to:

$$N_{pbt} = N_{pb0}e^{-\lambda_{pb}t} \qquad \text{(A-10)}$$

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where N_{pbt} is the number of atoms of ^{212}Pb on the filter at time *t* after the pump was shut off. For ^{212}Bi:

$$N_{bit} = N_{bi0} e^{-\lambda_{bi} t} + \lambda_{pb} \left[\frac{N_{pb0} e^{-\lambda_{pb} t}}{\left(\lambda_{bi} - \lambda_{pb}\right)} + \frac{N_{pb0} e^{-\lambda_{bi} t}}{\left(\lambda_{pb} - \lambda_{bi}\right)} \right] \quad \text{(A-11)}$$

where N_{bit} is the number of atoms of ^{212}Bi on the filter at time = *t* after the pump was shut off.
For *t* = 6 hours:

$$N_{pb6} = 1.512 \times 10^{7} \quad \text{(A-12)}$$

$$N_{bi6} = 1.583 \times 10^{6} \quad \text{(A-13)}$$

For *t* = 24 hours:

$$N_{pb24} = 4.681 \times 10^{6} \quad \text{(A-14)}$$

$$N_{bi24} = 4.903 \times 10^{5} \quad \text{(A-15)}$$

The corresponding activities are:

$$A_{pb6} = \lambda_{pb} N_{pb6} = 7{,}391 \text{ pCi} \quad \text{(A-16)}$$

$$A_{bi6} = \lambda_{bi} N_{bi6} = 8{,}163 \text{ pCi} \quad \text{(A-17)}$$

$$A_{pb24} = \lambda_{pb} N_{pb24} = 2{,}289 \text{ pCi} \quad \text{(A-18)}$$

$$A_{bi24} = \lambda_{bi} N_{bi24} = 2{,}528 \text{ pCi} \quad \text{(A-19)}$$

Referring back to Equations 2-8 and 2-9 and using the values for A_{bi6} and A_{bi24} calculated above using the GKE the results are:

$$A_{6} = A_{bi6} + A_{\alpha} = 8{,}163 \text{ pCi} + 1{,}080 \text{ pCi} = 9{,}243 \text{ pCi} \quad \text{(A-20)}$$

$$A_{24} = A_{bi24} + A_{\alpha} = 2{,}528 \text{ pCi} + 1{,}080 \text{ pCi} = 3{,}608 \text{ pCi} \quad \text{(A-21)}$$

which agree with the previously calculated values under the assumption of transient equilibrium.