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Determining postmortem interval : a preliminary examination of postmortem thorium actinium and radium isotopes in bone

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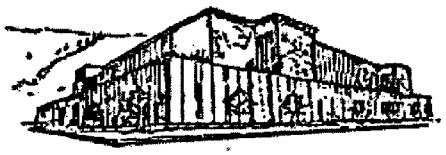

Determining Postmortem Interval: A Preliminary Examination of Postmortem Thorium, Actinium, and Radium Isotopes in Bone

By

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Abstract

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Determining Postmortem Interval: A Preliminary Examination of Postmortem Thorium, Actinium, and Radium Isotopes in Bone

Director: Randall R. Skelton

The estimation of postmortem interval (PMI) can aid law enforcement agents in helping to solve criminal cases that involve deceased individuals. When only the skeleton remains, the current methods used have severe limitations. For this reason, the following research project was undertaken.

Five samples with known time of death were analyzed for Th-228, Ra-228, and Ac-228 isotope concentrations. The method is similar to that utilized in current radiometric methods i.e. Potassium/Argon and C-14. An integration constant was determined to create an equation with the postmortem interval being the only unknown. Unfortunately, a common value for the integration constant could not be determined with the method described in this paper. A variety of possible errors in the design need to be addressed before this method can be thrown out. In conclusion, more research needs to be done.

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Chapter 1: Introduction

The estimation of postmortem interval (PMI) can aid law enforcement agents in determining whether a crime has been committed and who the potential suspects may be. Determining PMI can also aid in the identification of the deceased. Thus any advance in determining PMI could benefit law enforcement agents and the forensic scientists who must make this determination. Presently, a variety of methods are utilized to try and estimate PMI. Pathologists and medical examiners look at immediate and late changes of the body, and at the decomposition rates of soft tissue (Clark et al., 1997; Lyman, 1994). Entomologists and botanists examine the insects and plants found on or in the vicinity of the remains (Goff, 1993; Hall, 1997; Willey and Heilman, 1987). To analyze fully skeletonized remains, authorities call on forensic anthropologists and archaeologists to try and determine the PMI. At this point, there is no accurate way for forensic anthropologists to determine PMI. Thus, for forensic anthropology, an accurate method for determining PMI would be invaluable. The goal of this research project was to develop a preliminary method to estimate the PMI by means of the decay rates of three radioactive isotopes found in bone, Radium-228 (Ra-228), Actinium-228 (Ac-228) and Thorium-228 (Th-228). Since a large number of police cases involve decomposed and undocumented remains, determining the PMI can aid in solving many of them.

Immediate and Late Changes

A variety of changes occur immediately after death. These well-documented changes aid the pathologist or medical examiner in estimating PMI. The skin loses its normal color, the muscles relax, which may lead to the expelling of stomach and bowel

contents, and the eyes form an external dark band (Clark et al., 1997). Coagulated blood also starts to collect in the retinal vessels and eventually throughout the entire body (ibid., 1997). These initial stages are noticeable usually within the first two hours (Clark et al, 1997; McDonough, 1992; Sledzik, 1998). After the initial changes, the body undergoes through a series of late changes simultaneously: rigor mortis, algor mortis, and livor mortis (Clark et al., 1997; McDonough, 1992; Sledzik, 1998). Rigor mortis is the stiffening of the muscles due to chemical build up. After about 48 hours, the muscles relax as "the chemicals ... are consumed" (Clark et al. 152, 1997). Algor mortis is the cooling or heating of the body to equilibriate itself with the ambient temperature at approximately 1.5 degrees F per hour (Clark et al., 1997). The final late change is livor mortis. This change involves the settling of blood due to gravity. The blood pools in the areas closest to the ground turning the skin purple or red in color. Rigor mortis, algor mortis, and livor mortis all are noticeable within the first 2 - 4 hours after death (Clark et al., 1997; McDonough, 1992; Sledzik, 1998). Although rigor mortis may last up to 48 hours or two days, it is the initial and late changes that help the medical examiner determine PMI from death up to only one day (Sledzik, 1998 and Clark et al., 1997).

Decomposition

With the onset of death, the body begins a decomposition process. Several studies have been conducted to determine decomposition rates for a variety of environments (Rhine and Dawson, 1998; Rodriguez, 1997; Rodriguez and Bass, 1985; Sledzik, 1998; Galloway et al., 1989; Mann et al., 1990). Depending on the environmental conditions, the decomposition rates can vary from days to months

(Sledzik, 1998). From these decomposition studies, PMI can be determined in a variety of locations.

Although decomposition rates can help to determine PMI, many factors can alter the rate of decomposition. Insect, carnivore, and rodent activity play crucial factors in accelerating the rates of decomposition (Sledzik, 1998; Clark et al., 1997). Trauma can also increase the decomposition factor which comes about by insect activity increasing at the trauma location (Sledzik, 1998). Sledzik (118, 1998) claims that "Given the highly variable nature of decomposition, it is unlikely that highly accurate methods will be developed." (Sledzik 118; 1998) Although the accuracy of decomposition rates can be debated by forensic scientists, the method becomes inapplicable when skeletonization occurs. Depending on environmental factors, the soft tissue can completely decompose and skeletonization can occur within days to months.

Forensic Entomology

Forensic entomology is also commonly used to determine PMI. Insects usually arrive within a few hours of death to utilize the body as a food source and forensic entomologists concentrate their research on these insects (Catts and Goff, 1992; Goff et al., 1988). Because insects' development and succession rates are so well documented, comprehensive studies of the insects present can help entomologists pinpoint PMI. (Goff, 1993; Catts and Goff, 1992; Goff, et al., 1988)

Forensic entomology is most useful during the time of decomposition (Goff, 1993). However once skeletonization has occurred, the food source or soft tissue is no longer present. Therefore, the insects have no reason to stay on the body and they leave in search of new food sources. Although insects are no longer on the skeletonized body, entomologists can sometimes still determine the PMI with the remains that insects have left behind (i.e. pupae cases). Entomologists compare the insects that were found with the remains because different species will arrive at different decomposition stages and during different seasons (Goff, 1993; Catts and Goff, 1992; Goff et al., 1988). The developmental stages of insects found with the remains can also help to pinpoint season and minimal amount of time the body needed to be at the location (Goff, 1993). This can help to determine a wide time frame for PMI estimation.

A variety of factors can potentially cause problems for the entomologist's estimation of PMI. "In general, the parameters for the PMI estimate will become wider as the time since death increases" (Goff, 1993). In essence, the longer a body has been deceased, the wider the PMI estimate. Many environmental factors can hinder an entomologist's ability to determine PMI. Factors such as concealment of remains, cloud cover, temperature, and rainfall can delay the arrival of insects (Goff, 1993; Catts and Goff, 1992). This delay can cause the estimated PMI to be shorter than it actually is.

Forensic Botany

Forensic botany can also aid in determining PMI (Hall, 1997; Willey and Heilman, 1987; Lane et al., 1990). Botanists look at the growth patterns and

decompositional rates of plants or other floral materials that are found in the vicinity of the body. In certain cases, plants are found growing through the remains (Hall, 1997; Willey and Heilman, 1987). Through determination of plant growth rate and age, botanists can predict the minimum amount of time the body would have to lay in a certain spot for the plant to have grown through it (Willey and Heilman, 1987). Time frames such as these can help narrow the assessment of PMI. Plants found with a buried body are also beneficial in estimating PMI (Hall, 1997). The species of plants found can help pinpoint the season in which the body was buried and the decompositional rates of these plants can aid in determining PMI (Hall, 1997). Although Hall (1997) does not discuss the limitations of forensic botany, one major limitation would be the lack of any plant material present to aid in the PMI estimation. Of course, a major limitation of forensic botany would be its dependence on environment.

Current Radiometric Methods

Currently, Carbon-14 (C-14) and Potassium/Argon (K-40/Ar-40) are used to determine PMI for ancient skeletal remains and both techniques are based on the same principles. Radioactive isotopes decay at known rates into other isotopes. A radioactive or parent isotope decays into what is referred to as a daughter isotope (Eisenbud, 1987: Friedlander et al., 1981; Lederer et al., 1967). The daughter isotope can be either another radioactive isotope, or a stable one (Eisenbud, 1987; Friedlander et al., 1981). By knowing the decay rate, the parent/daughter relationship, and the ratio of the isotopes present, a postmortem interval can be determined. Carbon-14 (C-14) looks at the decay rate of C-14 into Nitrogen-14 (N-14) (Leute, 1987; Herz and Garrison, 1998). At the rate

of 5730 +/- 40 years, half of the existing C-14 in a sample will decay into N-14. This decay rate is referred to as the half-life of the isotope. This known half-life is applied to the ratio between C-14 and N-14. From this information, a postmortem interval can be determined. One of the benefits of this method is that C-14 is found in the remnants of all living things, thus, any organic material can be individually dated.

K-40/Ar-40 also looks at the rate of decay of radioactive isotopes but looks at the decay of K-40 into Ar-40 (Leute, 1987; Herz and Garrison, 1998). K-40 actually decays into two separate isotopes: Calcium-40 (Ca-40) and Ar-40 at a ratio of 89% and 11% respectively (Herz and Garrison, 1998). Because the ratio of Ca-40 and Ar-40 are known, only one of the two isotopes needs to be looked at. Ca-40 is a naturally occurring isotope found in abundance in almost everything on earth and almost any sample will contain a large amount of Ca-40 that is independent of the K-40 decay (Herz and Garrison, 1998). Therefore, Ar-40 is the isotope used. Although the methods, C-14 and K-40/Ar-40, are similar, K-40/Ar-40 has two major differences. The first is the decay rate. The rate for half of the existing concentration of K-40 to decay into Ar-40 is 1.25 x 10^9 years (Herz and Garrison, 1998). This allows for much more ancient remains to be dated. The other major difference is that K-40 is not found in organic remains (Leute, 1987; Herz and Garrison, 1998). K-40 is found in minerals, and a strong correlation between the minerals and the artifacts must be established. If this can not be accomplished, the date derived from the minerals can not be transferred to the artifacts. These methods are useful for determining the age of ancient remains, but can not be

transferred to modern remains due to the long half-lives of the isotopes. My research stemmed from these methods, but utilized isotopes with a much shorter half-life.

My Research

Since the determination of PMI is crucial and many of the current methods have limitations, a new method of determining PMI could only be beneficial to the forensic science community. A commonly recovered item in almost all homicides is the skeleton. Therefore, alternate methods to determine PMI for modern skeletal remains would be extremely valuable. This research combined the premise of current radiometric dating methods and isotopes with much shorter half-lives. By using isotopes with shorter half-lives, a new method can be determined to date modern remains. Ra-228 and Th-228 have very abbreviated half-lives and documented presences in bone (Harley and Fisenne, 1990; Ibrahim et al., 1983; Tandon et al., 1998). The half-lives of Ra-228 and Th-228 are 6.7 years and 1.9 years, respectively (Eisenbud, 1987; Lederer et al., 1967; Friedlander et al., 1981). Ra-228 decays into Ac-228, which has a half-life of 6.13 hours. Ac-228 then decays into Th-228 (Eisenbud, 1987; Lederer et al., 1967; Friedlander et al., 1981) The following diagram shows how the series progresses and the half-life of each isotope:

Ra-228(6.7yrs)⇒ Ac-228(6.13 hrs)⇒ Th-228(1.9 yrs)

*⇒ = decay; numbers in ( ) is the half-life

Although no documentation could be found to show that Ac-228 is found in bone, the fact that it is the intermediate step between Ra-228 and Th-228 points to the likelihood that it is present in bone. The half-lives for these and all isotopes is derived from

observed decay rates and assume the exponential law of radioactive decay (Krane, 1996). In other words, the half-life remains constant because both the amount of the given isotope and the decay rate decrease exponentially over time. The goal of this research was to be able to use the measurement of the decay rate at the time of testing to be able to determine PMI.

This research method was based on three premises that other research supports. The first is that environment has no affect on decay rates. According to Friedlander (1981), decay rates can not be altered by "variation of ordinary experimental conditions, such as temperature, chemical change, pressure, and gravitational, magnetic, or electric fields" (5). Leute (1987) also confirms that environmental factors do not affect the decay rates. Thus, this method would have the singular advantage of not being affected by environmental conditions.

The second premise was that all of the Th-228 found in bone is due to the decay of Ra-228 and is not also being taken up independently by the non-living bone. If the human bone takes up Th-228, the amount present in the skeleton would include both Th-228 from Ra-228 decay and independent uptake from the environment. Since there would be no way to determine how much of the Th-228 was actually from Ra-228 decay, a dating technique would be unreliable. The estimated PMI would be higher than the actual PMI due to the new Th-228. According to Dr. Isabel Fisenne (personal communication, 2000), the human skeleton does take up Ra-228 during the lifecycle but

Th-228 in not taken up independently by pre- or postmortem human bone. Therefore, any Th-228 found in the bone is due to the decay of total Ra-228 at the time of death.

The final premise was that Th-228 exists in an equilibrium with the environment prior to death. If random accumulation were to happen, a dating method using isotope decay may not be able to be used to assess PMI. Research points to the probability that Th-228 does exist in an equilibrium in the bone throughout the lifecycle. Harley and Fisenne (1990) tested for Th-228 but suggested that the Th-228 concentration "would be more representative of the buildup from Ra-228 in the intervening years than of the concentration of this radionuclide at the time of death" (1990: 517). Although not conclusive, this passage suggests the possibility that the Th-228 concentration is due to postmortem accumulation.

Another research project points more conclusively to the fact that Th-228 is not stored in pre-mortem bone. Ibrahim et al. (1983) also examined Th-228 isotope concentration in bone. With the goal of determining whether Th-230 was found in higher concentration in body tissues in regions where uranium mill tailings were present. For comparison, Th-228 and Th-232 were also assayed in the tissue samples. Although a variety of tissue samples were tested, this discussion will only focus on the results from the bone samples. Ibrahim et al., (1983) studied two groups, which included a sample of 22 individuals from Grand Junction, Colorado with a median age of 67 and a sample of 10 individuals from Washington, D.C. with a median age of 33. All samples were harvested postmortem. The difference between the two groups for both Th-230 and Th-

232 was significant ($p<0.1$). The Grand Junction population had higher concentrations of both isotopes. The conclusion as to why significant differences were seen was undetermined but two hypotheses were offered. The two possibilities included the older age of the Grand Junction population and/or the higher concentration of uranium mill tailings in the environment.

On the other hand, the difference between Th-228 concentrations in the two populations was not significant ($p>.01$). The Washington, D.C. population had a median concentration of .60 pCi/kg, and the Grand Junction population had a median concentration of .54 pCi/kg. Although the Washington, D.C. population had lower concentrations of the other isotopes, they had an insignificantly higher concentration of Th-228. No attempt at an explanation was given for this data. A possible explanation for the higher concentration could be longer travel time needed to arrive at the Los Alamos National Laboratory testing facility from Washington, D.C. than from Grand Junction, Colorado. While the difference in the two populations was not significant, Th-228 was still found in the samples. This could either be attributed to post-mortem or pre-mortem build up. The essentially similar levels may point to a pre-mortem equilibrium that exists with the environment. Basically, Th-228 concentration in pre-mortem bone would be at a constant level in all individuals. Post-mortem build up could also be the explanation for the Th-228 concentrations. Th-228 could have accumulated in the interval between death and the actual testing. These hypotheses are in contradiction to each other. Because Harley and Fisenne's hypothesis (1990) and the second hypothesis are in agreement, I favor Harley and Fisenne's (1990) argument that there is no pre-mortem accumulation of

Th-228. A possible explanation for the lack of Th-228 before death would be that live bone dispels the isotope by some unknown mechanism and only after death, does the bone start to accumulate Th-228. If the premise that the Th-228 concentration is zero at the time of death is found to be true, the postmortem interval would be determined by examining the decay rate for each of the three isotopes simultaneously.

This research was guided by the following hypotheses:

H_0 (null hypothesis): There is no relationship between the concentration of Th-228, Ac-228, and Ra-228 or their simultaneous decay rates are the length of the PMI.

H_1: There is a significant correlation between Th-228, Ra-228, and/or Ac-228 decay rate or concentration and the length of the PMI.

Chapter 2: Materials and Methods

The main isotopes looked at were Th-228 and Ra-228, due to the fact that their presence is documented in bone, and they have relatively short half-lives. These half-lives will allow for an accurate determination of PMI from present to approximately twenty years. Although research does not confirm the presence of Ac-228, it was also assayed. Its intermediacy between Ra-228 and Th-228 makes the inclusion of Ac-228 crucial for an accurate determination of PMI. If Ac-228 were ignored, this method would be uniformly inaccurate. The determination of the integration constant, C was contingent on the Th-228 concentration being zero or existing at a constant ratio with Ra-228 at death.

The research was carried out using a sample of 5 individuals. The samples were harvested by Dr. Gary Dale, the Montana State Medical Examiner. Samples consisted of the sternal end of the rib and were harvested during the fall of 2000. For analysis, the sternal end of the rib was chosen for two reasons:

1. Sternal rib ends can be harvested with ease.
2. Rib contains both trabecular and cortical bone (Harley and Fisenne, 1990).

The time and date of death were recorded for each specimen, and the age and sex were also recorded where available. The following table presents the demographic data for the samples used in the study. It includes the sex, age, time and date of death for each sample.

Sample	Age	Sex	Date of Death	Time of Death
1	34	Female	11/8/00	~1500 hrs
2	39	Male	11/25/00	0200 hrs
3	57	Male	10/24/00	0830 hrs
4	67	Male	11/10/00	~1800 hrs
5	35	Male	11/17/00	1700 hrs

Table 1: Demographic Data

Samples were tested at the Pacific Northwest National Laboratory in Washington State. Ra-228 and Ac-228 were derived from testing the sample on an ICP-MS, which counts the number of isotopes present. The samples were first removed from the plastic wrapping and placed overnight in a muffle furnace at 500 degrees C. The samples were cooled and then ground to an ash with a mortar and pestle. The ash was then dissolved in 15 ml of acid, similar to the acid digestion method described in Singh et al. (1979). One ml was then analyzed on the ICP-MS. The ICP-MS tested for the Th-232 isotope, which is the parent isotope of Ra-228 (Eisenbud, 1987; Lederer et al., 1967; Friedlander et al., 1981). Because the half-life of Th-232 is 1.41×10^{10} years (Eisenbud, 1987; Lederer et al., 1967; Friedlander et al., 1981), the half-life of Ra-228 is so much shorter, and the activity of Th-232 is "negligible over the course of the observation period" (Wagenaar, 2.2.3.2, 1995), the Bateman equation for secular equilibrium can be used to determine the

concentration of both Ra-228 and Ac-228 (Wagenaar, 1995). The following is the Bateman equation for secular equilibrium (Wagenaar, 1995):

$$A_d(t) \approx A_p(1 - e^{-\lambda_d t})$$

A= decay rate p = parent t = time λ = decay constant
d = daughter e = base of natural logarithms

Th-228 was assayed on a radon-free, ultralow background, high purity germanium spectrometer. This testing method determines decay rate and is non-invasive; no preparation method was used. The samples were sealed in an air-tight aluminum can, which keeps outside radon and its daughter products out. It also keeps radon and its daughter products generated by the sample inside the can. Because the isotopes were tested on different equipment at different times, an equation was used to correct the isotope concentrations for one standard time. Ra-228 was corrected to reflect the time that Th-228 was tested. Since Ra-228 was tested by isotope counting, the following formulas was used to determine the amount of isotopes present at death (Wagenaar,1995; Krane, 1996).

$$N = N_0 e^{-\lambda t_1}$$

N=number of isotopes e=base of natural logarithms t_1=time Ra-228 was tested
N_0 =number of isotopes at t = 0 (at death) λ = decay constant

Ra-228 was then determined for the time that Th-228 was tested. Since the decay rate was determined for Th-228, the following equation was used to estimate the decay rate

from the isotope count for Ra-228 at the time that Th-228 was tested (Krane, 1996; Wagenaar, 1995).

$$R = \lambda N_0 e^{-\lambda t_2}$$

R = decay rate	λ = decay constant e=base of natural logarithms
N_0 = # of isotopes at death (t = 0)	t_2 = time Th-228 was tested

The decay rates of Ra-228 and Th-228 for one standard time will be used to estimate the integration constant, C. The integration constant was determined for each sample. This research rested on the premise that C was constant for all samples. If a common C could be established, then an equation could be derived to estimate PMI. The following equation was used to determine C (Krane, 1996):

$$C = (R_B(t) - \frac{R_A(t)\lambda_B}{\lambda_B - \lambda_A})(\frac{1}{-\lambda_B e^{-\lambda_B t}})$$

A = Ra-228	λ = decay constant	e = base of natural logarithms	
B = Th-228	C = integration constant	t = time	R = decay rate

The preceding equation only takes into account the decay rates of Ra-228 and Th-228. Because this research was only a preliminary study, Ac-228 was excluded from the equation. Ac-228 did not need to be included in the equation to determine whether or not this method would work. In further research, Ac-228 will be included in the equation for an accurate determination of the integration constant. If Ac-228 were to be included in this preliminary study, the equations would have become extremely cumbersome. By only looking at Th-228 and Ra-228, determining whether or not this new method was

viable became much easier. The goal of this study was to decide whether or not this new method for PMI determination was viable and worth future research.

Chapter 3: Results

The following table contains all of the data derived at the Pacific Northwest National Laboratory:

Sample	1	2	3	4	5
Weight	3.80 g	4.465 g	4.62 g	6.63 g	3.51 g
Date Tested: Ra-228	8/28/01	8/28/01	8/28/01	8/28/01	8/28/01
Dates Tested: Th-228	2/9/01 to 2/13/01	2/24/01 to 3/7/01	3/8/01 to 3/19/01	3/30/01 to 3/27/01 and 3/29/01 to 4/13/01	4/14/01 to 4/24/01
Days Tested: Th-228	5.03 days	11.90 days	12.08 days	22.68 days	11.24 days
Ra-228/ Ac-228	.084 pCi/g	.098 pCi/g	.090 pCi/g	.048 pCi/g	.082 pCi/g
Th-228	.12 pCi/g	.12 pCi/g	.13 pCi/g	.039 p Ci/g	.036 pCi/g
K-40	3.29 pCi/g	3.91 pCi/g	2.20 pCi/g	2.34 pCi/g	4.88 pCi/g
Ra-226	.15 pCi/g	.16 pCi/g	.055 pCi/g	.0805 pCi/g	.17 pCi/g

Table 2: Data derived by PNNL

By using the information contained in table one and the equations in the paper, the following two tables contain the data used to derive the integration constant and the resulting integration constant:

Th-228 Decay Constant	$9.99 \times 10^{-4}\ d^{-1}$
Ra-228 Decay Constant	$2.83 \times 10^{-4}\ d^{-1}$
Base of Natural Logarithms (e)	2.7182

Table 3: Constants

Sample	**1**	**2**	**3**	**4**	**5**
Isotope count (A)	3.60×10^6	4.94×10^6	4.70×10^6	3.59×10^6	3.25×10^6
Isotope count (B)	1.30×10^6	1.71×10^6	1.92×10^6	8.27×10^5	4.04×10^5
Decay rate-A (decays/day)	1019.52	1398.82	1329.23	1017.35	920.10
Decay rate-B (decays/day)	1382.4	1712.85	1920	826.60	403.95
t-Time*	97 days	102 days	146 days	154 days	158 days
C- Integration Constant	1.57×10^6	1.77×10^6	2.15×10^6	1.22×10^6	1.10×10^6

Table 4: Derived data

*Due to the long testing period of Th-228, the last day of testing was used to determine time. Although this date is arbitrary, it will allow for consistency in the equations.

The resulting integration constants were then used to determine the average integration constant of 1.56×10^6. The average integration constant was

then used in the equation to get the derived PMI. The following table compares the derived PMI with the actual PMI.

Sample	Actual PMI	Derived PMI
1	97	90.5
2	102	226.5
3	146	175.4
4	154	93
5	158	194

Table 5: Comparison of Actual and Derived PMI
Derived PMI is reported as an absolute value.

Chapter 4: Discussion

As the preceding data suggests, a common value for C could not be determined with the method utilized in this paper. Because of the small sample size (N=5), accurate statistics could not be done. Instead, the average integration constant and the derived data for each sample were put back in the equation. By using the average integration constant, the unknown variable became time since death (t). When comparing the actual time with the estimated time, it becomes apparent that this method is not an accurate one. The derived PMI is grossly inaccurate. While the actual PMI gets larger from sample 1 to 5, the similar is not seen in the derived PMI.

The question then arises as to whether there was a flaw in the design or the basic premise of the research. Before the entire method can be thrown out, the design of the research has to be examined. Certain steps in this process may be flawed, while the method is sound. A variety of areas need to be examined for possible flaws. One of the major areas could be the assumption that Ra-228 can be extrapolated from the testing of Th-232. The basis of this assumption is that Th-232 and Ra-228 exist in secular equilibrium. Th-232 exists in secular equilibrium with Ra-228 in the environment. This paper assumed that the same would be seen in bone. If newly formed bone does not mirror its environment and begins with an absence of naturally occurring isotopes, Th-232 and its daughter products will follow a linear growth rate for about 40 years. After forty years have lapsed, the isotopes will then fall into secular equilibrium (Arthur, 2002)

If Th-232 is not in secular equilibrium with the environment, the Ra-228 determined from the Bateman equation would not be an accurate representation of the actual Ra-228 found in the bone. A method to directly test for Ra-228 would need to be found.

If Th-232 and Ra-228 are found to be in secular equilibrium at birth, other flaws in the research design need to be addressed. The sample preparation method for the ICP/MS could have some major flaws. The grinding of the dried bone with a mortar and pestle could introduce outside contamination to the ashed sample. Another problem with the sample preparation method is the assumption that all of the Th-232 goes into solution during the acid digestion method. If any of the Th-232 is lost during the digestion method, the derived and actual numbers of Th-232 could be vastly different. The ICP-MS preparation method needs to be examined for possible contamination and also for potential loss of Th-232 isotopes.

When the sternal rib ends were harvested from the corpses, some fluids and soft tissue remained on the samples. In a normal situation, the bones would have been cleaned by either dermestid beetles or by boiling. To avoid possible contamination, these methods were not utilized. Instead, the excess fluids and soft tissue were removed with a scalpel. Although all attempts were made to remove all fluids and tissue, miniscule amounts still remained. During the testing on the germanium spectrometer, the small amounts of tissue and fluid were still attached to the bone. The preparation method for the ICP-MS included the use of a muffle furnace, and this would have dried any tissue or evaporated any fluid. The inclusion of the tissue and fluid in one process and the

exclusion of it in the second could have skewed the results. The isotope results on the germanium spectrometer could potentially be higher than the actual isotope amount present in the bone. A thorough cleaning method for the sternal rib ends needs to be determined. All of the previously mentioned areas need to examined before the basic premises can be ruled as faulty and the method thrown out.

Conclusion

The hypothesis of this paper states that there is a significant correlation between Ra-228, Th-228, and Ac-228 decay rate or correlation and the length of the PMI. The data from Pacific Northwest National Laboratory and the results extrapolated from the data does not support this hypothesis. The null hypothesis can not be rejected. Therefore, the method as described in this paper does not seem to work. Whether it is the basic premises or design that is flawed warrants future study. Before the entire method is discarded, the possible design flaws needs to be examined. If areas of the design are determined to be flawed, these areas need to be corrected and research in this method should be continued. If the design does not seem to be flawed, the method can be discarded for these isotopes. Future research could include examination by the same method utilizing alternate isotopes.

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