Int J Legal Med
DOI 10.1007/s00414-014-0970-8

REVIEW ARTICLE

Radioactive isotope analyses of skeletal materials in forensic science: a review of uses and potential uses

Gordon T. Cook · Angus B. MacKenzie

Received: 21 November 2013 / Accepted: 21 January 2014

Abstract A review of information that can be provided from measurements made on natural and anthropogenic radionuclide activities in human skeletal remains has been undertaken to establish what reliable information of forensic anthropological use can be obtained regarding years of birth and death (and hence post-mortem interval (PMI)). Of the anthropogenic radionuclides that have entered the environment, radiocarbon (^{14}C) can currently be used to generate the most useful and reliable information. Measurements on single bones can indicate whether or not the person died during the nuclear era, while recent research suggests that measurements on trabecular bone may, depending on the chronological age of the remains, provide estimates of year of death and hence PMI. Additionally, ^{14}C measurements made on different components of single teeth or on teeth formed at different times can provide estimates of year of birth to within 1–2 years of the true year. Of the other anthropogenic radionuclides, ^{90}Sr shows some promise but there are problems of (1) variations in activities between individuals, (2) relatively large analytical uncertainties and (3) diagenetic contamination. With respect to natural series radionuclides, it is concluded that there is no convincing evidence that ^{210}Pb dating can be used in a rigorous, quantitative fashion to establish a PMI. Similarly, for daughter/parent pairs such as $^{210}Po/^{210}Pb$ (from the ^{238}U decay series) and $^{228}Th/^{228}Ra$ (from the ^{232}Th decay series), the combination of analytical uncertainty and uncertainty in activity ratios at the point of death inevitably results in major uncertainty in any estimate of PMI. However, observation of the disequilibrium between these two daughter/parent pairs could potentially be used in a qualitative way to support other forensic evidence.

G. T. Cook (✉) · A. B. MacKenzie
Scottish Universities Environmental Research Centre, Scottish Enterprise Technology Park, Rankine Avenue, East Kilbride G75 0QF, Scotland, UK
e-mail: Gordon.Cook@glasgow.ac.uk

Introduction

One of the fundamental tasks often required of a forensic anthropologist is to establish the identity of deceased individuals based only on skeletal remains. The determination of age at death is particularly important as an accurate assessment will enable certain people to be eliminated from any missing persons enquiry while conversely, it may serve to highlight others that could require additional investigation. When dealing with the remains of juveniles, conventional determination of age at death can achieve the levels of accuracy required by the forensic anthropologist [1, 2]. However, age determination of adult remains is significantly less accurate, particularly in the post 40-year age range where the anthropologist is often only able to make limited statements such as 'mature adult' [3]. In adults, many procedures involving the examination of a range of skeletal characteristics have been proposed but unfortunately, most suffer from methodological bias and complex variability in the skeletal ageing process [4]. Even the best skeletal-based methods are often limited to the identification of broad age groupings [5]. In addition, time between death and discovery (post-mortem interval or PMI) can also be important in any investigation of human remains. There are a number of established techniques for estimating this but most are for relatively short-term intervals. Limitations in accuracy increase with increasing PMI and estimates based on bone morphology are strongly influenced by site factors throughout the PMI [6].

Radionuclides contained in human skeletal remains have some potential for estimating year of birth and year of death/PMI because they decay at known, fixed rates and in some cases their levels can be related to man's activities during

JnlID 414_ArtID 970_Proof#1 - 04/02/2014

known periods in time. During the last decade in particular, there have been a number of radionuclide studies designed to establish these parameters [6–16] and so the purpose of this review is to synthesise the relevant published work on the analysis of both natural and anthropogenic radionuclides in human skeletal remains and critically review their applicability in forensic science.

Discussion

Radiocarbon (^{14}C)

Radiocarbon (^{14}C) analysis is the most widely used technique for estimating either the year of birth or year of death of human remains. The traditional use of the radiocarbon dating method has been in archaeology where the technique has been applied to the dating of bone and teeth samples ranging between approximately 300 and 50,000 years. Here, the technique relies on a relatively constant rate of ^{14}C production in the upper atmosphere. This is followed by rapid oxidation to $^{14}CO_2$, subsequent mixing with the stable carbon isotope forms ($^{12}CO_2$ and $^{13}CO_2$) and uptake by green plants during photosynthesis, thus labelling all plant life with ^{14}C. Subsequent consumption of green plants by animals results in similar labelling of all animal life. All of these mixing and transfer processes occur very rapidly in comparison to the average lifetime of a ^{14}C atom (8,300 years approx.) and so all living organisms are labelled to a first approximation with the same ^{14}C-specific activity (becquerel per kilogram). During life, an organism will retain this equilibrium living value, however, on death, it ceases to assimilate ^{14}C and so the level decreases in accordance with its half-life. The radiocarbon age (i.e., the time that has elapsed since the organism died) is calculated according to:

$$t = \frac{1}{\lambda} \ln\left(\frac{A_o}{A_t}\right)$$

where t=time elapsed since death; λ=decay constant for ^{14}C=ln(2)/half-life=1.245×10^{-4} year^{-1}; A_0=equilibrium living value (based on measurement of a modern reference standard) and A_t=activity of the sample t years after death.

N.B. The true physical half-life of ^{14}C is 5,730 years but in radiocarbon dating the so-called Libby half-life of 5,568 years is used. This discrepancy is accounted for when calibrating radiocarbon ages to the calendar timescale.

Since the late nineteenth century, man's activities have influenced the atmospheric ^{14}C concentration in two contrasting manners. First, the onset of the Industrial Revolution was accompanied by massive burning of fossil fuels. These contain no ^{14}C because of their great age, and so releases of CO_2 are confined to $^{12}CO_2$ and $^{13}CO_2$. This has resulted in a reduction in the atmospheric ratio of $^{14}CO_2$:$^{12}CO_2$ (and $^{14}CO_2$:$^{13}CO_2$) such that from AD 1890 until the early 1950s, this dilution was measureable in annual tree rings from that period (Suess Effect) and by the late 1940s, this had resulted in an approximate 3 % reduction in the Northern Hemisphere [17].

Second, the atmospheric testing of nuclear devices (bomb effect) resulted in the production of ^{14}C. These tests began in 1945 and continued until the Partial Test Ban Treaty in1963 which most countries with a nuclear capability signed up to. The tests led to an almost doubling of the atmospheric ^{14}C activity of the Northern Hemisphere by 1963 and about a 65 % increase in the Southern Hemisphere. Since 1963, the atmospheric ^{14}C activity has declined as the excess has entered the biota and the oceans [18]. Figure 1 illustrates the atmospheric ^{14}C activity in the Northern Hemisphere between 1950 and 2010.

The Suess Effect had a detrimental influence on conventional radiocarbon dating because it made it impossible to differentiate between organisms (including humans) that died in the period between approximately 1890 and 1950 and those that died between the mid-1600s and early-1700s. However, even in the absence of the Suess Effect, radiocarbon dating could not have provided the chronological precision required to make this a useful forensic technique for pre-1950 skeletal remains. One of the main reasons is the slow turnover of carbon in bone collagen, particularly during adulthood [19]. Other reasons include natural variations in the ^{14}C production rate (Fig. 2), the error on the measurement and the very small annual reduction in ^{14}C relative to this error. In contrast, the ^{14}C produced by atmospheric nuclear weapons tests has provided opportunities to study carbon turnover in collagen (and other tissues) and to provide significant information of forensic interest.

Knight [20] stated that "no physico-chemical or morphological techniques have yet been devised that will determine date independently of environmental deterioration. The only exception is the radiocarbon estimation in bones of greater antiquity than those of medico-legal interest". However, since then, significant advances have been made both in the measurement of radiocarbon and our understanding of carbon turnover in various components of skeletal remains.

Radiocarbon measurements on bone collagen

Through reference to the ^{14}C bomb peak it has been recognised that the ^{14}C activity of human bone collagen lags significantly behind the activity in a range of organs and soft tissues [21, 22]. Hedges et al. [19] found that their data constrained models of collagen turnover in adult human femoral mid-shafts to ≤4 % between the ages of 20 and 80 years. During adolescent growth (10–15 years age), the turnover is higher at 5 to 15 % year^{-1}. Geyh [23] suggests a significant

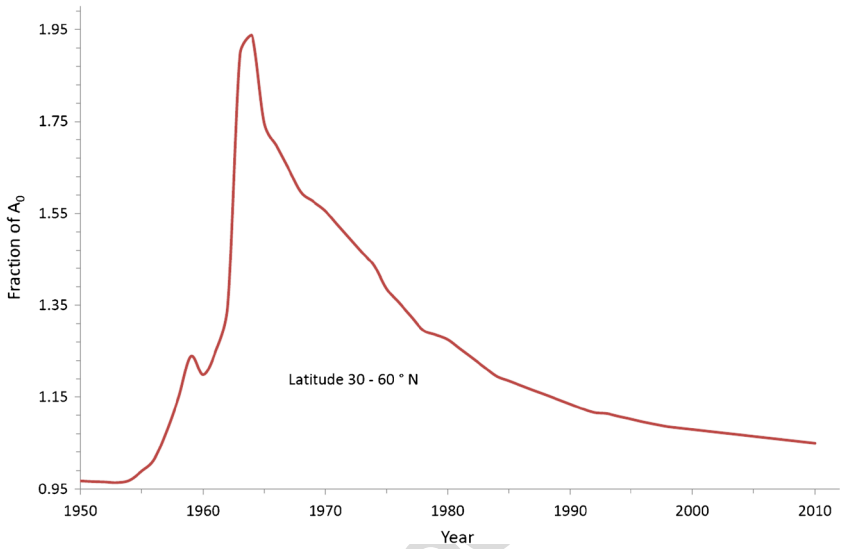


Fig. 1 Atmospheric ^{14}C activity of the Northern Hemisphere during the period 1950–2010 expressed as a fraction of the natural equilibrium living activity

decrease in collagen turnover to 1.5 % year^{-1} after the age of 19 (termination of puberty). He produced a modelled relationship between year of death and the ^{14}C activity of human bone collagen for several years of birth and plotted the measured ^{14}C values for individuals born with known dates of birth against these curves. The object here was to determine burial time. However, there were several outliers in the data and he concluded that burial time could seldom be estimated with a precision of <3 years and it could be as large as several decades. Furthermore, this work relies on knowing the year of birth, information that is often lacking. Taylor et al. [24] proposed that three time segments could be assigned to the recent past: (1) a pre-1650 period (non-modern), (2) a 1650–1950 period (pre-modern) and (3) a post-1950 period (modern), i.e. bomb period. They analysed the ^{14}C activity of human bone samples from five cases in which various coroner agencies from California had sought information on PMI. Those showing evidence of bomb ^{14}C were assigned to the modern period and deemed to be of further interest.

Therefore, in the absence of other information, use of single ^{14}C measurements on bone collagen from juveniles and adults can provide limited information, i.e. whether or not the person died during the nuclear era (because their ^{14}C activity was enhanced relative to the natural equilibrium living value).

Consumption of marine [25, 26] or freshwater resources from certain water bodies [27, 28] can lead to reduced ^{14}C activities in human remains due to ^{14}C activity offsets between these carbon reservoirs and the terrestrial biosphere [29–31]. Georgiadou and Stenström [32] used UN Food and Agriculture Organisation data on fish consumption to model changes in age. They calculated a shift of <2.5 years for all populations studied. The one exception was where a person was deriving their marine resources from close to the

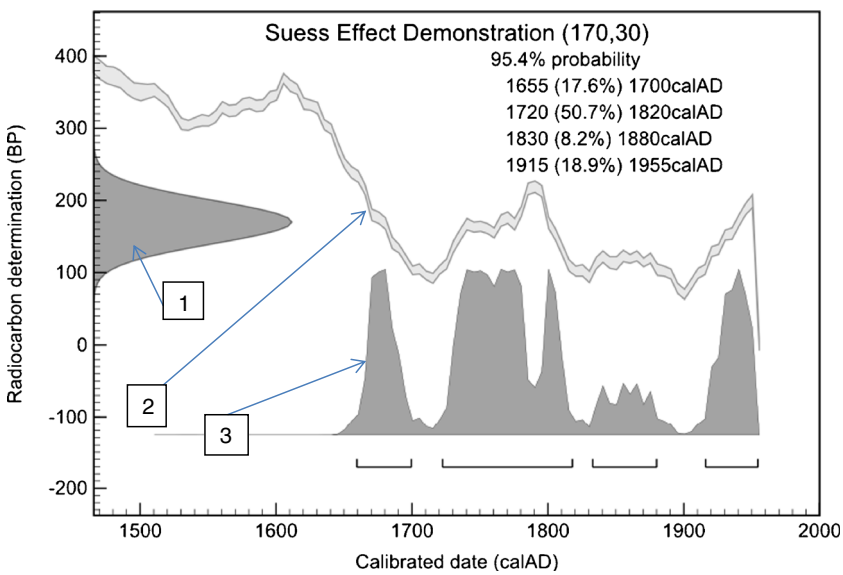


Fig. 2 Calibration plot for a radiocarbon age of 170 ^{14}C years $\pm 1\sigma$ of 30 years, indicating the inability to discriminate between samples from the second half of the seventeenth century through to the first half of the twentieth century. *1* Gaussian distribution of the radiocarbon age and associated error, *2* calibration curve (95 % confidence interval) based on radiocarbon measurements made on known age tree rings, *3* probability distribution for the calendar age ranges (figure based on Bronk Ramsey [107])

Sellafield nuclear fuel reprocessing plant in Cumbria, England. Due to the significant discharges of ^{14}C to the Irish Sea [33], the shift in age was −26.3 years.

There is one exception in which dating of single modern-period bone samples can produce high-precision estimations of year of birth. This is where the skeletal remains are of new-born or close to new-born babies. The shape of the ^{14}C bomb peak has been well constrained through extensive measurements of ^{14}C activities [34–37] and the rapid annual changes provide the potential for a chronologically precise methodology. This applies to components of human remains that exhibit either very fast or no carbon turnover. The bone collagen in infants is formed from the mother's dietary intake, and here, the ^{14}C will be relatively close to equilibrium with atmospheric levels. Broecker et al. [38] derived an average value of <1 year for the period between initial fixation of carbon by plants and human consumption and a maximum lag of <6 months between carbon consumption and appearance in the blood. Therefore, a radiocarbon measurement made on the bone collagen should represent the ^{14}C activity of the atmosphere 1–2 years earlier than the year of death.

Radiocarbon measurements on bone apatite

Up to 1.2 % by weight of carbon is substituted within the bioapatite component of fresh bone as structural carbonate [39]. In archaeological studies, radiocarbon dating of the carbon contained in this fraction was abandoned decades ago due to anomalous ages caused by post-mortem contamination effects [40–42]. Cremation of bone results in total loss of collagen and some reduction in structural carbonate but accompanying changes in the crystallinity produces a more compact structure resistant to carbonate substitution [43]. Lanting and Brindley [44] were the first to demonstrate the validity of dating this structural carbonate in cremated bone, however, recent studies have demonstrated the potential for exchange of carbon between CO_2 derived from the pyre fuel and the bone apatite [39, 45]. Therefore, while in principle it is possible to obtain similar information to that gained from ^{14}C activity measurements on collagen from single bones, it is important to have information on the manner in which the body was cremated (likely cremation temperature, pyre fuel). Furthermore, additional studies are required to assess the importance of carbon exchange under different cremation conditions.

Radiocarbon measurements on teeth

Until the development of accelerator mass spectrometry (AMS) in the 1980s [46, 47], analysis of the ^{14}C activity in single teeth was impossible. The standard analysis techniques typically required gram quantities of carbon whereas AMS requires approximately 1 mg. Teeth offer an attractive source of information with regards to age determination due to their survivability, the lack of carbon turnover (particularly in the enamel component) and our ability to determine a year of formation of this component through reference to the bomb peak [11]. Dentition will survive both heat and chemical degradation more readily than bone and are therefore found more frequently in a suitable condition for ^{14}C analysis. In juveniles, age determination based on the relationship between tooth mineralisation, eruption, emergence and deciduous tooth loss is the most accurate method [7]. However, in mature adults, indications of age based on morphology are extremely limited and ^{14}C has a significant role to play here. The enamel component contains approximately 0.4 % carbon and because there is no turnover, the ^{14}C activity reflects that of the atmosphere close to the time when enamel formation was occurring [11]. In addition, the crown of each tooth forms at a well-defined time during childhood. Combined with the well-constrained bomb peak, these factors provide us with a very powerful forensic tool for estimating year of birth for individuals who were born post-1950. Spalding et al. [11] first proposed this method and determined ages to within 1.6± 1.3 years, i.e. with better precision than other available methods [48]. To resolve the ambiguity of which side of the peak the results fell, they measured the ^{14}C activity in two teeth that form at different times. Cook et al. [7] proposed that an unambiguous year of birth could be determined from separate ^{14}C analyses on the enamel component and the collagen from the combined dentine and cementum. Since the crown enamel forms before the root then if the combined dentine/cementum ^{14}C activity is greater, the age must lie on the up-slope of the curve and if lower, the age must lie on the down-slope. Similarly, Kondo-Nakamura et al. [9] derived unambiguous ages on single teeth by separately measuring ^{14}C in enamel from the occlusal and cervical regions as they form at different times. Wang et al. [14] proposed that by selecting enamel from close to the cervix of the tooth, this reduced the error caused by the difference between sample formation time and the considerable time for formation of the entire enamel component. They also measured the ^{14}C content of the organic component of the root and found much lower ^{14}C activities. They proposed that these activities could be a potential tool for estimating age at death; however, the ^{14}C activities are almost always lower than would be possible for a dentine sample which must, by definition, be younger than the enamel component from the same tooth, i.e. the organic components of the root were lower than even the present day atmospheric ^{14}C activity. The only possible explanation would be an age offset due to consumption of non-terrestrial resources (as discussed above) but that would also have influenced the enamel. Several studies [49–51] have established a close correlation between the ages of dentine in teeth and the extent of aspartic acid racemization. Similarly, Ohtani et al. [52] studied aspartic acid racemization in cementum and

concluded that it remains stable throughout an individual's life. These studies indicate that there is little or no carbon turnover in the dentine and cementum which contradict the assertion of Wang et al. [14] but supports the conclusion of Cook et al. [7] that the dentine/cementum can be used to establish where the enamel ^{14}C activities fall on the bomb curve.

Radiocarbon measurements on bone and teeth from single individuals

Ubelaker et al. [12] proposed that an unambiguous year of birth could be gained by measuring the ^{14}C activity in cortical and trabecular bone, which remodel at different rates, in addition to tooth enamel. They also proposed that some information could be obtained on PMI using all 3 measurements but this depended on the age of the individual and the relationship between the bomb curve and death date. Ubelaker and Parra [13] studied date of birth and death of 4 individuals of varying age at death from Andean Peru. In the 3 older individuals (27, 44 and 56 years of age at death) they found: (1) tooth enamel ^{14}C activities that were consistent with years of birth, (2) significant lags (≥11 years) between death and the ^{14}C formation value of cortical bone but (3) minimum lags between trabecular bone formation and death and (4) ^{14}C analysis of enamel in the youngest individual (16 years of age at death) again enabled accurate determination of the year of birth and both the cortical and trabecular bone ^{14}C activities were consistent with the death date.

Natural decay series radionuclides

Uranium and thorium are environmentally ubiquitous radioelements (approximate crustal abundances of 2.7 and 9.6 mg kg^{-1}, respectively) and each has three natural isotopes: ^{238}U ($t_{1/2}=4.47\times10^9$ year), ^{235}U ($t_{1/2}=7.04\times10^8$ year) and ^{234}U ($t_{1/2}=2.45\times10^5$ year) for uranium and ^{232}Th ($t_{1/2}=1.4\times10^{10}$ year); ^{230}Th ($t_{1/2}=7.5\times10^4$ year) and ^{228}Th ($t_{1/2}=1.9$ year) for thorium. ^{238}U, ^{235}U and ^{232}Th are the parents of natural radioactive decay series (Fig. 3) and the systematics of ingrowth of daughter activity in these series is described by the Bateman equations [53]. In a mineral that has been isolated from external influences for about 2 million years, each of the decay series will be in secular equilibrium, with the decay rate of each member of a chain being equal to that of the parent. However, in open systems, differential behaviour during physical, chemical and biological processes can result in separation of parent-daughter pairs within the series. Isolation of a daughter nuclide will result in simple radioactive decay of the unsupported daughter nuclide, whereas isolation of a parent nuclide will be followed by ingrowth of the activity of the daughter nuclide. Such radioactive disequilibria have been widely used to characterise the rates and mechanisms of environmental processes on timescales ranging from millions of years [e.g. 54–56] to days [e.g. 57, 58]. Four radionuclides in the natural decay series which have been used successfully to characterise environmental processes have half-lives appropriate to investigating PMI, namely ^{210}Pb ($t_{1/2}=22.3$ year) and ^{210}Po ($t_{1/2}=138.4$ day) in the ^{238}U decay series, and ^{228}Ra ($t_{1/2}=5.8$ year) and ^{228}Th ($t_{1/2}=1.9$ year) in the ^{232}Th series.

^{210}Pb dating

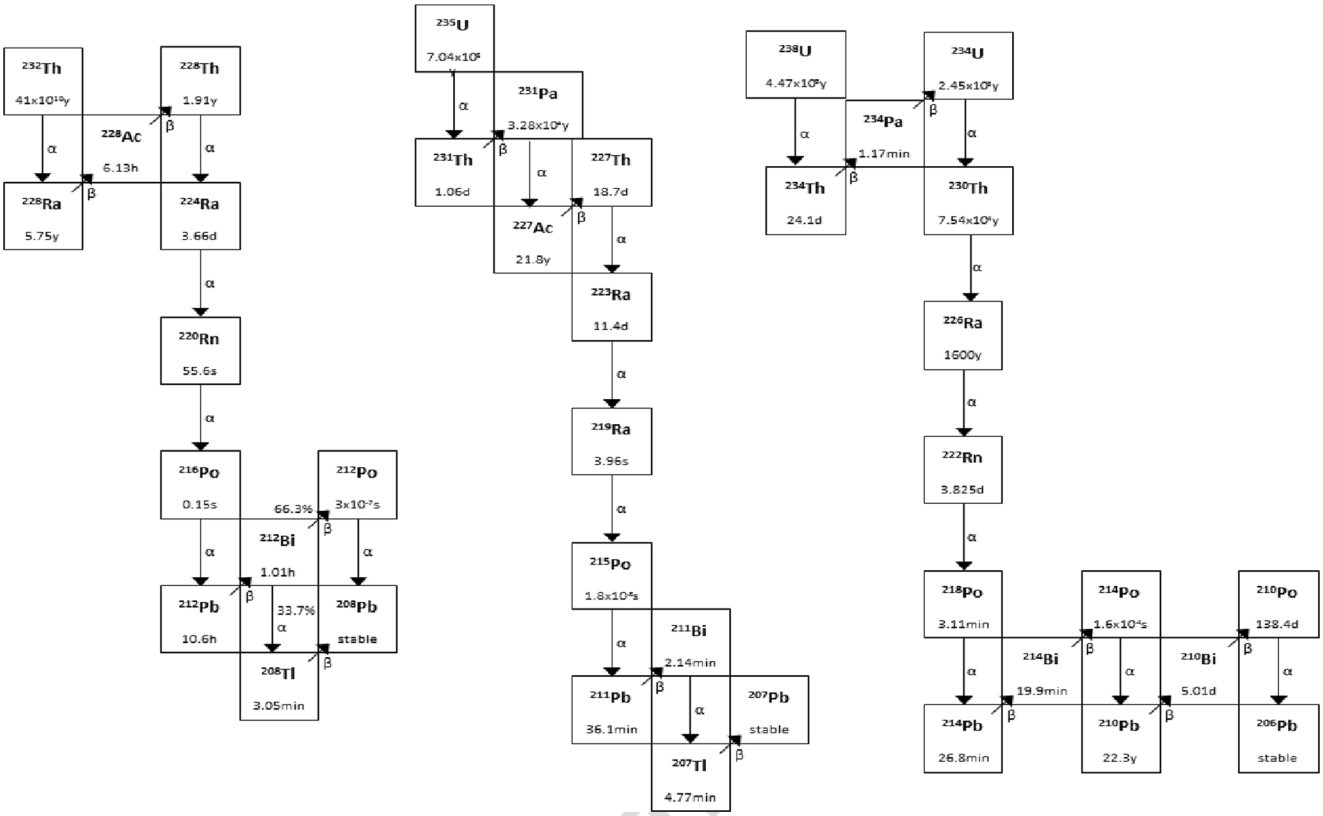
^{210}Pb dating has been widely used to establish short-term chronologies (approximately less than 150 years) in natural systems [e.g. 59–64]. The underlying principle is that ^{210}Pb, produced in the atmosphere from decay of the radioactive inert gas ^{222}Rn, is deposited on the surface of the earth, mainly through wet deposition. Progressive radioactive decay in layers of increasing depth (age) results in an exponential distribution of unsupported ^{210}Pb in accumulating sediments, from which chronologies can be derived. In ^{14}C dating, the initial specific activity in living organisms is a well-characterised constant value, whereas the initial specific activity of ^{210}Pb varies between different systems, depending upon site-specific conditions. Thus, ^{210}Pb dating gives ages relative to an assumed age of zero for the surface layer, rather than absolute ages. Moreover, most environmental systems contain minerals in which the ^{238}U decay series is in secular equilibrium, giving a supported ^{210}Pb component which must be subtracted from the total activity to quantify the unsupported activity.

^{210}Pb in the human body is preferentially incorporated in skeletal material, paralleling the behaviour of stable Pb. Thus, in principle, decay of unsupported ^{210}Pb could potentially be used to date bones or teeth, but for this to be a viable approach in forensic investigations:

- The specific activities of both ^{210}Pb and ^{226}Ra at the point of death in the bone or tooth sample analysed would have to be well-characterised, constant values applying across the population in which the individual lived.
- There must be no post-mortem diagenetic loss or gain of either ^{210}Pb or ^{226}Ra.

Studies of radionuclides in human skeletal material have dominantly been undertaken from a health perspective and biokinetic models are of major value in such work [e.g. 65, 66]. However, these models are not of sufficient accuracy to provide unambiguous, quantitative values for the initial specific activities of ^{210}Pb and ^{226}Ra for forensic dating.

The limited data available confirm that unsupported ^{210}Pb can be detected in bone, but do not provide a convincing case for ^{210}Pb dating as a robust forensic method. The requirement for accurate knowledge of constant initial activities of ^{210}Pb and ^{226}Ra presents a problem, since there is a very limited data



Q2 **Fig. 3** ^{232}Th, ^{235}U and ^{238}U natural decay series indicating decay modes and half-lives

set for these radionuclides in human bone. Blanchard [67] presented ^{210}Pb results for vertebrae from 14 infant children, all of whom died in 1962, with observed specific activities ranging from 2.9 to 8.8 Bq kg^{-1}. Ladiskaya et al. [68] observed significant differences between the ^{210}Pb content of different human bones obtained from autopsies of adults killed in street accidents, as shown in Table 1.

In data reported by Fisenne [69] for analysis of vertebrae from adults in the USA, summarised in Table 2, the specific activity of ^{210}Pb varied from 0.26±0.19 to 8.26±0.35 Bq kg^{-1}, while the ^{210}Pb/^{226}Ra activity ratio ranged from 0.3±0.2 to 15.8±1.2.

The samples had been stored for a period of years, but exact times since death were not provided since the objective of the study was to develop an appropriate analytical method, rather

t1.1 **Table 1** Average ^{210}Pb and ^{210}Po specific activities (Bq kg^{-1}) and ^{210}Po/^{210}Pb activity ratios for adult human bones [68]

t1.2 Sample type (number of analyses)	^{210}Pb	^{210}Po	^{210}Po/^{210}Pb
t1.3 Skull (15)	4.07±0.39	2.2±0.3	0.5±0.1
t1.4 Vertebrae (15)	1.6±0.3	0.66±0.11	0.4±0.1
t1.5 Sternum (21)	1.4±0.2	0.74±0.06	0.5±0.1
t1.6 Femur (15)	3.7±0.4	1.7±0.1	0.5±0.1
t1.7 Rib (21)	2.6±0.4	1.6±0.2	0.5±0.1

Table 2 ^{210}Pb and ^{226}Ra specific activities (Bq kg^{-1}) and ^{210}Pb/^{226}Ra activity ratios for archived samples of ashed human vertebrae [69]

t2.1 ^{210}Pb	^{226}Ra	^{210}Pb/^{226}Ra t2.2
3.01±0.25	0.93±0.11	3.3±0.4 t2.3
3.05±0.17	0.33±0.02	9.2±0.4 t2.4
3.64±0.22	0.48±0.02	7.6±0.3 t2.5
4.21±0.23	0.41±0.07	10.3±1.9 t2.6
2.88±0.17	0.52±0.04	5.6±0.4 t2.7
2.97±0.19	0.52±0.04	5.8±0.4 t2.8
2.43±0.16	0.88±0.03	2.8±0.1 t2.9
2.59±0.16	0.89±0.04	2.9±0.1 t2.10
8.26±0.35	0.52±0.04	15.8±1.2 t2.11
7.21±0.32	0.52±0.04	13.9±1.0 t2.12
6.90±0.26	0.41±0.04	17.0±1.6 t2.13
1.63±0.16	1.00±0 .22	1.6±0.4 t2.14
4.77±0.24	0.85±0.07	5.6±0.5 t2.15
4.42±0.24	0.56±0.04	8.0±0.6 t2.16
2.0±0.20	0.89±0.19	2.2±0.5 t2.17
3.20±0.15	0.96±0.07	3.3±0.3 t2.18
2.33±0.29	0.67±0.04	3.5±0.2 t2.19
1.16±0.20	0.37±0.15	3.1±1.3 t2.20
1.24±0.23	0.52±0.18	2.4±0.9 t2.21
0.26±0.19	0.81±0.07	0.3±0.2 t2.22
4.35±0.30	0.59±0.04	7.3±0.5 t2.23
1.83±0.24	0.56±0.11	3.3±0.7 t2.24
5.15±0.34	0.96±0.11	5.4±0.6 t2.25

than investigate PMI. Nevertheless, even allowing for potential differences in storage time, the results reveal significant variations in specific activities of both radionuclides. Also, a significant fraction of the ^{210}Pb is supported by ^{226}Ra, so activities of both would have to be analysed in any attempt to date bone.

Johnston et al. [70] used low-background gamma spectroscopy for in vitro analysis of ^{210}Pb in bone samples from three elderly individuals. For individual 1, total bone analyses were performed as summarised in Table 3.

A single whole bone analysis from the skull of individual 2 gave a value of 2.4±0.5 Bq kg^{-1}, while for individual 3, whole bone analyses gave 5.3±0.16 Bq kg^{-1} for fibula head and 3.8±0.6 for patella.

Separate analyses of cortical and trabecular bone were undertaken for individuals 2 and 3 as shown in Table 4.

The variability of ^{210}Pb-specific activity between different bones in an individual and between the same bones in different individuals highlights the problem of attempting to define an initial value for dating of bone. The data also reveal no systematic variation between trabecular and cortical bone.

Salmon et al. [71] reported an interesting feature of the distribution of ^{210}Pb in bone, with enhanced concentrations in a surface layer of depth <3 μm. Specific activities of ^{210}Pb (measured indirectly via ^{210}Po) in the surface layer of four cranium samples and one femur sample were about four times higher than those in bulk bone. However, given the small contribution of the enriched layer to the total bone mass, it is unlikely to have a significant effect on the analysis of these radionuclides in bulk bone samples.

Based on a study of children's deciduous teeth and permanent teeth of juveniles, James et al. [72] observed the highest concentrations of ^{210}Pb (inferred from ^{210}Po) in the highly calcified outer enamel surface, representing cumulative environmental exposure. In contrast, the highest concentrations of ^{226}Ra were in the circumpulpal region. James et al. [72] summarised data for several studies of teeth and the combined data revealed ^{210}Pb-specific activities ranging from 1.38 to 5.3 Bq kg^{-1} and ^{226}Ra varying from 0.14 to 1.96 Bq kg^{-1}. In general ^{210}Po was in equilibrium with ^{210}Pb. The problem of assigning an initial ^{210}Pb-specific activity as a basis for dating is again emphasised by the variability between teeth of different individuals and in different regions of any given tooth.

Further work by James et al. [73] investigating geographical variations of ^{210}Pb in the permanent teeth of juveniles, concluded that both oral intake and inhalation were implicated in ^{210}Pb uptake. Vehicle exhaust emissions were identified as a significant source, resulting in differences in ^{210}Pb-specific activities in juvenile teeth from different environments, with mean values of 7.83 Bq kg^{-1} for rural, 7.94 Bq kg^{-1} for small town and 8.48 Bq kg^{-1} for urban environments. Anomalously high values were observed in Devon as a consequence of regionally high ^{222}Rn levels. Thus, geographical and environmental variations would have to be taken into account in any attempted forensic use of ^{210}Pb dating.

The complexity of modelling ^{210}Pb uptake and distribution in the human body was highlighted by Salmon et al. [66], who compared modelled and observed levels of ^{210}Pb in bones from people of different ages. A clear age dependence was observed and, while reasonable agreement (better than order of magnitude) was found between modelled and observed specific activities of ^{210}Pb in human bone, the level of agreement was not good enough to provide a basis for quantitative forensic use of ^{210}Pb dating.

The few studies that have attempted to use ^{210}Pb dating to investigate PMI have revealed further complications. Swift et al. [74] presented data for 15 samples of human skeletal material from Portugal. The bodies had been subject to conventional burial in soil for a period of 5 to 6 years, after which the skeletal material had been disinterred, "re-coffined and transferred into a drawer" and stored for a period of up to decades before eventual reburial. Samples of diaphyseal compact bone, obtained after the period of storage in the "drawers", had unsupported ^{210}Pb-specific activities (at the time of analysis) ranging from 3.64±0.07 Bq kg^{-1} to a non-detectable level, with a decreasing trend as a function of time since death that gave a reasonable fit to an exponential

Table 3 ^{210}Pb-specific activities for bone samples from individual 1 [70]

Sample type	^{210}Pb (Bq kg^{-1} dry weight)
Femur	3.2±0.12
Femur head	2.5±0.5
Fibula	2.8±0.4
Patella	3.0±0.6
Tibia	1.9±0.3
skull	3.3±0.9

Table 4 ^{210}Pb-specific activities for bone samples from individuals 2 and 3 [70]

Sample (individual)	Cortical	Trabecular	Cortical/trabecular ratio
Femur (2)	2.0±0.3	2.4±0.4	0.83
Femur (3)	1.8±0.3	2.4±0.7	0.75
Femur head (3)	2.8±0.5	3.4±0.5	0.82
Fibula (2)	0.97±0.23	1.8±0.7	0.54
Fibula (3)	1.3±0.3	< 6	
Hip (3)	1.8±0.4	2.9±0.7	0.62
Patella (2)	2.4±0.6	2.9±0.7	0.83
Tibia (2)	1.8±0.3	2.4±0.3	0.75
Tibia (3)	1.7±3	2.8±0.1	0.61
Tibia head (3)	4.0±0.7	3.5±0.7	1.1
skull	2.0±0.4	2.1±0.5	0.95

function. However, if a correction is applied for decay over the period since the known time since death, the specific activities still show an irregular, logarithmically decreasing trend with increasing age, clearly implying post-mortem loss of ^{210}Pb by processes other than radioactive decay. Swift et al. [74] observed a corresponding trend of decreasing uranium concentrations with increasing PMI for these samples which implies post-mortem chemical loss, similar to that of ^{210}Pb. Such post-mortem loss violates one of the fundamental conditions for quantitative use of ^{210}Pb in dating.

In a carefully conducted study, Schrag et al. [10] investigated the relative contributions of diagenetic and biogenic ^{210}Pb and ^{90}Sr in the vertebrae of individuals who had been buried in 1999 and exhumed in 2007. The study revealed the presence of diagenetic ^{210}Pb in the vertebrae and demonstrated that use of selective dissolution in a "solubility profile" method, could potentially be used to isolate an uncontaminated sample of trabecular bone for ^{210}Pb analysis. The problem remained, however, of defining accurately the initial ^{210}Pb activity in bone for the living population.

In summary, it has been established that unsupported ^{210}Pb does occur in human bone, but that there are significant differences in specific activities between: (1) different bones, and different parts of the same bone, in an individual; (2) different individuals within the same age group; (3) individuals in different age groups; and (4) individuals living in different environments. The studies summarised above reveal that there is no basis for assuming a constant initial specific activity of ^{210}Pb or ^{210}Pb/^{226}Ra activity ratio in human bone. In addition, diagenetic processes have been shown to result in post-mortem alteration of ^{210}Pb activities in buried bone. Thus, there is no convincing evidence that ^{210}Pb dating can be used in a rigorous, quantitative way to establish PMI. Nevertheless, unsupported ^{210}Pb will be present in bone and teeth immediately after death, so its presence could, in principle, be used in a qualitative or semi-quantitative way to support other evidence for PMI.

^{210}Po/^{210}Pb disequilibrium

^{210}Pb/^{210}Po disequilibrium can potentially be used to investigate processes taking place on a timescale of up to about 2 years and the systematics of ingrowth in the ^{210}Pb-^{210}Bi-^{210}Po system are summarised in Fig. 4. Use of ^{210}Po/^{210}Pb disequilibrium for dating in environmental applications has been restricted to studies of very recent volcanic events, based upon assumed initial, high-temperature, total outgassing of ^{210}Po [e.g. 57, 75, 76].

Swift [77] proposed that the ^{210}Po/^{210}Pb activity ratio could be used to estimate PMI, with the assumption that during life ^{210}Po is present at a lower specific activity than ^{210}Pb in bone, but that after death ^{210}Po will grow in towards transient equilibrium with ^{210}Pb. Starting from pure ^{210}Pb, it would take 830 days for the ^{210}Po activity to become equal to that of ^{210}Pb, after which there would be a small excess ^{210}Po activity. However, some ^{210}Po is present in human bones during life due to decay of ^{210}Pb, so the initial ^{210}Po/^{210}Pb activity ratio would have to be known in order to calculate a PMI. Swift [77] observed that estimates of the initial value for this ratio ranged from 0.1 to 1.0 and suggested that the IAEA estimate of 0.8 could be used for the initial value. This contrasts both with the assumption by Schrag et al. [10] that ^{210}Po and ^{210}Pb are in equilibrium in bone during life and with the age-dependent trend of this ratio used in the DOSE210 model [66]. Thus, the fact that the initial ratio is not well-defined presents a fundamental limitation in application of this method.

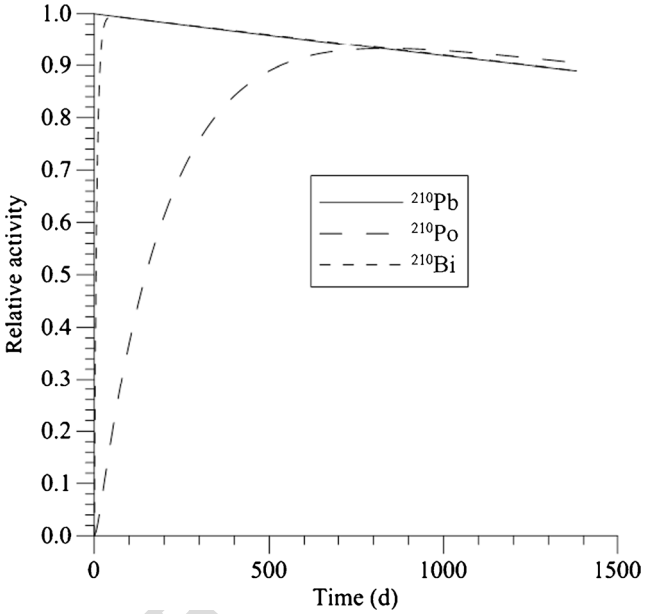


Fig. 4 Ingrowth of ^{210}Pb and ^{210}Po activities from the decay of ^{210}Pb

Ziad et al. [15] attempted to use ^{210}Po/^{210}Pb disequilibrium to calculate PMIs for seven samples of human skeletal remains of known time of death. Reasonable agreement was obtained for three adult samples, but discrepancies of over 20 years were found for two samples from adolescents, while samples from the twelfth and thirteenth centuries, respectively, indicated 1983 and 1980 as the years of death.

Use of ^{210}Po/^{210}Pb disequilibrium for dating bone requires analysis of ^{226}Ra, ^{210}Pb and ^{210}Po and there is an unavoidable analytical uncertainty in each of these measurements. Using state-of-the-art gamma spectroscopy equipment in an underground laboratory to minimise background, Johnston et al. [70] reported analytical uncertainties of: counting statistics (2–35 %), detection efficiency (5–10 %), γ ray emission probability (~0.15 %) and mass (<0.35 %). Combining such analytical uncertainties with the uncertainty in the initial ^{210}Po/^{210}Pb activity ratio would inevitably result in major uncertainty in any estimate of PMI.

$^{228}Th/^{228}Ra$ disequilibrium

The timescale that could potentially be covered by ingrowth of ^{228}Th towards transient equilibrium with ^{228}Ra is shown in Fig. 5, but there have been relatively few uses of ^{228}Th/^{228}Ra disequilibrium in environmental studies. Applications have included assessing the ages of carbonate deposits [78] and of newly formed minerals in hydrothermal and volcanic systems [76, 79–82], and characterising sediment mixing processes [83–85]. In one investigation of potential forensic relevance, Brunnermeier et al. [86] attempted to estimate the period since death of an elephant by analysis of ^{228}Th in its tusk, observing a systematic increase in specific activity from 1.4±0.2 to 7.3±0.3 Bq kg^{-1} and in the ^{228}Th/^{232}Th activity ratio, from 7±2 to 63±22, from the tip towards the root of the tusk.

Some results for concentrations of ^{228}Th in human bone have been reported for occupationally exposed individuals [e.g. 87] or for patients who had received Thorotrast injections [e.g. 88], but data for the general population are sparse. Studies undertaken from a radiological perspective have indicated that ^{228}Th in human bone is present at very low levels and is dominantly derived from the decay of ^{228}Ra. For example, Takizawa et al. [89] reported ^{228}Th-specific activities in the range 4.4–69.1 mBq kg^{-1} for human bones from Northern Japan, while specific activities for ^{232}Th were in the range 0.63–5.7 mBq kg^{-1}. Analysis of such low levels of thorium isotopes is challenging and analytical uncertainties are relatively high, as highlighted by Martinez-Canet et al. [90] who used state-of-the-art gamma spectrometry for the analysis of bone. The magnitude of analytical uncertainty is illustrated (Table 5) for bones from one individual who died in January 1999, with analysis performed in November 2000. The data confirmed that specific activities of ^{228}Ra and ^{228}Th were very low and indicated that the ^{228}Th/^{228}Ra activity ratios could not be distinguished from unity within error.

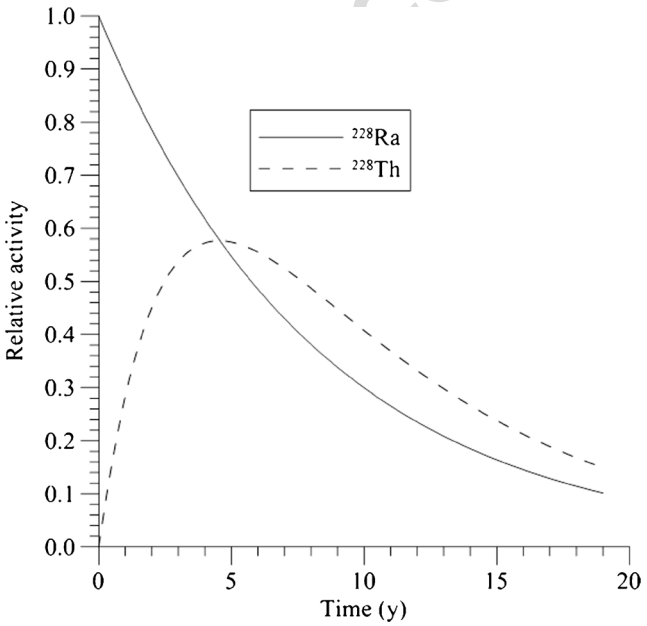


Fig. 5 Decay and ingrowth curves for the ^{228}Ra-^{228}Ac-^{228}Th system with the assumption that ^{228}Ac is in secular equilibrium with ^{228}Ra

Table 5 ^{228}Ra- and ^{228}Th-specific activities (mBq kg^{-1}) and activity ratios for samples of human bone [90]. Errors are fully propagated 1σ values

Sample	^{228}Ra	^{228}Th	^{228}Th/^{228}Ra
Skull	56±15	87±14	1.55±0.48
Femur head	<140	<215	
Femur bone	74±15	88±15	1.19±0.32
Patella	110±40	105±25	0.95±0.41
Tibia	86±22	64±27	0.74±0.37
Fibula	78±30	<138	

Kandlbinder and co-workers [6, 8, 16] suggested that ^{228}Th in human bone is derived almost entirely from in situ decay of ^{228}Ra and that for living individuals the ^{228}Th/^{228}Ra activity ratio is less than unity. On this basis, as the system grows towards transient equilibrium after death, the ^{228}Th/^{228}Ra activity ratio could potentially be used for estimation of PMI. This approach would have the advantage of being insensitive to variations in the initial specific activity of ^{228}Ra, but would require that: (1) the initial ^{228}Th/^{228}Ra activity ratio was known accurately, (2) the initial disequilibrium was larger than the uncertainty on the analytical data and (3) there was no post-burial diagenetic influence on either ^{228}Ra or ^{228}Th. In order to test the proposed method, Kandlbinder et al. [91] analysed 13 samples of human femur or humerus using gamma spectroscopy for ^{228}Ra and alpha spectroscopy for ^{228}Th. Specific activities of ^{228}Ra ranged from 0.16±0.12 to 1.04±0.65 mBq kg^{-1} and ^{228}Th from 0.10±0.08 to 0.52±0.1 mBq kg^{-1}, both being on an ashed weight basis. Corresponding ^{228}Th/^{228}Ra activity ratios at time of death were in the range 0.12±0.12 to 0.72±0.36. In an extension of this work, Kandlbinder et al. [92] identified a significant blank contribution in the ^{228}Th analytical procedure and introduced a new tracer in an attempt to overcome this problem. Results for new analysis of human femur samples from autopsies or exhumed bodies gave blank-corrected specific activities in the range 0.13±0.11 to 0.75±0.26 mBq kg^{-1}, but estimated ^{228}Th/^{228}Ra activity ratios at time of death ranged from −1047.9±210.83 to 0.75±0.23 mBq kg^{-1}, relative to a value of 0.37, based upon a biokinetic model. Problems of thorium addition or removal as a consequence of soil diagenetic processes were identified for the exhumed samples. Kandlebinder et al. [92] also reported an improved gamma spectroscopy method for analysis of ^{228}Ra in human bone ash by chemical removal of potassium to reduce the ^{40}K contribution to the Compton continuum in the gamma spectrum. The improvement in analytical uncertainty was illustrated for three samples of bone

ash which yielded results of $(3.6\pm2.0)\times10^{-4}$, $(3.0\pm2.0)\times10^{-4}$ and $(3.8\pm1.9)\times10^{-4}$ mBq kg^{-1}, respectively, without potassium separation but values of $(3.8\pm1.1)\times10^{-4}$, $(3.1\pm1.0)\times10^{-4}$ and $(4.4\pm0.7)\times10^{-4}$ mBq kg^{-1}, respectively, after potassium separation.

On the basis of: (1) variations in initial ^{228}Th/^{228}Ra activity ratio in human bone, (2) the large analytical uncertainties and (3) the influence of soil diagenetic processes, it is apparent that the ^{228}Th/^{228}Ra activity ratio does not provide a basis for quantitative dating of bone in forensic applications. However, as concluded above for unsupported ^{210}Pb, observation of radioactive disequilibrium between ^{228}Ra and ^{228}Th could potentially be used in a qualitative way to support other forensic evidence.

Anthropogenic radionuclides

Atmospheric testing of nuclear weapons was carried out mainly between 1945 and 1963, with very few tests since then and the last having been in 1980 [93]. As noted above for ^{14}C, deposition of weapons testing fallout followed a well-characterised trend with a pronounced peak in 1963, as illustrated in Fig. 6 for ^{90}Sr. Releases from the Chernobyl nuclear accident in the Ukraine in 1986 resulted in high levels of contamination in areas close to the plant and lower levels in more distant areas of Europe and Scandinavia [93]. These temporal variations in input have resulted in anthropogenic radionuclide distributions that can be used to establish chronologies in accumulating sediments [94] and, in principle, variations in the specific activity of anthropogenic radionuclides could potentially be used for establishing PMI for skeletal material from people who lived in the nuclear era. Prerequisites for this to be practicable for any given anthropogenic radionuclide are:

- The specific activity of the radionuclide in bone or teeth would have to vary with time in a characteristic way that can be related to the fallout deposition pattern.
- There would have to be a sufficiently large data base of empirical results to provide confidence in relating the specific activity for a given sample to the fallout trend.
- Post-burial diagenetic processes would have to have a negligible influence on the radionuclide.

Most analyses of anthropogenic radionuclides in human skeletal material have been undertaken from a radiological perspective, often in combination with biokinetic models, in attempts to assess the health implications of radioactive contamination. Attention has dominantly been focussed on radionuclides that are preferentially concentrated in the skeleton, notably ^{90}Sr ($t_{1/2}$=28.5 year) and the alpha-emitting isotopes of plutonium (^{238}Pu, $t_{1/2}$=87.7 year; ^{239}Pu, $t_{1/2}=2.4\times10^4$ y; ^{240}Pu, $t_{1/2}=6.56\times10^3$ year) [e.g. 67, 95–99]. Specific activities of anthropogenic radionuclides in bone are low and a review by Tandon et al. [96] indicated a range of 1.5–25.7 mBq kg^{-1} for Pu and 19–180 Bq ^{90}Sr kg Calcium^{-1}. Analysis of such low concentrations in small samples is analytically challenging, especially for Pu, and Tandon et al. [96] stressed the importance of rigorous analytical quality control procedures.

Jones and Prosser [95] evaluated the accuracy of biokinetic models for plutonium using an extensive suite of analyses of post-mortem tissues in the US, spanning the period 1953–1985. Based on results for 2,633 measurements for a variety of tissue types, it was concluded that biokinetic models represent the movement of plutonium through the body reasonably well, but that there were order of magnitude variations between different model estimates of plutonium concentrations depending upon the choice of absorption parameters. Moreover, results for 529 analyses of plutonium in bone displayed order of magnitude variations between samples from different individuals and between measured and model values. Biokinetic models thus represent a powerful and important technique in radiological applications, with ongoing improvements in the accuracy of the models and associated databases [e.g. 100], but the uncertainty associated with model-derived concentrations means that they are of restricted value in potential forensic applications.

Various studies have demonstrated temporal variations in specific activities of ^{90}Sr and Pu in skeletal material, particularly trabecular bone and teeth, with trends reflecting fallout deposition in combination with environmental and body

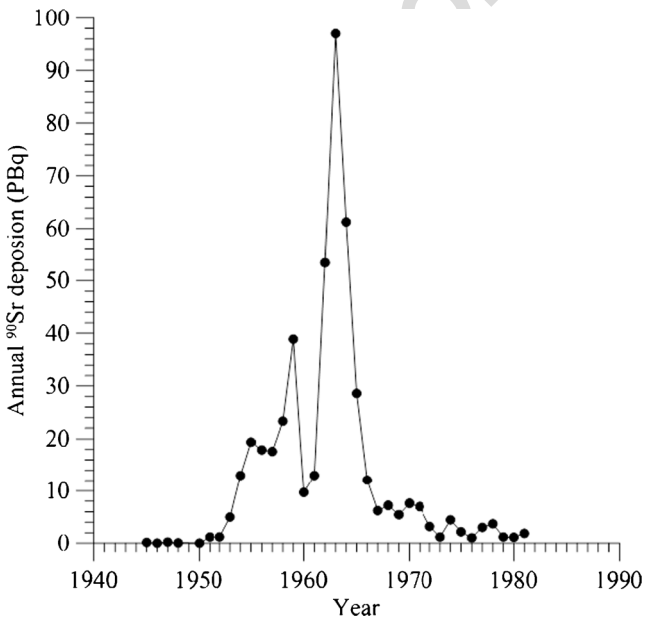


Fig. 6 Annual deposition (PBq) of ^{90}Sr in the Northern Hemisphere as a result of atmospheric testing of nuclear weapons (data for 1945–1957 based on calculations; data for 1958–1981 based on measurements) (UNSCEAR, 2000)

residence times [e.g. 97, 99, 101–104]. Geographical variations are also important, with Froidevaux et al. [102] reporting little influence of Chernobyl-derived ^{90}Sr in deciduous teeth in Switzerland, but Schmitz et al. [105] observing specific activities for Ukranian children's teeth approximately twice those for children in Germany.

Thus, variations in concentrations of anthropogenic radionuclides in bone and teeth appear to have potential for investigation of PMI, but significant problems have been encountered in the few attempts that have been made to exploit this potential.

MacLaughlin-Black et al. [106] analysed ^{90}Sr in femora from a mediaeval skeletal collection and from contemporary post-mortem examinations in an attempt to distinguish between forensic and archaeological provenance, with an assumed demarcation point of 75 years before present. The contemporary samples had specific activities in the range 2.11–3.15 Bq kg^{-1} calcium, consistent with input from weapons testing fallout, but values in the range 0.70–1.51 Bq kg^{-1} calcium for the mediaeval samples provided clear evidence of diagenetic contamination. MacLaughlin-Black et al. [106] observed that for this approach to be useful: (1) data would have to be available for ^{90}Sr levels in bone samples spanning a suitable timescale and (2) the effects of contamination would have to be defined for different burial conditions. Nevertheless, they concluded that the technique could potentially prove of value in determining whether or not an individual was alive before or after the 75-year demarcation point.

Swift et al. [74] presented plutonium data for the 15 femur samples discussed above in the context of ^{210}Pb dating, with dates of death ranging from 1921 to 1983. Plutonium was only detected in samples corresponding to dates of death after 1945, indicating negligible contamination under the conditions affecting these remains (burial in soil for 5 to 6 years then recovery and storage of the skeletal remains in a drawer). This observation indicates that plutonium analysis has potential to distinguish between bones of people who lived in the nuclear era from older remains. However, the results for samples from the nuclear era had a relatively wide range (6–67 mBq kg^{-1}), with large analytical uncertainties and no trend matching temporal variations in fallout deposition. Thus, the data appear to be of value in distinguishing nuclear era samples from pre-nuclear era samples but do not provide a means of more accurate definition of time within the nuclear era.

As noted above, Schrag et al. [10] analysed ^{90}Sr and ^{210}Pb in bones of individuals who had been buried in 1999 and exhumed in 2007 and attempted to relate observed ^{90}Sr activities to the calibration curve for ^{90}Sr in vertebrae [103]. The study highlighted that diagenetic contamination presented a significant problem in this approach. Consequently, a "solubility profile" approach was developed, in which sequential selective dissolutions were performed in an attempt to isolate uncontaminated trabecular bone. Schrag et al. [10] applied this method to human remains excavated from a construction site in Switzerland in 2008 and concluded that the observed ^{90}Sr activities in the purified trabecular bone did correspond to the nuclear era. However, it was noted that the observed activity could correspond to either the rising or the falling section of the fallout curve, so ^{210}Pb data for the purified bone were used to infer that the result applied to the rising component. However, they cautioned that even with rigorous analysis, the use of ^{90}Sr still had problems of: (1) variations in ^{90}Sr between individuals as a consequence of geographical and dietary effects and (2) relatively large analytical uncertainties (often>15 %).

Conclusions

Currently, anthropogenic radiocarbon (^{14}C) appears to be the only radionuclide capable of providing information that could be of quantitative use in forensic pathology. Measurements on single bones can be used to determine whether or not the person died during the nuclear era (post-1954 approx.) while recent studies have demonstrated that ^{14}C measurements on trabecular bone have some potential for estimating year of death but this is influenced by the biological age of the remains, with older ages resulting in significant lag times. There has been significant research on dating various components of teeth and estimations of year of birth to within 1–2 years of the true year are entirely possible.

There is no convincing evidence that ^{210}Pb dating can be used in a rigorous, quantitative way to establish PMI. There are significant variations in ^{210}Pb-specific activities between: (1) different bones and different parts of the same bone in an individual, (2) different individuals within the same age group, (3) individuals in different age groups and (4) individuals living in different environments. Nevertheless, unsupported ^{210}Pb will be present in bone and teeth immediately after death, so its presence could, in principle, be used in a qualitative or semi-quantitative way to support other evidence for PMI.

On the basis of: (1) variations in initial activity ratios of daughter/parent pairs from the natural series radionuclides (^{228}Th/^{228}Ra and ^{210}Po/^{210}Pb) in human bone, (2) the large analytical uncertainties and (3) the influence of soil diagenetic processes, it is apparent that the determination of activity ratios does not provide a basis for quantitative estimation of PMI in forensic applications. However, as concluded above for ^{210}Pb, observation of radioactive disequilibrium between ^{210}Pb and ^{210}Po, and ^{228}Ra and ^{228}Th could potentially be used in a qualitative way to support other forensic evidence.

Of the non-^{14}C anthropogenic radionuclides that have entered the environment via atmospheric nuclear weapons tests, discharges from nuclear fuel cycle activities or accidental

releases, ^{90}Sr shows the most promise but still has problems of (1) activity variations between individuals as a consequence of geographical and dietary effects and (2) relatively large analytical uncertainties (often>15 %). Currently, none can provide any more information than the most basic radiocarbon analysis of a single bone and this information has the potential to be misleading if any post-mortem diagenetic contribution is not fully removed prior to analysis.

References

1. Reventlid M, Mörnstad H, Teivens A (1996) Intra- and inter-examiner variations in four dental methods for age estimation of children. Swed Dent J 20:133–139
2. Scheuer L, Black S (2000) Developmental Juvenile Osteology. Elsevier, London
3. Foti B, Adalian P, Signoli M, Ardagna Y, Dutour O, Leonetti G (2001) Limits of the Lamendin method in age determination. Forensic Sci Int 122:101–106
4. Schmitt A, Murail P, Cunha E, Rougé D (2002) Variability of the pattern of aging on the human skeleton: evidence from bone indicators and implications on age at death estimation. J Forensic Sci 47:1203–1209
5. Corsini M-M, Schmitt A, Bruzek J (2005) Aging process variability on the human skeleton: artificial network as an appropriate tool for age at death assessment. Forensic Sci Int 148:163–167
6. Zinka B, Kandlbinder R, Schupfner R, Hass G, Wolfbeis O, Graw M (2012) The activity ratio of ^{228}Th to ^{228}Ra in bone tissues of recently deceased humans: a new dating method in forensic examinations. Anthropol Anz 69:147–157
7. Cook GT, Dunbar E, Black SM, Xu S (2006) A preliminary assessment of age at death determination using the nuclear weapons testing ^{14}C activity of dentine and enamel. Radiocarbon 48:305–313
8. Kandlbinder R, Geißler V, Schupfner R, Wolfbeis O, Zinka B (2009) Analysing of ^{228}Th, ^{232}Th, ^{228}Ra in human bone tissues for the purpose of determining the post mortal interval. J Radioanal Nucl Chem 280:113–119
9. Kondo-Nakamura M, Fukui K, Matsu'ura S, Kondo M, Iwadate K (2011) Single tooth tells us the date of birth. Int J Legal Med 125:873–877
10. Schrag B, Uldin T, Mangin P, Froidevaux P (2012) Dating human skeletal remains using a radiometric method: biogenic versus diagenetic ^{90}Sr and ^{210}Pb in vertebrae. Forensic Sci Int 220:271–278
11. Spalding KL, Buchholz BA, Bergman L-E, Druid H, Frisen J (2005) Age written in teeth by nuclear tests. Nature 437:333–334
12. Ubelaker DH, Bucholz BA, Stewart JEB (2006) Analysis of artificial radiocarbon in different skeletal and dental tissue types to evaluate date of death. J Forensic Sci 51:484–488
13. Ubelaker DH, Parra RC (2011) Radiocarbon analysis of dental enamel and bone to evaluate date of birth and death: perspective from the southern hemisphere. Forensic Sci Int 208:103–107
14. Wang N, Shen CD, Ding P, Yi WX, Sun WD, Liu KX, Ding XF, Fu DP, Yuan J, Yang XY, Zhou LP (2010) Improved application of bomb carbon in teeth for forensic investigation. Radiocarbon 52:706–716
15. Ziad N, Zarki R, Benmansour M, Sayerh T, Laissaoui A (2012) Determination of ^{210}Pb in human skeletal remains from Morocco: implications for time since death assessment. J Radioanal Nucl Chem 292:315–319
16. Zinka B, Kandlbinder R, Hass G, Schupfner R, Wolfbeis O, Graw M (2011) Radionuklidanalyse von ^{228}Th und ^{228}Ra. Rechtsmedzin 21:124–130
17. Suess H (1955) Radiocarbon concentration in modern wood. Science 122(3166):415–417
18. Hua Q, Barbetti M (2004) Review of tropospheric bomb C-14 data for carbon cycle modeling and age calibration purposes. Radiocarbon 46:1273–1298
19. Hedges REM, Clement JG, Thomas CDL, O'Connell TC (2007) Collagen turnover in the adult femoral mid-shaft: modeled from anthropogenic radiocarbon tracer measurements. Am J Phys Anthropol 133:808–816
20. Knight B (1985) The examination of skeletal remains. In: Wecht CH (ed) Legal medicine. Praeger, New York, pp 1–19
21. Libby WF, Berger R, Mead JF, Alexander GV, Ross JF (1964) Replacement rates for human tissue from atmospheric radiocarbon. Science 146:1171–1173
22. Harkness DD, Walton A (1972) Further investigations of the transfer of bomb ^{14}C to man. Nature 240:302–303
23. Geyh MA (2001) Bomb radiocarbon dating of animal tissues and hair. Radiocarbon 43:723–730
24. Taylor RE, Suchey JM, Payen LA, Slota PJ Jr (1989) The use of radiocarbon (^{14}C) to identify human skeletal materials of forensic science interest. J Forensic Sci 34:1196–1205
25. Harkness DD (1983) The extent of natural ^{14}C deficiency in the coastal environment of the United Kingdom. In: Mook WG, Waterbolk HT (eds) PACT 8 - First Int Symp14C and Archaeol, Groningen 1981. 351–364
26. Tauber H (1983) 14C dating of human beings in relation to dietary habits. In: Mook WG, Waterbolk HT (eds) PACT 8 - First Int Symp 14C and Archaeol, Groningen 1981. 365–375
27. Lanting JN, van der Plicht J (1998) Reservoir effects and apparent ages. J Ir Archaeol 9:151–165
28. Cook GT, Bonsall C, Hedges REM, McSweeney K, Boroneant V, Pettitt PB (2001) A freshwater diet-derived reservoir effect at the Stone Age sites in the Iron Gates gorge. Radiocarbon 43:453–460
29. Stuiver M, Pearson GW, Braziunas T (1986) Radiocarbon age calibration of marine samples back to 9000 cal yr BP. Radiocarbon 28:980–1021
30. Stuiver M, Braziunas TF (1993) Modelling atmospheric ^{14}C influences and ^{14}C ages of marine samples to 10,000 BC. Radiocarbon 35:137–189
31. Ascough PL, Cook GT, Church MJ, Dunbar E, Einarsson A, McGovern TH, Dugmore AJ, Perdikaris S, Hastie H, Friðriksson A, Gestsdóttir H (2010) Temporal and spatial variations in freshwater ^{14}C reservoir effects: lake Mývatn, Northern Iceland. Radiocarbon 52:1098–1112
32. Georgiadou E, Stenström K (2010) Bomb-pulse dating of human material: modeling the influence of diet. Radiocarbon 52:800–807
33. Cook GT, MacKenzie AB, Naysmith P, Anderson R (1998) Natural and anthropogenic ^{14}C in the UK coastal marine environment. J Environ Radioact 40:89–111
34. Levin I, Kromer B, Schoch-Fischer H, Bruns M, Münnich M, Berdau D, Vogel JC, Münnich KO (1994) $\delta^{14}CO^2$ record from Vermunt. In Trends: a Compendium of Data on Global Change. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn, USA
35. Levin I, Kromer B (1997) Twenty years of atmospheric CO_2 observations at Schauinsland. Radiocarbon 39:205–218
36. Manning MR, Melhuish WH (1994) Atmospheric ^{14}C record from Wellington. In Trends: a Compendium of Data on Global Change. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn, USA
37. McGee EJ, Gallagher D, Mitchell PI, Baillie M, Brown D, Keogh SM (2004) Recent chronologies for tree rings and terrestrial

archives using ^{14}C bomb fallout history. Geochim Cosmochim Acta 68:2509–2516

38. Broecker WS, Schulert A, Olson EA (1959) Bomb Carbon-14 in human beings. Science 130:331–332
39. Hüls CM, Erlenkeuser H, Nadeau M-J, Grootes PM, Andersen N (2010) Experimental study on the origin of cremated bone apatite carbon. Radiocarbon 52:587–599
40. Berger R, Horney AG, Libby WF (1964) Radiocarbon dating of bone and shell from their organic components. Science 144:999–1001
41. Hassan AA, Termine JD, Haynes CV (1977) Mineralogical studies on bone apatite and their implications for radiocarbon dating. Radiocarbon 19:364–374
42. Tamers MA, Pearson FJ (1965) Validity of radiocarbon dates on bone. Nature 208:1053–1055
43. Van Strydonck M, Boudin M, Hoefkens M, De Mulder G (2005) ^{14}C-dating of cremated bones, why does it work? Lunula Archaeologia Protohistoric 13:61–63
44. Lanting JN, Brindley AL (1998) Dating cremated bone: the dawn of a new era. J Ir Archaeol 9:1–7
45. Van Strydonck M, Boudin M, De Mulder G (2010) The carbon origin of structural carbonate in bone apatite of cremated bones. Radiocarbon 52:578–586
46. Litherland AE (1984) Accelerator mass spectrometry. Nucl Inst Methods Phys Res B 5:100–108
47. Purser KH, Litherland AE, Rucklidge JC (1979) Secondary ion mass spectrometry at close to single-atom concentration using dc accelerators. IEEE Trans Nucl Sci NS-26(1):1338–1346
48. Cunha E, Baccino E, Martrille L, Ramsthaler F, Prieto J, Schuliar Y, Lynnerup N, Cattaneo C (2009) The problem of aging human remains and living individuals: a review. Forensic Sci Int 193:1–13
49. Helfman PM, Bada JL (1976) Aspartic acid racemisation in dentine as a measure of ageing. Nature 262:279–281
50. Ogino T, Ogino H, Nagy B (1985) Application of aspartic acid racemization to forensic odontology: post mortem designation of age at death. Forensic Sci Int 29:259–267
51. Ritz S, Schütz H-W, Schwarzer B (1990) The extent of aspartic acid racemization in dentin: a possible method for a more accurate determination of age at death. Z Rechtsmedizin 103:457–462
52. Ohtani S, Sugimoto H, Sugeno H, Yamamoto S, Yamamoto K (1995) Racemization of aspartic acid in human cementum with age. Arch Oral Biol 40:91–95
53. Bateman H (1910) Solution of a system of differential equations occurring in the theory of radio-active transformations. Proc Camb Philos Soc 15:423–450
54. Thomson J, Walton A (1972) Redetermination of chronology of Aldabra Atoll by ^{230}Th/ ^{234}U dating. Nature 240:145–146
55. Henderson GM, Cohen AS, O'Nions RK (1993) 234U 238U ratios and 230Th ages for Hateruma Atoll corals: implications for coral diagenesis and seawater 234U/238U ratios. Earth Planet Sci Lett 115:65–73
56. Scott RD, MacKenzie AB, Alexander WR (1992) The interpretation of ^{238}U-^{234}U-^{230}Th-^{226}Ra disequilibria produced by rock-water interactions. J Geochem Explor 45:323–343
57. Rubin KH, Smith MC, Perfit MR, Christie DM, Sacks LF (1998) Geochronology and geochemistry of lavas from the 1996 North Gorda Ridge eruption. Deep-Sea Res Part II Top Stud Oceanogr 45: 2571–2597
58. Buesseler KO, McDonnell AMP, Schofield OME, Steinberg DK, Ducklow HW (2010) High particle export over the continental shelf of the west Antarctic Peninsula. Geophys Res Lett 37:L22606
59. Goldberg ED (1963) Geochronology with ^{210}Pb in "Radioactive dating". International Atomic Energy Agency, Vienna, pp 121–131
60. Robbins JA, Edgington DN (1975) Determination of recent sedimentation rates in Lake Michigan using ^{210}Pb and ^{137}Cs. Geochim Cosmochim Acta 39:285–304
61. Appleby PG, Oldfield F (1978) The calculation of Lead-210 dates assuming a constant rate of supply of unsupported ^{210}Pb to the sediment. Catena 5:1–8
62. MacKenzie AB, Farmer JG, Sugden CL (1997) Isotopic evidence of the relative retention and mobility of lead and radiocaesium in Scottish ombrotrophic peats. Sci Total Environ 203:115–127
63. MacKenzie AB, Hardie SML, Farmer JG, Eades LJ, Pulford ID (2011) Analytical and sampling constraints in ^{210}Pb dating. Sci Total Environ 409:1298–1304
64. Appleby PG (2008) Three decades of dating recent sediments by fallout radionuclides: a review. The Holocene 18:83–93
65. Leggett RW (1993) An age specific kinetic model of lead metabolism in humans. Environ Health Perspect 101:598–616
66. Salmon PL, Bondarenko OA, Henshaw DL (1999) DOSE210, a semi-empirical model for prediction of organ distribution and radiation doses from long-term exposure to ^{210}Pb and ^{210}Po. Radiat Prot Dosim 82:175–192
67. Blanchard RL (1966) Correlation of Lead-210 with strontium-90 in human bones. Nature 211:995–996
68. Ladiskaya LA, Parfenov YD, Popov DK, Fedorova AV (1973) ^{210}Pb and ^{210}Po content in air, water, foodstuffs and the human body. Arch Environ Health 27:254–258
69. Fisenne IM (1994) Lead-210 in animal and human bone: a new analytical method. Environ Int 20:427–432
70. Johnston PN, Hult M, Gasparro J, Martinez-Canet M-J, Vasseli R, McKenzie R, Solomon SB, Lambrichts I (2005) The distribution of ^{210}Pb in human bone and its impact on methods for the retrospective estimation of ^{222}Rn exposure from in vivo measurements. J Environ Radioact 80:245–257
71. Salmon PL, Henshaw DL, Keitch PA, Allen JE, Fews AP (1994) TASTRAK spectroscopy of Polonium-210 alpha-particle activity at bone surfaces: evidence for a concentrated surface deposit less than 3 μm deep. Radiat Res 140:63–71
72. James PR, Close JJ, Keitch PA, Allen JE, Fews AP, Henshaw DL (2004) Morphological features of the microdistribution of naturally occurring ^{210}Pb/^{210}Po and ^{226}Ra in the teeth of children and juveniles. Int J Radiat Biol 80:185–198
73. James PR, Close JJ, Keitch PA, Allen JE, Fews AP, Henshaw DL (2004) Aspects of the geographical variations of naturally occurring ^{210}Pb/^{210}Po in permanent teeth of juveniles in the UK. Int J Radiat Biol 80:199–208
74. Swift B, Lauder I, Black S, Norris J (2001) An estimation of the post-mortem interval in human skeletal remains: a radionuclide and trace element approach. Forensic Sci Int 117:73–87
75. Rubin KH, Macdougal JD, Perfit MR (1994) ^{210}Po-^{210}Pb dating of recent volcanic eruptions on the sea floor. Nature 368:841–844
76. Sims KWW, Pichat S, Reagan MK, Kyle PR, Dulaiova H, Dunbar NW, Graham Layne D, Blichert-Toft J, Gauthier PJ, Charette MA, Elliott TR (2013) On the time scales of magma genesis, melt evolution, crystal growth rates and magma degassing in the Erebus volcano magmatic system using the ^{238}U, ^{235}U and ^{232}Th decay series. J Petrol 54:235–271
77. Swift B (1998) Dating human skeletal remains: investigating the viability of measuring the equilibrium between ^{210}Po and ^{210}Pb as a means of estimating the post-mortem interval. Forensic Sci Int 98: 119–126
78. Schmidt S, Cochran JK (2010) Radium and radium-daughter nuclides in carbonates: a brief overview of strategies for determining chronologies. J Environ Radioact 101:530–537
79. Voltaggio M, Andretta D, Tadddeucci A (1987) Dating of newly formed minerals in geothermal fields through ^{232}Th series short lived isotopes: check on minerals of known age and implications to fluid-rock interactions. Geothermics 16:255–261
80. Moore WS, Stakes D (1990) Ages of barite-sulfide chimneys from the Mariana Trough. Earth Planet Sci Lett 100:263–267

81. Reyes A, Moore WS, Stakes DS (1995) ^{228}Th/^{228}Ra ages of a barite rich chimney from the Endeavour segment of the Juan de Fuca Ridge. Earth Planet Sci Lett 131:99–113
82. Ditchburn RG, de Ronde CEJ, Barry B (2012) Radiometric dating of volcanogenic massive sulphides and associated iron oxide crusts with an emphasis on ^{226}Ra/Ba and ^{228}Ra/^{226}Ra in volcanic and hydrothermal processes at intraoceanic arcs. Econ Geol 107:1635–1648
83. Legeleux F, Reyss J-L, Schmidt S (1994) Particle mixing rates in sediments of the northeast tropical Atlantic: evidence from ^{210}Pb$_{xs}$, ^{137}Cs, ^{228}Th$_{xs}$ and ^{234}Th$_{xs}$ downcore distributions. Earth Planet Sci Lett 128:545–562
84. Hancock GJ, Hunter JR (1999) Use of excess ^{210}Pb and ^{228}Th to estimate rates of sediment accumulation and bioturbation in Port Phillip Bay, Australia. Mar Freshw Res 50:533–545
85. Ruiz-Fernandez AC, Hillaire-Marcel C, Ghaleb B, Paez-Osuna F, Soto-Jimenez M (2001) Isotopic constraints (^{210}Pb, ^{228}Th) on the sedimentary dynamics of contaminated sediments from a subtropical coastal lagoon (N W Mexico). Environ Geol 41:74–89
86. Brunnermeier M, Schmied AK, Schupfner R (2012) Distribution of ^{14}C, ^{90}Sr and ^{228}Ra in an elephant tusk. J Radioanal Nucl Chem 292:1285–1290
87. Stehney AF, Lucas HF (2000) Thorium isotopes in autopsy samples from thorium workers. Health Phys 78:8–14
88. McInroy JF, Gonzales ER, Miglio JJ (1992) Measurement of thorium isotopes and ^{228}Ra in soft tissues and bones of a deceased Thorotrast patient. Health Phys 63:54–71
89. Takizawa Y, Qingmei H, Hisamatsu S, Abe T (1997) Concentration of ^{232}Th, ^{230}Th and ^{228}Th in various tissues of Japanese subjects. J Radioanal Nucl Chem 226:191–194
90. Martinez-Canet MJ, Hult M, Johnston PN, Lambrichts I (2001) Ultra-low level gamma ray spectrometry of thorium in human bone samples. Radiat Prot Dosim 97:169–172
91. Kandlbinder R, Schupfner R, Wolfbeis OS, Zinka B (2010) A study on thorium levels in human femur bones. J Radioanal Nucl Chem 285:187–192
92. Kandlbinder R, Schupfner R, Wolfbeis OS (2010) Determination of ^{228}Ra in human bone ash containing significant quantities of ^{40}K and Ca^{2+}. J Radioanal Nucl Chem 283:69–73
93. UNSCEAR (2000) Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 2000 Report to the General Assembly
94. Eades LJ, Farmer JG, MacKenzie AB, Kirika A, Bailey-Watts T (1998) High resolution profile of radiocaesium deposition in Loch Lomond sediments. J Environ Radioact 39:107–116
95. Jones KA, Prosser SL (1997) A comparison of $^{239+240}$Pu post-mortem measurements with estimates based on current ICRP models. J Radioanal Nucl Chem 226:129–133
96. Tandon L, Iyengar GV, Parr RM (1998) A review of radiologically important trace elements in human bones. Appl Radiat Isot 49:903–910
97. Hodgson SA, Ham GJ, Youngman MJ, Etherington G, Stradling GN (2004) A review of measurements of radionuclides in members of the public in the UK. J Radiol Prot 24:369–389
98. Luciani A, Polig E (2007) Surface-seeking radionuclides in the skeleton: current approach and recent developments in biokinetic modelling for humans and beagles. Radiat Prot Dosim 127:140–143
99. Mietelski JW, Golec EB, Tomankiewicz E, Golec J, Nowak S, Szczygiel E, Brudecki K (2011) Human bones obtained from routine joint replacement surgery as a tool for studies of plutonium, americium and ^{90}Sr body-burden in general public. J Environ Radioact 102:559–565
100. Leggett RW (2003) Reliability of the ICRP's dose coefficients for members of the public. III. Plutonium as a case study of uncertainties in the systemic biokinetics of radionuclides. Radiat Prot Dosim 106:103–120
101. Neuzil EF, Dysart ME (1984) The determination of ^{90}Sr and ^{228}Ra in human teeth by age groups and in other substances. Int J Appl Radiat Isot 35:1113–1115
102. Froidevaux P, Geering J-J, Valley J-F (2006) ^{90}Sr in deciduous teeth from 1950 to 2002: the Swiss experience. Sci Total Environ 367:596–605
103. Froidevaux P, Bochud F, Haldimann M (2010) Retention half times in the skeleton of plutonium and ^{90}Sr from above-ground nuclear tests: a retrospective study of the Swiss population. Chemosphere 80:519–524
104. Froidevaux P, Haldimann M (2008) Plutonium from above-ground nuclear tests in milk teeth: investigation of placental transfer in children born between 1951 and 1995 in Switzerland. Environ Health Perspect 116:1731–1734
105. Schmitz M, Schmitz K, Aumann DC (2004) A simple radiochemical method for determining bone-seeking radionuclides in bone and teeth: ^{90}Sr in teeth of children from Ukraine and Germany. J Radioanal Nucl Chem 260:451–457
106. MacLaughlin-Black SM, Herd RJM, Willson K, Myers M, West IE (1992) Strontium-90 as an indicator of time since death: a pilot investigation. Forensic Sci Int 57:51–56
107. Bronk Ramsey C (2009) Bayesian analysis of radiocarbon dates. Radiocarbon 51:337–360

AUTHOR QUERIES

AUTHOR PLEASE ANSWER ALL QUERIES.

Q1. Keywords are desired. Please provide if necessary.
Q2. Figure 3 contains poor quality of text. Please provide replacement. Otherwise, please advise if okay to proceed with the figures as is.