Gamma dosing and shielding of a human tooth by a mandible and skull cap: Monte Carlo simulations and implications for the accuracy of ESR dating of tooth enamel

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Introduction

In recent years, ESR analysis has been used for the direct dating of human tooth enamel (e.g. Thorne et al. 1999, Grün et al. 2003). During the refereeing process of a paper on the ESR dating of an enamel fragment from Skhul, the dating results were objected to because of the unknown dose rate contributions to the tooth from the human mandible and the skull cap, which are in close proximity to the sample. Here we address the problem of dosing and shielding of a human tooth by a mandible and skull cap, and present model calculations for three cases: Skhul, Mungo and Border Cave.

Skull radiation modelling

Dosimetry was modelled using the Monte Carlo radiation transport code MCNP4C (Briesmeister 2000). Input files are employed which contain: a) spatially-resolved materials; b) radiation emission spectra and source locations; c) radiation detector tallies; and d) additional systemic information. MCNP follows the history of each gamma source photon (and any secondary particles generated) by stochastically sampling initially a starting position, direction and energy and then interactions based on tabulated data. Secondary electrons were assumed to be deposited locally. The strength of Monte Carlo methods arises from their ability to calculate radiation transport in complex environments with little increase in systematic uncertainties.

To assess the dose rate contributions of a mandible and a skull cap, we used a computer tomography scan (axial slices) of the Skhul V specimen (see home site of the Peabody Museum, Harvard University) as the basis for the geometry. The data were re-sampled to yield voxels (short for ***volume pixel***, the smallest distinguishable box-shaped part of a three-dimensional image) of approximately 4mm isotropic dimension. Further processing was needed to insert the skull geometry into the Monte Carlo model. We designed a program to divide up the skull and surrounding sediment into several thousand "cells" optimised for computational efficiency. The calculations were carried out for "typical" dry sediment (Garrels and MacKenzie 1971) as 1.775 g cm^{-3} and "typical" wet sediment of 2 g cm^{-3} with water present 25% by weight following Aitken (1985). During the life time of an individual, the average density of dense human bone sections is around 1.9 g cm^{-3} (Cameron et al. 1999, p. 96). During diagenesis, bones may either lose mineral component, i.e. become less dense, or continue to mineralise, i.e. become more dense (Hedges 2002). Calculations were carried out for densities of 1.6 g cm^{-3} and 2.7 g cm^{-3} to cover the possible density range of fossil bones. To calculate the overall dose rates to a tooth from the mandible and skull cap, it is important to know how much of the sediment dose rate from Th and K is shielded. These values were also calculated for the above conditions.

The gamma spectra were downloaded 21 April 2003 from the Evaluated Nuclear Structure Data File (ENSDF) database at the Brookhaven National Laboratory http://www.nndc.bnl.gov. All models were run for approximately 20 million photon histories. MCNP provides energy deposition in detectors normalised to one source particle. The infinite matrix dose (IMD) for each decay group was estimated by determining the energy deposited in a known mass embedded inside a large volume of uniform fossil material. The ratio of tooth self-dose to IMD self-dose, Ψ, can be written:

$$\Psi = \frac{D_{\mathrm{tooth}}}{D_{\mathrm{IMD}}} \frac{m_{\mathrm{skull}}}{m_{\mathrm{tooth}}} \frac{m_{\mathrm{IMD\ detector}}}{m_{\mathrm{IMD\ volume}}} \quad (1)$$

where D is the energy deposited and m is the mass. In an infinite matrix, there exists a state of "radiation equilibrium", such that energy entering a small mass is equal to the energy exiting. In this case, 1-Ψ provides a measure of the attenuation of the IMD within the tooth. Although this is not strictly the case for the models used here, the composition of sediment (average Z-value) is sufficiently similar to the composition of the skull to warrant this approximation and the uncertainties in the attenuation estimates are incorporated in the results (see e.g. gamma attenuation factors of Hubbell 1982).

Results of the modelling

The gamma dose contributions are tabulated in Tables 1a-c ($\rho = 1.6$ g cm^{-3}) and 2a-c ($\rho = 2.7$ g cm^{-3}) as fractions of the bone IMD. Uncertainties are associated with counting statistics of the model.

The most obvious and somewhat surprising result is that the IMD fraction of the complete skull to a specific tooth (Tables 1a and 2a) is not simply the sum of the skull cap (Tables 1b and 2b) and mandible (Tables 1c and 2c). This is caused for a tooth in the mandible (i) by the greater distance to the skull cap compared to a tooth in the upper jaw, and (ii) by partial shielding of the dose rate from the skull cap by the teeth in the upper jaw. The equivalent symmetrical relationships apply to a tooth in the upper jaw. The self-dosing/shielding of a row of four teeth is in the range of 5% of the IMD.

For the U-chains in equilibrium, the wetness of the sediment has virtually no influence on dose rate calculations. The fractions of the whole skull's (ρ = 1.6 g cm^{-3}) uranium IMD received by a lower incisor are 0.0731 (dry sediment) and 0.0737 (wet sediment, see Table 1a), i.e. the differences are less than 1%.

The contribution of the skull to the dose rate of a tooth is strongly dependent on the density of the skull (compare Tables 1 and 2). The percentages of the whole skull's uranium IMD received by a lower incisor are 0.0731 (ρ = 1.6 g cm^{-3}, dry sediment) and 0.1137 % (ρ = 2.7 g cm^{-3}, dry sediment), the latter being about 56% higher.

To calculate the effect of gamma dosing from the mandible and skull cap, two processes have to be considered:

1) U-series disequilibrium. About 93% of the total gamma dose rate from the U-decay chains are generated by the decay of ^{214}Pb and ^{214}Bi. Therefore, the ^{230}Th/^{238}U ratio has to be considered for dose rate calculations.

The activity of ^{230}Th is expressed by:

$$^{230}Th = {}^{234}U(1 - e^{(-\lambda_{230} t)}) = {}^{238}U(1 - e^{(-\lambda_{230} t)})$$

$$if \frac{^{234}U}{^{238}U} = 1 \quad (2)$$

where λ_{230} is the decay constant of ^{230}Th. Equation (2) requires that ^{234}U and ^{238}U are in secular equilibrium at t=0 (i.e. when the uranium is incorporated into the sample). This is usually not the case because most natural waters have an excess of ^{234}U over ^{238}U (e.g. Chapter V in Cherdyntsev 1971). This is due to the fact that ^{234}U is produced by -decay of ^{238}U: when an alpha particle is emitted, the decaying atom recoils, leading to a weakening of its lattice position. Dissolution of minerals starts preferentially at weakened lattice sites, as a consequence, these solutions are enriched with ^{234}U. Alternatively, the recoiled atom can be directly ejected from the mineral surface into the solution. Because of its long half-life of about 244 ka, the excess ^{234}U activity has to be taken into account:

$$\frac{^{230}Th}{^{238}U} = (1 - e^{(-\lambda_{230} t)}) + \frac{\lambda_{230}}{\lambda_{230} - \lambda_{234}} \left(\frac{^{234}U}{^{238}U} - 1\right)(1 - e^{-(\lambda_{230} - \lambda_{234})t}) \quad (3)$$

where λ_{234} is the decay constant of ^{234}U.

2) U-uptake. Most bones and teeth experience U-uptake after burial, and this has a strong influence on dose rate calculations (e.g. Grün 1989). To simplify calculations, it is convenient to calculate the dose a tooth has received from a bone that represents a closed system for U-series (i.e. applying the CSUS-ESR model of Grün 2000), rather than iteratively evaluating average dose rates using the p-value system (Grün et al. 1988). This has the advantage that the dose calculation becomes independent of the actual age of the sample. The differences between

CSUS-ESR and open system p-value dose calculations are small (see Grün 2000).

We now present the gamma dose calculations for three cases:

1) Skhul II (Grün et al., in preparation)
2) Mungo 3 (Thorne et al. 1999) and
3) Border Cave 5 (Grün et al., 2003)

Skhul II (partial mandible, partial skull): To calculate the possible effects of U-dosing of the skull and shielding of sediment gamma dose by the skull, the two extreme ^{230}Th/^{238}U values can be used, that of the dentine of the tooth from Skhul II (Model 1) and the surface layer of a bone from Skhul IX (Model 2). Skhul II consist only of a partial mandible and fragments of the skull (Garrod and Bate 1937, p. 98), therefore, the overall effect of dosing will be smaller than calculated for a whole mandible and skull cap.

Model 1: U-series data were evaluated on the dentine: U(DE) = 60 ppm; ^{234}U/^{238}U = 1.073 ± 0.003; ^{230}Th/^{238}U = 0.275 ± 0.006, resulting in a closed system U-series age of 32.1 ± 0.8 ka and an initial ^{234}U/^{238}U value of 1.080 ± 0.003. The time-averaged ^{230}Th/^{238}U ratio is 0.144 and the ^{231}Pa/^{235}U ratio is 0.274. The average ^{234}U/^{238}U ratio does not have to be considered as the gamma intensities of ^{234}U are very small. Assuming that the U-series data apply to the whole skull, using the values from Tables 1a and 2a and the dose rate values of Adamiec and Aitken (1998), total dry gamma doses of 3.73 Gy (ρ = 1.6 g cm^{-3}) and 5.59 Gy (ρ = 2.7 g cm^{-3}) are obtained.

At the same time, the tooth is shielded by the skull and mandible from the sediment gamma dose rate generated by the radioactive isotopes in the sediment, namely the U and Th decay chains in equilibrium and K. The average composition of the sediment at Skhul is 2.00 ± 0.68 ppm U, 2.38 ± 1.4 ppm Th and 0.45 ± 0.15% K. The total shielded sediment gamma dose in 32.1 ka is 1.22 ± 0.30 Gy (ρ = 1.6 g cm^{-3}) and 1.90 ± 0.48 Gy (ρ = 2.7 g cm^{-3}). Assuming an age of about 120 ka, the shielded doses are 4.56 ± 1.12 and 7.10 ± 1.79 Gy for ρ = 1.6 g cm^{-3} and 2.7 g cm^{-3}, respectively. The net effect of the presence of the skull would lead to corrections of less than 1.6 Gy, or less than 1.6% on a measured dose value of 98.7 ± 7.8 Gy.

Model 2: A bone surface sample from Skhul IX yielded the highest closed system U-series age with 8.7 ppm U, ^{234}U/^{238}U = 1.052 ± 0.002; ^{230}Th/^{238}U = 0.742 ± 0.006, resulting in a closed system U-series age of 131 ± 2 ka and an initial ^{234}U/^{238}U value of 1.075 ± 0.002. The time-averaged ^{230}Th/^{238}U ratio is 0.446 and the ^{231}Pa/^{235}U ratio is 0.662. Using the same procedures as above, we obtain total U-doses of 5.55 and 8.53 Gy for densities of 1.6 and 2.7 g cm^{-3}, respectively. The total shielded gamma doses over 131 ka are 4.98 ± 1.22 and 7.75 ± 1.95 Gy for densities of 1.6 and 2.7 g cm^{-3}, respectively, requiring net dose corrections of less than 0.8 Gy.

As mentioned above, because of the fragmentary nature of the mandible and skull, the overall effect on a tooth from Skhul II is most probably much less than 1.6 Gy (the higher correction value derived from model 1).

Mungo 3 (mandible and skull cap): The average U-concentration in the bone material is 5.5 ± .9 ppm, ^{234}U/^{238}U = 1.319 ± 0.035 (average of TIMS measurements on bones); ^{230}Th/^{238}U = 0.610 ± 0.067 (average of TIMS and gamma spectrometric measurements of the bones and skull), resulting in a closed system U-series age of 65.6 ± 10 ka and an initial ^{234}U/^{238}U value of 1.384 ± 0.035. The time averaged ^{230}Th/^{238}U ratio is 0.339 and the ^{231}Pa/^{235}U ratio is 0.461. This results in a gamma dose from the skull (over the time period of 65.6 ka) of 1.38 and 2.11 Gy for densities of 1.6 and 2.7 g cm^{-3}, respectively. The average composition of the Mungo sediment is 0.27 ± 0.05 ppm U, 0.82 ± 0.04 ppm Th and 0.15 ± 0.01% K. The shielded doses for the same time are 0.57 ± 0.16 and 0.90 ± 0.5 Gy for densities of 1.6 and 2.7 g cm^{-3}, respectively, necessitating corrections of less than 1.21 Gy, or about 4.5% on a measured dose value of 26 ± 2 Gy (the mean EU age would be reduced from 63 to 60 ka). This is smaller than the quoted uncertainty of 6 ka for this age determination (Thorne et al. 1999).

Border Cave 5 (mandible only). The age of the sample has been estimated as 74 ka. The U-concentrations in bones in Border Cave are very low, in the range of 0.2 ppm resulting in a dose rate value from the mandible of less than 1 µGy a^{-1}. A sediment sample in the vicinity of the mandible had a composition of 2.05 ± 0.50 ppm U, 10.8 ± 0.2 ppm Th and 2.87 ± 0.10 % K. The mandible shields about 70 µGy a^{-1} for ρ = 1.6 g cm^{-3}. The bones from Border Cave are not mineralised so that calculations for ρ = 2.7 g cm^{-3} would be misleading. As a result, the total dose rate of 2026 µGy a^{-1} would decrease by 3.5 % and the age of the tooth would increase from about 74 to 77 ka.

Summary

We have calculated the overall effect of uranium gamma dosing and U, Th, and K shielding of the environmental dose rate by a human skull on the total

	Sediment type	^{238}U-^{234}Th	^{230}Th-^{206}Pb	^{235}U-^{231}Th	^{231}Pa-^{207}Pb	U	Th	K
IMD (µGy a^{-1} per ppm U, Th; %K)		2.0	109.0	0.5	1.4	113	48	243
Lower incisor	Dry Wet	0.1657 ± 0.0017 0.1689 ± 0.0017	0.0696 ± 0.0009 0.0701 ± 0.0008	0.2536 ± 0.0013 0.2560 ± 0.0013	0.1502 ± 0.0010 0.1533 ± 0.0010	0.0731 ± 0.0009 0.0737 ± 0.0008	0.0685 ± 0.0013 0.0690 ± 0.0012	0.0605 ± 0.0009 0.0611 ± 0.0012
Upper incisor	Dry Wet	0.1645 ± 0.0021 0.1669 ± 0.0021	0.0699 ± 0.0011 0.0699 ± 0.0011	0.2518 ± 0.0017 0.2570 ± 0.0017	0.1492 ± 0.0013 0.1535 ± 0.0013	0.0734 ± 0.0011 0.0735 ± 0.0011	0.0659 ± 0.0016 0.0661 ± 0.0011	0.0606 ± 0.0012 0.0612 ± 0.0010
Lower molar	Dry Wet	0.1917 ± 0.0011 0.1948 ± 0.0011	0.0858 ± 0.0006 0.0861 ± 0.0006	0.2817 ± 0.0009 0.2850 ± 0.0009	0.1788 ± 0.0007 0.1829 ± 0.0007	0.0897± 0.0006 0.0901 ± 0.0006	0.0839 ± 0.0009 0.0844 ± 0.0006	0.0738 ± 0.0006 0.0743 ± 0.0008
Upper molar	Dry Wet	0.1912 ± 0.0014 0.1947 ± 0.0014	0.0860 ± 0.0007 0.0866 ± 0.0007	0.2796 ± 0.0011 0.2846 ± 0.0011	0.1784 ± 0.0008 0.1831 ± 0.0008	0.0899 ± 0.0007 0.0906 ± 0.0007	0.0831 ± 0.0011 0.0838 ± 0.0015	0.0749 ± 0.0008 0.0755 ± 0.0009

Table 1a: Gamma dose contribution (columns 2-5) and gamma shielding (columns 6-8, U and Th chains in equilibrium) of a **whole skull** as percentage of the infinite matrix dose, IMD (**skull density 1.6 g cm^{-3}**)

	Sediment type	^{238}U-^{234}Th	^{230}Th-^{206}Pb	^{235}U-^{231}Th	^{231}Pa-^{207}Pb	U	Th	K
Upper incisor	Dry	0.1457 ± 0.0019	0.0591 ± 0.0010	0.2327 ± 0.0015	0.1294 ± 0.0011	0.0623 ± 0.0010	0.0573 ± 0.0015	0.0494 ± 0.0010
Upper molar	Dry	0.1618 ± 0.0011	0.0676 ± 0.0006	0.2512 ± 0.0009	0.1474 ± 0.0007	0.0711 ± 0.0006	0.0645 ± 0.0009	0.0584 ± 0.0006

Table 1b: same as Table 1a for the **skull cap.**

	Sediment type	^{238}U-^{234}Th	^{230}Th-^{206}Pb	^{235}U-^{231}Th	^{231}Pa-^{207}Pb	U	Th	K
Lower incisor	Dry	0.1309 ± 0.0004	0.0444 ± 0.0002	0.2163 ± 0.0004	0.1122 ± 0.0003	0.0475 ± 0.0002	0.0425 ± 0.0034	0.0374 ± 0.0024
Lower molar	Dry	0.1454 ± 0.0003	0.0517 ± 0.0001	0.2303 ± 0.0002	0.1255 ± 0.0002	0.0551 ± 0.0001	0.0497 ± 0.0022	0.0441 ± 0.0016

Table 1c: same as Table 1a for the **mandible.**

	Sediment type	**^{238}U-^{234}Th**	**^{230}Th-^{206}Pb**	**^{235}U-^{231}Th**	**^{231}Pa-^{207}Pb**	**U**	**Th**	**K**
Lower incisor	Dry	0.2211 ± 0.0021	0.1096 ± 0.0011	0.3111 ± 0.0016	0.2105 ± 0.0012	0.1137 ± 0.0011	0.1076 ± 0.0013	0.0951 ± 0.0008
Upper incisor	Dry	0.2164 ± 0.0026	0.1106 ± 0.0014	0.3072 ± 0.0020	0.2053 ± 0.0016	0.1145 ± 0.0014	0.1038 ± 0.0014	0.0949 ± 0.0011
Lower molar	Dry	0.2589 ± 0.0014	0.1347 ± 0.0007	0.3496 ± 0.0010	0.2509 ± 0.0008	0.1393 ± 0.0007	0.1313 ± 0.0010	0.1157 ± 0.0007
Upper molar	Dry	0.2571 ± 0.0014	0.1364 ± 0.0007	0.3479 ± 0.0011	0.2475 ± 0.0008	0.1409 ± 0.0007	0.1307 ± 0.0013	0.1179 ± 0.0012

Table 2a: Gamma dose contribution (columns 2-5) and gamma shielding (columns 6-8, U and Th chains in equilibrium) of a **whole skull** as percentage of the infinite matrix dose, IMD (**skull density 2.7 g cm^{-3}**)

	Sediment type	**^{238}U-^{234}Th**	**^{230}Th-^{206}Pb**	**^{235}U-^{231}Th**	**^{231}Pa-^{207}Pb**	**U**	**Th**	**K**
Upper incisor	Dry	0.1901 ± 0.0023	0.0912 ± 0.0012	0.2790 ± 0.0018	0.1769 ± 0.0013	0.0948 ± 0.0012	0.0885 ± 0.0015	0.0757 ± 0.0015
Upper molar	Dry	0.2145 ± 0.0014	0.1065 ± 0.0008	0.3036 ± 0.0011	0.2003 ± 0.0008	0.1105 ± 0.0008	0.0990 ± 0.0006	0.0902 ± 0.0011

Table 2b: same as Table 2a for the **skull cap**.

	Sediment type	**^{238}U-^{234}Th**	**^{230}Th-^{206}Pb**	**^{235}U-^{231}Th**	**^{231}Pa-^{207}Pb**	**U**	**Th**	**K**
Lower incisor	Dry	0.1728 ± 0.0006	0.0707 ± 0.0003	0.2581 ± 0.0004	0.1539 ± 0.0003	0.0744 ± 0.0003	0.0687 ± 0.0033	0.0594 ± 0.0021
Lower molar	Dry	0.1953 ± 0.0004	0.0832 ± 0.0002	0.2779 ± 0.0003	0.1756 ± 0.0002	0.0872 ± 0.0002	0.0798 ± 0.0025	0.0702 ± 0.0021

Table 2c: same as Table 2a for the **mandible**.

dose rate received by a tooth. The presence of bones near a tooth used for ESR dating may influence the resulting age estimate either way, depending on the relative balance between uranin in the bones and radioactive elements on the surrounding sediment, the ages may either increase or decrease (see calculated examples above). As it happens, the net effect is small for the three real cases presented in this study (< 5%). The average uranium dose rate from a whole cap is between about 7 and 14% of the skull's IMD (ρ = 1.6 g cm^{-3} and 2.7 g cm^{-3}). The average shielding of the total environmental dose rate by a complete skull is between about 7 and 12% (ρ = 1.6 g cm^{-3} and 2.7 g cm^{-3}). The effect of the presence of a skull is maximised in cases such as Border Cave, where the sediment dose rate is high and the uranium concentrations in the bones is very low. In other environments, the effect of increased U-concentrations in the bones of the skull (which are usually free of Th and K) is offset by (i) disequilibrium of the U-decay chains in the bones, (ii) delayed U-uptake by the bones, and (iii) shielding of the U, Th and K dose rates from the sediment.

It may be worth mentioning that when applying Tables 1 and 2, the true errors of any calculation will be rather large, as the dose rates are critically dependent on the precise position of the mandible and skull cap relative to the tooth. In most cases, the mandible and skull cap will have separated to some extent and parts of the bones may be missing (e.g. Skhul II). Nevertheless, it is useful to calculate the overall effect of dosing and shielding to estimate the magnitude of possible systematic errors.

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References

Adamiec, G. and Aitken, M.J. (1998). Dose-rate conversion factors: update. *Ancient TL* **16**, 37-50.

Aitken, M.J. (1985) *Thermoluminescence Dating*. Academic Press, New York.

Briesmeister, J.F. (2000) MCNP - A General Monte Carlo N-Particle Transport Code Version 4C. Report LA-13709-M, Los Alamos National Laboratory, USA.

Cherdyntsev, V.V. (1971) *Uranium-234*. Israel Program for Scientific Translations, Jerusalem.

Cameron, J.R., Skofronick, J.G. and Grant, RM. (1999). Physics of the Body. 2nd Ed., Medical Physics Publishing, Madison, WI.

Garrels, R.M., and MacKenzie, F.T. (1971) Evolution of sedimentary rocks. Norton, NY.

Garrod, D. and Bate, D. (1937) *The Stone Age of Mount Carmel. Vol. 1*, Oxford University Press, Oxford.

Grün, R. (1989) Electron spin resonance (ESR) dating. *Quaternary International* **1**, 65-109.

Grün, R. (2000) An alternative model for open system U-series/ESR age calculations: (closed system U-series)-ESR, CSUS-ESR. *Ancient TL* **18**: 1-4.

Grün, R., Beaumont, P., Tobias, P.V. and Eggins, S. (2003) On the age of Border Cave 5 human mandible. *Journal of human Evolution* **45**: 155-167.

Grün, R., Schwarcz, H.P. and Chadam, J.M. (1988) ESR dating of tooth enamel: Coupled correction for U-uptake and U-series disequilibrium. *Nuclear Tracks and Radiation Measurements* **14**: 237-241.

Grün, R., Stringer, C.B., Pike, A.W.G., McDermott, F., Nathan, R., Porat, N., Robertson, S., Taylor, L., Mortimer, G., Eggins, S., and McCulloch, M. (in preparation). Dating the human burials from Skhul.

Hedges, R.E.M (2002) Bone diagenesis: An overview of processes. *Archaeometry* **44**: 319-328.

Hubbell, J.H. (1982) Photo Mass Attenuation and Energy-absorption Coefficients from 1 keV to 20 MeV. *Applied Radiation and Isotopes* **33**: 1269-1290.

Thorne, A., Grün, R., Mortimer, G., Spooner, N.A., Simpson, J.J., McCulloch, M.T., Taylor, L. and Curnoe, D. (1999) Australia's oldest human remains: age of the Lake Mungo 3 skeleton. *Journal of Human Evolution* **36**: 591-612.

Reviewer

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