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ON THE CONTRIBUTION OF RADIOACTIVE FALLOUT ISOTOPES TO THE TOTAL DOSE RATE IN DATING OF YOUNG SEDIMENTS

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Abstract: Palaeodosimetric methods of dating, especially the OSL method, are now capable of dating very recent sediments, that if formed within the last 100 years, experienced a contribution to the absorbed dose from the fallout radioisotopes, the contribution that forms a significant portion of the total absorbed dose. The paper presents results of calculations of this contribution from the radioisotopes other than ^{137}Cs and finds it of approximately the same magnitude as dose received from ^{137}Cs alone, meaning that it should be taken into account in the case of high precision dating of recently formed sediments.

1. INTRODUCTION

The palaeodosimetric methods of dating are now capable of estimating the dose absorbed by mineral grains in sediments with both good accuracy and precision. For example, the total absorbed dose may be measured by means of the OSL SAR protocol with an accuracy of 1% or better, at the level down to tenths of a Gray. That means that the resulting accuracy of dating the sediment is now limited by the accuracy of estimating the average annual dose (or the absorbed dose rate). In the previous paper (Moska, *et al.*, 2004), we have shown that for young sediments the contribution of ^{137}Cs present in the sediment as result of radioactive fallout may amount to a significant figure of few to more than a dozen percent of the total dose; that means more than a typical uncertainty of dose rate estimates.

In this paper we continue along these lines and show that other fallout radioisotopes present in the global fallout and in the local fallout following the Chernobyl nuclear accident contribute to the total dose at the similar level.

Because the presence and numerical estimates of concentrations of fallout radioisotopes other than ^{137}Cs is less straightforward, we propose the way to obtain reliable estimates that eventually should improve the quality of the dating result.

2. FALLOUT COMPOSITION

The main components of the atmospheric fallout are gathered in **Table 1** together with data relevant to absorbed dose rate assessment (because in the previous paper, *ib.*, the references have been misquoted, we repeat this table here). The total contribution to the absorbed dose are given for the assumed coarse grains with 90% effectiveness of beta dose caused by the attenuation. The source of the numerical data and details of calculations have been already given *ib.*, so we do not repeat them here. From these isotopes only two (^{137}Cs, ^{90}Sr) are still present in the fallout and in the soils or sediments. While the presence of ^{137}Cs in the sediment is easily detected by gamma spectrometry, the detection of ^{90}Sr (or its daughter ^{90}Y) requires a more complicated chemical extraction and then beta spectrometry or beta counting techniques (Schimmack *et al.*, 2003). Because of that we quote the relative amounts of fallout isotopes after Mellander (1986) and after Haak and Rydberg (1998). If the specific activity of ^{90}Sr (^{90}Y) is not measured directly, then its amount may be estimated basing on given data.

Because the half-lives and dose rates values of these isotopes represent quite wide ranges we have calculated the total potential absorbed dose for each of them. This quantity is defined here as the total dose delivered to the sediment when the isotope undergoes a complete decay.

$$D_t = \frac{\tau_{1/2}}{ln\,2} \cdot c_0 \cdot d_f \qquad (2.1)$$

where: $\tau_{1/2}$ – the half-live of the radioisotope in years, c_0 – the initial specific activity in sediment in Bq/kg, d_f – the annual dose in μGy/a per unit specific activity of 1 Bq/kg. To compare them directly and to assess the importance of each of radioisotopes for the dosimetry of the dated sediment we have calculated the total potential doses D_t for the assumed 100 Bq/kg initial specific activity of ^{137}Cs and further assuming that initial specific activities of other isotopes presented in **Table 1** are given by relative coefficients a_r

$$D_{it} = \frac{\tau_{i1/2}}{ln\,2} \cdot a_{ir} \cdot c_{Cs} \cdot d_{if} \qquad (2.2)$$

where: i – indexes isotopes listed in **Table 1**, c_{Cs} – the initial specific activity of ^{137}Cs in the sediment (equal to 100 Bq/kg for the sake of further calculations).

In **Table 2,** we give calculated values of effective annual dose rates (calculated from the total contribution in **Table 1**), total potential dose rates and time after deposition when the actual absorbed dose for a given isotope reaches the indicated fraction of the total potential dose.

It is easily seen from the data given in **Table 2** that the contributions of ^{125}Sb, ^{106}Ru, ^{144}Ce and ^{131}I may be neglected for any practical purpose. If ^{137}Cs is present in the recent sediment (i.e., less than ca 60 years old) the contributions from ^{137}Cs and ^{90}Sr should be taken into account. The contribution of ^{134}Cs may be significant only for sediments originated shortly after the Chernobyl accident and only for areas with the actual post-Chernobyl fallout.

3. POSSIBLE SCENARIOS

For practical purposes we may consider two simplified scenarios. The first scenario accounts for nuclear test fallout only, that is it assumes no fallout following Chernobyl accident. The second scenario assumes certain proportion of both fallout sources. The period of intense nuclear weapon tests had ended around mid sixties so we also assume that after the year 1966 the global fallout consisted of isotopes injected earlier to the stratosphere.

1-st scenario

Unless the age of the sediment coincides with the period of the nuclear weapon tests we may further neglect the contribution from ^{134}Cs and either use the measured ^{90}Sr specific activity or double the dose resulting from ^{137}Cs content. In this scenario the modified age equation (cf. Moska *et al.*, 2004) takes one of the following forms:

$$D_n T + \frac{\tau_{Cs} C_{Cs} d_{Cs}}{ln(2)} \left(exp\left[\frac{ln(2)}{\tau_{Cs}} T \right] - 1 \right) + \frac{\tau_{Sr} C_{Sr} d_{Sr}}{ln(2)} \left(exp\left[\frac{ln(2)}{\tau_{Sr}} T \right] - 1 \right) = D_e \qquad (3.1)$$

or

$$D_n T + 2\frac{\tau_{Cs} C_{Cs} d_{Cs}}{ln(2)} \left(exp\left[\frac{ln(2)}{\tau_{Cs}} T \right] - 1 \right) = D_e \qquad (3.2)$$

where: T – is the age of the sediment in years, τ_{Cs}, τ_{Sr} – are the half-lives of ^{137}Cs and ^{90}Sr respectively expressed in years, C_{Cs}, C_{Sr} – the specific activities as measured now and d_{Cs}, d_{Sr} – are annual doses per unit specific activity. Doubling the ^{137}Cs dose contribution is justified by the fact that typically the amount of ^{90}Sr in the fallout is 2/3 of ^{137}Cs (Mellander, 1986) while its specific annual dose is about 30% higher (cf. **Tables 1** and **2**).

Table 1. *Data on selected radioactive fallout isotopes.*

Isotope	Half-life $\tau_{1/2}$	Amount relative to ^{137}Cs[1], a_r	Total energy released per parent disintegration (MeV)		Total contribution[2] to the dose (MeV)
			E_γ	E_β	
^{137}Cs	30.07 ± 0.03 a	1	0.565	0.245	0.786
^{90}Sr – ^{90}Y	28.79 ± 0.06 a	0.66[3] 0.01[3]	0.043	1.130	1.060
^{125}Sb	2.75856 ± 0.00025 a	0.05	0.434	0.097	0.521
^{134}Cs	2.0648 ± 0.0010 a	0.61	1.555	0.164	1.703
^{106}Ru	373.59 ± 0.15 d	0.26	0.000	0.010	0.009
^{144}Ce	284.91 ± 0.05 d	0.07	0.019	0.092	0.102
^{131}I	8.02070 ± 0.00011 d	2.95	0.382	0.192	0.555

[1] the composition of the Chernobyl fallout relative to ^{137}Cs (except for ^{90}Sr) after Mellander (1986); [2] total contribution is calculated as $0.90 \cdot E_\beta + E_\gamma$; [3] data for ^{90}Sr - ^{90}Y are quoted after Haak and Rydberg (1998): the higher value is for the nuclear test fallout, the lower value is for the Chernobyl fallout; a=years, d=days; E_γ, E_β – energies of gamma and beta rays.

The obtained year of sedimentation should fall between 1966 and 1986. For sediments deposited after 1986 the 2-nd scenario should be considered. Sediments deposited during the period of nuclear tests require more complicated treatment that is beyond the scope of this paper (see a comment at the end of this chapter).

2-nd scenario

The modified age equation contains now one more term:

$$D_n T + \frac{\tau_{Cs} C_{Cs} d_{Cs}}{ln(2)} \left(exp\left[\frac{ln(2)}{\tau_{Cs}} T \right] - 1 \right) + \frac{\tau_{Sr} C_{Sr} d_{Sr}}{ln(2)} \left(exp\left[\frac{ln(2)}{\tau_{Sr}} T \right] - 1 \right) + \frac{\tau''_{Cs} C''_{Cs} d''_{Cs}}{ln(2)} \left(exp\left[\frac{ln(2)}{\tau''_{Cs}} T \right] - 1 \right) = D_e \quad (3.3)$$

where: $\tau''_{Cs}, C''_{Cs}, d''_{Cs}$ - are as above but for ^{134}Cs.

The activity of ^{134}Cs cannot be detected at present so we may assume it was deposited in the proportion to ^{137}Cs as quoted in **Table 2**. Then its specific activity can be calculated as:

$$C''_{Cs} = 0.61 a C_{Cs}\, exp\left[\left(\frac{1}{30.07} - \frac{1}{2.06} \right) ln(2)(Y_0 - 1986) \right] \quad (3.4)$$

where: the numerical factor 0.61 is taken from **Table 2**, a – is the proportion of after Chernobyl ^{137}Cs fallout in the total ^{137}Cs fallout, and Y_0 – is the year of measurements. The proportion a should be evaluated regionally basing on available fallout data (Dubois and Bossew, 2003; Playford *et al.*, 1990; Aarkrog *et al.*, 1992; Strzelecki *et al.*, 1994; Aoyama and Hirose, 2001; Haak and Rydberg, 1998; Mellander, 1986).

If ^{90}Sr is not measured in the sediment then again it may be assumed to be in proportion to the nuclear test ^{137}Cs (**Table 1**). The measured ^{90}Sr activity may be used as indication of the nuclear test fallout since it was almost not present in the Chernobyl fallout (Haak and Rydberg, 1998).

The resulting year of sedimentation should be now equal or later than 1986.

Let us consider now a numerical example of a hypothetical sediment that was sampled and measurements were done at the end of 2005 yielding the following results: the equivalent dose D_e = 60 mGy, natural annual dose D_n = 2 mGy/a and the specific activity of ^{137}Cs C_{Cs} = 30 Bq/kg. These values are quite typical for diluvial sediments deposited within the last 50 years. If we neglect the possible contribution of the fallout dose then the age is given by the ratio D_e / D_n = 30 years.

If we consider the numerical values assumed in the example above then only the *1-st scenario* yields consistent results. The estimated age is equal to 26 years when the activity of ^{90}Sr is assumed to be in proportion to ^{137}Cs given in tables, and differs by about 1 year from the value obtained when ^{137}Cs contribution is doubled.

Table 2. *Effective dose rates, total potential doses (cf. Eq. 2.2)*

Isotope	^{137}Cs	^{90}Sr - ^{90}Y	^{134}Cs	^{125}Sb	^{106}Ru	^{144}Ce	^{131}I
annual dose d_f in µGy/a per 1 Bq/kg	3.976	5.139	8.607	2.641	0.046	0.474	2.803
amount a_r relative to ^{137}Cs	1	0.66	0.61	0.05	0.26	0.07	2.95
total potential dose for 100 Bq/kg ^{137}Cs, (mGy)	17.25	14.09	1.56	$5.26 \cdot 10^{-02}$	$1.76 \cdot 10^{-03}$	$3.73 \cdot 10^{-03}$	$2.62 \cdot 10^{-02}$
fraction of total dose	time in years after which the given fraction of the total dose is delivered						
0.1	4.6	4.4	0.3	0.4	0.2	0.1	0.003
0.2	9.7	9.3	0.7	0.9	0.3	0.3	0.007
0.3	15.5	14.8	1.1	1.4	0.5	0.4	0.011
0.4	22.2	21.2	1.5	2.0	0.8	0.6	0.016
0.5	30.1	28.8	2.1	2.8	1.0	0.8	0.022
0.6	39.8	38.1	2.7	3.6	1.4	1.0	0.029
0.7	52.2	50.0	3.6	4.8	1.8	1.4	0.038
0.8	69.8	66.8	4.8	6.4	2.4	1.8	0.051
0.9	99.9	95.6	6.9	9.2	3.4	2.6	0.073
0.95	130	124	8.9	11.9	4.4	3.4	0.095
0.99	200	191	13.7	18.3	6.8	5.2	0.146

If the period of sedimentation coincides with the peak fallout between 1956 – 1966 then a more elaborate model should be used, one that takes into account detailed fallout data available, for example, from Aoyama and Hirose (2001).

4. CONCLUSIONS

The proposed approach and modified age equations should be used in case of recent sediments, younger than 50 years, that contain measurable amounts of ^{137}Cs indicating the presence of fallout radioisotopes in the sediment. The results of this study show that obtained ages of sediments may differ by about 10% when the fallout radioactivity of ^{137}Cs and ^{90}Sr in the sediment is neglected.

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