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A note on overcounting in alpha-counters and its elimination

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Summary

Overcounting in alpha-counters is found to depend on the grain size distribution of the sample rather than from radon escape. The U- and Th- activity of several sediments was measured by alpha-counting and neutron activation analysis. Overcounting was detected in samples with a considerable grain size fraction larger than 20 µm. Further decrease of the grain size by grinding resulted in good agreement of the alpha dose derived from alpha-counting with the alpha dose calculated from neutron activation data. Concentrations of Th- and K in loess samples increase with decreasing grain size of the unpowdered sample, whereas the U-distribution appears to be more homogenous.

Introduction

Overcounting in alpha-counters has been observed in numerous cases. The most striking example is the "Hong Kong sand" with a count rate of ca. 160% higher than expected from concentrations of U and Th, which were determined by neutron activation analysis (NAA) (cf. Pernicka & Wagner, 1982). In our laboratory, α-overcounting proved to be a serious problem for TL-dating of some loess samples from Rotenberg and Nussloch, south of Heidelberg, West Germany (Zöller et al., 1988). The α-dose calculated from NAA data was up to about 30% lower than the values obtained from α-counting. If the β- and γ-dose rates are derived from the α-count rate, the error of TL age determination is even larger. There was no systematic difference between the count rates of sealed and unsealed samples, which is usually taken as indication of radon escape from the sample. In some cases, the sealed count rate was even slightly lower than the unsealed one. This may be explained by electrostatic attraction of radon atoms to the perspex holder immediately after filling in the ground sample. After some hours or days of delay this effect disappears. We observed this effect in many other loess samples, too, and it may also be the cause for some low sealed/unsealed count-rate ratios published by Wintle (1987) and Juvignè & Wintle (1988). Since radon escape obviously cannot account for α-overcounting satisfactorily, we looked for other possible explanations.

Sedimentological characteristics of the loess samples

The loesses from the sections near Rotenberg (RO) and Nussloch (NU) south of Heidelberg are characterized by different grain-size distributions with a larger median diameter compared with "normal" German loesses. Most of them can be classified as sandy loess. This is readily explained by a short transportation distance from the source area, namely the large plain of the Upper Rhine valley, which was drained by a braided river system during the pleniglacial (Bente & Löscher, 1987). In some parts of the Middle Würmian (=lower pleniglacial) loess sequences even aeolian sands occur. The mean clay content of the loesses (<10%) is unusually low, while sand contents often exceed 10%. The maximum of the grain size distribution lies within the middle and coarse silt fractions (cf. fig.1, after Bente & Löscher, 1987). However, in paleosols decomposition of mineral grains due to soil formation results in higher clay contents at the expense of sand and coarse silt. In the case of para-brownearths, pores of the Bt-horizon are coated with clay (NU-4). In these samples overcounting is less pronounced.

Experimental details

Two scintillation α–counters (Littlemore Eng. Co.) were used for thick source α– counting. Round screens with a diameter of 42 mm and a coating of ZnS were covered with ca. 1 mm of loess. Both α-counters were calibrated with two standards, whose U and Th contents have been determined by NAA. Secular equilibrium was confirmed by α- and γ-spectrometry. These standards were: O-110 (obsidian, grain size <50 µm, expected count-rate 4.54 ± 0.10 min^{-1}), and TONY (sedimentary clay, expected count-rate 1.11 ± 0.03 min^{-1}). Most samples were counted on both counters (1000 counts minimum). After a first count the sample holders were sealed and the α- activity counted again after a delay of several days. Originally the loess aggregates were only slightly crushed using an agate mortar without breaking up single grains. The α-counting procedure was repeated after powdering the samples for 15 min in an agate ball mill. U and Th have been determined by instrumental neutron activation (Pernicka & Wagner, 1982). Then alpha-dose-rates were calculated from NAA data using conversion factors calculated by Nambi & Aitken (1986).

Results

Count-rates of selected samples, together with concentrations of U and Th, are listed in table 1. Overcounting is most prominent with those loess samples which consist essentially of middle and coarse silt with relatively high sand and low clay contents (NU- and RO-series, except for NU-4 from a strongly altered soil horizon).

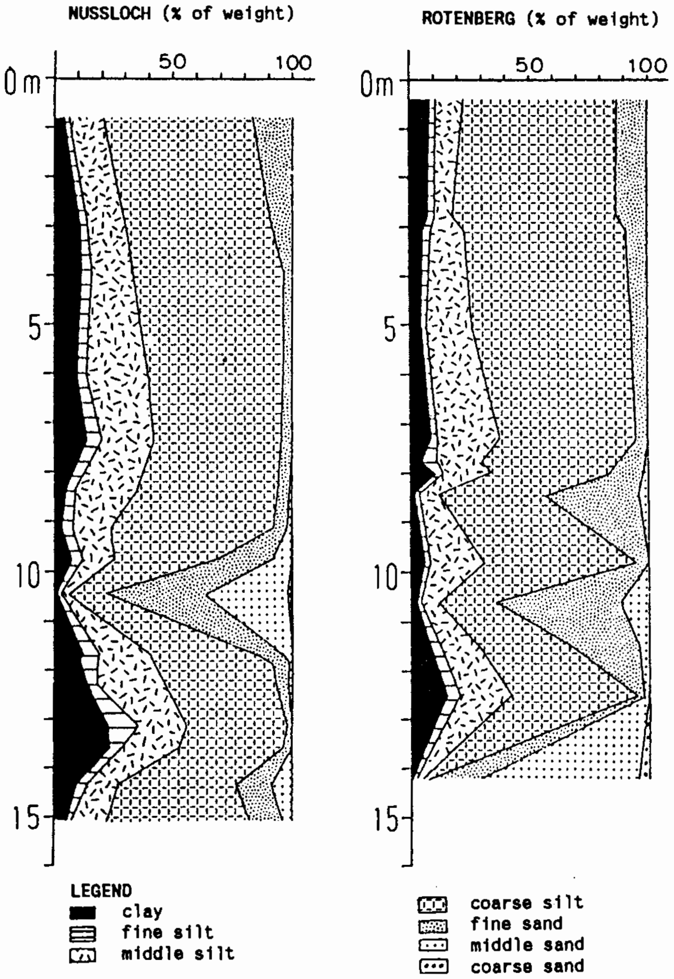

Figure 1 Grain size distributions in the loess sections of Nussloch and Rotenberg south of Heidelberg, W-Germany. Except for sandy layers, the coarse silt fraction (20-63 μm) is the main grain size fraction of these loesses. The high clay content at a depth of 12.5 to 14 m in the Nussloch section is due to interglacial weathering and soil formation.

Samples of "normal" fine grained loess (KAE-series, RD-8) from the Kaerlich pit and from Rheindahlen do not exhibit overcounting; in the case of KAE-4, the α–count-rate is even 10.5% lower than expected from NAA data, assuming secular equilibrium. In a sandy silt from tidal deposits (WA-4) from the Wacken pit α–overcounting amounts to 49%, whereas for powdered quartz grains from the same exposure (WA-2) the results are in excellent agreement with NAA despite their extremely low α–activity. Table 2 demonstrates the effect of powdering on the α–count-rate. The count-rates of coarse grained samples of the RO-series are about 10-46% higher than the count-rates of the same samples after they have been powdered. The maximum value for overcounting was found for the "Hong Kong sand" (see Pernicka & Wagner, 1982). Even this sample after grinding to a grain size <20 μm comes to an α–count-rate within 5% of the expected count-rate from U and Th contents. On the other hand α–count-rates of samples from "normal" grained loesses (SO-series) from the Bad Soden pit are much less affected by powdering. Overcounting ranges between 2 and 15% only. In the case of sample RO-8p (powdered) , the α–dose-rate derived from the α–count-rate agrees excellently with the α–dose-rate calculated from NAA data. This sample was also analyzed by α–spectrometry and found to be in secular equilibrium.

Obviously it is not necessary to immobilize radon for α–counting using a resin (Murray, 1982) or a glassy melt (Jensen & Prescott, 1983, Huntley et al.,1986,). Therefore it seems that overcounting is not primarily caused by diffusion of radon to the ZnS screen but by the gaps filled with air, which result when the grain size of the sample is of the same order as the range of the α–particles in the solid (ea. 25 μm), and when there are few smaller grains to fill the gaps.

In order to test this hypothesis, we separated different grain size fractions of the sample RO-8 with equivalent diameters of <2 μm, 2 - 6.3 μm, 6.3 - 20 μm, and 20-63 μm by sedimentation in Atterberg glass cylinders. In these four grain size fractions U-, Th-, and K- concentrations were measured by NAA and compared with their α–activities. The results listed in table 3 show a significant increase of Th- and K-concentrations with decreasing grain size, whereas U-concentrations are relatively constant (NAA data). The slightly higher U-concentration in the fraction >20 μm may be due to zircon grains, because the maximum zircon-concentration in sediments is normally observed within the fine sand fraction, 63-200 μm. The α–dose-rates calculated from NAA data and from the α–count-rates of the four grain size fractions agree within 10% only after grinding. The agreement is less without grinding. These results confirm that:

i) within the range of α-particles there exists an inhomogeneous distribution of radioactivity in loess,
ii) radioactivity in loess is concentrated in the finest grain size fractions, and
iii) overcounting is probably dominated by a geometric effect due to large grain sizes, because the air filling the gaps between large grains has a significantly lower stopping power for α-particles. Therefore in many cases overcounting can not be detected by counting the sample sealed and unsealed.

Conclusions

If α–counting is used to estimate dose-rates of U- and Th-decay chains for TL dating, significant underestimates of TL ages may result from α-overcounting, which may remain undetected by the usual practice of counting a sample with the container open and closed. For example, 60.5% of the effective dose-rate derives from U- and Th-decay chains in loess, if typical values (3 ppm U, 10 ppm Th, 1.5% K, cosmic dose-rate = 0.15 Gy/ka, *a* value = 0.09, and δ=1,15 = moist weight/dry weight) are assumed. Loesses of local origin, i.e. with short transport distances, frequently contain coarser grains and hence α-overcounting can be expected for unpowdered sample aliquots. These problems can be overcome by simple powdering of the loess before α-counting. Melting of the sample as was suggested by Huntley et al. (1986) does not seem to be necessary in the case of loess.

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PI. Reviewer's comments (Martin Aitken)

It is now nearly 10 years since strong evidence for overcounting was reported at the 1980 Specialist Seminar (Murray 1982; Pernicka and Wagner, 1982). That thick source alpha counting continues in extensive routine use nevertheless is indicative of its high sensitivity, its convenience, and its direct relevance to the evaluation of alpha dose-rate. The above authors reported the effect primarily in respect of pottery though it was also evident in soil and sand. The present authors report that whereas in some samples of 'normal' fine-grains loess the effect was absent, in some coarser loess the overcounting was strong; they conclude that by powdering the sample (following Jensen and Prescott, 1983) the effect can be avoided though giving direct comparison with neutron activation only for 3 samples: RO8, WA2 and HS. Overcounting was also reported recently by Wintle and Dijkmans (1988) in respect of some Dutch sands; although they too found the effect to be substantially reduced by crushing, the crushed count-rate remained substantially in excess of that predicted by gamma spectrometry (by between 20% and 40% for their 6 samples measured). On the face of it, this suggests that crushing is effective in some samples but not in others. However it must be borne in mind that unless the state of radioactive equilibrium has been measured, any comparison with prediction is far from definitive. Z & P state that RO8 has been found to be in equilibrium but there is no information given on the Dutch sands.

The finding that the potassium and thorium contents are substantially higher in the O - 2 μm fraction of sample RO8 parallels that for a sample of pottery as reported by Murray (1982a), who refers to a similar finding by Scott (1968) in respect of river sediment. I agree that this unhomogeneity in radioactivity is likely to contribute to overcounting but I think that radon emanation may be of major importance too. Unfortunately the sealed/unsealed test for emanation does not appear to be working in the present work and so there is no reliable evidence as to whether these samples emanate. In tables 1 - 3 the sealed count-rate is never more than 10% high than the unsealed, except in one instance (the O - 2 μm fraction of RO8). However the samples include the notorious Hong Kong sand (HS) for which strong emanation is well attested: Pernicka and Wagner (1982) report a radon escape of 21%; at Oxford, Huxtable observes a sealed/unsealed ratio 1.3 - 1.4 depending on the degree of powdering. The authors find a ratio of only 1.05.

In any quantitative assessment of the degree of overcounting, the basis used for calibration is relevant. The present authors mention use of two standards for calibration but for both these the observed count-rate is in excess of that predicted by neutron activated by 8% and 6%. There is now a wide range of geological standards available (mostly as fine powder) and it would seem sense for all alpha counting laboratories to quote the value obtained for at least one of these when publishing; results for some of these standards have been given, for instance, by Goedicke (1983), Aitken (1985) and Huntley et al (1986). The Oxford labaratory still has limited supplies available for send-out, of Sand 105A (10.2ppm U; pitchblende in silica) and Sand 109 (104 ppm Th; 3.7 ppm U; monazite in silica) from the New Brunswick Laboratory.

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Neutron activation analysis				**alpha counter II**			
	U (ppm)	Th (ppm)	Dα (Gy/ka)	count-rate unsealed	Dα	count-rate sealed	Dα
NU1	2.9	9.6	12.56	0.766	16.47	0.788	16.94
NU2	2.9	10.6	13.19	0.708	15.22	0.699	15.03
NU3	3.5	11.0	14.78	0.806	17.33	-	-
NU4	3.3	14.9	16.82	0.829	17.82	0.823	17.69
NU5	2.7	8.7	11.54	0.774	16.64	-	-
RO1	2.9	8.2	11.67	0.734	15.78	-	-
RO8	2.8	9.5	12.27	0.719	15.46	-	-
KAE1	3.2	12.0	14.75	0.626	13.46	0.679	14.60
KAE2	3.4	11.5	14.88	0.661	14.21	0.695	14.94
KAE4	3.5	15.0	17.33	0.722	15.52	0.680	14.62
RD8	3.3	12.5	15.29	0.670	14.41	-	-
WA4	1.58	4.82	6.58	0.456	9.80	-	-
WA2p	0.09	0.17	0.31	0.0127	0.27	-	-
HS	3.78	16.4	18.84	2.220	47.73	2.33	49.63
Standards (Errors are 3% for Th in both standards and 3% for U in O-110 and 5% for U in TONY.)							
O110	28.3	54.5	97.52	-	-	4.916	105.7
TONY	5.79	17.15	23.80	-	-	1.179	25.35

Table 1 Comparison of alpha count-rates and neutron activation analysis data (α dose-rates in Gy/ka, α-count-rates in min^{-1} on 42 mm diameter ZnS screen). NU=Nussloch loess or soil, RO=Rotenberg loess or soil, KAE=Kaerlich loess or soil, RD =Rheindahlen decalcified loess, WA4=Wacken, sandy silt, WA2=Wacken quartz sand, HS=Hong Kong sand, p=powdered in agate mill.

Sample	**alpha counter I**				**alpha counter II**			
	unsealed		sealed		unsealed		sealed	
	c - r	Dα	c - r	Dα	c - r	Dα	c - r	Dα
RO2	-	-	-	-	0.836	17.97		
RO2p	0.556	11.95	0.541	11.84	0.577	12.29	0.557	11.86
RO3	-	-	-	-	0.673	14.47	-	-
RO3p	0.606	13.03	-	-	0.611	13.14	-	-
RO6	-	-	-	-	0.798	17.16		
RO6p	0.583	12.54	0.487?	10.46?	0.563	12.11	0.571	12.16
RO7	-	-	-	-	0.757	16.28	0.707	15.20
RO7p	0.581	12.49	-	-	0.557	11.98	0.548	11.79
RO8	0.667	14.34	0.680	14.49	0.734	15.78	-	-
RO8p	0.585	12.58	0.568	12.10	-	-	0.590	12.57
SO2	-	-	-	-	0.74	15.90	-	-
SO2p	0.665	14.30	0.692	14.88	-	-	0.703	15.11
SO3	-	-	-	-	0.706	15.18	0.687	14.77
SO3p	0.571	12.28	0.556	12.17	-	-	0 599	12.88
SO4	-	-	-	-	0.653	14.04	-	-
SO4p	0.602	12.94	0.578	12.43	0.642	13.80	0.697	14.99
HS	-	-	-	-	2.220	47.73	2.330	49.63
HSp	-	-	-	-	0.984	20.96	-	-

Table 2 Effect of powdering of loess samples on the α-count-rates ("p" after sample code means powdered in an agate mill). SO=Bad Soden loess or soil.

	NAA				**αcount rate (I)**			
fraction (μm)	U (ppm)	Th (ppm)	K (%)	Dα (Gy/ka)	unsealed	Dα (Gy/ka)	sealed	Dα (Gy/ka)
0-2	2.78	22.8	2.81	20.69	0.949	20.40	1.045	22.47
2-6.3	2.34	12.7	2.61	13.28	0.732	15.74	0.726	15.61
6.3-20	2.79	7.8	1.59	11.17	0.590	12.69	-	-
6.3-20p					0.500	10.75	-	-
20-63	3.00	7.8	0.93	11.64	0.703	15.11	-	-
20-63p					0.605	13.01	-	-

Table 3 Neutron activation analysis data and alpha-count-rates from different grain size fractions (clay to coarse silt fraction) of sample RO-8 (p=powdered fraction).