COSMIC BACKGROUND REDUCTION IN THE RADIOCARBON MEASUREMENTS BY LIQUID SCINTILLATION SPECTROMETRY AT THE UNDERGROUND LABORATORY OF GRAN SASSO

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ABSTRACT. Radiocarbon measurements by two 1220 Quantulus™ ultra low background liquid scintillation spectrometers were performed at the underground laboratory of Gran Sasso and the Radiocarbon Laboratory of E.N.E.A.-Bologna to study the efficiency and background variations related to measurement sites. The same configuration setup, i.e. the same center of gravity of the ^{14}C spectrum (SQP(I) = 410 ± 1) was obtained in both instruments. Many different background and modern standards with pure analytical benzene were used and spectra for 40 one-hour periods were obtained. The data indicates a background reduction of approximately 65% between the surface and underground laboratories, with no differences in the efficiency. Recording similar efficiencies in both spectrometers is probably due to fairly identical photomultiplier characteristics. The cosmic noise reduction observed at the laboratory of Gran Sasso makes it possible to perform high precision ^{14}C measurements and to extend for these idealized samples the present maximum dating limit from 58,000 BP to 62,000 BP (5 mL, 3 days counting).

INTRODUCTION

The underground laboratory of Gran Sasso is located in the central Apennines (Italy) at 963 m above the sea level. The maximum thickness of the rock overburden is 1400 m, corresponding to 3800 m of water. The rock has a density of 2.71 g/cm³, a mean nuclear charge number (Z) equal to 9.4 and a low rate of natural radioactivity. The underground laboratory is characterized by a cosmic-ray flux that is extremely reduced compared with the external flux; in fact, only neutrinos and high energy muons can penetrate through the thick rock. Due to their low interaction probability, incoming neutrinos can filter through the rock independently of their energy, but of the 100 muons per square meter per second that arrive on the earth's surface, only those that have a sufficiently high energy (>1.4 TeV) can reach the underground laboratory. The resulting muon flux in the underground laboratory is 1 per square meter per hour (Bettini 2000).

These characteristics are optimal to investigate the contribution of the environmental radiation to the residual background signal of the 1220 Quantulus™.

In the first step in our investigations, we performed measurements using idealized samples to study the efficiency and background variations between the surface laboratory located at E.N.E.A.-Bologna and the underground laboratory of Gran Sasso.

RESULTS

For the background, two sets of three teflon vials with benzene volumes ranging from 1 mL to 5 mL filled with pure analytical benzene were used. For the modern standards two sets of three teflon vials with the same benzene volumes filled with pure analytical benzene enriched with radiocarbon to give the same activity as the standard sucrose ANU (sucrose ANU/modern = 1.0866) were used. The scintillation cocktail comprised 15 mg butyl-PBD/mL benzene (Gupta and Polach 1985). For each of these standards, spectra were obtained for 40 one-hour periods.

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Cylindrical teflon-S vials designed by ISTA Ltd (Faenza, Italy) with Delrin cap sealed with epoxy resin have been used. The vials characteristics are: height of 50 mm, external diameter 27 mm, thickness of the bottom teflon base 12 mm, capacity of 9 mL.

Table 1 The count rate of the set A of teflon vials with benzene volumes of 1, 3, and 5 mL related to surface and underground (italic) laboratories. The labels L and H indicate background and modern standards, respectively.

Sample	Count rate (cpm)	Count error (cpm)	Modern activity (dpm)	Modern activity error (dpm)	Eff (%)	FM	fM	T_{max} (BP)
L1A	0.278	0.022						
H1A	12.949	0.148	8.853	0.088	80.93	25,531.503	17	48,200
L1A	*0.059*	*0.010*						
H1A	*12.282*	*0.144*	*8.540*	*0.094*	*76.76*	*99,734.000*	*35*	*54,000*
L3A	0.398	0.026						
H3A	39.140	0.257	27.068	0.161	83.28	17,419.104	43	55,900
L3A	*0.150*	*0.016*						
H3A	*38.235*	*0.254*	*26.609*	*0.166*	*81.35*	*44,052.587*	*69*	*59,600*
L5A	0.655	0.033						
H5A	65.206	0.332	45.101	0.209	83.60	10,676.865	57	58,000
L5A	*0.235*	*0.020*						
H5A	*63.874*	*0.328*	*44.464*	*0.215*	*81.89*	*28,580.867*	*92*	*61,900*

Table 2 The count rate of the set B of teflon vials with benzene volumes of 1, 3, and 5 mL related to surface and underground (italic) laboratories. The labels L and H indicate background and modern standards, respectively.

Sample	Count rate (cpm)	Count error (cpm)	Modern activity (dpm)	Modern activity error (dpm)	Eff (%)	FM	fM	T_{max} (BP)
L1B	0.273	0.021						
H1B	12.646	0.146	8.645	0.087	79.03	22,856.284	17	48,100
L1B	*0.096*	*0.013*						
H1B	*12.020*	*0.142*	*8.331*	*0.090*	*75.13*	*58,659.673*	*27*	*51,900*
L3B	0.442	0.027						
H3B	38.463	0.255	25.565	0.159	81.84	15,152.806	40	55,300
L3B	*0.157*	*0.016*						
H3B	*38.181*	*0.254*	*26.567*	*0.166*	*81.24*	*42,038.809*	*67*	*59,400*
L5B	0.590	0.32						
H5B	64.470	0.330	44.628	0.208	82.65	11,570.346	59	58,300
L5B	*0.230*	*0.020*						
H5B	*64.176*	*0.329*	*44.678*	*0.216*	*82.28*	*29,488.245*	*94*	*62,100*

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The count rate of the two sets (A, B) of teflon vials with benzene volumes of 1, 3 and 5 mL related to surface and underground (italic) laboratories is shown in Tables 1 and 2. The data are related to optimized soft window in channels 5–540 and PAC equal to 200. Labels L and H indicate background and modern standards, respectively. Also the modern activity, the efficiency (Eff), the figure of merit (FM), the factor of merit for ^{14}C dating (fM) and the maximum determinable age (T_{max}) for a 2-σ detection criterion and a counting time of 4742 min are shown in Tables 1 and 2 (page 158).

Figure 1 shows the anticoincidence count rate of guard with sample events recorded at surface (a) and underground (b) laboratories. Also, Figure 2 shows the coincidence count rate of guard with sample events recorded at surface (a) and underground (b) laboratories.

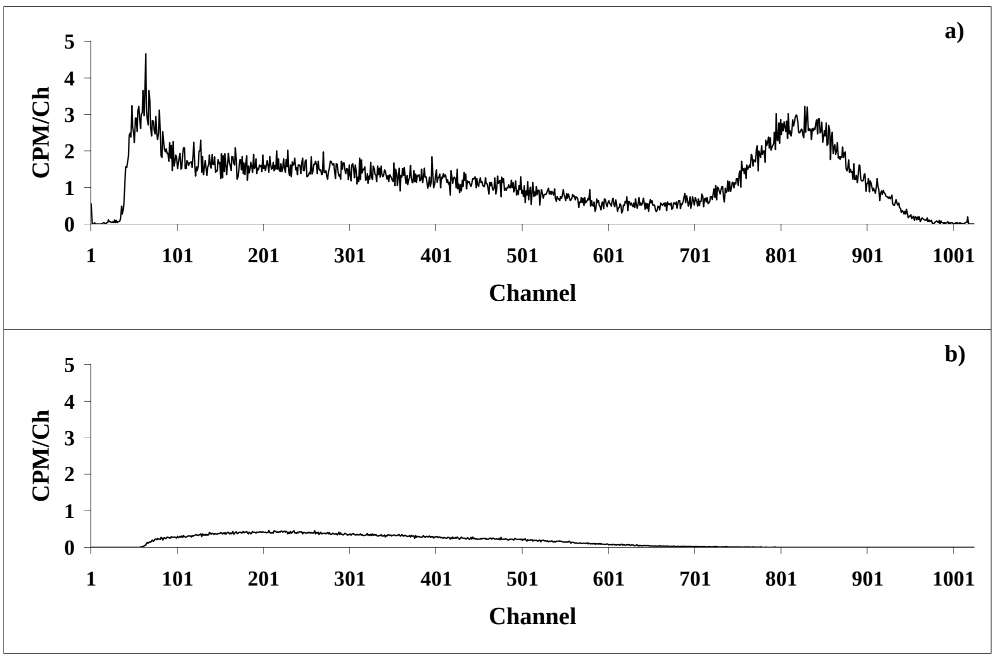

Figure 1 The anticoincidence count rate of guard with sample events recorded at the surface (a) and the underground (b) laboratories

DISCUSSION

From the spectra shown in Figure 1, it may be observed that the cosmic muon flux is missing at the underground laboratory (0.071 cpm) compared to the surface one (414 cpm). The anticoincident Compton continuum spectrum shows 669 and 161 cpm at the surface and the underground laboratories, respectively. This portion of guard spectrum is related to the flux of the environmental gamma photons and secondary cosmic particles. The remaining Compton signal in Gran Sasso is due to the guard phototubes, which are fully embedded, in the scintillating cocktail filling the guard (Figure 3).

The coincidence count rate of the guard with sample events shows at the surface laboratory a cosmic muon flux of 10.9 cpm and Compton continuum spectrum of 1.06 cpm while at the underground laboratory we have recorded 0.0 cpm and 0.063 cpm, respectively. The coincident guard events are active background reducing events. The data indicates a background reduction of approximately 65% at the underground laboratory compared to the surface one, with no differences in the efficiency

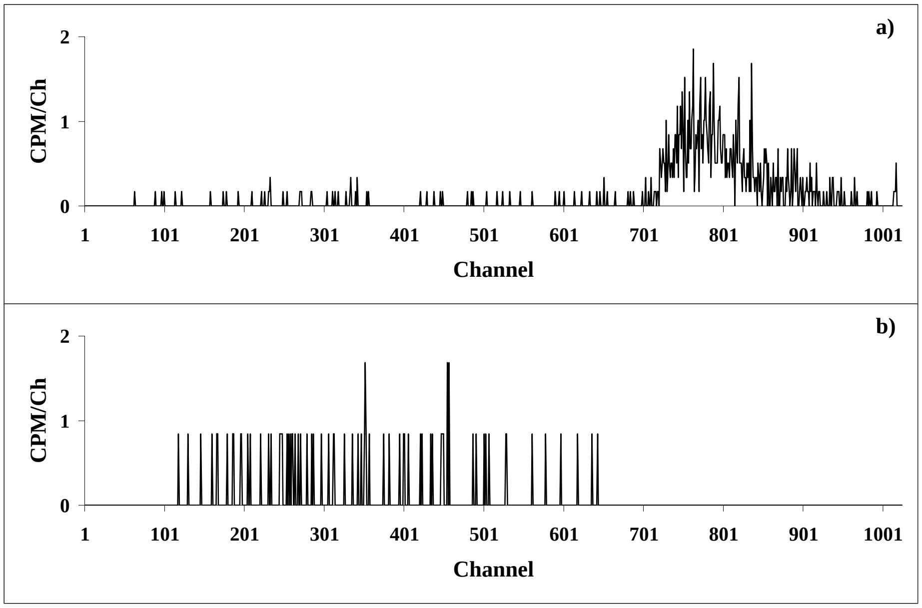

Figure 2 The coincidence count rate of guard with sample events recorded at the surface (a) and the underground (b) laboratories. To a better graphical representation the CPM/Ch of surface (a) and underground (b) laboratories are multiplied by factors 10E+01 and 10E+03, respectively.

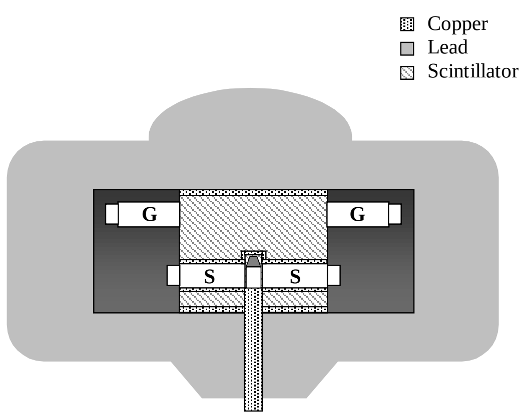

Figure 3 Schematic diagram of 1220 Quantulus™ lead shield assembly and phototubes. The labels S and G indicate sample and guard phototubes, respectively.

particularly for the 3 and 5 mL benzene volumes of set B. This background reduction results from the low environmental radiation flux in Gran Sasso.

The vials are fully symmetric with no extra reflectors to cut off the light path from one phototube to another and no metallic cap on top (Shibata et al. 1983; Polach et al. 1988; Kaihola et al. 1992). Epoxy resin is used to seal the cap to minimize benzene loss. The surface activator and the epoxy resin may emit light when hardening with the background signal increasing. High bias threshold was selected in the measurements to cut off low energy emission signal and therefore interference from epoxy resin does not appear in the results.

CONCLUSION

The first step of measurements carried out in the surface and underground laboratories, with the 1220 Quantulus™, have shown a background reduction of 65% in the underground laboratory.

Thus, the characteristics of the underground laboratory of Gran Sasso make it possible to perform high precision radiocarbon measurements, to improve fM by 60% and to extend for these idealized samples the dating limit from 58,000 BP to 62,000 BP (5 mL, 3 days counting). However, experiments on real material have emphasized that the problems in extending the limit of detection back in time, have much more to do with benzene synthesis and contaminants introduced through lithium, surface reaction catalysts and impurities than the counter background level (Radnell and Muller 1980; Lowe 1989; McCormac et al. 1993).

Coincidence count rate of guard and sample events is almost negligible in Gran Sasso indicating that the contribution from the external radiation has vanished. Vial design does not affect the background in Gran Sasso, but is very important on the surface. The residual background is only dependent on the sample volume in Gran Sasso, approaching nil at zero sample volume. This means that the sample acts as a target of photons from the small inherent residual radioactivity of sample phototubes.

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