LIQUID SCINTILLATION SPECTROMETRY AT GRAN SASSO NATIONAL LABORATORY: RADIOCARBON MEASUREMENTS

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ABSTRACT. Radiocarbon measurements using a Quantulus™ ultra low-level liquid scintillation spectrometer were performed at the Gran Sasso National Laboratory (National Institute for Nuclear Physics) to study the efficiency and background related to the measurement site (Plastino et al. 2001). Cosmic background and its variation have been removed in the Gran Sasso laboratory by its 1400-m rock overburden. Stable, high-performance liquid scintillation counting conditions are obtained when any remaining variable components of the environmental background, such as radon, are eliminated. The ultra low-level liquid scintillation spectrometer Quantulus has an anti-Compton guard detector, which allows for the monitoring of gamma radiation in the background. Guard detector efficiency in ^{14}C background reduction is 8% at Gran Sasso, while 80% is observed on the surface (Plastino and Kaihola 2004). The big difference in the guard detector efficiency between surface laboratories and Gran Sasso is due to the absence of cosmic and associated lower energy Compton radiation (Plastino and Kaihola 2006). The cosmic noise reduction observed at the Gran Sasso laboratory makes it possible to perform high-precision ^{14}C measurements and to extend for these ideal samples the present maximum dating limit from 58,000 to 62,000 BP (5 mL, 3 days of counting) (Plastino et al. 2001).

INTRODUCTION

Underground laboratories provide environments where cosmic radiation is attenuated to a great extent. Previously, Kalin and Long (1989) reported liquid scintillation radiocarbon measurements in Tucson, and Schäfer et al. (2000) measured tritium at the Felsenkeller underground laboratory at VKTA Rossendorf e.V., near Dresden. The Arizona laboratory is covered by 10 m of concrete, the Dresden laboratory by 47 m of hornblende monzonite rock. At the Arizona lab, the ^{14}C background reached 0.16 cpm at 71% counting efficiency in 3-mL copper Teflon® vials. The tritium background in the Felsenkeller laboratory was half of that at TU Bergakademie Freiberg.

The most attenuation of cosmic radiation thus far is provided deep underground in Gran Sasso, Italy, where the cosmic muon flux has been estimated to be 1 muon m^{-2} hr^{-1} (Arpesella 1992). A Quantulus™ operated from 2000 to 2004 but was later replaced with a new unit. Experiments were carried out to evaluate ^{14}C performance values in Gran Sasso and to compare with the same samples measured in Bologna using another Quantulus.

EXPERIMENTAL AND DISCUSSION

The Quantulus anti-cosmic guard detector (Plastino and Kaihola 2006) is an effective detector of cosmic radiation. The multichannel analyzer allows for monitoring coincidences between sample and cosmic muon events and Compton scattered electrons in the guard to evaluate the performance of the guard in background reduction. Guard detector efficiency in ^{14}C background reduction is 8% at Gran Sasso, while 80% is observed on the surface (Plastino and Kaihola 2004). This difference is due to the absence of a cosmic signal in the underground laboratory.

Instrument temperature at Gran Sasso is 9 °C and no cooling unit was mounted on the instrument. The guard phototube and cocktail single-photon noise is negligible (Plastino and Kaihola 2006). The laboratory is basically a steel container with no Rn removal devices. It is in a well-ventilated tunnel, but water containing radon flows into the tunnel.

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Optimized Vial for Reduced Sample Volumes

Special vial designs eliminate cross-talk events, as in 3-, 7-, 15-, and 20-mL copper Teflon vials (Wallac), where unused volume is replaced by a massive, non-transparent cap and base (Polach et al. 1983). This masked vial design leads to the linear dependency of sample ^{14}C background on vial volume. Figure 1 shows a graph down to 0.3-mL masked vial size with an extrapolated 0.05-cpm empty vial background in surface laboratories (Kaihola et al. 1992). A special adapter of 20-mL vial size is required for the smaller vials.

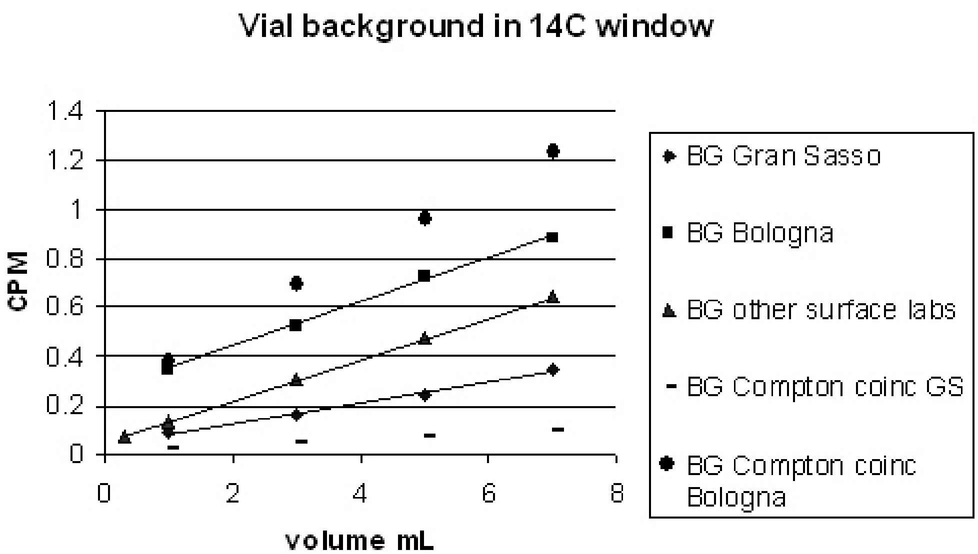

Figure 1 ^{14}C background and removed background dependence on benzene volume in a 9-mL unmasked Teflon® vial in Gran Sasso National Laboratory (cpm = 0.0422 × V + 0.0422); 9-mL unmasked Teflon vial in Radiocarbon Laboratory ENEA-Bologna (cpm = 0.0908 × V + 0.2598); 3-, 7-, 15-, and 20-mL masked copper Teflon vials and 0.3-mL Teflon vial in a black masking Delrin adapter in other surface laboratories (cpm = 0.0844 × V + 0.0507).

Minivials have also been made of silica (Haas and Trigg 1991; Hogg 1992). Buzinny et al. (1995) designed a set of small vials that are mounted on a 28-mm base plate and have a hole in the cap to release benzene pressure, minimizing losses during counting. The hole is blocked during counting.

When a large 9-mL Teflon vial, such as the one used at Bologna, is used for variable volumes, background dependence is again linear, but an additional contribution remains due to the empty portion of the vial, which is the source of air scintillations. The count rate of 9-mL Teflon vials with benzene volumes of 1, 3, and 5 mL related to surface (Bologna) and underground (Gran Sasso) laboratories is shown in Figure 2. The surface laboratory has extrapolated a 0.26-cpm background at zero benzene volume in a 9-mL unmasked vial, while the background value in the underground laboratory is only 0.04 cpm (Plastino et al. 2001; Plastino and Kaihola 2004).

The background of a masked vial filled with benzene and measured on the surface approaches the same zero-volume background as an unmasked vial with equivalent benzene volumes in Gran Sasso. Cross-talk and air scintillations in the ^{14}C window are thus not observed in Gran Sasso at a high bias threshold. Absolute background values in Gran Sasso are about one-half of the surface values for the ideal masked vials. Copper Teflon vials have not been tested yet underground.

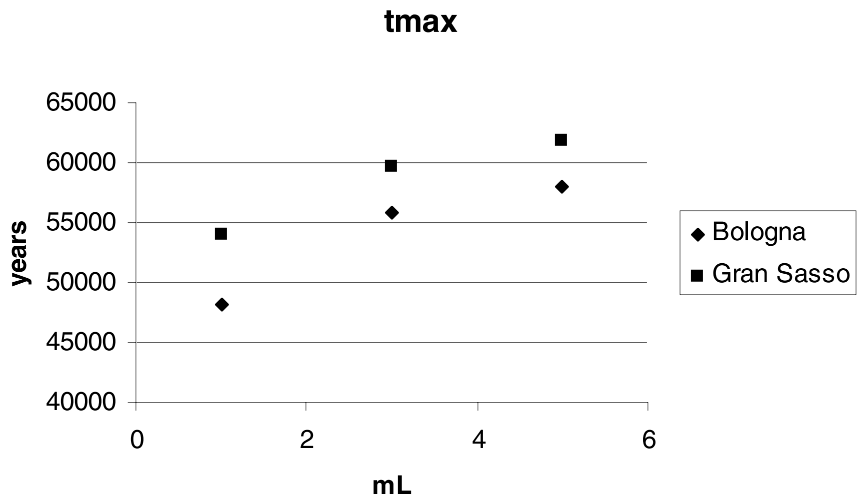

Figure 2 Background variation: the maximum age in surface (Bologna) and underground (Gran Sasso) laboratories for benzene volumes of 1, 3, and 5 mL in 9-mL Teflon vials. Counting time is 3 d.

A pulse amplitude comparator was applied at levels of 100, 180, and 200. The figure of merit did not improve beyond 100, indicating that there are no cross-talk events in the ^{14}C spectrum range in the Gran Sasso low-background environment.

Count-rate variation of background samples in Gran Sasso is larger than that of active samples. Also, the guard count rate varies excessively, indicating the presence of variable Rn concentration in this laboratory, where a variable cosmic particle flux is missing. A 19-d record from 19 October to 10 November 1999 for the guard detector showed $\chi^2 = 650$ in 472 repetitions of 60 min each; the mean count rate was 157.6 cpm. The observed distribution of measurements does not fit the expected one. Neither a diurnal nor weekly pattern in the variation was found. Direct Rn measurement of the laboratory air using a 20-mL plastic vial with a MeltiLex® scintillating plastic sheet on the inside walls indicated a Rn concentration of 1250 Bq/m^3 in May 2003. An open vial inside the instrument showed a concentration of 650 Bq/m^3.

CONCLUSIONS

Excellent performance for liquid scintillation spectrometry of ^{14}C is achieved underground in the Gran Sasso National Laboratory. Unmasked vials in Gran Sasso show better background than masked vials on the surface; absolute backgrounds are about half that of the surface figures. We attribute the sample background to be fully derived from the instrument's phototube radioactivity and external gamma radiation, as the cosmic flux is negligible in Gran Sasso laboratory. The negligible cosmic background at the Gran Sasso National Laboratory allows for high-precision ^{14}C measurements and extension of the present maximum dating limit from 58,000 to 62,000 BP (5 mL, 3 d counting).

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