Measurement of the ^{14}C Abundance in a Low-Background Liquid Scintillator

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Borexino-CTF Collaboration

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Abstract

The ^{14}C/^{12}C ratio in 4.8 m^3 of high-purity liquid scintillator was measured at $(1.94 \pm 0.09) \times 10^{-18}$, the lowest ^{14}C abundance ever measured. At this level the spectroscopy of low-energy solar neutrinos, in particular a measurement of the ^{7}Be neutrino flux, will not be obstructed by the ^{14}C β decay intrinsic to a liquid scintillator detector. A comprehensive study of the deviation of the shape of the ^{14}C β spectrum from the allowed statistical shape reveals consistent results with recent observations and calculations. Possible origins of the ^{14}C in the liquid scintillator are discussed.

1 Introduction

Organic liquid scintillators are widely used in physics research as a detection medium for nuclear and particle radiation. Detectors employing large amounts of liquid scintillator (hundreds of tons) will be used in upcoming experiments that will search for rare events with low energy deposition. These experiments include the spectroscopy of solar neutrinos, the search for a neutrino magnetic moment and searches for non-baryonic dark matter. The feasibility of these experiments depends on a low background counting rate at energies typically below a few hundred keV. Since organic liquid scintillators are predominantly composed of carbon, the intrinsic concentration of the radioisotope ^{14}C

(β^- decay, Q = 156 keV, $t_{1/2} = 5,730$ years) can constitute the main background at low energies and hence can restrict the sensitivity of these experiments.

Liquid scintillator mixtures contain aromatic solvents and these in turn are usually synthesized from petroleum. While modern carbon of biological origin has an isotopic ratio $^{14}C/^{12}C$ of about 10^{-12}, it is expected that petroleum derivatives have a ^{14}C abundance that is orders of magnitude lower. Petroleum deposits are mostly found deep underground and are shielded from the cosmic ray flux present at the Earth's surface responsible for the continuous production of ^{14}C from ^{14}N, via the reaction $^{14}N(n,p)^{14}C$. Consequently, the cosmogenic production of ^{14}C in underground petroleum is minimized and any ^{14}C originally present in the precursor organic material to the petroleum will have had millions of years to decay away. Thus one would expect the ^{14}C isotopic abundance in an organic liquid scintillator to be extremely low. In fact, the abundance of ^{14}C in a petroleum-derived scintillator has never been determined.

In the Borexino solar neutrino experiment [1], low-energy neutrinos (< 1 MeV) are detected via neutrino–electron scattering in a large target mass of liquid scintillator. The expected neutrino–electron interaction rate in Borexino, for electron recoils above 250 keV, is dominated by ^7Be solar neutrinos and amounts to ~0.5 events/day/ton, assuming the neutrino flux calculated in Standard Solar Models [2, 3]. Though the maximum β energy for ^{14}C is 156 keV, finite energy resolution and pile-up can create a tail in the β energy spectrum that extends to higher energy. Thus, the intrinsic level of ^{14}C in the liquid scintillator becomes an important consideration in setting the low-energy threshold for neutrino events in Borexino. From these considerations, it is desirable in Borexino that the $^{14}C/^{12}C$ ratio not substantially exceed 10^{-18}.

Trace concentrations of ^{14}C are usually measured with accelerator mass spectroscopy (AMS). The sensitivity of this technique is limited by sample preparation at the 10^{-15} level [4], corresponding to a radiocarbon age of about 60,000 years. In one atypical measurement, CO samples were prepared from purified methane from a natural gas well and underwent an enrichment process to potentially increase the $^{14}C/^{12}C$ isotope ratio. When analyzed with AMS at the IsoTrace Laboratory in Toronto, Canada, by comparing with blank values, an upper limit of $< 1.6 \times 10^{-18}$ was obtained [5]. This result suggested a petroleum-derived scintillator would be satisfactory for the Borexino solar neutrino experiment. However, a direct measurement of the ^{14}C abundance in a liquid scintillator was considered a necessary prerequisite.

This paper describes the measurement of the $^{14}C/^{12}C$ ratio in an organic liquid scintillator carried out in the Counting Test Facility (CTF) of the Borexino project, located in the Gran Sasso underground laboratory in Italy. In addition to the ^{14}C abundance measurement, the low background rate of the detector together with its large target mass and low energy threshold allowed a comprehensive study of the ^{14}C β shape to be made. The results obtained for the deviation from the allowed statistical shape are compared with the contradictory experimental findings in recent literature.

2 Measurement of ^{14}C with the Counting Test Facility

In the case of liquid scintillators, sensitivities better than 10^{-18} can be achieved by direct measurement of the β^- decay of ^{14}C. This is only possible if the scintillation detector has a mass of several tons and is constructed with stringent requirements on the selection of materials with low radioactivity (high radiopurity). The detector size is crucial in order to provide for an adequate event rate. A large size also leads to a lower surface-to-volume ratio which assists in lowering the background rate from external sources. In addition, the scintillating volume must be installed in an environment with sufficient shielding against cosmic rays and ambient radioactivity.

A prototype for the Borexino experiment, the Counting Test Facility (CTF) [6], was constructed in the Gran Sasso National Laboratory in Italy, at an underground depth of ~3500 meters-water-equivalent. The primary objective of this instrument was to measure the abundances of trace radioactive contaminants in a liquid scintillator detector, in particular the abundances of ^{14}C, U and Th. Sensitivities for the $^{14}C/^{12}C$ ratio in the 10^{-19} range were achievable. The CTF is the only instrument presently available that is capable of measuring the ^{14}C abundance in a petroleum-derived scintillator.

2.1 The Detector

The CTF is an unsegmented liquid scintillation detector with three-dimensional light collection and graded shielding against external background sources. A schematic drawing of the detector is shown in Fig. 1 and a complete description is published in Ref. [6]. Only a short summary is given here.

A transparent, spherical nylon vessel forms the active volume of the CTF, and contains 4.8 m^3 of liquid scintillator with density $\rho = 0.88$ g/cm^3. The scintillator solvent is 1,2,4-trimethylbenzene (pseudocumene), C_9H_{12}, with 2,5-diphenyloxazole (PPO) as the fluor at a concentration of 1.5 grams/liter. The scintillating volume is shielded in all directions by about 4.5 m of high-purity water which serves as a buffer against the ambient gamma rays and neutrons at the underground site. Nylon monofilament strings hold the nylon vessel in place against the upward buoyant force that exists when the vessel is filled with scintillator and surrounded by water. Strain gauges attached to the strings measure the buoyant force and these readings indicate the volume (and mass) of scintillator in the detector. One hundred photomultipliers equipped with light concentrators are mounted on a spherical open structure. They are set back in the water buffer 2.3 m from the active volume. A light yield of $\sim$ 300 photoelectrons per MeV of energy deposited in the scintillator is observed.

During normal data taking, the trigger threshold for recording scintillation events is established at a coincidence level of six hit photomultipliers, corresponding to about 25 keV (for β and γ interactions). For each event, the collected photomultiplier signal charges and photon arrival times are recorded. From this information, the total energy deposition and spatial location of an event can be deduced in an off-line analysis.

The energy scale in the detector was established from isotopes in the decay chain of ^{222}Rn and from ^{85}Kr. These activities were present at low levels in the scintillator due to an initial air exposure and were likely to be uniformly distributed throughout the scintillating volume, making them suitable calibration sources. The Rn daughter ^{214}Po ($t_{1/2} = 164$ μs, 7.68 MeV) has an α decay that can be identified unambiguously through its delayed β–α coincidence with the parent isotope ^{214}Bi. The β decay of ^{85}Kr feeds a 0.43% branch to a 514 keV excited state with lifetime $t_{1/2} = 1.0$ μs, giving rise to an identifiable β–γ delayed coincidence.

2.2 Data Taking

The total event rate in the detector for energy deposits greater than 25 keV was $\sim$1 s^{-1}, dominated by the rate coming from ^{14}C β decay. Thus, even a short data-taking period of several days allowed for accumulation of sufficient counts in the ^{14}C β-decay spectrum.

The first data set (Period 1) analyzed for ^{14}C abundance had a data collection live time of 13.6 days and corresponds to the period soon after the CTF detector was initially filled with 4.24 tons of scintillator (and following a short waiting period to allow for some short-lived radioactive backgrounds to decay away). The second data set (Period 2) corresponds to a collection time of 10.5 days and was taken after the scintillator was stripped with nitrogen and extracted with ultra-pure water. The N_2 stripping was performed in order to extract gaseous contaminants from the scintillator while the water extraction aimed at the removal of ionic species. The third data set (Period 3) had a collection time of 14.2 days and was acquired after vacuum distillation of the scintillator.

Additional ^{14}C data were taken during a final phase of the experiment when the PPO concentration in the scintillator was reduced from 1.5 to 1.3 grams/liter. The purpose of this procedure was to investigate whether the fluor might be the dominant carrier of ^{14}C. A comprehensive description of the various scintillator operations is given in Ref. [7, 8].

Several data-taking periods were dedicated to background characterization. The "water run" consists of data taken with the CTF filled with water, before the introduction of scintillator into the active volume. This spectrum characterizes the background from Čerenkov light emission by γ-ray and muon interactions in the water buffer. During another period that served as an external source calibration, data were taken after the deliberate introduction of radon into the surrounding water buffer. This served to characterize the background in the scintillator coming from external γ sources in the water (coming mostly from ^{214}Bi decay, a daughter of ^{222}Rn).

Table 1 compares the count rates from various data sets. The entry "radon in water buffer" is an estimate of the background event rate during normal data taking periods, originating from residual Rn in the water buffer (concentration of ~ 25 mBq/m^3), as deduced by the external calibration data runs described above. The background count rate from muons and from other identified, internal background radioactivities are negligibly small compared to the raw data rate dominated by ^{14}C decays.

Table 1: *Count rate comparison.*

Data Set	Count Rate [events/day] between 70–150 keV
first scintillator runs	$30,613 \pm 48$
water run	$1,051 \pm 40$
radon in water buffer	$1,558 \pm 248$

3 Data Analysis and Results

The theoretical β spectrum used for fitting the data has the following general form [9]:

$$N(W_e)\,dW_e = p_e\,W_e\,(W_0 - W_e)^2\,F(Z,W_e)\,C(W_e)\,dW_e, \qquad (1)$$

where p_e, W_e are the momentum and total energy of the emitted electron, W_0 is the endpoint, $F(Z,W_e)$ is the Fermi function which accounts for the influence of the nuclear Coulomb field on the β spectrum, and $C(W_e)$ is the "shape factor" for the β decay.

A recent theoretical calculation by García and Brown [10] of the ^{14}C β spectrum shape predicts a shape factor correction, $C(W_e)$, of:

$$C(W_e) = 1 - (0.37 \pm 0.04)W_e,$$

where W_e is the total energy of the electron in MeV.

A non-statistical shape factor for ^{14}C was first measured by Sonntag *et al.* [11], with the following form:

$$C(W_e) = 1 - 9.14W_e + 1.5/W_e + 8W_e^2,$$

with the coefficients converted to the same W_e units expressed above. The size of this correction is in disagreement with the more recent measurement by Wietfeldt *et al.* [12], which found a smaller shape factor:

$$C(W_e) = 1 - (0.45 \pm 0.04)W_e,$$

and which is closer to agreement with the recent theoretically-calculated value. This work was a refinement of earlier studies [13] of the ^{14}C β spectrum, including re-analysis of previous data after several detector problems were resolved. Aside from [13], the above two results constitute the only published measurements of the ^{14}C shape factor.

The raw data were fit with a function $S(W_e)$ given by

$$S(W_e) = \int N(W_e')\,g(W_e', W_e)\,dW_e' + B(W_e) \qquad (2)$$

where $N(W_e')$ is the theoretical β spectrum according to (1), $g(W_e', W_e)$ is the detector response function and $B(W_e)$ is a polynomial which accounts for the background contribution.

The measurements from the background-characterization runs provided information about the shape of the background energy spectrum. Monte Carlo simulations, which included the detector response, were performed to confirm our understanding of the background rates and normalizations. These simulations (based on measurements) indicate that the shape of the background underlying the ^{14}C β

spectrum has a small, constant slope for energies between 60–500 keV. Thus, in fitting each β spectrum, the background $B(W_e)$ was parameterized by fitting a first-order polynomial above the ^{14}C β endpoint, between 250 keV and 500 keV. With these parameters then fixed, the fitting procedure extrapolated the background polynomial into the ^{14}C energy range.

From calibration measurements using the ^{214}Po α line, it is known that $g(W_e', W_e)$ has a Gaussian shape. Its width σ depends on the energy and scales approximately with its square root. For the Fermi function, the tabulated values from Ref. [14] were taken and parameterized.

A χ^2 analysis was performed in an energy interval between 60 keV and 250 keV. During minimization, the β endpoint for ^{14}C was fixed at 156 keV while the following four parameters were allowed to vary:

1. The overall normalization factor corresponding to the ^{14}C activity.

2. The shape factor $C(W_e)$ describing the deviation of the ^{14}C β spectrum from the allowed statistical shape.

3. The proportionality constant determining $\sigma(E) \propto \sqrt{E}$, where E is the energy deposition.

4. The energy scale factor relating the measured photomultiplier charge signal to the energy deposition.

The latter two parameters were left free in the fit as they were not determined with the necessary accuracy at these low energies.

Table 2: *Results obtained from fitting the low energy CTF data with Eq. (2). E is the kinetic energy of the β particle and W_e is the total energy. Q corresponds to the measured photomultiplier charge in units of photoelectrons. The parameter, a, is the linear shape factor in $C(W_e) = 1 + a\,W_e$, and 90% CL limits are quoted. During the three data taking periods, a different number of photomultipliers were operational, affecting both the energy scale Q/E and the resolution $\sigma(E)/\sqrt{E}$. The first uncertainty in the ratio $^{14}C/^{12}C$ corresponds to the statistical error from the fit and the second to the systematic error due to target mass uncertainty.*

	Period 1	Period 2	Period 3
Data collection time [d]:	13.6	10.5	14.2
χ^2/ndf :	236/201	195/185	136/154
^{14}C activity [Bq]:	1.425 ± 0.007	1.384 ± 0.009	1.369 ± 0.007
Shape factor a [MeV^{-1}]:	> -0.10	> -0.72	> -0.72
$\sigma(E)/\sqrt{E}$ [keV$^{1/2}$]:	2.88 ± 0.01	2.99 ± 0.04	3.13 ± 0.04
Q/E [keV^{-1}]:	0.273 ± 0.001	0.252 ± 0.001	0.205 ± 0.001
Detector mass [tons]:	4.24 ± 0.20	4.09 ± 0.20	4.07 ± 0.20
Ratio ^{14}C/^{12}C [10^{-18}]:	$1.94 \pm 0.01 \pm 0.09$	$1.95 \pm 0.01 \pm 0.08$	$1.94 \pm 0.01 \pm 0.09$

Over the three ^{14}C data-taking periods, several fundamental detector parameters changed: the target mass was reduced during the scintillator purification operations and the number of operating photomultipliers decreased. The latter affected the overall photoelectron yield and consequently changed the energy scale and the energy resolution, as can be seen in Table 2.

The limitation for the precise determination of the shape factor arose from the fact that the energy scale and energy resolution were left as free parameters during minimization. The variation of these fit parameters can counterbalance, to some degree, the shape factor $C(W_e) = 1 + a\,W_e$. Nevertheless, our fit results can exclude certain values for the shape parameter — our data are incompatible with values for $a < -0.72$ MeV^{-1} (90% CL).

In Fig. 2, the data of Period 1 are shown together with the fit ($a = -0.4$ MeV^{-1}). In the lower part of the plot the ratio data/model is depicted. The fitted spectra from Period 2 and Period 3 are shown in Fig. 3 and 4, respectively ($a = -0.4$ MeV^{-1}) and the results are given in Table 2.

The $^{14}C/^{12}C$ ratio, R, is given by:

$$R = A\,\frac{\tau\, M}{9\, N_A\, m}, \tag{3}$$

where A is the ^{14}C activity, τ is the mean lifetime of ^{14}C, M is the molecular weight of pseudocumene, 120.2 g/mol, N_A is the Avogadro constant, m is the scintillator mass, and the 9 in the denominator comes from the 9 atoms of carbon in C_9H_{12}.

The largest uncertainty in the absolute determination of the isotopic abundance of ^{14}C arises from the detector mass determination, which was known within an accuracy of 5%. The mass decrease between the different periods amounted to 150 ± 10 kg between Period 1 and Period 2, due to cleaning and preparation of the scintillator purification plant and to 20 ± 10 kg between Period 2 and Period 3, due to cleaning and sampling.

4 Possible Origins of ^{14}C Content

At some level, depending on the underground depth, cosmogenic production of ^{14}C still occurs, resulting indirectly from neutrons emitted following μ^- capture, neutrons produced by spallation, and (more rarely but depending on depth) from nuclear fragmentation directly following an inelastic muon-nucleus collision. However, at sufficient depth underground cosmogenic production becomes negligible compared to other production mechanisms involving natural radioactivity.

Production of ^{14}C deep underground can occur through nuclear reactions involving neutrons and α particles emitted by the surrounding natural radioactivity. The neutron flux deep underground originates chiefly from (α,n) reactions [15] on Al, Mg, Na and other elements in the surrounding rock. The uranium and thorium decay chains supply most of these α decays (neutrons are also emitted in spontaneous fission of uranium). Consequently, it is the abundance of uranium and thorium in the rock containing the petroleum that governs the abundance of ^{14}C therein.

The reactions expected to contribute the most to ^{14}C production in deep underground geological formations are [16]–[18]:

1. $^{17}O(n,\alpha)^{14}C$
2. $^{14}N(n,p)^{14}C$
3. $^{13}C(n,\gamma)^{14}C$
4. $^{11}B(\alpha,n)^{14}C$
5. direct ^{14}C emission from tripartition of ^{226}Ra

with the reactions listed in order of importance for production in typical underground ores (and with the significance of the fourth reaction strongly dependent on the boron content of the material being considered).

As a rough estimate of the $^{14}C/^{12}C$ ratio in petroleum, consider the production (by neutrons) of ^{14}C in limestone ($CaCO_3$). Knowing the typical uranium and thorium content of limestone, one can estimate the underground thermal neutron flux in this rock. The main reaction for producing ^{14}C is $^{17}O(n,\alpha)^{14}C$, since the thermal neutron capture cross section on ^{17}O is 235 mb, compared to the capture cross section on ^{13}C of 1.4 mb (and due to the absence of nitrogen). Using an estimate of the thermal neutron flux in limestone [16] and the above cross sections, Florkowski *et al.* have estimated an equilibrium activity level of: 4.4×10^{-8} ^{14}C decays per minute in one gram of limestone [15]. If all of the ^{14}C atoms in this one gram of rock were swept into one gram of adjacent petroleum (CH_2–), this would give an isotopic ratio for $^{14}C/^{12}C$ in the petroleum of $\sim 5 \times 10^{-21}$. There are no direct experimental data confirming the isotopic abundance at this level.

Our measurement of the ^{14}C isotopic abundance in the scintillator is much higher than the value estimated above (for petroleum). In the processing to synthesize scintillator solvent, petroleum is passed through fractional distillation and the selected cut undergoes catalytic cracking (to break any long chains), followed by catalytic reforming. Aromatic ring compounds are produced in the reforming, with hydrogen passing over the catalyst. The final step in producing pseudocumene might include polymerizing lightweight hydrocarbon gases and alkylation to add the methyl groups onto the benzene rings. During this final synthesis step, it is possible that some organic residuals containing modern carbon might be introduced, though this is not the common practice.

Another possible explanation for the higher ^{14}C abundance in our liquid scintillator mixture might be the exposure of the scintillator to CO_2. Though unlikely, CO_2 could have been introduced during the production process or during storage at the company site. Furthermore, CO_2 could have diffused into the scintillator from the water buffer of the CTF. Various scintillator purification operations were performed; these did not affect the the $^{14}C/^{12}C$ ratio. This is noteworthy as the nitrogen stripping of the scintillator had the potential for removing CO_2. Furthermore the ^{14}C abundance did not change over the course of the various measuring periods, which spanned about a year.

The presence of 1.5 grams/liter of PPO as a fluor in the scintillator mixture might have been the source of the ^{14}C in the mixture (if we accept that pseudocumene contains ^{14}C at the level estimated above). However, a specific experiment was conducted to test this hypothesis. The scintillator mixture was distilled to remove PPO, thereby reducing the PPO concentration. Following this experiment, again no change in the ^{14}C abundance was observed.

5 Conclusions

By direct counting of the ^{14}C β decays in a 4.8 m^3, low-background liquid scintillation detector, sensitivities at the 10^{-19} level were achieved. A $^{14}C/^{12}C$ ratio of $(1.94 \pm 0.09) \times 10^{-18}$ was found in the scintillator investigated. An examination of the shape of the β spectrum found results consistent with the theoretical prediction [10] and with the recent experimental result [12] for the deviation from the allowed statistical shape. The shape values reported in Ref. [11] can be excluded.

The ^{14}C activity has been stable over a period of more than one year. Various scintillator purification operations aimed at the removal of trace contaminants did not affect the ^{14}C abundance. A crude estimate of the abundance of ^{14}C in petroleum cannot account for the level observed in our scintillator. We conclude that it is likely that samples from different petroleum sources and variations in processing techniques might impact the ^{14}C isotopic abundance found in liquid scintillators.

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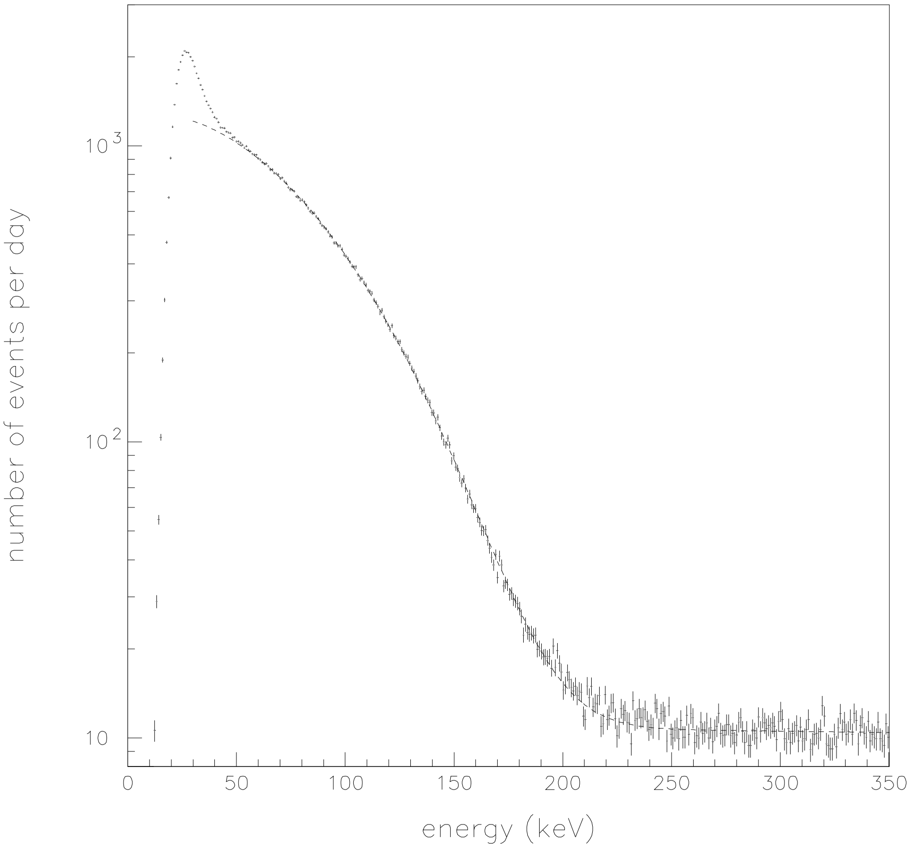
Figure 1: *Schematic drawing of the Counting Test Facility (CTF) of the Borexino experiment at Gran Sasso.*



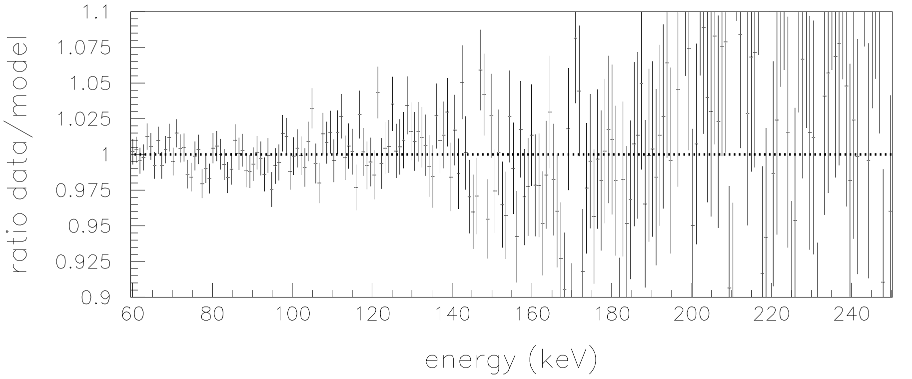


Figure 2: *Data and fit of Period 1. The solid line in the upper plot shows the fit of the* ^{14}C *spectrum to the raw data and the plot below displays the ratio between the data and the model in the appropriate energy range. The bin width is 0.92 keV and the data collection period was 13.6 days.*

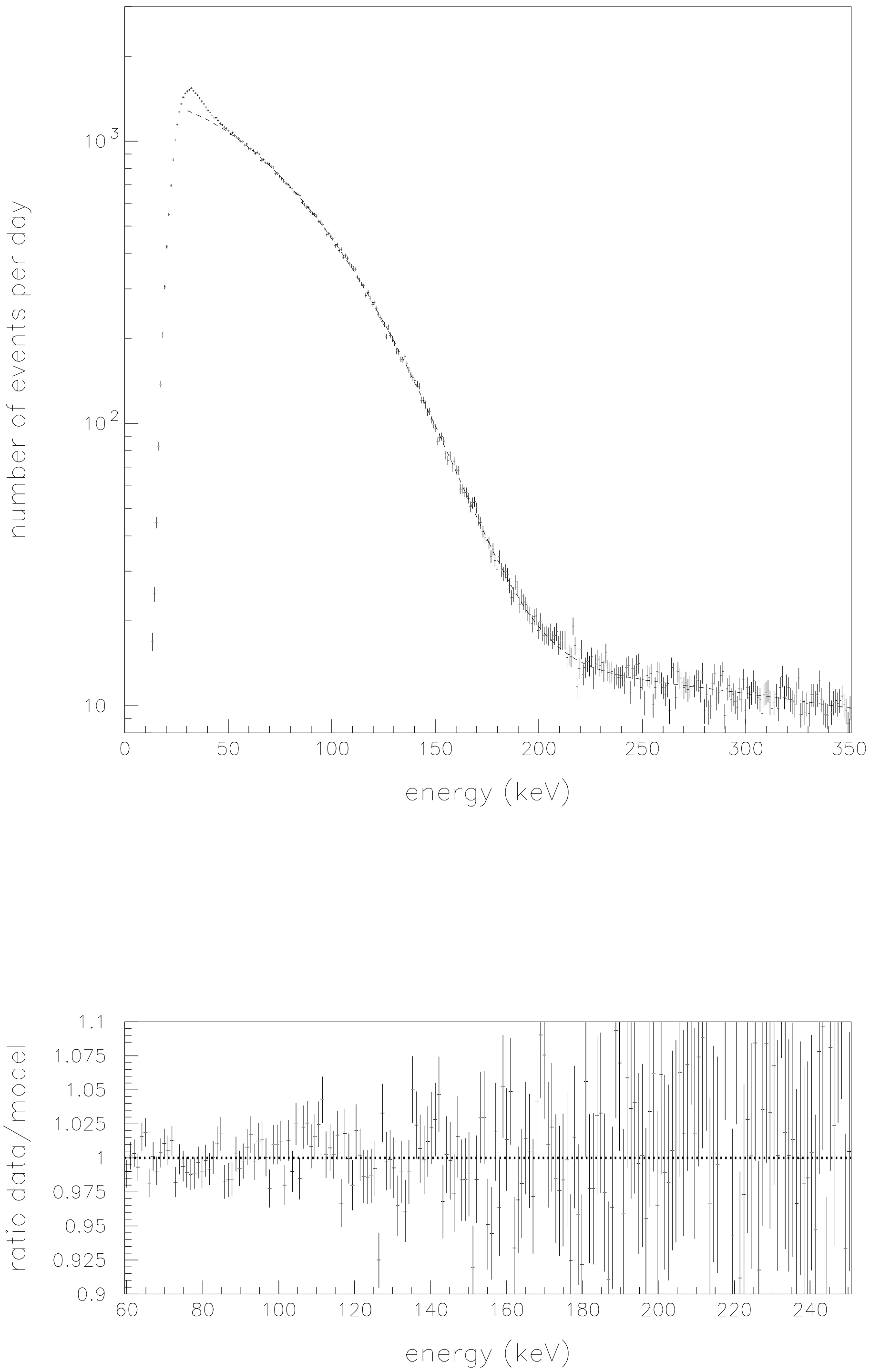
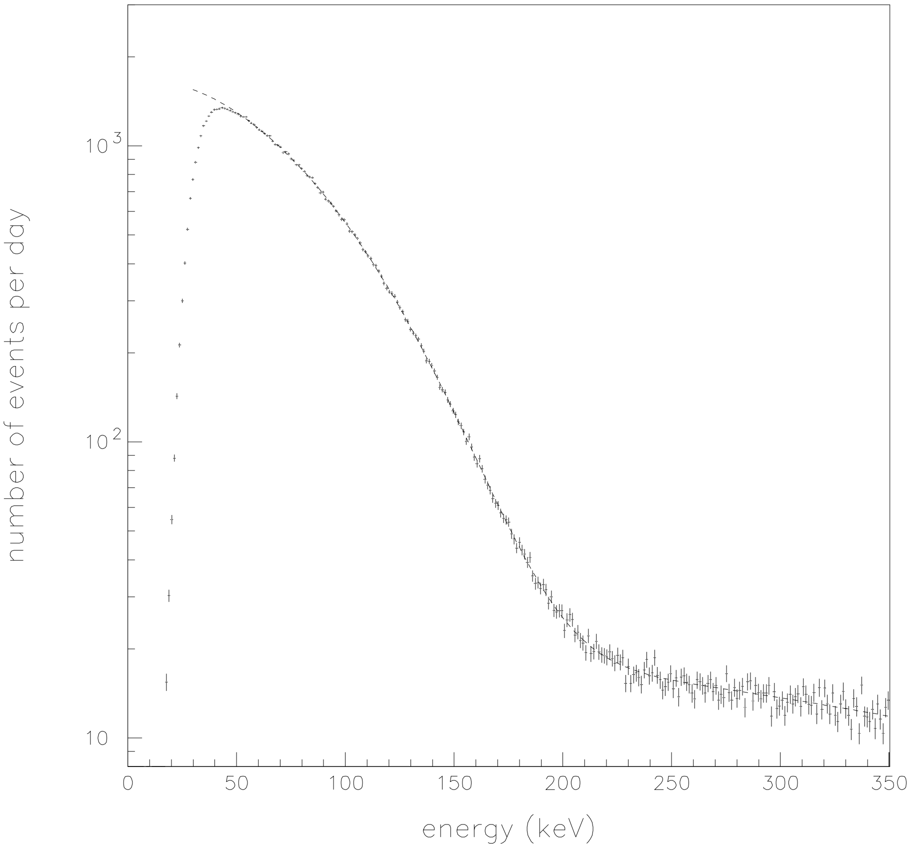


Figure 3: *Data and fit of Period 2. The bin width corresponds to 0.99 keV and the data collection time comprises 10.6 days.*



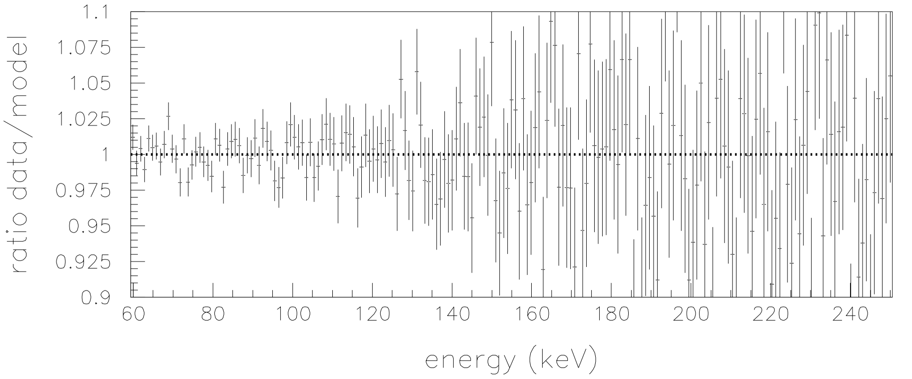


Figure 4: *Data and fit of Period 3. The low energy peak visible in Period 1 and Period 2 has disappeared due to the increase of the trigger threshold. Here, the bin width is 1.22 keV and the data collection time corresponds to 14.2 days.*