On concentration of $^{42}$Ar in liquid argon

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Abstract. Data from the DBA liquid argon ionization chamber experiment were used to obtain an estimate on the concentration of $^{42}$Ar in the Earth’s atmosphere, $9.2^{+2.2}_{-2.2} \times 10^{-21}$ atoms of $^{42}$Ar/atom of $^{40}$Ar. This corresponds to a specific activity of freshly produced liquid argon of $92^{+22}_{-46} \mu$Bq/kg.

1. Introduction
The long-lived isotope of argon, $^{42}$Ar, is a potential background source in argon-based low background detectors. The half-life of this isotope is 32.9 years and the beta decay of its daughter isotope, $^{42}$K, has the maximum electron energy of 3.52 MeV. The decay scheme is shown in Fig. 1. The potential background problem from $^{42}$Ar was first identified in connection with the ICARUS experiment [1, 2]. Ray Davis noted that a significant amount of $^{42}$Ar could be formed in the Earth’s atmosphere due to nuclear bomb tests in the upper atmosphere carried out at the end of the 1950th - beginning of the 1960th [3]. The first experimental limit on the contents of $^{42}$Ar in atmospheric argon was obtained in 1992, $<10^{-18}$ atoms $^{42}$Ar/atom $^{40}$Ar [4]). In 1995 estimates on the $^{42}$Ar concentration in atmospheric argon were carried out using the available information on nuclear tests in the atmosphere [5, 6]. During such tests $^{42}$Ar can be produced via a two-step neutron capture reaction:

$$^{40}\text{Ar}(n,\gamma)^{41}\text{Ar} \ (1)$$
$$^{41}\text{Ar}(n,\gamma)^{42}\text{Ar} \ (2)$$

Given the half-life of $^{41}$Ar ($T_{1/2} = 1.83$ h) the effective production of $^{42}$Ar requires a very high neutron flux, the condition fulfilled during nuclear tests in the atmosphere. It was shown that nuclear tests could not account for a concentration greater than $\sim 10^{-22} - 10^{-23}$ atoms $^{42}$Ar/atom $^{40}$Ar. It was suggested in 1997 [7] that a substantially greater contribution to the $^{42}$Ar concentration could be expected via the reaction $^{40}$Ar ($\alpha,2p$) $^{42}$Ar resulting from cosmic-ray interactions in the upper atmosphere. The corresponding $^{42}$Ar/$^{40}$Ar ratio was estimated to be $\sim 10^{-20}$.

A new experimental estimate of the $^{42}$Ar content came from the DBA experiment that ran in Gran Sasso Underground Laboratory during 1995-2000 [8, 9]. A liquid argon ionization chamber was used to search for the double beta decay of $^{100}$Mo. The first limit on $^{42}$Ar content was obtained from a partial data set, $< 6 \cdot 10^{-21}$ atoms $^{42}$Ar/atom $^{40}$Ar [10], followed by an updated limit from the full data set, $< 4.3 \cdot 10^{-21}$ atoms $^{42}$Ar/atom $^{40}$Ar at 90% C.L. [9]. It should be noted that a surplus of events above the expected background in a single electron energy spectrum was observed in the DBA experiment. However due to the lack of a clear
signature in this event topology in the detector it was difficult to attribute these events exactly to $^{42}$Ar. A conservative approach was therefore taken and consequently only an upper limit established.

The GERDA collaboration started data taking with the GERDA-I detector in 2011. High-purity germanium (HPGe) detectors were immersed directly in a vessel with $\sim 90$ tons of liquid argon. The 1525 keV gamma line from $^{42}$K was used to estimate the $^{42}$Ar content. In the first measurements the resulting counting rate exceeded the rate expected from [9, 10] by approximately factor 20. In addition, $^{42}$K produce a sizeable increase in background to neutrinoless double beta decay (in the energy region of $\sim 2039$ keV). It is important to note that the above estimates were obtained using the assumption of a uniform distribution of $^{42}$Ar atoms in liquid argon. It is known that as a result of $^{42}$Ar decay positive $^{42}$K ions are formed. Their spatial distribution in liquid argon is determined by the electric field configuration in the detector. A high voltage of ($\sim 4$ kV) is applied to HPGe detectors. As a result, $^{42}$K ions drift in the electric field and are accumulated in the vicinity of the detectors leading to a higher counting rate around 1525 keV gamma line than that expected from a uniform distribution of $^{42}$K in liquid argon.

The GERDA collaboration carried out a thorough study of the influence of the electric field on the level of background and methods of its reduction. Thin-walled copper containers, mini-shrouds, were constructed around HPGe detectors to isolate them from the main liquid argon volume. It resulted in a significant reduction of the background in the $0\nu$ region and allowed the $^{42}$Ar concentration in liquid argon to be reassessed, giving a range of $(7 - 12) \cdot 10^{-21}$ atoms $^{42}$Ar/atom $^{40}$Ar [11]. This value is approximately two times higher than the limit obtained in [9].

2. New DBA result

The data obtained in the DBA experiment [9] have been reanalyzed to estimate $^{42}$Ar content in the Earth’s atmosphere and the results are presented in this report. The detector was located in the Gran Sasso Underground Laboratory at a depth of 3500 m of water equivalent. The experimental setup consists of a liquid Ar ionization chamber placed in a 15 cm lead passive

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1 The GERDA low-background test facility LArGe was also used to study the $^{42}$Ar background. Its concentration was found to be $(2.2 \pm 1.0) \cdot 10^{-21}$ atoms $^{42}$Ar/atom $^{40}$Ar assuming a uniform distribution of $^{42}$K in liquid argon [12]. The effect of the electric field from PMTs immersed in the liquid argon volume was not taken into account in the estimation. As a result, the $^{42}$Ar concentration could have been underestimated, which was mentioned in [12].
Figure 2. Spectrum of single electron events in energy region 0-5.5 MeV (a) and 2.9-5.5 MeV (b) (from [9]). Measurement time is 2706 h.

shielding, an Ar purification and delivery system, readout electronics and data acquisition system. The active detection portion of the chamber is composed of alternating circular planes of anodes and cathodes with Frisch screening grids placed between them. The cathodes are made of a molybdenum foil approximately 50 mg/cm² thick. The chamber contains 14 cathodes, 15 anodes and 28 screening grids. The grid-anode distance is 5.5 mm and the grid-cathode is 14.5 mm. The sensitive volume diameter is 30 cm and its height is 56 cm. Each anode is connected to a charge-sensitive preamplifier, followed by an amplifier and a shaper. Shaped and unshaped signals are digitized by two 8-bit flash ADC with a 50 ns sampling time. Unshaped pulse analysis provides spatial information of the events. The trigger for data collection requires that at least one anode signal exceeds the threshold (~ 600 keV). The trigger causes digitized signals from all anodes to be recorded.

During the data taking period the electric fields was 1.9 kV/cm in the cathode-grid gap and 4 kV/cm for the anode-grid gap. The detector energy scale was calibrated using the $^{22}$Na ($E_\gamma = 1275$ keV) and $^{88}$Y ($E_\gamma = 1836$ keV) radioactive sources (see description in [9]). The energy resolution of the chamber was 6% (FWHM) at the $0\nu\beta\beta$ transition energy of $^{100}$Mo, 3 MeV. A single electron energy deposition signature was used to select candidate events from $^{42}$K decays. This signature requires a single energy deposition detected on one of the anodes with no signals on all the other anodes. Fig. 1 shows the spectrum of single electron candidate events.

The 3.0-3.5 MeV interval of the single electron spectrum with 201 events detected over 2706 hours was used to determine the $^{42}$Ar concentrations in liquid argon. After reanalyzing the experimental data it was noted that Monte Carlo (MC) simulations carried out in [9] did not fully take into account multiple interactions of $\gamma$ rays in a single cathode-grid volume. As a result, the energy calibration constants obtained with the $^{22}$Na and $^{88}$Y radioactive sources were
underestimated. Although the error was small and does not affect significantly low energies, in the high energy region above 3 MeV it results in a shift of approximately 100 keV. Consequently, the events counted in the 3.0-3.5 MeV in [9] should be actually attributed to the 3.1-3.6 MeV interval, which has a lower detection efficiency due to a rapidly dropping $\beta$-spectrum of $^{42}\text{K}$. New MC calculations were carried out to evaluate the detection efficiency in the 3.1-3.6 MeV energy interval for two possible distributions of $^{42}\text{K}$ in liquid argon. In the first case, positive ions of $^{42}\text{K}$ created in the $\beta$ decay of $^{42}\text{Ar}$ can maintain their charge for a sufficiently long time to be transported to negatively charged cathodes of the detector and undergo their $\beta$ decay from there, as was shown in [13]. In the second case, a uniform distribution of $^{42}\text{K}$ decay in the liquid argon volume of the detector was assumed. The efficiency was calculated to be 0.40% and 0.47% in the first and second case respectively. The calculations below assume the accumulation of positive $^{42}\text{K}$ ions on negatively charged cathodes of the chamber. However even in case of a uniform distribution of neutral $^{42}\text{K}$ atoms in the detector volume the final result will not differ by more than 15%. Recently, EXO-200 reported the fraction of $^{214}\text{Bi}^+$ ions in the $^{214}\text{Pb}$ $\beta$ decay (in liquid Xe) to be (76.4±5.7)% , while the remainder of $^{214}\text{Bi}$ atoms are neutral [14]. Assuming the same ratio of positive to neutral $^{42}\text{K}$ atoms from the $^{42}\text{Ar}$ decay (in liquid Ar) the mentioned above efficiency is estimated to be 0.42% (or only $\sim 5\%$ higher than for positively charged ions case).

Table 1 shows possible other contributions to the single electron events in the 3.1-3.6 MeV energy interval which represent a background to the $^{42}\text{Ar}$ events.

| Background                      | Number of events |
|--------------------------------|------------------|
| $\gamma$-rays from neutrons    | 45 ± 4           |
| $^{208}\text{Tl}$ (external)   | 1 ± 0.2          |
| $^{214}\text{Bi}$ (external)   | < 0.2            |
| $^{208}\text{Tl}$ (Mo)         | < 0.2            |
| $^{214}\text{Bi}$ (Mo)         | < 2              |

The contribution from $n\gamma$ reactions was evaluated by extrapolating a high energy flat spectrum down to lower energies. External background from $^{208}\text{Tl}$ and $^{214}\text{Bi}$ was estimated using information about the radio-purity of Titanium, which was the primary material of the detector vessel, as well as by fitting the Compton edge of the single electron around 2400 keV (see Fig. 2). Limits obtained from screening measurements with low-background HPGe detectors were used to constrain the background from the internal contamination inside the molybdenum cathode foils. Taking into account the above background sources the contribution of $^{42}\text{Ar}$ to the electron events observed in the 3.1-3.6 interval over the period of data taking was estimated to be $155 \pm 16 (stat) ^{+35}_{-75} (syst)$ events. The main source of the systematic error is a relatively weak constrain on $^{208}\text{Tl}$ in the Mo cathode foils (see table 1). Other sources of the systematic error include the energy scale calibration, the $^{42}\text{K}$ ion distribution in liquid argon and its effect on the detection efficiency.

The argon used in the detector had been produced $\sim 10$ years before the measurements were carried out. It means that concentration of $^{42}\text{Ar}$ in the Earth’s atmosphere is $\sim 20\%$ higher then in our Ar. Thus, given the half-life of $^{42}\text{Ar}$ (32.9 years) the concentration of the isotope in the detector’s volume is 20% lower than that in the Earth’s atmosphere.

Using the above parameters and accounting for the total fiducial Ar mass of 52 kg , the concentration of $^{42}\text{Ar}$ in the Earth’s atmosphere can be estimated as $9.2^{+2.2}_{-4.6} \times 10^{-21}$ atoms of
\( ^{42}\text{Ar} / \text{atom of} \ ^{40}\text{Ar} \). This corresponds to a specific activity of freshly produced liquid argon of \( 92^{+22}_{-46} \mu\text{Bq/kg} \), in agreement with the earlier GERDA-I result [11].

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