Measurement of the ⁸⁵Kr specific activity in the GERDA liquid argon

The GERDA collaboration^a,

M. Agostini¹⁰, A. Alexander¹⁰, G. Araujo²¹, A.M. Bakalyarov¹⁵, M. Balata¹,

I. Barabanov^{13,b}, L. Baudis²¹, C. Bauer⁹, S. Belogurov^{14,13,c}, A. Bettini^{18,19},

L. Bezrukov¹³, V. Biancacci³, E. Bossio^{17,i}, V. Bothe⁹, R. Brugnera^{18,19},

A. Caldwell¹⁶, S. Calgaro^{18,19}, C. Cattadori¹¹, A. Chernogorov^{14,15},

P.-J. Chiu²¹, T. Comellato¹⁷, V. D'Andrea³, E.V. Demidova¹⁴, N. Di Marco²,

E. Doroshkevich¹³, M. Fomina⁷, A. Gangapshev^{13,9}, A. Garfagnini^{18,19},

C. Gooch¹⁶, P. Grabmayr²⁰, V. Gurentsov¹³, K. Gusev^{7,15,17},

J. Hakenmüller^{9,d}, S. Hemmer¹⁹, W. Hofmann⁹, J. Huang²¹, M. Hult⁸,

L.V. Inzhechik^{13,e}, J. Janicskó Csáthy^{17,f}, J. Jochum²⁰, M. Junker¹,

V. Kazalov¹³, Y. Kermaïdic⁹, H. Khushbakht²⁰, T. Kihm⁹, K. Kilgus²⁰,

I.V. Kirpichnikov¹⁴, A. Klimenko^{9,7,g}, K.T. Knöpfle⁹, O. Kochetov⁷,

V.N. Kornoukhov^{13,c}, P. Krause¹⁷, V.V. Kuzminov¹³, M. Laubenstein¹,

M. Lindner⁹, I. Lippi¹⁹, A. Lubashevskiy⁷, B. Lubsandorzhiev¹³, G. Lutter⁸,

C. Macolino³, B. Majorovits¹⁶, W. Maneschg⁹, G. Marshall¹⁰, M. Misiaszek⁵,

M. Morella², Y. Müller²¹, I. Nemchenok^{7,g}, M. Neuberger¹⁷, L. Pandola⁴,

K. Pelczar⁸, L. Pertoldi^{17,19}, P. Piseri¹², A. Pullia¹², C. Ransom²¹,

L. Rauscher²⁰, M. Redchuk¹⁹, S. Riboldi¹², N. Rumyantseva^{15,7}, C. Sada^{18,19},

S. Sailer⁹, F. Salamida³, S. Schönert¹⁷, J. Schreiner⁹, A-K. Schütz^{20,h},

O. Schulz¹⁶, M. Schwarz¹⁷, B. Schwingenheuer⁹, O. Selivanenko¹³,

E. Shevchik⁷, M. Shirchenko⁷, L. Shtembari¹⁶, H. Simgen⁹, A. Smolnikov^{9,7},

D. Stukov¹⁵, S. Sullivan⁹, A.A. Vasenko¹⁴, A. Veresnikova¹³, C. Vignoli¹,

K. von Sturm^{18,19}, T. Wester⁶, C. Wiesinger¹⁷, M. Wojcik⁵, E. Yanovich¹³,

B. Zatschler⁶, I. Zhitnikov⁷, S.V. Zhukov¹⁵, D. Zinatulina⁷, A. Zschocke²⁰, K. Zuber⁶, and G. Zuzel⁵.

¹INFN Laboratori Nazionali del Gran Sasso, Assergi, Italy

²INFN Laboratori Nazionali del Gran Sasso and Gran Sasso Science Institute, Assergi, Italy

³INFN Laboratori Nazionali del Gran Sasso and Università degli Studi dell'Aquila, L'Aquila, Italy

⁴INFN Laboratori Nazionali del Sud, Catania, Italy

⁵Institute of Physics, Jagiellonian University, Cracow, Poland

 6 Institut für Kern- und Teilchenphysik, Technische Universität Dresden, Dresden, Germany

⁷Joint Institute for Nuclear Research, Dubna, Russia

⁸European Commission, JRC-Geel, Geel, Belgium

⁹Max-Planck-Institut für Kernphysik, Heidelberg, Germany

¹⁰Department of Physics and Astronomy, University College London, London, UK

¹¹INFN Milano Bicocca, Milan, Italy

¹²Dipartimento di Fisica, Università degli Studi di Milano and INFN Milano, Milan, Italy

¹³Institute for Nuclear Research of the Russian Academy of Sciences, Moscow, Russia

¹⁴Institute for Theoretical and Experimental Physics, NRC "Kurchatov Institute", Moscow, Russia

¹⁵National Research Centre "Kurchatov Institute", Moscow, Russia

¹⁶Max-Planck-Institut für Physik, Munich, Germany

¹⁷Physik Department, Technische Universität München, Germany

 18 Dipartimento di Fisica e Astronomia, Università degli Studi di Padova, Padua, Italy

¹⁹INFN Padova, Padua, Italy

²⁰Physikalisches Institut, Eberhard Karls Universität Tübingen, Tübingen, Germany

²¹Physik-Institut, Universität Zürich, Zurich, Switzerland

 $\mathbf{2}$

Abstract The radioactive isotope 85 Kr is found in sig-1 nificant quantities in the atmosphere largely due to 2 nuclear industry. Its β -decay with a half-life of 10.7 3 years and a Q-value of 687 keV is a dangerous back-4 ground source for low-threshold noble gas and liquid 5 detectors, which distill their detector medium from air. The GERDA experiment was operating high-purity germanium detectors immersed in a clean liquid argon bath 8 deep underground to search for neutrinoless double beta 9 decay with unprecedented sensitivity. The ⁸⁵Kr specific 10 activity in the liquid argon at the start of the second 11 phase of the experiment has been determined to be 12 $(0.36 \pm 0.03) \,\mathrm{mBq/kg}$ through an analysis of the full 13 subsequent data set that exploits the excellent γ -ray 14 spectroscopic capabilities of GERDA. 15

16 1 Introduction

¹⁷ The β -decay of ⁸⁵Kr ($T_{1/2} = 10.7 \text{ yr}, Q_{\beta} = 687 \text{ keV}$) is a ¹⁸ background in low-energy-threshold detectors employing ¹⁹ noble gases or liquids cryogenically distilled from the ²⁰ atmosphere as detector medium [1].

The presence of ⁸⁵Kr in the atmosphere is largely 21 anthropogenic: being a nuclear fission product, it can 22 reach the atmosphere in spent nuclear fuel reprocess-23 ing plants, nuclear weapon tests or accidents. As a re-24 sult, the average atmospheric ⁸⁵Kr specific activity¹ has 25 steadily increased since the beginning of the nuclear 26 industry era to an average global value of $1-2 \text{ Bq/m}^3$ [2-27 4]. Moreover, concentrations are typically higher nearby 28 nuclear reprocessing facilities and generally higher in the 29 northern hemisphere than in the southern hemisphere. 30 Specific meteorological conditions also induce regional 31 differences. 32

Since the ⁸⁵Kr activity in distilled gases or liquids 33 correlates to that in air at the production facility and at 34 distillation time, its initial value can significantly vary 35 across different batches. Moreover, ⁸⁵Kr can leak from 36 the atmosphere into the experiment over time, depend-37 ing on the detector technology. The WARP collaboration 38 has reported an activity of (0.12 ± 0.09) Bq/kg in atmo-39 spheric liquid argon (LAr) by directly constraining the 40

 $^1\mathrm{In}$ the following we use 'activity' in the sense of 'specific activity'.



Figure 1 Simplified ⁸⁵Kr decay scheme [13]. The decay channel studied in this work is highlighted in bold.

 85 Kr β -decay spectrum [5]. Using the same method, the 41 DarkSide collaboration has measured an unexpectedly 42 high activity of (2.05 ± 0.13) mBq/kg in underground 43 sourced LAr, potentially due to atmospheric leaks or 44 from natural fission underground [6]. Liquid xenon (LXe) 45 experiments typically remove ⁸⁵Kr by cryogenic distilla-46 tion [7, 8] or gas chromatography [9, 10]. A concentra-47 tion of natural krypton (^{nat}Kr) of 480 ppq (mol/mol) in 48 LXe (corresponding to $0.14 \,\mu Bq/kg$ of ^{85}Kr , assuming a $^{85}Kr/^{nat}Kr$ abundance of $2 \cdot 10^{-11}$) has been mea-49 50 sured using rare gas mass spectroscopy (RMGS) [11] 51 in the XENONnT detector after filling and reduced to 52 (56 ± 36) ppq through a krypton distillation column [8]. 53 144 ppq (g/g) of ^{nat}Kr (27 nBq/kg of ^{85}Kr) have been 54 measured through a liquid nitrogen cold trap in the 55 LZ detector after purification through gas chromatogra-56 phy [12]. 57

⁸⁵Kr decays to ⁸⁵Rb via β-decay with a half-life of 10.7 yr. In 0.43% of the cases an excited ⁸⁵Rb nucleus is produced, which de-excites to the ground state by emitting a 514 keV γ-ray with a half-life of 1 µs [13]. A simplified decay scheme is shown in fig. 1.

58

59

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

The GERmanium Detector Array (GERDA) experiment operated high-purity germanium (HPGe) detectors in a cryogenic bath of atmospheric LAr. Full absorption of 514 keV γ -rays in germanium following ⁸⁵Rb de-excitations produces a narrow peak-like signature in the GERDA HPGe detectors, thanks to their excellent energy resolution [14] and the fact that β particles are typically fully absorbed in argon. To measure the activity of ⁸⁵Kr nuclei in LAr, the number of events in the full energy peak (FEP) can be therefore extracted with a simple analytical model and then corrected by the detection efficiency, determined through a Monte Carlo simulation.

This article is organized as follows: we start by giving a summary of the experimental setup in section 2, then proceed to a description of the data set selection in section 3. The determination of the ⁸⁵Kr decay signature

^a correspondence: gerda-eb@mpi-hd.mpg.de

 $^{^{\}rm b}$ deceased

^c also at: NRNU MEPhI, Moscow, Russia

^d present address: Duke University, Durham, NC USA

^ealso at: Moscow Inst. of Physics and Technology, Russia

^fpresent address: Semilab Zrt, Budapest, Hungary

galso at: Dubna State University, Dubna, Russia

^hpresent address: Nuclear Science Division, Berkeley, USA

ⁱpresent address: IRFU, CEA, Université Paris-Saclay, Gif-sur-Yvette, France

and conversion factor between observed counts and ⁸⁵Kr 80 activity at filling time through Monte Carlo simulations 81 is presented in section 4. The statistical techniques em-82 ployed to analyze the data, including likelihood function 83 and fit model, are detailed in section 5, followed by an 84 assessment of systematic uncertainties in section 6. The 85 final results on the ⁸⁵Kr activity and their interpretation 86 are discussed in sections 7 and 8, respectively.

87

2 The GERDA experiment 88

The GERDA experiment, decommissioned at the begin-89 ning of 2020, was primarily aimed at searching for the 90 lepton number violating neutrinoless double beta $(0\nu\beta\beta)$ 91 decay [15] deep underground at the Laboratori Nazion-92 ali del Gran Sasso (LNGS) of the Istituto Nazionale 93 di Fisica Nucleare (INFN), in Italy. HPGe detectors, 94 isotopically-enriched in 76 Ge at $\sim 88\%$, were arranged 95 in a closely-packed, low-background string array and 96 operated in a 64 m³ LAr cryostat [16], inside a 590 m³ 97 water tank instrumented with photomultiplier tubes 98 (PMTs). The latter, together with scintillating panels 99 placed at the top of the experiment, constituted a passive 100 and active shield against laboratory and cosmic back-101 grounds [17]. The cryostat was filled between November 102 and December 2009 with 5.0-grade (i.e. 99.999% pure) 103 LAr, distilled from the atmosphere at the Linde 2 facility 104 in Trieste (Italy). At LNGS the LAr was transferred 105 from the Linde tanker via a 6.3 m^3 storage tank and 106 an about 30 m long vacuum-insulated pipe with an 107 ultra-fine filter at the end in the cryostat (see section 4.1 108 of ref. [16] for more details); no further distillation or 109 purification was done. After this first major filling, only 110 few minor top-ups took place during the lifetime of the 111 experiment. The fraction of the argon gas volume was 112 about 5% of the LAr volume. In consideration of the 113 measures taken to seal the LAr from the atmosphere, in 114 order to preserve its optical properties in the absence of 115 an online purification system, a ⁸⁵Kr re-contamination 116 during data taking is not expected. Moreover, the *in-situ* 117 ⁸⁵Kr production rate due to cosmic rays or spontaneous 118 fission of ²³⁸U is negligible at LNGS. 119

In the second phase of the experiment (Phase II), 120 the array consisted of 10 semi-coaxal (Coax) detectors 121 (including 3 detectors with natural isotopic abundance) 122 and 30 Broad Energy Germanium (BEGe) detectors, ar-123 ranged in a seven-string layout. The detector array was 124 125 surrounded by a scintillation light readout instrumentation made by two sub-systems: low-activity PMTs and 126 wavelength-shifting (WLS) fibers coupled to silicon pho-127 tomultipliers [18, 19]. Each HPGe detector string was 128



Figure 2 The energy spectrum of the GERDA data around the region of interest at the 85 Kr FEP (514 keV), with labels indicating the prominent spectral features and the expected contribution from the $2\nu\beta\beta$ decay in orange. The inset shows a zoom around the region of interest with a finer binning (0.2 keV), in linear y-scale.

enclosed in a nylon cylinder to limit the collection of ra-129 dioactive potassium ions on the detector surface [20]. A 130 cylindrical copper shroud was shielding the array against 131 ²²²Rn emanating from e.g. the cryostat walls. A detailed 132 description of the Phase II experimental setup has been 133 published in [18]. The three natural HPGe detectors and 134 one Coax detector were removed and five new inverted-135 coaxial (IC) detectors were deployed during a hardware 136 upgrade in spring 2018 [21]. The coverage and radio-137 purity of the WLS fibers was also improved. The energy 138 calibration of the HPGe detectors was performed during 139 dedicated weekly calibration runs in which the detectors 140 were exposed to three 228 Th sources [14]. 141

3 Data selection

To constrain the ⁸⁵Kr activity, we consider the full 143 Phase II physics data set, corresponding to data taken 144 from December 2015 to November 2019 with a short 145 interruption in May 2018 due to the hardware upgrade. 146 The accumulated HPGe exposure usable for analysis 147 is 105.5 kg yr (61.4 kg yr before and 44.1 kg yr after 148 the upgrade). Data from detectors made from natural 149 germanium is discarded due to detector operational 150 instabilities and low exposure contribution. 151

The standard HPGe detector digital signal process-152 ing pipeline [22], including a zero-area-cusp filter for 153 energy reconstruction [23], are applied. Non-physical 154 and pile-up events are identified and removed with an 155 estimated acceptance rate of (physical) events of more 156 than 99.9% [15]. Events classified as muon-like, as well 157 as those characterized by energy depositions in multiple 158

²https://www.linde-gas.it



Figure 3 The best-fit model superimposed to data. The highlighted top-left panel shows a combination of the entire GERDA Phase II data set. The remaining panels separately show data from different detector types (BEGe, Coax and IC) and before or after the May 2018 hardware upgrade. The continuum, the 511 keV peak and the ⁸⁵Kr FEP are plotted separately with dashed lines. The difference between data and best fit model for each bin, normalized by standard deviation expected from Poisson statistics, is shown in a panel below each spectrum.

detectors are removed from the data set. Data from the LAr instrumentation and HPGe signal pulse shapes have not been included in this analysis, as they do not improve the overall accuracy in determining the ⁸⁵Kr activity. This is due to the large uncertainties in the signal acceptance efficiencies of the LAr veto and HPGe pulse shape discrimination (PSD) at these energies.

Due to different detector properties, e.g. energy resolution and efficiency, and changes in the detector configuration during the upgrade, the data is divided into five data sets: pre-upgrade BEGe, pre-upgrade Coax, postupgrade BEGe, post-upgrade Coax and post-upgrade I70 IC.

The energy spectrum of the combined data set is 172 shown in fig. 2. The main backgrounds contributing in 173 the ~ 0.5 MeV energy region and below are the decay 174 of ³⁹Ar ($Q_{\beta} = 565 \text{ keV}$) in LAr, the $2\nu\beta\beta$ decay of 175 ⁷⁶Ge, ⁴²Ar decays in LAr and decays of ⁴⁰K, ²³⁸U and 176 $^{232}\mathrm{Th}$ chain isotopes in structural materials. A detailed 177 model of the GERDA Phase II energy spectrum has 178 been published in [24]. The inset shows a zoom in the 179 region around 514 keV with a finer binning. A prominent 180 peak-like structure is visible, constituted by ⁸⁵Rb de-181

excitation 514 keV γ -rays fully absorbed in the active 182 volume of the HPGe detectors and 511 keV γ -rays from 183 positron annihilation and the ²⁰⁸Tl decay cascade. The 184 positrons originate from pair production of high energy 185 gamma rays. The excellent energy resolution of the 186 GERDA detectors [14], in particular BEGe and IC post-187 upgrade, results into a separation between the two event 188 populations. 189

Data from GERDA Phase I is not considered in this ¹⁹⁰ analysis, as the energy resolution of HPGe detectors ¹⁹¹ was not at the level required to disentangle the 511 keV ¹⁹² and the 514 keV peaks ^[25]. ¹⁹³

4 ⁸⁵Kr signature and conversion factor

The GEANT4-based [26–28] application MAGE [29], ¹⁹⁵ which implements the full GERDA Phase II experimental ¹⁹⁶ setup to a high level of detail, is used to simulate ⁸⁵Kr ¹⁹⁷ decays and track their products. The decay vertices are ¹⁹⁸ uniformly distributed in the LAr volume enclosed by a ¹⁹⁹ cylinder (radius 70 cm, height 180 cm, $m_{\rm LAr} = 3835$ kg) ²⁰⁰ centered at the HPGe array. The dimensions of such ²⁰¹



Figure 4 Top panel: horizontal spatial distribution of a sample of simulated 85 Kr decays associated with an energy deposition of 514 keV in the HPGe array. Key structural components (the radon shroud, the WLS fiber shroud and the detectors) are pictorially shown. Bottom panel: expected signature of the decay in the GERDA energy spectrum, for each detector type, normalized by 85 Kr specific activity at the start of GERDA Phase II. 10^{10} total decays have been simulated in a LAr cylinder (see text). The energy windows used to calculate the full-energy peak efficiency are shown in gray (see text).

cylinder are significantly larger than the absorption 202 length of 514 keV γ -rays in LAr (of about 10 cm) and 203 do not bias the energy distribution of the HPGe detector 204 hits. Events that deposit energy in the HPGe detectors 205 are stored on disk for further offline processing. The 206 top panel of fig. 4 shows the xy spatial distribution of a 207 sample of simulated 85 Kr decays in which the 514 keV 208 γ -ray is fully absorbed in the active volume of the de-209 tectors. A representation of the salient features of the 210 setup (HPGe detectors, WLS fiber curtain and radon 211 shroud) is overlaid. 212

At the Monte Carlo event post-processing stage, the operational status of each individual detector in the considered data taking period is taken into account. Since the 85 Kr half-life is comparable to the GERDA



Figure 5 Visualization of the exponential decrease of the ⁸⁵Kr activity (continuous blue line) during the GERDA data taking. GERDA Phase II data taking (blue areas) started at $t_0 = 25$ December 2015. The time of LAr cryostat filling, 17 December 2009, and the Phase I data taking period (dashed area), not considered in this work, are indicated. The activity A_0 is obtained by a fit to the GERDA Phase II data (see section 7).

lifetime, the exponential decrease of its activity:

$$A(t) = A_0 \exp[-\lambda(t - t_0)]$$

where $A_0 = A(t_0)$ is the ⁸⁵Kr activity at time t_0 and ²¹³ $\lambda = \ln(2)/10.7$ yr, is also taken into account. The procedure is illustrated in fig. 5. The blue solid line represents ²¹⁶ the ⁸⁵Kr decay curve while each blue-filled region corresponds to a physics data taking run. The largest silent ²¹⁷ period corresponds to the hardware upgrade works. ²¹⁸

In addition to the hardware settings, a model of the 219 HPGe detector active volume is applied, as detailed 220 in [24]. The thickness of the dead layer at the n⁺ elec- 221 trode, measured during detector characterization before 222 deployment [30], is varied at the post-processing stage 223 according to its uncertainty to estimate the impact on 224 the conversion factor (see section 6). 225

After post-processing, probability density functions 226 (pdfs) of the energy deposited in HPGe detectors are ob-227 tained from the simulated event sample for each analysis 228 data set. The bottom panel of fig. 4 shows for the three 229 GERDA detector types the expected counts in Phase II 230 for 1 Bq/kg ⁸⁵Kr in 1 keV binning. The spectra are 231 smeared with effective energy resolution curves corre-232 sponding to the Phase II data set [14]. By scaling these 233 pdfs by the 85 Kr activity (in Bq/kg) A_0 , the expected 234 event distribution detected by GERDA is obtained. It 235 is characterized by a prominent peak at 514 keV, its 236 Compton continuum, and significantly fewer high-energy 237 events resulting from the simultaneous, unlikely detec-238 tion of a β -particle that reaches the HPGe active volume 239 and a delayed 85 Rb de-excitation γ -ray. The half-life of 240 the excited nucleus $(1 \ \mu s)$ is compatible with the time 241 scale of HPGe signals, leading to the observation of 242

Table 1 Summary of the effective energy resolution (full width at half maximum) at 514 keV and conversion factor ξ ($\xi > 1$, see text for exact definition) for each analysis data set, with start of the GERDA Phase II data taking $t_0 = 25$ December 2015. Reported uncertainties on ξ include active volume effects.

Dataset	$_{\rm (keV)}^{\rm FWHM}$	$\xi/10^5 ({ m Bq/kg})^{-1}$
— before upgrade BEGe Coax	$2.2 \pm 0.2 \\ 2.7 \pm 0.2$	$\begin{array}{c} 4.88 \pm 0.14 \\ 4.44 \pm 0.34 \end{array}$
— after upgrade BEGe Coax IC	$\begin{array}{c} 1.8 \pm 0.1 \\ 3.3 \pm 1.3 \\ 2.2 \pm 0.1 \end{array}$	$\begin{array}{c} 1.89 \pm 0.05 \\ 2.98 \pm 0.09 \\ 1.137 \pm 0.008 \end{array}$

²⁴³ a single pulse carrying the summed energy from the ²⁴⁴ β -particle and the γ -ray.

Table 1 reports the conversion factor ξ of 514 keV 245 FEP events which is defined as: the number of observed 246 events N for each analysis data set is $N = A_0 \xi$. It is 247 computed from the pdfs as the difference between the in-248 tegral in the signal window [509, 519] keV (solid gray re-249 gion in fig. 4) and the background window [504, 509] keV 250 (hatched gray region), taking into account the respective 251 widths. Note that, as the signal-to-background ratio is 252 very high, the actual window choice does not affect the 253 results. The uncertainty includes the mentioned HPGe 254 detector active volume effects, and has been estimated 255 by varying the dead-layer thicknesses in their experi-256 mental uncertainty. 257

258 5 Analysis method

The ⁸⁵Kr activity is extracted through maximum likeli-259 hood estimation from the combined analysis of the five 260 data sets, in the energy window [490, 535] keV using a 261 1 keV binning. Given the GERDA energy resolution, this 262 window fully contains the 85 Kr FEP at 514 keV, the 263 signal of interest. It has been checked that the choice of 264 bin width does not have any influence on the results by 265 repeating the analysis with smaller bins. 266

The sum of the backgrounds responsible for the continuum event distribution (³⁹Ar, $2\nu\beta\beta$ and Compton scatters) discussed in section 3 is modeled as a linear function, which provides a good approximation in the analysis window. The signal is modeled with a Gaussian peak, centered at the expected energy of 514 keV and with a width given by the data set energy resolution (reported in table 1). The latter is determined by combining all ²²⁸Th calibration data, as detailed in [14]. A second Gaussian peak is introduced to describe the sum of the e⁺e⁻ annihilation peak and the ²⁰⁸Tl γ -peak. It is centered at the expected energy of 511 keV, but an additional broadening factor f is introduced to account for possible deviation in the peak width from the reference energy resolution due to Doppler effects in e^+e^- annihilation. This width σ can therefore be written as:

$$\sigma^2(E_{\text{ann}}, f) = (\text{FWHM}(E_{\text{ann}})/2.355)^2 + f^2 ,$$
 (1)

where $E_{\text{ann}} = 511 \text{ keV}$, $2.355 \approx 2\sqrt{2 \ln 2}$ and the full ²⁶⁷ width at half maximum (FWHM) is calculated from the ²⁶⁸ effective energy resolution curves [14]. ²⁶⁹

The full likelihood reads as follows:

$$\mathcal{L}(\text{data} | A_0, \vec{\vartheta}) = \prod_i^{\text{ds}} \prod_j^{\text{bins}} \text{Pois}(\nu_{ij} | \mu_{ij}(A_0, \vec{\vartheta}_i)) \times \text{Pull}(\vec{\vartheta}_i) ,$$
(2)

where $\operatorname{Pois}(\nu \mid \mu)$ is the Poisson distribution pdf, the products run over the data sets *i* and bins *j* and Pull denotes additional pull terms described in the following. The likelihood depends on the ⁸⁵Kr activity A_0 , which is a common parameter among all the 5 data sets and the only parameter of interest, and on a set of nuisance parameters $\vec{\vartheta}$ that are data set specific and affect both the signal and background distributions. Finally, ν_{ij} denotes the number of observed events in the data set *i* and bin *j*, and μ_{ij} is the expectation value for the same data set and bin. The latter is given by the sum of signal and background contributions in that bin $\mu_{ij} = b_{ij} + s_{ij}$. The expected number of signal events can be written as:

$$s_{ij} = A_0 \,\xi_i \,\int_{\Delta E_j} dE \,\mathcal{N}(E \,|\, E_K + \delta_i(E_K), \sigma_i^2(E_K)) \,,$$

where $\mathcal{N}(E \mid \mu, \sigma^2)$ is the normal distribution and $E_K = 514$ keV. The expression depends on the ⁸⁵Kr activity A_0 , the ⁸⁵Kr FEP conversion factor ξ_i (defined in section 4), the energy scale systematic bias term $\delta_i(E_K)$, and the energy resolution

$$\sigma_i(E_K) = \text{FWHM}(E_K)_i / 2.355$$

The expected number of background events can be written as:

$$b_{ij} = \int_{\Delta E_j} dE \left[N_i^b \operatorname{Pol1}(E, \vec{p}_i) + N_i^s \mathcal{N}(E \mid E_{\operatorname{ann}} + \delta_i(E_{\operatorname{ann}}), \sigma_i^2(E_{\operatorname{ann}}, f)) \right],$$

i.e. the sum of a linear contribution $N_i^b \operatorname{Pol1}(E, \vec{p}_i)$, ²⁷⁰ which depends on the normalization of the linear distribution N_i^b and its parameters \vec{p}_i , both data set specific, ²⁷¹ and a normal contribution, describing the 511 keV peak, ²⁷³ which depends on the normalization N_i^s , the energy ²⁷⁴

scale bias $\delta_i(E_{\text{ann}})$, and the broadened energy resolu-275 tion given by equation 1. All the parameters entering 276 this last normal term are data set specific, except for the 277 broadening factor f, which is kept the same for all the 278 data sets. We verified that the first-order polynomial 279 function describes the continuum in this energy region 280 well and that a second-order polynomial function does 281 not improve the fit. 282

A product of normal pull terms

$$\operatorname{Pull}(\vec{\vartheta_i}) = \prod_k \mathcal{N}(\vartheta_{ik} \mid \ldots)$$

is included in the likelihood in eq. (2) to constrain some 283 of the nuisance parameters, namely the conversion factor 284 ξ_i , the energy scale bias δ_i and the energy resolution 285 σ_i , according to their expected distribution. These will 286 be discussed in more detail in section 6. All the other 287 nuisance parameters, namely the parameters of the lin-288 ear background, the number of events in the 511 keV 289 peak, and the broadening, are free and are left uncon-290 strained. To estimate the uncertainty on the parameters 291 of interest, the profile likelihood is used. 292

²⁹³ 6 Systematic uncertainties

In this section, the uncertainties affecting the conversion factor ξ are first discussed and evaluated. They can be categorized into: data quality cuts, cryostat top-ups, and HPGe active volume. Uncertainties affecting the energy scale and resolution are discussed in the last part of the section.

Data quality cuts — Physical events with an energy 300 in the region of interest are accepted with an efficiency 301 larger than 99.9% [15]. If both the β -particle and the sub-302 sequent 514 keV γ -ray from ⁸⁵Kr decay reach the active 303 volume of the same HPGe detector, the delay of the two 304 pulses (the half-life of the excited 85 Rb state is $\sim 1 \mu$ s) 305 in the digitized HPGe signal could be, in principle, large 306 enough to trigger the pile-up rejection algorithm, which 307 is not modeled in Monte Carlo simulations. In practice, 308 the rate of such coincident detection in the $^{85}\mathrm{Kr}$ FEP 309 region is so low (due to the much lower detection ef-310 ficiency of 85 Kr decay β -particles, see fig. 4) that the 311 impact on the detection efficiency is negligible. 312

Cryostat top up — The GERDA cryostat has been 313 periodically refilled with small amounts of LAr between 314 2009 and 2018. This argon might have had a different 315 ⁸⁵Kr activity, compared to that in the cryostat, and 316 might have therefore had an impact on the existing con-317 tamination. We estimated a mass of additional argon of 318 roughly 2.5 tons, corresponding to less than 3% of the 319 total argon volume, additionally deployed in the experi-320 ment. The impact on the estimated ⁸⁵Kr activity has 321

been evaluated by assuming that the whole amount was deployed during the hardware upgrade works (see fig. 5), a conservative assumption that largely corresponds to reality. In such a scenario, the initial ⁸⁵Kr activity would be overestimated by less than 2%, within this analysis. ²²²

HPGe active volume — Uncertainties on the size 327 of the HPGe active volume affect the conversion factor 328 ξ . Typical sizes of the detector dead layers are 1–2 329 mm known with an uncertainty of 5-30% [30]. The 330 contribution to ξ varies according to the data set: the 331 active volume of Coax detectors is poorly known, and 332 BEGe detectors suffer from a large uncertainty too, due 333 to dead-layer growing effects. The IC active volume is, 334 on the other hand, better constrained. To determine the 335 impact on each of the ξ_i , Monte Carlo simulations have 336 been re-processed while varying the dead-layer model 337 within the respective uncertainties. The uncertainties 338 reported in table 1 include these effects, which contribute 339 with about 3% in case of BEGe detectors, 8% in case of 340 Coax and less than 1% in case of IC. 341

Energy scale and resolution — The uncertainty on en-342 ergy calibration and resolution, parametrized in eq. (2)343 by pull terms on the δ_i and σ_i nuisance parameters, 344 respectively, can be estimated based on ²²⁸Th calibra-345 tion data. Such an evaluation has been carried out by 346 focusing at 2 MeV, in the context of the $0\nu\beta\beta$ decay 347 analysis [14]. In this work, we base our estimate of 348 the uncertainty in the 0.5 MeV energy region on those 349 results and on the analysis of special low-energy calibra-350 tion data taken at the end of the GERDA data taking. 351 This procedure has been documented in detail in [31], 352 for which the energy scale and resolution uncertainties 353 have been estimated in the same energy region. The 354 FWHM with uncertainty at 514 keV for each analysis 355 data set is reported in table 1. The adopted mean cali-356 bration bias δ_i is 0 keV, with a Gaussian uncertainty of 357 0.1 keV for all the 5 analysis data sets. 358

7 Results

Figure 3 shows the fit model at the profile likelihood 360 maximum superimposed to data from the 5 data sets. 361 Additionally, the contributions from the signal (⁸⁵Kr 362 peak at 514 keV) and the background (linear background 363 plus 511 keV peak) are separately shown with dashed 364 lines. The fit yields a p-value of 0.33. The difference 365 between data and best-fit model in units of standard 366 deviation is shown below each data set. 367

The best-fit value and 68% C.L. interval of the parameter representing the 85 Kr activity in LAr at the start of the GERDA Phase II data taking $t_0 =$

25 December 2015 is:

 $A_0 = (0.36 \pm 0.03) \,\mathrm{mBq/kg}$,

where the confidence interval boundaries have been estimated assuming Gaussianity of the likelihood around its maximum and include all systematic uncertainties discussed in section 6. An exponential extrapolation at cryostat filling time t = 17 December 2009 yields an activity of (0.53 ± 0.05) mBq/kg.

To determine the impact of the systematic uncer-374 tainties, the analysis has been repeated by removing the 375 Gaussian pull terms from the likelihood and fixing the 376 value of the conversion factor ξ and the energy scale and 377 resolution nuisance parameters to their central values: 378 the resulting activity is $(0.53 \pm 0.05) \,\mathrm{mBq/kg}$ at cryostat 379 filling time. It can be therefore deduced that the sta-380 tistical contribution dominates the global uncertainty 381 budget. 382

383 8 Conclusions

The GERDA collaboration has measured the activity of 384 β -emitting ⁸⁵Kr isotopes in the atmospheric LAr batch 385 deployed in the experiment, at cryostat filling time. 386 This result has been achieved by constraining the rate 387 of γ -rays following the decay of excited ⁸⁵Rb daughters, 388 as seen by the HPGe detector array immersed in the 389 liquid. This technique is made possible by the excellent 390 γ-spectroscopy capabilities of GERDA. 391

We find a significantly lower activity than the cen-392 tral value reported by the WARP collaboration: (0.12 \pm 393 (0.09) Bq/kg in atmospheric LAr [5]. Within experimen-394 tal uncertainties, however, the two results are compat-395 ible. Our measured value is also lower than the one 396 reported by the DarkSide collaboration in underground 397 LAr: $(2.05 \pm 0.13) \text{ mBq/kg}$ [6]. The latter, unexpect-398 edly high, has been attributed to in situ contamination. 399 Given the strong influence of the details of the processes 400 of distillation and handling of gas and liquids obtained 401 from the atmosphere, the ⁸⁵Kr activity is typically sub-402 ject to a high variability across different experiments. 403

The Large Enriched Germanium Experiment for
Neutrinoless-ββ Decay (LEGEND) collaboration is currently operating the LEGEND-200 experiment at LNGS,
within the existing GERDA cryostat. About 200 kg of
HPGe detectors, immersed in a fresh batch of atmospheric LAr, will offer the possibility to repeat the measurement.

Acknowledgements The GERDA experiment is supported financially by the German Federal Ministry for Education and
Research (BMBF), the German Research Foundation (DFG),
the Italian Istituto Nazionale di Fisica Nucleare (INFN), the

Max Planck Society (MPG), the Polish National Science Cen-415 tre (NCN, Grant number UMO-2020/37/B/ST2/03905), the 416 Polish Ministry of Science and Higher Education (MNiSW, 417 Grant number DIR/WK/2018/08), the Russian Foundation 418 for Basic Research, and the Swiss National Science Founda-419 tion (SNF). This project has received funding/support from 420 the European Union's Horizon 2020 research and innovation 421 programme under the Marie Sklodowska-Curie Grant agree-422 ments no 690575 and no 674896. This work was supported 423 by the Science and Technology Facilities Council, part of the 424 UK Research and Innovation (Grant no. ST/T004169/1). The 425 institutions acknowledge also internal financial support. The 426 GERDA collaboration thanks the directors and the staff of 427 the LNGS for their continuous strong support of the GERDA 428 experiment. 429

References

 J. A. Formaggio and C. J. Martoff, "Backgrounds 431 to sensitive experiments underground," Ann. Rev. 432 Nucl. Part. Sci., vol. 54, pp. 361–412, 2004. DOI: 433 10.1146/annurev.nucl.54.070103.181248. 434

- [2] A. Kersting et al., "Krypton-85 datasets of the 435 northern and southern hemisphere collected 436 over the past 60 years," Data in Brief, vol. 33, 437 p. 106 522, 2020. DOI: https://doi.org/10. 438 1016/j.dib.2020.106522. [Online]. Available: 439 https://www.sciencedirect.com/science/ 440 article/pii/S2352340920314049. 441
- K. Winger et al., "A new compilation of the atmo-[3]442 spheric ⁸⁵krypton inventories from 1945 to 2000 443 and its evaluation in a global transport model," 444 Journal of Environmental Radioactivity, vol. 80, 445 no. 2, pp. 183-215, 2005. DOI: https://doi.org/ 446 10.1016/j.jenvrad.2004.09.005. [Online]. 447 Available: https://www.sciencedirect.com/ 448 science/article/pii/S0265931X04002887. 449
- [4] J. Ahlswede et al., "Update and improvement of the global krypton-85 emission inventory," Journal of Environmental Radioactivity, vol. 115, pp. 34– 42, 2013. DOI: https://doi.org/10.1016/ j.jenvrad.2012.07.006. [Online]. Available: https://www.sciencedirect.com/science/ article/pii/S0265931X12001816.
- [5] P. Benetti *et al.*, "Measurement of the specific 457 activity of ³⁹Ar in natural argon," *Nucl. Instr.* 458 and Meth. A, vol. 574, no. 1, pp. 83–88, 2007. DOI: 459 10.1016/j.nima.2007.01.106. 460
- [6] P. Agnes *et al.* (DARKSIDE Collaboration), "Results from the first use of low radioactivity argon in a dark matter search," *Phys. Rev. D*, vol. 93, no. 8, p. 081 101, 2016, [Addendum: Phys.Rev.D 95, 069901 (2017)]. DOI: 10.1103/PhysRevD.93. 081101. arXiv: 1510.00702 [astro-ph.CO].

536

542

- E. Aprile et al. (XENON Collaboration), "Remov-|7|467 ing krypton from xenon by cryogenic distillation 468 to the ppq level," Eur. Phys. J. C, vol. 77, no. 5, 469 p. 275, 2017. DOI: 10.1140/epjc/s10052-017-470 4757-1. arXiv: 1612.04284 [physics.ins-det]. 471
- E. Aprile et al. (XENON Collaboration), "The 8 472 XENONnT dark matter experiment," Eur. Phys. 473 J. C, vol. 84, no. 8, p. 784, 2024. DOI: 10.1140/ 474 epjc/s10052-024-12982-5. arXiv: 2402.10446 475 [physics.ins-det]. 476
- A. Ames (LZ Collaboration), "Krypton removal 9 477 via gas chromatography for the LZ experiment," 478 AIP Conf. Proc., vol. 2908, no. 1, p. 070 001, 2023. 479 DOI: 10.1063/5.0161400. 480
- [10] D. S. Akerib et al. (LZ Collaboration), "The 481 LUX-ZEPLIN (LZ) Experiment," Nucl. In-482 strum. Meth. A, vol. 953, p. 163047, 2020. 483 DOI: 10.1016/j.nima.2019.163047. arXiv: 484 1910.09124 [physics.ins-det]. 485
- [11] S. Lindemann and H. Simgen, "Krypton assay 486 in xenon at the ppq level using a gas chro-487 matographic system and mass spectrometer," 488 Eur. Phys. J. C, vol. 74, p. 2746, 2014. DOI: 489 10.1140/epjc/s10052-014-2746-1. arXiv: 490 1308.4806 [physics.ins-det]. 491
- [12]J. Aalbers et al. (LZ Collaboration), "Background 492 determination for the LUX-ZEPLIN dark mat-493 ter experiment," Phys. Rev. D, vol. 108, no. 1, 494 p. 012010, 2023. DOI: 10.1103/PhysRevD.108. 495 012010. arXiv: 2211.17120 [hep-ex]. 496
- B. Singh and J. Chen, "Nuclear Data Sheets for 497 13 A=85," Nucl. Data Sheets, vol. 116, pp. 1–162, 498 2014. DOI: 10.1016/j.nds.2014.01.001. 499
- M. Agostini et al. (GERDA Collaboration), "Cal-[14]500 ibration of the GERDA experiment," Eur. Phys. 501 J. C, vol. 81, no. 8, p. 682, 2021. DOI: 10.1140/ 502 epjc/s10052-021-09403-2. arXiv: 2103.13777 503 [physics.ins-det]. 504
- M. Agostini et al. (GERDA Collaboration), "Final [15]505 Results of GERDA on the Search for Neutrinoless 506 Double- β Decay," Phys. Rev. Lett., vol. 125, no. 25, 507 p. 252 502, 2020. DOI: 10.1103/PhysRevLett. 508 125.252502. arXiv: 2009.06079 [nucl-ex]. 509
- K. T. Knöpfle and B. Schwingenheuer, "Design [16]510 and performance of the GERDA low-background 511 cryostat for operation in water," JINST, vol. 17, 512 no. 02, P02038, 2022. DOI: 10.1088 / 1748 -513 0221 / 17 / 02 / P02038. arXiv: 2202 . 03847 514 [physics.ins-det]. 515
- M. Agostini *et al.* (GERDA Collaboration), "Flux [17]516 modulations seen by the muon veto of the GERDA 517 experiment," Astropart. Phys., vol. 84, pp. 29-35, 518

2016. DOI: 10.1016/j.astropartphys.2016.08. 519 002. arXiv: 1601.06007 [physics.ins-det]. 520

- M. Agostini et al. (GERDA Collaboration), "Up-[18]521 grade for Phase II of the GERDA experiment," 522 Eur. Phys. J. C, vol. 78, no. 5, p. 388, 2018. DOI: 523 10.1140/epjc/s10052-018-5812-2. arXiv: 524 1711.01452 [physics.ins-det]. 525
- [19]J. Janicsko-Csathy et al., "Development of an 526 anti-Compton veto for HPGe detectors operated 527 in liquid argon using silicon photo-multipliers," 528 Nucl. Instrum. Meth. A, vol. 654, pp. 225–232, 529 2011. DOI: 10.1016/j.nima.2011.05.070. arXiv: 530 1011.2748 [physics.ins-det]. 531
- A. Lubashevskiy et al., "Mitigation of ⁴²Ar/⁴²K [20]532 background for the GERDA Phase II experiment," 533 Eur. Phys. J. C, vol. 78, no. 1, p. 15, 2018. DOI: 10. 534 1140/epjc/s10052-017-5499-9. arXiv: 1708. 535 00226 [physics.ins-det].
- [21]M. Agostini et al. (GERDA Collaboration), "Char-537 acterization of inverted coaxial ⁷⁶Ge detectors in 538 GERDA for future double- β decay experiments," 539 Eur. Phys. J. C, vol. 81, no. 6, p. 505, 2021. DOI: 540 10.1140/epjc/s10052-021-09184-8. arXiv: 541 2103.15111 [physics.ins-det].
- M. Agostini, L. Pandola, and P. Zavarise, "Off-line [22]543 data processing and analysis for the GERDA exper-544 iment," J. Phys. Conf. Ser., vol. 368, L. Teodor-545 escu et al., Eds., p. 012047, 2012. DOI: 10.1088/ 546 1742-6596/368/1/012047. arXiv: 1111.3582 547 [physics.data-an]. 548
- M. Agostini et al. (GERDA Collaboration), "Im-[23]549 provement of the energy resolution via an opti-550 mized digital signal processing in GERDA Phase 551 I," Eur. Phys. J. C, vol. 75, no. 6, p. 255, 2015. 552 DOI: 10.1140/epjc/s10052-015-3409-6. arXiv: 553 1502.04392 [physics.ins-det]. 554
- M. Agostini et al. (GERDA Collaboration), "Mod-[24]555 eling of GERDA Phase II data," JHEP, vol. 03, 556 p. 139, 2020. DOI: 10.1007/JHEP03(2020)139. 557 arXiv: 1909.02522 [nucl-ex]. 558
- M. Agostini et al. (GERDA Collaboration), "Re-[25]559 sults on Neutrinoless Double- β Decay of ⁷⁶Ge from 560 Phase I of the GERDA Experiment," Phys. Rev. 561 Lett., vol. 111, no. 12, p. 122503, 2013. DOI: 10. 562 1103/PhysRevLett.111.122503. arXiv: 1307. 563 4720 [nucl-ex]. 564
- S. Agostinelli et al., "Geant4 a simulation [26]565 toolkit," Nucl. Instrum. Meth. A, vol. 506, 566 no. 3, pp. 250-303, 2003. DOI: 10.1016/S0168-567 9002(03)01368-8. 568
- [27]J. Allison et al., "Geant4 developments and appli-569 cations," IEEE Trans. Nucl. Sci., vol. 53, no. 1, 570

571		pp. 270–278, 2006. DOI: 10.1109/TNS.2006.
572		869826.
	[00]	

- J. Allison *et al.*, "Recent developments in Geant4,"
 Nucl. Instrum. Meth. A, vol. 835, pp. 186–225,
 2016. DOI: 10.1016/J.NIMA.2016.06.125.
- M. Boswell *et al.*, "MaGe a Geant4-Based Monte Carlo Application Framework for Low-Background Germanium Experiments," *IEEE Trans. Nucl. Sci.*, vol. 58, no. 3, pp. 1212–1220, 2011. DOI: 10.1109/TNS.2011.2144619.
- [30] M. Agostini *et al.* (GERDA Collaboration), "Characterization of 30 ⁷⁶Ge enriched Broad Energy Ge detectors for GERDA Phase II," *Eur. Phys. J. C*, vol. 79, no. 11, p. 978, 2019. DOI: 10.1140/
 epjc/s10052-019-7353-8. arXiv: 1901.06590 [physics.ins-det].
- M. Agostini *et al.* (GERDA Collaboration), "An improved limit on the neutrinoless double-electron capture of ³⁶Ar with GERDA," *Eur. Phys. J. C*, vol. 84, no. 1, p. 34, 2024. DOI: 10.1140/epj c/s10052-023-12280-6. arXiv: 2311.02214
 [nucl-ex].