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Production of ^{26}Al and ^{10}Be via neutron reactions in SiO_2 rocks: implications for burial dating related to the Cradle of Humankind–UNESCO World Heritage Site

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Abstract

The Cradle of Humankind World Heritage Site (COHWHS) in South Africa hosts one of the highest concentrations of hominin fossils globally. Burial Dating offers a unique opportunity to understand the formation and chronology of the complex chambers systems in the COHWHS, that is essential to reconstruct early human history, likely rooted in this region. This contribution presents the theoretical motivations and the experimental approach used to measure the production rate by neutrons of the ^{10}Be and ^{26}Al isotopes, used in the context of burial dating. In addition, the preliminary results from the measurement campaign carried out at the n_TOF facility at CERN are discussed.

Introduction

The Cradle of Humankind World Heritage Site (COHWHS), located in South Africa, represents one of the world's most important archaeological and paleoanthropological regions. The area hosts the largest concentration of hominin fossils discovered to date, including species such as Australopithecus, Paranthropus, and Homo naledi, preserved within complex dolomitic karst cave systems [1]. Understanding the formation processes, accessibility, and chronology of these subterranean environments is essential for reconstructing the evolutionary history of early hominins. Indeed, the origins of the human lineage, and consequently of our shared biological and cognitive identity, are likely deeply rooted in this region. Among the most remarkable discoveries from the COHWHS is Homo naledi, first identified in the Dinaledi Chamber in 2013 [2]. This species appears difficult to fit within the current scheme of human evolution, as it combines anatomical features resembling those of Homo sapiens (500–200 ka) with a markedly smaller endocranial volume, comparable to that of Australopithecus (~ 2 Ma). The evolutionary placement and chronology of Homo naledi therefore remain one of the open questions in paleoanthropological research.

Despite the exceptional scientific relevance of the COHWHS, many subsurface cavities remain only partially mapped, while the chronological attribution of several key fossil assemblages is still affected by significant uncertainties. Conventional excavation and stratigraphic approaches are often limited by the restricted accessibility of underground chambers and by the intrinsic difficulties associated with dating dolomitic cave sediments. In this context, the MUSTAR project was established in 2024 through a collaboration between INFN and the University of Johannesburg. The project aims to address these challenges by applying muography techniques to investigate the geological structures surrounding the caves and by developing a more rigorous physics-based framework for burial dating.

Burial dating based on cosmogenic isotopes, particularly ^{26}Al and ^{10}Be, provides a widely used method for determining the age of fossil-bearing sediments through the radioactive decay of nuclides generated by cosmic-ray interactions [3]. However, the reliability of this approach depends critically on assumptions regarding local muon, neutron fluxes and production rates, which are generally inferred from theoretical models rather than direct experimental measurements. The principal innovation proposed within MUSTAR is the in-situ measurement of cosmic-ray-induced muon and neutron fluxes at key

paleoanthropological sites, thereby improving the accuracy of chronological estimates and enabling, for the first time, the implementation of a fully experimental burial-dating protocol. Accurate burial dating also requires a precise determination of the production rates of ^{26}Al and ^{10}Be. These cosmogenic nuclides are produced through cosmic-ray interactions: fast neutrons induce spallation processes near the surface, while fast and negative muons contribute at greater depths. Although muon-induced production rates are relatively well constrained, substantial uncertainties still affect the neutron-induced component because of the limited availability of experimental nuclear cross-section data. These uncertainties propagate directly into the dating errors associated with fossil-bearing sediments.

To address these limitations, MUSTAR project carried out the first direct measurement campaign of ^{26}Al and ^{10}Be production in quartz exposed to a high-intensity neutron spectrum at the n_TOF Collaboration facility at CERN [4][5]. The n_TOF source provides a white neutron spectrum closely resembling that of cosmic rays, but with intensities several orders of magnitude higher, allowing statistically significant activation of quartz samples collected from the COHWHS in short times. Following irradiation, the resulting nuclide concentrations will be measured by Accelerator Mass Spectrometry at CIRCE Laboratory [6].

Theoretical Model

Cosmogenic nuclide burial dating has become an increasingly valuable tool in archaeology and paleoanthropology. As a geochronological technique applicable over timescales of up to approximately 5 million years, it fills an important chronological gap in these disciplines. In particular, it offers significant potential for dating Earlier Stone Age (ESA) archaeological contexts and constraining the timing of early hominin presence beyond East Africa. This technique is especially valuable in regions where conventional radiometric methods cannot be applied.

Among the established dating methods, the ^{40}Ar/^{39}Ar technique provides highly precise age determinations but requires the presence of volcanic materials, which are absent in many sedimentary contexts. Other approaches, including paleomagnetism, uranium-series dating, and electron spin resonance (ESR), are often limited by constraints related to sediment composition, applicable age ranges, or geochemical conditions. Similarly, luminescence and ESR methods are affected by uncertainties associated with dose-rate estimation and uranium uptake histories. On contrary, burial dating method is based on the radioactive decay of rare cosmogenic nuclides generated within rocks through interactions with cosmic rays. These nuclides progressively accumulate in materials exposed near the Earth's surface. Once the material is buried, for example within caves or alluvial deposits, exposure to cosmic radiation is strongly reduced or completely suppressed, and the cosmogenic nuclide concentrations subsequently decrease through radioactive decay over time [1][3].

Although several cosmogenic nuclides are produced naturally, only a limited number are suitable for geochronological applications. To be effective, a cosmogenic nuclide must be distinguishable from

naturally occurring isotopes within the sample and must be generated in sufficiently measurable quantities. Commonly used cosmogenic nuclides and their half-lives are summarized in Table 1.

Table 1
Nuclides produced by cosmic rays, used for geochronology.

Nuclide	Primary Targets	Half-life	Primary Production Mechanism
^{10}Be	O, C	1.39 Ma	Spallation, μ− capture
^{14}C	O	5730 yr	Spallation, μ− capture
^{26}Al	Si	0.71 Ma	Spallation, μ− capture
^{36}Cl	Ca, K, ^{35}Cl	0.3 Ma	Spallation, μ− capture, n capture
^{3}He	Al	Stable	Spallation
^{21}Ne	Si	Stable	Spallation, μ− capture

Among the in situ-produced cosmogenic nuclides employed in archaeology and paleoanthropology, ^{26}Al and ^{10}Be are the most extensively used. Both isotopes are produced within quartz, which represents the preferred mineral phase for cosmogenic nuclide dating. Quartz is widespread, chemically stable, and highly resistant to weathering, making it readily available in a broad range of geological environments. Its relatively simple mineralogical composition also facilitates the accurate measurement of cosmogenic nuclides, even at very low concentrations. Furthermore, quartz can be efficiently isolated and purified from contaminant phases, thereby improving analytical reliability. Its frequent occurrence within archaeological sediments and lithic materials further enhances its suitability for chronological investigations.

Quartz-bearing rocks, soils, and sediments exposed at the Earth's surface progressively accumulate ^{26}Al and ^{10}Be through interactions with cosmic rays, particularly secondary neutrons. Although the absolute concentration of these nuclides varies according to the specific exposure history of each sample, both isotopes experience the same exposure conditions; consequently, their concentration ratio evolves according to a predictable relationship. When quartz is suddenly buried—whether in a cave, river terrace, colluvial wedge, or under cultural deposits—its inherited nuclide concentrations begin to decay, following this equation:

$N_i(t) = N_{i,inh}\, e^{-t/\tau_i} + N_{i,pb}$ (1) where the subscript *inh* refers to the inherited cosmogenic nuclide concentration at the time of burial, *pb* denotes post-burial production, and *i* identifies the nuclide under consideration. For the fossil-bearing rocks relevant to this study, accelerator mass spectrometry (AMS) measurements indicate ^{10}Be concentrations ranging from 5×10^5 to 1×10^6 atoms/g and ^{26}Al concentrations ranging from 8×10^5 to 5×10^6 atoms/g.

The burial age is obtained by simultaneously solving the governing equations for ^{26}Al and ^{10}Be:

$(N_{26} - N_{26,pb})/(N_{10} - N_{10,pb}) = R_{inh}\, e^{-t/\tau_bur}$ (2) where $\tau_bur = 1/(1/\tau_{26} - 1/\tau_{10})$ is a decay constant and $Rinh = (N_{26,inh} / N_{10,inh})$. This equation constitutes the theoretical basis of cosmogenic burial dating, and the various dating approaches are derived from specific simplifications or modifications of this relationship. Three principal methods are commonly employed [3].

The first approach, known as the Simple Burial Dating method, assumes that post-burial cosmogenic nuclide production is negligible. The second approach, referred to as the Min/Max Burial Dating method, estimates a minimum burial age using the assumptions of the Simple Burial method, while the maximum burial age is obtained by accounting for post-burial production through muon-induced production profiles. The third approach, the Isochron Burial Dating method, relies on multiple samples to explicitly constrain post-burial production. Since the latter method cannot be applied to the fossil-bearing rocks considered in the present study, the analysis must focus on the first two approaches.

Within the Simple Burial Dating framework, the samples are assumed to be buried at sufficient depth such that post-burial production by muons can be neglected. Under these conditions, the governing equation simplifies to:

$N_{26}/N_{10} = R_{inh}\, e^{-t/\tau_bur}$ (3) assuming $\tau_{10} \approx \tau_{26}$, the inherited ratio can be expressed in terms of the surface production rates yieldin:

$N_{26}/N_{10} = [(P_{26}/P_{10})/(1 + N^{*}_{10}\, e^{t/\tau_10})]\, e^{-t/\tau_bur}$ (4) where P_{10} and P_{26} represent the surface production rate of ^{10}Be and ^{26}Al respectively and $N^{*}_{i} = N_i /(P_i\tau_i)$.

The Min/Max Burial Dating method is applied when burial depths are insufficient to neglect post-burial cosmogenic production. In this case, the minimum burial age is derived using the Simple Burial approach, while the maximum age is calculated by considering the highest plausible post-burial production rates experienced by the sample.

For both methods, accurate determination of production rates is essential. In the Simple Burial approach, the relevant parameter is the surface production rate, whereas in the Min/Max method the production rate at burial depth must also be evaluated to estimate post-burial contributions. Published production rates are generally obtained by combining the contributions from the different physical mechanisms responsible for cosmogenic nuclide generation, namely:

- spallation reactions induced by secondary cosmic-ray neutrons interacting with rocks;
- interactions between cosmic-ray muons and rocks, including negative muon capture and fast muon reactions.

The total production rate can be approximated as:

$P_i = \Sigma_j A_{i,j} e^{-z/L_j}$ (5) where $A_{i,j}$ represents the production rate coefficient associated with reaction j for nuclide i, and L_j denotes the corresponding attenuation length.

Using the parameterization proposed by R. Braucher et al. [7], Fig. 1a illustrates the production rates of ^{10}Be as a function of depth (similar for ^{26}Al), together with the relative contributions from the different production mechanisms. At the Earth's surface, neutron-induced spallation constitutes the dominant production source. Consequently, accurate knowledge of neutron interactions with rocks is of primary importance for reliable calculation of burial ages. The energy threshold above which the production of these isotopes can happen in quartz allows to set the energy region of interest (E_n > 10 MeV) for the measurement the cross sections. Nowadays, there are only few measurements available for reactions that produce ^{10}Be and ^{26}Al by neutron spallation [8]. To overcome the lack of experimental measurements for the cross sections, direct experimental determination of these production rates could significantly improve the reliability of burial dating by replacing currently adopted approximations with experimentally constrained values.

Experimental procedure

An accurate estimate of the production rates of ^{10}Be and ^{26}Al would require detailed knowledge of the neutron-induced reaction cross sections as a function of neutron kinetic energy. In principle, the measurement of differential cross sections would necessitate multiple irradiations using monoenergetic neutron beams covering a wide energy range, from approximately 10 MeV up to 1 GeV.

The approach adopted in this work aims instead at directly measuring the production rates of ^{10}Be and ^{26}Al by reproducing, at the n_TOF facility at CERN, the integrated effect of approximately 1 Ma of exposure to the cosmic-ray neutron flux. The n_TOF facility provides a neutron energy spectrum that, in the energy region of interest (E_n > 10 MeV), closely resembles the cosmic-ray neutron spectrum, as shown in Fig. 1b. The facility delivers three comparable neutron fluxes in the experimental areas EAR1, EAR2, and NEAR [9][10], characterized by slight variations in the position of the intra-nuclear cascade peak, located between approximately 80 and 150 MeV.

The experimental campaign involved the irradiation of samples collected from the site of interest, the Dinaledi Chamber in the Cradle of Humankind, in all three experimental areas. The ongoing activity foresees the measurement of the produced ^{10}Be and ^{26}Al isotopes at the CIRCE laboratory in Caserta [6], where Accelerator Mass Spectrometry (AMS) measurements are being performed. Furthermore, in order to relate the irradiation time at n_TOF to the equivalent natural exposure time under cosmic rays, an integral measurement of the neutron flux in the relevant energy region will be carried out in proximity to the cave site. With this approach the formula for the production rates of ^{10}Be and ^{26}Al is:

$P_{Al,Be} = \#Isotope_{Al,Be}$ *produced/Exposure time (⇔ #neutrons)* (6)

The irradiations performed with three slightly different neutron spectra allow an estimation of the systematic uncertainty associated to the limited knowledge of the cosmic-ray neutron flux. In addition, three different types of quartz samples were irradiated: chemically purified quartz from the Dinaledi Chamber, non-purified quartz from the same site, and commercially available ultra-pure quartz. All samples were prepared in powder form. The first sample underwent a chemical purification procedure, whereas the second consisted of mechanically crushed rock material. The irradiation of these representative samples enables the evaluation of possible contributions to the production rates of the cosmogenic isotopes due to contaminants present in the natural rock matrix.

For the irradiations performed at the NEAR experimental station, the three samples were exposed both with and without a B_4C filter placed in front of the beam line. The B_4C filter suppresses the low-energy component of the neutron spectrum, thereby allowing the investigation of possible contributions to the isotope production rates arising from other reactions below the energy threshold considered in the theoretical model and therefore neglected in the analysis.

The samples were prepared by pressing the quartz powder into capsules made of PEEK and sealed with Kapton foil. The mass of each sample was measured using an analytical balance. The capsules were mounted on dedicated supports designed to position the samples at the center of the neutron beam. In addition, high-purity ^{197}Au foils were placed in front of the samples. Gold is commonly used as a reference material in neutron measurements because the cross sections of neutron-induced reactions on gold are accurately known. The activation of the gold foils allows the determination of the neutron fluence impinging on the samples. Several neutron-induced reactions on gold were investigated, as reported in Table 2. The reactions most relevant for the present study are $^{197}Au(n,2n)^{196}Au$ and $^{197}Au(n,4n)^{194}Au$.

Table 2
List of the studied gold isotopes with the main decay characteristics

Isotopes	Half-life	Energy of emitted gamma	Intensity of the emitted gamma
^{198}Au	2.69 days	411.8 keV	0.960
		675.9 keV	0.008
^{196}Au	6.16 days	355.7 keV	0.87
		333 keV	0.23
^{194}Au	38 hr	328.5 keV	0.63

The threshold energies of the $^{197}Au(n,2n)^{196}Au$ and $^{197}Au(n,4n)^{194}Au$ reactions are approximately 8 MeV and 24 MeV, respectively. Consequently, these reactions provide information on the high-energy component of the neutron spectrum, which is the energy region relevant for the production of beryllium

and aluminium cosmogenic isotopes. In order to monitor also the thermal component of the neutron spectrum, the neutron-capture reaction ^{197}Au(n,γ) ^{198}Au was investigated.

All activation products are radioactive isotopes that decay through the emission of characteristic gamma rays. These gamma rays were measured using a High-Purity Germanium (HPGe) detector. By determining the number of emitted gamma rays over a fixed acquisition time, it is possible to evaluate the activity of the activated gold foils and consequently determine the neutron fluence received by the samples:

$$N_{neutrons} = (C \cdot M_{Au} \cdot \lambda \cdot t_{irr} \cdot I_{flux})/(I_{\gamma} \cdot I \cdot f_1 \cdot f_2 \cdot f_3 \cdot m \cdot N_A) \quad (7)$$

Where C is the result of the HPGe measurements, M_{Au} is the molar mass of gold, λ is the activity of the studied isotopes, t_{irr} is the irradiation time, I_{flux} is the integral of the neutron flux in the region of interest (10 MeV < E < 1 GeV), I_{γ} is the intensity of the studied gamma emission (see Table 2), I is the integral in the interest region of the convolution between the neutron flux and the cross section, $f_1 = (1 - e^{-\lambda t_irr})$, $f_2 = e^{-\lambda t_w}$, $f_3 = (1 - e^{-\lambda t_m})$, t_w is the time between the end of the irradiation and the start of the measurement inside the HPGe, t_m is the measurement time, m is the mass of the gold sample, N_A is the Avogadro number.

As complementary method, it's possible to calculate the neutron flux also starting from the nominal number of protons impinging on the spallation target to produce neutrons during the irradiation time. These two methods will be compared to check the reliability of proposed procedure.

Results

After the irradiation campaign, the first step of the analysis was focused on the gold activation to calculate the number of neutrons impinging on the samples. In the gold spectrum obtained with the HPGe, showed in Fig. 2a, the characteristic peaks corresponding to the gamma emissions listed in Table 2 are clearly visible. Each peak was fitted with a Gaussian plus a background function (see Fig. 2b).

The integrals of the Gaussian functions represent the number of emitted gammas during the measurement time. Figure 3 shows the number of neutrons impinging on each sample calculated with the Formula 7 and using the (n,2n) reaction. Figure 3 shows the results for the various samples at the NEAR station in the region of energy of interest (10 MeV < E < 1 GeV).

The first result obtained from this study was the confirmation of the irradiation uniformity across all three samples. Secondly, a first estimate of the neutron flux was obtained. This result will be validated using the (n,4n) reaction and the second method described previously and will subsequently be used to determine the equivalent exposure time required for the calculation of the production rate ^{10}Be and ^{26}Al (Formula 6).

Conclusions

This work presents the theoretical and experimental framework of the MUSTAR project, aimed at improving the reliability of cosmogenic nuclide burial dating within the Cradle of Humankind World Heritage Site. In particular, the study addresses one of the main sources of uncertainty affecting burial dating techniques, namely the knowledge of the production rates of ^{10}Be and ^{26}Al.

The irradiation campaign performed at the CERN n_TOF facility represents the first direct attempt to experimentally reproduce the production of ^{10}Be and ^{26}Al under a neutron spectrum closely resembling that of cosmic rays. The use of multiple experimental areas characterized by slightly different neutron spectra, together with the irradiation of different quartz materials, will allow the investigation of systematic uncertainties related to the neutron energy distribution as well as the possible contribution of impurities present in the natural rock matrix.

Preliminary results obtained from the analysis of gold activation foils confirmed the uniformity of the irradiation conditions across the samples and provided a first estimate of the neutron fluence in the energy region relevant for cosmogenic nuclide production. These measurements constitute a crucial step toward the determination of the production rates of ^{10}Be and ^{26}Al, which will be obtained through Accelerator Mass Spectrometry measurements at the CIRCE laboratory.

Overall, this study establishes the basis for an experimentally constrained determination of cosmogenic production rates, reducing the dependence on theoretical approximations currently adopted in burial dating models. The proposed approach has the potential to significantly improve the accuracy of burial age estimates in paleoanthropological contexts of the Cradle of Humankind and, more broadly, to strengthen the application of cosmogenic nuclide burial dating in archaeology and geochronology.

Declarations

Author Contribution

S. Rabaglia and N. Pieretti wrote the main manuscript text and reviewed it.

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Figures

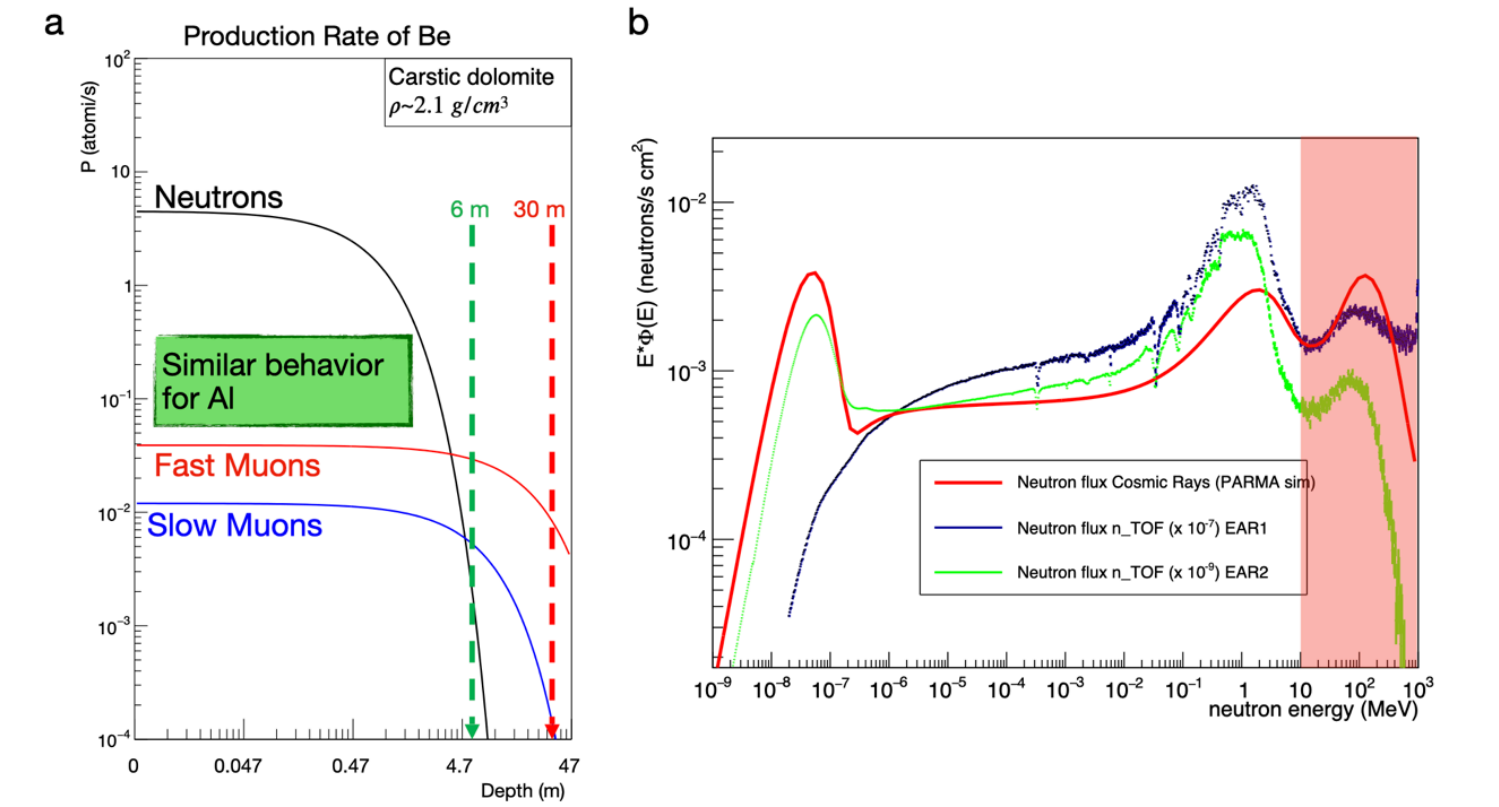


Figure 1

(a) Production rate of Berillium as fuction of depth in meter. (b) Integral neutron spectrum of the n_TOF EAR1 and EAR2 experimental locations and of simulated cosmic ray neutrons (in red the energy region of interest).

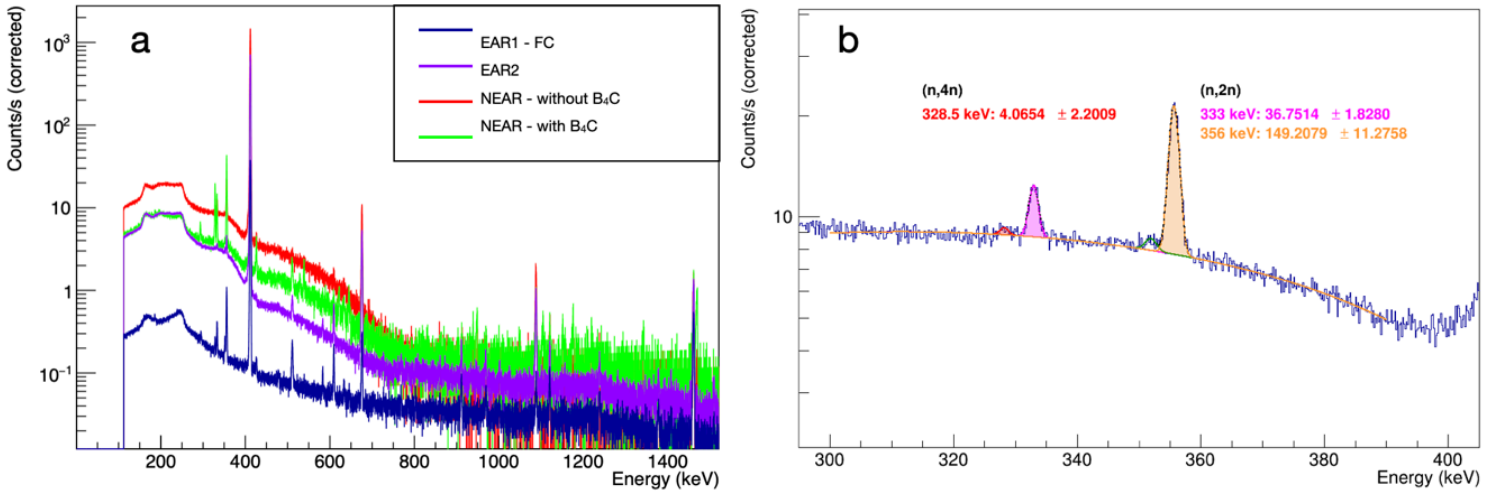


Figure 2

(a) Emission Spectrum measured by the HPGe as function of the energy. (b) Fit of the spectrum: Gaussian function for both the (n,2n) and (n,4n) reactions plus a polynomial function to describe the background.

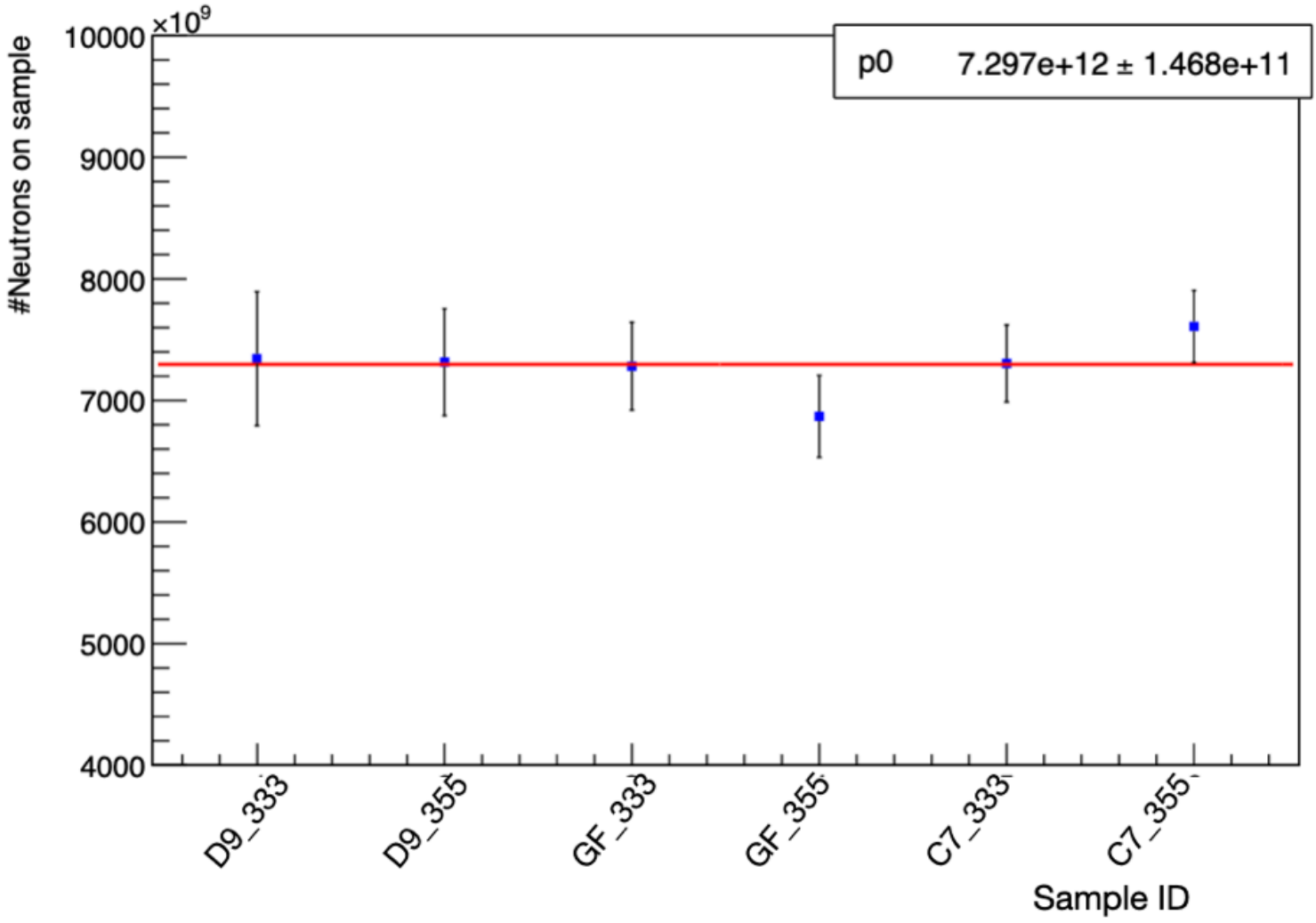


Figure 3

Number of neutrons impinging on the various samples in the region of interest (10 MeV < E < 1 GeV) in the NEAR station calculated with the reaction (n,2n).