



UNIVERSITÀ DEGLI STUDI DI TORINO

AperTO - Archivio Istituzionale Open Access dell'Università di Torino

# Cosmogenic radionuclides in the Cavezzo meteorite: Gamma-ray measurement and detection efficiency simulations

 This is a pre print version of the following article:

 Original Citation:

 Availability:

 This version is available http://hdl.handle.net/2318/1908835
 since 2023-06-06T08:25:43Z

 Published version:

 DOI:10.1016/j.apradiso.2023.110651

 Terms of use:

 Open Access

 Anyone can freely access the full text of works made available as "Open Access". Works made available under a Creative Commons license can be used according to the terms and conditions of said license. Use of all other works requires consent of the right holder (author or publisher) if not exempted from copyright protection by the applicable law.

(Article begins on next page)

# Cosmogenic radionuclides in the Cavezzo meteorite: $\gamma$ -ray measurement and detection efficiency simulations

Ilaria Bizzarri<sup>a,\*</sup>, Dario Barghini<sup>a,b</sup>, Paolo Colombetti<sup>a</sup>, Daniele Gardiol<sup>b</sup>, Sara Rubinetti<sup>c</sup>, Salvatore Mancuso<sup>b</sup>, Mario Di Martino<sup>b</sup>, Giovanni Pratesi<sup>d,e</sup>, Vanni Moggi Cecchi<sup>f</sup>, Nora Groschopf<sup>g</sup>, Andrea Aquino<sup>h</sup>, Matthias Laubenstein<sup>i</sup>, Narendra Bhandari<sup>j</sup>, Carla Taricco<sup>a</sup>

<sup>a</sup>Universitá degli Studi di Torino, Via Pietro Giuria 1, Torino, 10125, Italy

<sup>b</sup>INAF – Osservatorio Astrofisico di Torino, Via Osservatorio, 20, Pino Torinese (TO), 10025, Italy

<sup>c</sup>Dipartimento di Scienze Ambientali, Informatica e Statistica, Universitá Ca'Foscari di Venezia, Via Torino, 155, Mestre Venezia, 30172, Italy

<sup>d</sup>Dipartimento di Scienze della Terra, Università di Firenze, Via Giorgio La Pira, 4, Firenze, 50121, Italy

<sup>f</sup>Museo di Storia Naturale, Università degli Studi di Firenze, Via Romana, 17, Firenze, 50125, Italy

<sup>h</sup>Dipartimento di Scienze della Terra, Università di Pisa, Via Santa Maria, 53, Pisa, 56126, Italy

<sup>i</sup>Istituto Nazionale di Fisica Nucleare - Laboratori Nazionali del Gran Sasso, Via Giovanni Acitelli, 22, Assergi (AQ), 67100, Italy

<sup>j</sup>Science and Spirituality Research Institute, Navrangpura, Ahmedabad, 380009, India

#### Abstract

The Cavezzo meteorite was recovered on January 4<sup>th</sup>, 2020, just three days after the fall observed over Northern Italy by the all-sky cameras of the Italian PRISMA fireball network. Two specimens, weighing 3.1 g (F1) and 52.2 g (F2), were collected in the predicted strewn-field and the meteorite has been classified as an L5 anomalous chondrite. The  $\gamma$ -activity of F2 sample was measured at the Monte dei Cappuccini underground Research Station (Torino, Italy) with a large-volume HPGe-NaI(Tl) spectrometer. Thanks to the high efficiency, selectivity and low background of the spectrometer, we were able to detect many cosmogenic radioisotopes with activities even below 0.1 decay per minute (dpm). The presence of nuclides with half-lives down to few days ( $^{47}$ Ca,  $^{52}$ Mn and  $^{48}$ V) undoubtedly confirmed the recent fall of the sample. The very low activity of  $^{44}$ Ti and  $^{60}$ Co was revealed with a particular coincidence between the HPGe and NaI(Tl) detectors. To obtain the detection efficiency, we have simulated the response of the detector with the GEANT4 toolkit, once the spectrometer's dead layer thickness was estimated using standards of known activity. Moreover, the simulation of the Dhajala meteorite (H3/4 chondrite) measurement allowed us to verify that the self-absorption of the sample is correctly taken into account and validate our simulations. In this contribution, we focus on the coincidence optimization techniques and the detection efficiency computation.

Keywords: Meteorites, Cosmogenic radionuclides,  $\gamma$ -ray spectrometry

## 1. Introduction

A large number of stable and radioactive isotopes is produced in meteoroids by the interaction of cosmic rays (CR) in the interplanetary space. When a meteoroid enters the Earth's atmosphere and a meteorite sample is collected on the ground, the activity of such radionuclides can be measured. The production of these cosmogenic isotopes ends as soon as the meteorite falls, when the CR flux irradiation ceases. Freshly-fallen meteorites are of great interest in planetary science since they give

\*Corresponding author

Email address: ilaria.bizzarri@unito.it (Ilaria Bizzarri)

Preprint submitted to Applied radiation and isotopes

the opportunity to reveal cosmogenic radioisotopes with short half-lives (days or weeks), that otherwise cannot be revealed in any other natural sample.

The activity of a cosmogenic radioisotope in a meteorite mainly depends on the primary galactic cosmic rays (GCR) flux in space, roughly over few half-lives of the isotope before the meteorite falls on the Earth. It is therefore possible to study GCR flux intensity and its variations over time through the measurement of gamma-activity in meteoritic material. On the other hand, cosmogenic isotopes concentration is an important proxy of the solar activity, which is anticorrelated to GCR flux (Beer et al., 2012).

<sup>&</sup>lt;sup>e</sup> INAF – Istituto di Astrofisica e Planetologia Spaziali, Via del Fosso del Cavaliere, 100, Roma, 00133, Italy

<sup>&</sup>lt;sup>8</sup>Department of Geosciences, Johannes Gutenberg University, J.-J.-Becher-Weg 21, Mainz, 55128, Germany

Freshly-fallen meteorites can be recovered thanks to the observation of their atmospheric transit and the consequent estimation of the strewn-field of survived fragments. This is the operational principle of fireball networks, which usually deploy optical all-sky cameras dedicated to the recovery of such samples. Thanks to the dynamic and photometric analysis of the bright flight, it is also possible to estimate the pre-atmospheric size of the meteoroid and other relevant physical parameters. In this context, the measurement of long-lived cosmogenic radioisotope activity in meteorite samples provides an independent estimation of the meteoroid size. In addition, this measurement allows to define the average shielding conditions of the meteorite during its CR exposure age.

On January 1st, 2020, the PRISMA all-sky camera network recorded a brilliant fireball in the skies of Northern Italy. Thanks to these observations, the expected strewn-field was confined in an area of about 5 km<sup>2</sup> near the municipality of Cavezzo, Modena (Gardiol et al., 2021). Such analysis allowed to recover two meteorite specimens in the predicted area just three days after the event was observed. The analysis of geochemical, mineralogical and petrographic properties of both fragments supported the classification of the Cavezzo meteorite as an L5 anomalous chondrite (Pratesi et al., 2021), being the first of this class. Similar observations about the atmospheric path of fallen meteorites are available only for 35 among all the officially classified meteorites (Colas et al., 2020; Gardiol et al., 2021). The analysis of fresh-fallen meteorites, for which the pre-atmospheric orbit has been determined, is of utmost importance in planetary science, enabling for instance investigations between particular meteorite groups and their source region in the Solar System.

To determine the activities of cosmogenic radionuclides in Cavezzo, we used a large-volume and highefficiency HPGe-NaI(Tl) spectrometer located in the underground Laboratory of Monte dei Cappuccini (Torino, Italy). Such detectors allow for a non-destructive and highly selective measurement of the  $\gamma$ -activity of the counted sample. Several meteorites have been measured in this facility (see e.g. Taricco et al., 2008; Colombetti et al., 2008, 2013; Taricco et al., 2016). In particular, the activity of <sup>44</sup>Ti measured in 22 meteorites fallen to Earth in the last 250 years, has been used to probe the GCR flux and solar activity long-term modulation (Asvestari et al., 2017; Mancuso et al., 2018, 2019).

The measurement of the main mass of the Cavezzo meteorite indicated the presence of fifteen cosmogenic isotopes with half-life down to few days. In this paper, we focus on the methods developed to identify radioiso-



Figure 1: The bigger (larger) recovered specimen (F2) of the Cavezzo meteorite. An evident fusion crust is visible on most of the surface. This specimen also presents a less pronounced secondary fusion crust that may be related to fragmentation during its atmospheric transit. A small portion of the surface does not show any fusion features probably because of the impact on the ground (Gardiol et al., 2021).

topes with low gamma-activity and numerical simulations to estimate the spectrometer detection efficiency. Section 2 gives a brief review of the Cavezzo meteorite recovery, chemical analysis and classification. In Sect. 3 we describe the instrumental setup, while Sect. 4 presents the measured spectrum and the counting rates of identified cosmogenic radioisotopes. Section 5 describes the optimization technique for the coincidence between the HPGe and the NaI(TI) detector signals, used to reveal the faint activity of <sup>44</sup>Ti and <sup>60</sup>Co. In Sect. 6 we describe numerical simulation implemented with GEANT4 for the estimation of the dead layer thickness and the detection efficiency of the HPGe spectrometer. We draw our conclusions in Sect. 7.

### 2. Cavezzo meteorite: recovery and classification

The PRISMA network, born in 2016, was conceived to achieve systematic surveillance of the Italian skies to monitor fireballs and bolides (Gardiol et al., 2016; Gardiol, 2019) and is a partner of the FRIPON collaboration (Colas et al., 2020). To date, the network deploys almost 70 stations, including operating ones and those in the installation phase, distributed all over the country. Each station is equipped with an all-sky camera operated at 30 Hz to capture the passage of bright meteors (Gardiol, 2019).

On January 1st 2020 at 18:26:53 UT, eight PRISMA stations detected a brilliant fireball over Northern Italy. The analysis of these observations pointed out that a meteorite residue was most likely to have survived the atmospheric transit and to be found on the ground. Analysis of the light-curve profile also suggested that the meteoroid body underwent fragmentation at about 30 km height from the ground, so that smaller samples could be expected along the final part of its trajectory. Three days after the bolide was observed, two meteorite specimens were found in the territory of the municipality of Cavezzo (Modena province) at coordinates 44°49'43".7 N 10°58'19".5 E, within the predicted area of fall. Specimen n.1 (F1) weighs 3.1 g while specimen n.2 (F2), the largest one, weighs 52.2 g. A picture of the biggest samples (F2) is shown in Fig 1. More details about Cavezzo search and recovery and fireball data analysis are reported in Gardiol et al. (2021).

The analysis for classification and characterization of the two samples of the Cavezzo meteorite has been carried out at the Department for Earth Science of the Firenze University (Pratesi et al., 2021). The texture, crystal chemistry, and modal mineralogy of specimen F2 are typical of an ordinary L chondrite. However, specimen F1 shows different lithological and geochemical characteristics, oxygen isotopic composition and rare element patterns. F1 sample also presents anomalous texture, structure, and modal mineralogy compared to typical L chondrites. Because of these discrepancies, the Cavezzo meteorite has been classified as an anomalous L5 ordinary chondrite.

# 3. Experimental setup

The  $\gamma$ -activity of the F2 sample was measured at the Monte dei Cappuccini underground Research Station in Torino with a large-volume, high-efficiency HPGe-NaI(Tl) spectrometer, named GEM90 (Taricco et al.). This systems consists of a hyperpure germanium (HPGe) crystal (2 kg, 95% relative efficiency, resolution ~ 2 keV) operating in coincidence with an umbrella of NaI(Tl) scintillator (55 kg). Figure 2 shows the F2 specimen located on the top of the Ge crystal, surrounded by the NaI(Tl) annulus. A passive Pb-Cd-Cu shield surrounds the spectrometer and nitrogen is continuously flushed inside this cavity to minimize the contribution of the ambient radon and its decay products, to prevent humidity and avoid condensation on the cold parts (including



Figure 2: GEM90 HPGe detector surrounded by the NaI(Tl) annulus. The F2 specimen is placed on the top of the germanium crystal. The six photomultiplier tubes on the top of the NaI annulus are also visible.

electronics). The natural shielding of the rock under which the laboratory is located (70 meter water equivalent) provides a cosmic  $\mu$  rate 30 times less than at the surface level, thus considerably reducing the background counts.

The GEM90 multi-parametric acquisition system allows for the independent recording of HPGe and NaI(Tl) signals which are stored together with their timestamps (Colombetti et al., 2008; Colombetti, 2009), allowing to perform coincidence between Ge and NaI events by post-processing these data (see Sect. 5).

# 4. Gamma-activity measurement

The  $\gamma$ -activity measurement of F2 specimen of Cavezzo took place about three weeks after the sample was recovered and lasted ~ 45 days. The counted  $\gamma$ ray spectrum in normal mode (HPGe alone) is shown in Fig. 3. Several peaks are visible, both related to natural (black) and cosmogenic (red) radioisotopes. Thanks to the high selectivity and low background of the system, and to the coincidence between the two detectors, we were able to measure the activity of fifteen cosmogenic radionuclides, down to values lower than 0.1 decay per minute (dpm). All the other peaks that are visible in the spectrum are due to the  $\gamma$  decay of naturally occurring radioisotopes in both the sample and the surrounding environment, such as <sup>238</sup>U, <sup>232</sup>Th and their daughters along the decay chain in secular equilibrium (e.g. <sup>212</sup>Pb, <sup>214</sup>Bi, <sup>226</sup>Ra, <sup>214</sup>Pb, <sup>228</sup>Ac and <sup>208</sup>Tl). The most intense photo-peak, at the energy of ~ 1461 keV<sup>1</sup>, is due to the

<sup>&</sup>lt;sup>1</sup>All energy levels, transition energies and branching ratios are retrieved from the Nuclear Structure and Decay Data database (www. nndc.bnl.gov/nudat3)



Figure 3: The Cavezzo meteorite  $\gamma$ -ray spectrum in normal mode (HPGe alone, ~ 45 days counting time). Some peaks are highlighted and associated with the related cosmogenic (red) or natural occurring (black) radionuclide.

Nuclide	Decay mode	Half-life	$E_{\gamma}$ [keV]	BR [%]	Counts per day [cpd]
<sup>47</sup> Ca	$\beta^{-}(100\%)$	4.5 d	1297.09	67	$137 \pm 71$
<sup>52</sup> Mn	$\epsilon(70.6\%)$ - $\beta^+(29.4\%)$	5.6 d	1434.09	100	$50 \pm 42$
$^{48}V$	$\epsilon(50.1\%)$ - $\beta^+(49.9\%)$	16.0 d	983.53	99.98	$51 \pm 7$
	· · · · ·		1312.11	98.2	$35 \pm 8$
<sup>51</sup> Cr	$\epsilon(100\%)$	27.7 d	320.08	9.91	$25 \pm 6$
<sup>7</sup> Be	$\epsilon(100\%)$	53.2 d	477.60	10.44	$31 \pm 3$
<sup>58</sup> Co	$\epsilon(85.1\%)$ - $\beta^+(14.9\%)$	70.9 d	810.76	99.45	$13 \pm 2$
<sup>56</sup> Co	$\epsilon(80.3\%)$ - $\beta^+(19.7\%)$	77.2 d	856.77	99.94	$15 \pm 2$
	· · · · · ·		1238.29	66.5	$7 \pm 2$
<sup>46</sup> Sc	$\beta^{-}(100\%)$	83.8 d	889.28	99.98	$21 \pm 2$
	• • •		1120.55	~100	$8 \pm 2$
<sup>57</sup> Co	$\epsilon(100\%)$	271.4 d	122.06	85.6	$28 \pm 3$
<sup>54</sup> Mn	$\epsilon(100\%)$	312.20 d	834.85	99.98	$195 \pm 3$
<sup>22</sup> Na	$\epsilon(9.6\%)$ - $\beta^+(90.4\%)$	2.6 y	1274.54	99.94	$130 \pm 2$
<sup>60</sup> Co*	$\beta^{-}(100\%)$	5.3 y	1173.23	99.85	$1.52 \pm 0.36$
	• • •	·	1332.49	99.98	$1.54 \pm 0.44$
<sup>44</sup> Ti*	$\epsilon(100\%)$	59.2 y	1157.02*	99.9	$4.4 \pm 0.8$
<sup>26</sup> A1	$\epsilon(18.3\%)$ - $\beta^{-}(81.7\%)$	717 ky	1129.67	2.5	$3 \pm 1$
		2	1808.65	99.8	$64.6 \pm 0.9$
<sup>40</sup> K	$\epsilon$ (10.7%) - $\beta^-$ (89.3%)	1248 My	1460.82	10.66	$327 \pm 4$

Table 1: Activity of cosmogenic radionuclides measured in the Cavezzo meteorite, reported to the date of fall. (\*) Measured with the coincidence technique. ( $\star$ ) Gamma emitted by its short-lived daughter <sup>44</sup>Sc.

decay of <sup>40</sup>K.

Table 1 lists all the detected cosmogenic isotopes, together with the relevant decay information and the measured count per day (cpd) corrected by the decay factor, that is reporting the measured activity to the date of fall. Short-lived radionuclides with half-lives up to few days, such as <sup>47</sup>Ca, <sup>52</sup>Mn and <sup>48</sup>V, are detected, thus undoubtedly confirming the recent fall of the meteorite and its link with the New Year's Eve fireball.

The measured activities of  ${}^{54}$ Mn,  ${}^{22}$ Na,  ${}^{26}$ Al and  ${}^{40}$ K are determined within an uncertainty of about 1%, while most of the other radionuclides were counted on less intense photo-peaks thus resulting in a relative error of the order of 15%.

On the other hand, <sup>47</sup>Ca and <sup>52</sup>Mn activities have been estimated with a fairly high uncertainty, since their halflife is as short as 5 days so that they were halved already 4 times at the start of the measure. We determined the activity of these radionuclides on the partial acquisition of 17 days because the two peaks were totally submerged by the background after this period. Particular attention must be paid to the detection of <sup>52</sup>Mn. The two other major lines of the <sup>52</sup>Mn decay (744.23 and 935.54 keV), with a branching ratio (BR) greater than 90%, were never detected in any partial spectrum. Two photo-peaks are indeed visible close to these energies, but their counts can be fully explained by the activities of naturally occurring radioisotopes <sup>234m</sup>Pa and <sup>214</sup>Bi<sup>2</sup>. The <sup>52</sup>Mn line at 1434.09 keV indeed suffers from background interference of  $^{234m}$ Pa at 1434.14 keV (BR = 0.00973%), but the subtraction of the two peak integrals results in a small positive residue which could be possibly attributable to <sup>52</sup>Mn decay. Concerning <sup>47</sup>Ca, we determined its activity with a relative error of approximately 50% and no background peaks were expected in the neighboring channels of the spectrum.

The peaks of  ${}^{44}$ Ti and  ${}^{60}$ Co are not visible at all in the normal HPGe spectrum shown in Fig. 3. Therefore, we exploited the higher selectivity of our spectrometer provided by the coincidence between HPGe and NaI(Tl) detectors, as described in details in the following section.

# 5. Coincidence optimization for <sup>44</sup>Ti and <sup>60</sup>Co detection

The GEM90 acquisitions on HPGe and NaI(Tl) scintillator are independently recorded by the digital acquisition chain of the detector (Colombetti et al., 2008;



Figure 4: Surface representation of the two-dimensional Ge-NaI spectrum. White circles highlight the two regions of higher counts due to the coincidence between the annihilation  $\gamma$ 's and the 1809 keV line of <sup>26</sup>Al.

Colombetti, 2009), allowing us to obtain independent spectra for the two detectors. Figure 4 plots the twodimensional spectrum, that is the number of events recorded in the HPGe detector in coincidence with events on the NaI scintillator, as a function of their energies. For instance, the sharp vertical lines at 1275 and 1809 keV represent the major  $\gamma$ 's of the <sup>22</sup>Na and <sup>26</sup>Al decays detected on the HPGe in coincidence with the annihilation radiation detected on the NaI scintillator, with two regions of higher counts in correspondence of 511 and 1022 keV, as highlighted by the white circles in Fig. 4 for <sup>26</sup>Al. Oblique lines originates from partial energy depositions on both detectors. The most intense one starts from an energy of ~ 1461 keV on both axes and is due to the major line of <sup>40</sup>K.

By the integration of the two-dimensional spectrum on the whole NaI energy range, one can obtain again the normal spectrum shown in Fig.3. On the other hand, integrating over limited ranges allows to compute coincidence spectra.

Coincidence between HPGe and NaI detectors was needed to measure <sup>44</sup>Ti and <sup>60</sup>Co activities, since it provides a substantial reduction of the background level and allows to perform event selection.

<sup>44</sup>Ti has a half-life of  $T_{1/2}$ = 59.2 ± 0.6 yr (Ahmad et al., 1998; Taricco et al.) and decays by electron capture (100%) into <sup>44</sup>Sc. Relaxation to the ground state corresponds to the emission of two  $\gamma$ s of ~ 78 keV (96.4%) and ~ 68 keV (93%) or a single  $\gamma$  of ~ 146

<sup>&</sup>lt;sup>2</sup>Line of <sup>234m</sup>Pa at 742.81 keV (BR = 0.11%) and <sup>214</sup>Bi at 934.06 keV (BR = 3.09%)



Figure 5: Ge total spectrum (panel a) and corresponding Ge coincidence spectrum obtained using both 511 and 1022 keV windows (panel b) in the energy range of 1148–1163 keV. Red and green dashed lines represent <sup>214</sup>Bi peak at 1155.21 keV and <sup>44</sup>Sc peak at 1157.02 keV, respectively. The Levemberg-Marquardt parametric fit for the <sup>214</sup>Bi peak in the normal spectrum (red curve) and for the <sup>44</sup>Sc peak in the coincidence spectrum (green curve) are also shown.

keV energy (0.092%). These low-energy peaks lie in a spectral region with high background level and cannot be accurately revealed. Therefore, to measure the activity of <sup>44</sup>Ti we can rely on the decay of its short-lived daughter <sup>44</sup>Sc ( $T_{1/2} \sim 4$  h) which is in secular equilibrium with its parent. <sup>44</sup>Sc turns into <sup>44</sup>Ca by  $\beta^+$  decay (94%) emitting a 1157.02 keV  $\gamma$  (99.9%).

Unfortunately, the  $\gamma$  line of <sup>44</sup>Sc at 1157.02 keV suffers from the interference of an intense peak at 1155.21 keV, only ~ 2 keV away, originated by the  $\beta^-$  decay of the natural occurring <sup>214</sup>Bi radioisotope. Figure 5a shows the normal Ge spectrum in the  ${}^{44}$ Sc $-{}^{214}$ Bi peak region. It is evident that the signal from <sup>44</sup>Sc is totally submerged by <sup>214</sup>Bi counts (red curve) and a confident estimation of <sup>44</sup>Ti activity is not achievable on this spectrum. To overcome this issue, we can exploit the difference between the decay of <sup>44</sup>Sc ( $\beta^+$  decay) and <sup>214</sup>Bi ( $\beta^-$  decay). The positron emitted by the  $\beta^+$  decay of <sup>44</sup>Sc annihilates with one electron within the sample to produce two collinear  $\gamma$ s of 511 keV energy simultaneously to the emission of the 1157.02 keV  $\gamma$ . Therefore, we can perform event selection by using the coincidence between the 1157 keV  $\gamma$  revealed in the Ge detector and the 511 and 1022 keV annihilation  $\gamma$ 's detected by NaI(Tl) scintillator.

To obtain the coincidence spectrum, it is crucial to determine the optimal energy windows for NaI(Tl) signals, which must be as wide as several tens of keV due to the poor resolution of such scintillators. Furthermore, the choice of the 511 keV window is particularly tricky due to a 609.32 keV (45.44%)  $\gamma$  emitted by <sup>214</sup>Bi (simultaneously to the 1155.21 keV  $\gamma$ ), that must not be included in the 511 keV window (Taricco et al., 2007; Colombetti et al., 2008).

To optimize the choice of the coincidence windows for the detection of <sup>44</sup>Ti, we applied the approach introduced in Gardiol et al. (2017). In particular, we consider the whole ensemble of all possible coincidence windows (for both 511 and 1022 keV), parametrized through their centre *c* and their half-width *w*. Then, we extract the coincidence Ge spectrum for each (*c*, *w*) and compute the <sup>44</sup>Ti and <sup>214</sup>Bi counting rates through a double peak-fit (Simonits et al., 2003; Colombetti et al., 2008). The final coincidence window is chosen according to a particular figure of merit, looking for a simultaneous minimization of <sup>214</sup>Bi interference and maximization of <sup>44</sup>Ti signalto-noise ratio.

This optimization algorithm is performed independently for both 511 and 1022 keV windows. Figure 5b shows the sum of the coincidence spectra obtained in these two cases. It is evident that the <sup>214</sup>Bi interference is strongly reduced and the <sup>44</sup>Ti peak (green curve) is well visible above background level, which has been reduced by a factor more than 20 with respect to the normal spectrum. Finally, in order to correct for the count loss due to the application of the coincidence windows, the <sup>44</sup>Ti activity measured in the coincidence spectrum is normalized to the C/N ratio of the <sup>26</sup>Al peak at 1808.65 keV (Taricco et al.), which is the ratio between the peak counts in the coincidence (C) and normal (N) spectra. This enables us to estimate the <sup>44</sup>Ti counting rate of 4.4 ± 0.8 cpd.

A similar approach is applied to measure the faint activity of <sup>60</sup>Co. This radionuclide decays through the  $\beta^-$  channel by emitting two  $\gamma$ 's at 1173.23 keV ( $\gamma_1$ , BR = 99.85%) and 1332.49 keV ( $\gamma_2$ , BR = 99.98%) from the de-excitation of the daughter <sup>60</sup>Ni. The coincidence spectra are built in two cases: the  $\gamma_1$  peak detected on the HPGe in coincidence with the  $\gamma_2$  peak detected on the NaI(TI) scintillator and vice versa.

Figure 6 shows the results of the coincidence window optimization for <sup>60</sup>Co detection. The peaks related to  $\gamma_1$ and  $\gamma_2$  are not visible in the Ge total spectrum (panels a and c) but become visible in the coincidence spectra thanks to the significant reduction of the background level (panels b and d). By selecting the optimized windows, the normalized counts of the  $\gamma_1$  peak is equal to  $1.52\pm0.36$  cpd, while for the  $\gamma_2$  line we have  $1.54\pm0.44$ cpd. In this case, the counts of both peaks are normal-



Figure 6: Coincidence window optimization for  $^{60}$ Co detection. (a) Ge normal spectra in the energy ranges centered at 1173.23 keV (panel a) and 1332.49 keV (panel c). Panel b shows the coincidence spectrum between the detection of the 1173.23 keV peak on the Ge crystal and the 1332.49 keV peak on the NaI scintillator. Vice versa for panel d. A fit of the peaks is also shown (red curves). Red dashed lines are in correspondence of the 1173.23 and 1332.49 keV peaks of  $^{60}$ Co. A two-channel binning is performed in this case over both spectra.

ized to their C/N ratio, which are estimated from a coincidence analysis performed on the measurement of the activity of our <sup>60</sup>Co standard (see Sect. 6). The measured counts of the two peaks are consistent, as a countercheck of the accuracy of our coincidence optimization method and count loss normalization, even in this challenging experimental conditions. In fact, these two <sup>60</sup>Co lines have very similar BRs (see Table 1) and the detector efficiency does not vary significantly over this energy range.

#### 6. Detection efficiency simulations with GEANT4

In order to deduce the activity of a radionuclide from the  $\gamma$  spectrum, it is necessary to estimate the detection efficiency. For this purpose, we used the Monte Carlo simulation toolkit Geant4<sup>3</sup> (Agostinelli et al., 2003; Allison et al., 2006, 2016), useful to simulate the passage of particles through the matter and developed by the Geant collaboration at CERN. It provides an extended library of virtual classes, to be implemented for the specific case study, allowing for a custom modelling of the experiment to be simulated. Thanks to this approach, Geant4 is currently used in several research areas, including high energy, nuclear and accelerator physics, as well as studies in medical and space science.

Standard	Energy [keV]	cpm	
	1129.67	$0.40 \pm 0.01$	
26 A 1	1808.65	$8.86 \pm 0.02$	
AI	1808.65 - 511*	$0.26 \pm 0.01$	
	2938.32	$0.051 \pm 0.002$	
40 1	1460.82	$74.05 \pm 0.07$	
K	1460.82 - 511*	$0.59 \pm 0.03$	
	1173.23	$1314 \pm 9$	
<sup>60</sup> Co	1332.49	$1207 \pm 13$	
	2505.69	$66 \pm 2$	

Table 2: Results of the measurements of the  ${}^{26}$ Al,  ${}^{40}$ K and  ${}^{60}$ Co standards. The counts per minute (cpm) of each considered peak are shown with their uncertainties. We also report the result on single-escape peaks of the main lines of  ${}^{26}$ Al and  ${}^{40}$ K (\*).

Energy [keV]	measured cpd	simulated cpd
1129.67	$15.1 \pm 0.7$	$16.0 \pm 0.8$
1808.65	$434.3 \pm 0.6$	$450 \pm 4$
1808.65 - 511	$11.5 \pm 0.6$	$13.2 \pm 0.8$
2938.32	$1.6 \pm 0.1$	$1.3 \pm 0.2$

Table 3: Comparison between measured and simulated cpd of <sup>26</sup>Al peaks with their uncertainties for the Dhajala meteorite.

To estimate the detection efficiency, we need to determine the actual dead layer (DL) thickness of the HPGe detector. In fact, as a consequence of years of usage and thermal cycles of the apparatus, it can increase, thus enlarging the insensitive volume of the Ge crystal. Therefore, we deduced the actual DL thickness by comparing simulation results with measurements of <sup>26</sup>Al, <sup>40</sup>K, and <sup>60</sup>Co standards. Table 2 summarizes the results of such measurements. We simulated these activities by increasing the DL thickness from its nominal value (0.7 mm, provided by the constructor) up to 5 mm, with a step of 0.3 mm. The result of such procedure for the <sup>60</sup>Co standard is shown in Fig. 7. For each peak, the DL thickness is estimated as the intersection of the simulated values (red curve) with the experimental results (black horizontal line).

The results of such procedure are shown in Fig.8 for  ${}^{26}$ Al (blue),  ${}^{40}$ K (orange), and  ${}^{60}$ Co (green) standards. Our best estimate is the weighted mean of all these values, i.e.  $3.29 \pm 0.06$  mm (red horizontal line in Fig.8).

It should be emphasized that the obtained DL thickness could not correspond with the actual thickness of the outer contact layer of the Ge crystal. On the contrary, it must be addressed as an equivalent dead layer thickness, which takes into account also possible inaccuracies in the modelling of the HPGe geometry and allows for a fine-tuning of the simulation. For example, the end and

<sup>&</sup>lt;sup>3</sup>Geant4 version 10.2 patch-2 (geant4.web.cern.ch)



Figure 7:  $^{60}$ Co standard simulated cpm as a function of the DL thickness of the Ge crystal. The red lines plot the 2<sup>nd</sup>-degree polynomial fit and the red bands mark the 68% confidence intervals of the fit. The vertical dashed black lines indicate the intersection intervals with the experimental result, represented by the horizontal black line.



Figure 8: Dead layer estimate with associated uncertainty for  $^{26}$ Al,  $^{40}$ K, and  $^{60}$ Co standards in blue, orange and green, respectively. The red horizontal lines is the weighted mean value and the shaded red bar represents its uncertainty.

hole bullet radius of the Ge crystal are given by their nominal values but their size is not precisely known, affecting the active volume determination. Furthermore, the simulation accounts for the energy deposition within the active volume, but the charge collection process is not modelled. The detection efficiency can be lowered by an incomplete charge collection at the contact layers, especially in the low-field regions at the edges of the Ge crystal.

The estimated DL is then used to simulate the measurement of the Dhajala meteorite, an H3/4 chondrite fallen on 1976 in India (Graham, 1978). This also allows us to verify that the self-absorption of the sample is correctly taken into account in the simulation. The meteorite specimen was counted at our laboratory (Taricco et al., 2008). It has a volume of 245 cm<sup>3</sup>, a mass of 706 g ( $\rho = 2.88$  g cm<sup>-3</sup>) and a roughly squared form, which



Figure 9: Snapshot of the Geant4 simulation of Cavezzo activity. The meteorite is placed over the end cap window of the HPGe detector.

was modelled as a parallelepiped of sides 7.0/6.5/5.4 cm. Its composition was obtained from Gupta et al. (1978) and normalized to 100% neglecting elements under 1% of mass fraction. The <sup>26</sup>Al activity of our sample was measured to be 52.3  $\pm$  0.22 dpm kg<sup>-1</sup>. The Dhajala spectrum was simulated 100 times.

Table 3 shows the comparison between measured and simulated counts of the  $^{26}$ Al peaks with their uncertainties, which result to be overall in agreement, except for the 1808.65 keV peak where however the discrepancy is small (3%).

Finally, the detection efficiency for each radionuclide of interest is obtained by simulating the measurement of the Cavezzo meteorite. Figure 9 shows a snapshot of the activity simulation of Cavezzo, with the meteorite placed above the end cap window of the HPGe detector. The meteorite, having a roughly cubic form, was modelled as a cube of 2.5 cm sides. For each of the revealed radioisotopes, we simulated one million decays



Figure 10: Results of th detection efficiency simulation of the <sup>57</sup>Co line at 122.06 keV. The blue histogram shows distribution of the efficiency obtained by running 100 times the decay simulation. The mean value is shown by the vertical red line and the shaded red bar represents the associated uncertainty.

for 100 times. We calculated the net counts as the mean of the 100 runs and, finally, we deduced the detection efficiency ( $\eta_d$ ). An example of such simulation for the 122 keV peak of <sup>57</sup>Co is shown in Fig.10. The blue histogram, which is fitted with a gaussian distribution (black curve), represents the statistics of the peak integral counts. The mean value is shown by the vertical red line, corresponding to an efficiency value of  $\eta_d = 4.299 \pm 0.002$  %.

# 7. Conclusions

Cavezzo is an L5 anomalous chondrite felt on January 1<sup>st</sup> 2020 in Northern Italy and is the first meteorite recovered by the Italian PRISMA all-sky camera network.

The  $\gamma$ -activity measurement of such meteorite revealed the presence of cosmogenic radionuclides with half-lives down to few days, thus confirming the recent fall of the sample, and we were able to measure activities below 0.1 dpm, by virtue of the high efficiency and high selectivity of our  $\gamma$ -ray spectrometer at the Monte dei Cappuccini underground Research Station in Torino (Italy).

The measurement of the faint activities of <sup>44</sup>Ti and <sup>60</sup>Co has been performed thanks to the coincidence optimization between the HPGe and the NaI(Tl) detectors. The optimization of the coincidence window allowed to almost completely remove the interfering signal due to the natural occurring <sup>214</sup>Bi, thus enabling us to estimate a <sup>44</sup>Ti counting rate of  $4.4 \pm 0.8$  cpd. By applying a similar approach, we were able to reduce the background level and to detect the 1173 and 1332 keV peaks of <sup>60</sup>Co, which normalized counts turned out to be equal to  $1.52 \pm 0.36$  cpd and  $1.54 \pm 0.44$  cpd, respectively. In general, this optimization technique is useful whenever the coincidence between any pair of events can be used to detect the presence of a given radionuclide.

We estimated the detection efficiency of our spectrometer thanks to Monte Carlo simulations with the GEANT4 toolkit. The detector equivalent DL thickness was estimated by simulating the activity of three different standards by varying its value from 0.7 mm (nominal value) up to 5 mm. The comparison between measured and simulated activities allowed as to obtain an estimation of such parameter as  $3.29 \pm 0.06$  mm.

We simulated the measurement of the Dhajala meteorite (H3/4 chondrite fallen on 1976 in India) to verify the correct modelling of the self-absorption of the sample. The overall agreement between measured and simulated counts for the main peaks of <sup>26</sup>Al provided the validation of simulation. A small but still significant bias, of about 3%, was found only for the 1808.65 keV line of the Dhajala spectrum. This may be attributable to the fact that, at present time, we are not considering the uncertainty contribution due to the indetermination of our estimate of the DL thickness. This can lead to an underestimation of the error associated to final values of decay efficiency given by our simulation, and will be addressed in future work.

Finally, the detection efficiency for each radioisotope has been evaluated by performing Monte Carlo simulations of the Cavezzo measurement with the GEANT4 toolkit. The activity of all cosmogenic radionuclides measured Cavezzo will be discussed in a forthcoming publication.

#### References

- Agostinelli, S., Allison, J., Amako, K.a., Apostolakis, J., Araujo, H., Arce, P., Asai, M., Axen, D., Banerjee, S., Barrand, G., et al., 2003. GEANT4—a simulation toolkit. Nuclear instruments and methods in physics research section A: Accelerators, Spectrometers, Detectors and Associated Equipment 506, 250–303.
- Ahmad, I., Bonino, G., Castagnoli, G.C., Fischer, S., Kutschera, W., Paul, M., 1998. Three-laboratory measurement of the <sup>44</sup>Ti half-life. Physical Review Letters 80, 2550.
- Allison, J., Amako, K., Apostolakis, J., Araujo, H., Dubois, P.A., Asai, M., Barrand, G., Capra, R., Chauvie, S., Chytracek, R., et al., 2006. Geant4 developments and applications. IEEE Transactions on nuclear science 53, 270–278.
- Allison, J., Amako, K., Apostolakis, J., Arce, P., Asai, M., Aso, T., Bagli, E., Bagulya, A., Banerjee, S., Barrand, G., et al., 2016.

Recent developments in Geant4. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 835, 186–225.

- Asvestari, E., Usoskin, I.G., Kovaltsov, G.A., Owens, M.J., Krivova, N.A., Rubinetti, S., Taricco, C., 2017. Assessment of different sunspot number series using the cosmogenic isotope <sup>44</sup>Ti in meteorites. Monthly Notices of the Royal Astronomical Society 467, 1608–1613.
- Beer, J., McCracken, K., Steiger, R., 2012. Cosmogenic radionuclides: theory and applications in the terrestrial and space environments. Springer Science & Business Media.
- Colas, F., Zanda, B., Bouley, S., Jeanne, S., Malgoyre, A., Birlan, M., Blanpain, C., Gattacceca, J., Jorda, L., Lecubin, J., et al., 2020. FRIPON: a worldwide network to track incoming meteoroids. Astronomy & Astrophysics 644, A53.
- Colombetti, P., 2009. Measurement of Cosmogenic Radionuclides in Meteorites by Gamma-ray Spectrometry: Heliospheric Modulation of Cosmic Rays over the Last Three Centuries. Ph.D. thesis. University of Turin.
- Colombetti, P., Taricco, C., Bhandari, N., Romero, A., Verma, N., Vivaldo, G., 2008. Experimental set-up for gamma-activity measurements of astromaterials, in: 2008 IEEE Nuclear Science Symposium Conference Record, IEEE. pp. 1802–1805.
- Colombetti, P., Taricco, C., Bhandari, N., Sinha, N., Di Martino, M., Cora, A., Vivaldo, G., 2013. Low γ activity measurement of meteorites using HPGe–NaI detector system. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 718, 140–142.
- Gardiol, D., 2019. News from the Italian PRISMA fireball network, in: International Meteor Conference, Pezinok-Modra, Slovakia, pp. 81–86.
- Gardiol, D., Barghini, D.and Buzzoni, A., Carbognani, A., et al., 2021. Cavezzo, the first Italian meteorite recovered by the PRISMA fireball network. Orbit, trajectory, and strewn-field. Monthly Notices of the Royal Astronomical Society 501, 1215–1227.
- Gardiol, D., Barghini, D., Colombetti, P., Taricco, C., Mancuso, S., Rubinetti, S., Di Martino, M., 2017. Improvement of the extraction method of faint signals in γ-activity measurements of meteorites. The European Physical Journal Plus 132, 1–9.
- Gardiol, D., Cellino, A., Di Martino, M., 2016. PRISMA, Italian network for meteors and atmospheric studies, in: Roggemans, A., Roggemans, P. (Eds.), International Meteor Conference Egmond, the Netherlands, 2-5 June 2016, p. 76.
- Graham, A., 1978. Meteoritical Bulletin, No. 55. Meteoritics 13.
- Gupta, S.D., Gupta, P.S., Dube, A., Gupta, N.S., Gupta, D.D., 1978. The Dhajala meteorite 1. Mineralogical Magazine 42, 493–497.
- Mancuso, S., Taricco, C., Colombetti, P., Rubinetti, S., Sinha, N., Bhandari, N., 2018. Long-term evolution of the heliospheric magnetic field inferred from cosmogenic <sup>44</sup>Ti activity in meteorites. Astronomy and Astrophysics 610, A28.
- Mancuso, S., Taricco, C., Colombetti, P., Rubinetti, S., Sinha, N., Bhandari, N., Barghini, D., Gardiol, D., 2019. Long-term heliomagnetic field variation based on cosmogenic <sup>44</sup>Ti in meteorites. Nuovo Cimento C Geophysics Space Physics C 42, 43.
- Pratesi, G., Moggi Cecchi, V., Greenwood, R.C., Franchi, I.A., Hammond, S.J., Di Martino, M., Barghini, D., Taricco, C., Carbognani, A., Gardiol, D., 2021. Cavezzo—The double face of a meteorite: Mineralogy, petrography, and geochemistry of a very unusual chondrite. Meteoritics & Planetary Science 56, 1125–1150.
- Simonits, A., Östör, J., Kálvin, S., Fazekas, B., 2003. HyperLab: A new concept in gamma-ray spectrum analysis. Journal of Radioanalytical and Nuclear Chemistry 257, 589–595.
- Taricco, C., Bhandari, N., Cane, D., Colombetti, P., Verma, N., . Galactic cosmic ray flux decline and periodicities in the interplanetary space during the last 3 centuries revealed by <sup>44</sup>Ti in meteorites.

Journal of Geophysical Research: Space Physics .

- Taricco, C., Bhandari, N., Colombetti, P., Verma, N., 2008. Mid 19th century minimum of galactic cosmic ray flux inferred from <sup>44</sup>Ti in Allegan meteorite. Advances in Space Research 41, 275–279.
- Taricco, C., Bhandari, N., Colombetti, P., Verma, N., Vivaldo, G., 2007. Experimental set-up and optimization of a gamma-ray spectrometer for measurement of cosmogenic radionuclides in meteorites. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 572, 241–243.
- Taricco, C., Sinha, N., Bhandari, N., Colombetti, P., Mancuso, S., Rubinetti, S., Barghini, D., 2016. Early 18th century cosmic ray flux inferred from <sup>44</sup>Ti in Agen meteorite. Astrophysics and Space Science 361, 1–5.