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Study of cosmogenic radionuclides in the COSINE-100 NaI(Tl) detectors

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\textbf{Abstract}

COSINE-100 is a direct detection dark matter search experiment that uses a 106 kg array of eight NaI(Tl) crystals that are kept underground at the Yangyang Underground Laboratory to avoid cosmogenic activation of radioisotopes by cosmic rays. Even though the cosmogenic activity is declining with time, there are still significant background rates from the remnant nuclides. In this paper, we report measurements of cosmogenic isotope contaminations with less than one year half-lives that are based on extrapolations of the time dependent activities of their characteristic energy peaks to activity rates at the time the crystals were deployed underground. For longer-lived $^{109}$Cd ($T_{1/2} = 1.6$ y) and $^{22}$Na ($T_{1/2} = 2.6$ y), we investigate time correlations of characteristic $\gamma$-X-ray peaks. The inferred sea-level production rates are compared with calculations based on the ACTIVIA and MENDL-2 model calculations and experimental data. For $^3$H, which has a long, 12.3 year half-life, we evaluated the activity levels from the exposure times and determined a cosmogenic activation rate that is consistent with other measurements.

\textit{Keywords:} Cosmogenic radionuclide, activity, production rate, COSINE-100

\section{1. Introduction}

There are a number of experiments that search for direct evidence for dark matter particles in the halo of our Galaxy by looking for nuclei recoiling from dark matter–nucleus scattering \cite{1, 2} and report null results. One notable exception is the DAMA/LIBRA experiment that has consistently reported the observation of an annual event-rate modulation, that could be interpreted as dark-matter signal, in an array of NaI(Tl) crystal detectors with a statistical significance that is now more than 12.9 $\sigma$ \cite{3, 4}. Although this signal has persisted for over two decades and for three different configurations of the detector, it remains controversial because it is in conflict with the bounds from other direct detection experiments using different target materials \cite{5, 6, 7, 8} and indirect searches \cite{9}. However, since these conflicts depend on the details of the models for dark matter-nucleus scattering \cite{10} and the properties of the galactic dark matter halo \cite{11, 12, 13}, a conclusive statement about the DAMA/LIBRA signal can only be made by conducting an independent experiment using the same NaI(Tl) target material. This is the prime motivation of COSINE-100 and a number of other NaI(Tl)-crystal-based experiments \cite{14, 15, 16, 17, 18}.

COSINE-100 is a dark matter direct detection exper-
Figure 1: (a) The COSINE-100 detector. From outside inward, the four shielding layers include: 3 cm thick plastic scintillator panels (green), 20 cm of lead (khaki), a 3 cm thick copper box (orange), and liquid scintillator (light blue). (b) A side view of the detector geometry used in the Geant4 simulations.

The COSINE-100 experimental setup

The experimental setup of COSINE-100, shown in Fig. 1(a), is described in detail in Ref. [19]. Eight NaI(Tl) crystals, arranged in two layers, are located in the middle of a four-layer shielding structure. From outside inward, this comprises plastic scintillator panels, a lead-brick castle, a copper box, and a tank of scintillating liquid. The eight NaI(Tl) crystal assemblies and their support table are immersed in the scintillating liquid that serves both as an active veto and a passive shield. The eight NaI(Tl) crystals were grown from powder provided by Alpha Spectra (AS). Two crystals (Crystal-5 and Crystal-8) are not considered in this paper because their low light yields result in poorer energy resolution and because of their relatively high background contamination levels, especially at low energies.

The six crystals that are considered in this analysis have light yields of about 15 photoelectrons/keV; the energy threshold for an accepted signal from a crystal is 2 keV. Selection criteria that are used to isolate scintillation-light generated signals from photomultiplier tube noise are described in detail in Ref. [19]. Events that have above-threshold signals in only one of the crystals and none in any of the other crystals or the liquid scintillator are classified as single-hit events. Those with above-threshold signals in more than one crystal and/or the liquid scintillator are classified as multiple-hit events.

Monte Carlo simulations based on the Geant4 toolkit [24] are used to better understand the background spectra from the cosmogenic isotopes in the crystals; the geometry used for these simulations is shown in Fig. 1(b).

This paper is organized as follows: The COSINE-100 detector is described in section 2. In section 3, the cosmogenic isotopes that are produced in NaI(Tl) are listed and extrapolations of the crystals’ measured activity levels to values at the time of their initial deployment underground at Y2L are described. The use of these initial activity levels to infer production rates for cosmogenic isotopes at sea level and comparing them with ACTIVIA and MENDL-2 calculations [22, 23] and with experimental data is discussed in section 4. The fitted activities of $^3$H and $^{129}$I from the background modeling are evaluated in section 5 and conclusions are provided in section 6.
Table 1: (a) Cosmogenic radionuclides in the NaI(Tl) crystals; (b) surface exposure and underground radioactivity cooling times.

(a)

| Cosmogenic isotopes | Half-life (days) | Production rate at sea level [25] (counts/kg/day) | Decay type & Energy |
|---------------------|------------------|-------------------------------------------------|--------------------|
| $^{125}$I           | 59.4             | 221                                             | EC, 30-40 and 60-70 keV |
| $^{121}$Te          | 19.17            | 93                                              | 30-40 keV in multiple-hit events |
| $^{121m}$Te         | 164.2            | 93                                              |                    |
| $^{123m}$Te         | 119.2            | 52                                              |                    |
| $^{125m}$Te         | 57.4             | 74                                              |                    |
| $^{127m}$Te         | 106.1            | 93                                              |                    |
| $^{113}$Sn          | 115.1            | 9.0                                             |                    |
| $^{109}$Cd          | 461.4            | 4.8                                             | EC, 25.5 keV and 88 keV |
| $^{22}$Na           | 951              | 66                                              | $\beta^+$, 511 keV and 1274.6 keV |
| $^{3}$H             | 4500             | 26                                              | $\beta^-$, 0-20 keV |
| $^{129}$I           | $1.57 \times 10^7$ yr | -                                               |                    |

(b)

| Crystal | Exposure time (see text) (years) | Radioactivity cooling time at Y2L (years) |
|---------|----------------------------------|------------------------------------------|
| Crystal-1 | 2                               | 3                                       |
| Crystal-2 | 0.75                            | 2.75                                    |
| Crystal-3 | > 0.75                          | 1.2                                    |
| Crystal-4 | 1.7                             | 0.5                                    |
| Crystal-6 | 0.3                             | 0.6                                    |
| Crystal-7 | 0.3                             | 0.6                                    |

3. Cosmogenic radionuclides

Although the eight NaI(Tl) crystals had underground radioactivity cooling times that range from several months to three years, there are still backgrounds from the long-lived cosmogenic isotopes that were activated by cosmic rays while they were on the surface.

To understand these backgrounds, we first considered the list of cosmogenic radioactive isotopes that are produced in NaI(Tl) reported in Ref. [25, 26, 27, 28]. In Table 1 (a), we list the contributing cosmogenic isotopes with their half lives; short-lived isotopes, for which half lives are less than a year, are $^{125}$I, $^{121}$Te, $^{121m}$Te, $^{123m}$Te, $^{125m}$Te, $^{127m}$Te, and $^{113}$Sn and long-lived isotopes are $^{109}$Cd, $^{22}$Na, $^{3}$H, and $^{129}$I.

Since the detailed cosmic ray exposure history of each crystal is unknown, we estimated the time period for each crystal’s exposure, listed in Table 1 (b), from the time between the powder production by Alpha Spectra at Grand Junction, Colorado, to the date delivered to Y2L. Since Crystal-3 has a complicated exposure history, having been repaired once before deployment at Y2L, we can only be certain that the corresponding period is more than 9 months. The radioactivity cooling time for each crystal between delivery to Y2L and the start of data-taking is also listed in Table 1 (b). The short-lived ($T_{1/2} < 1$ y) isotopes are not expected to contribute significantly to either Crystal-1 or Crystal-2 because their cooling times are long enough to reduce these activities to a negligible level. However, we expect some backgrounds from the short-lived isotopes in other crystals because their production rates at sea level, as listed in Table 1 (a), are high and their cooling times are less than or equal to a year.

Data points in Fig. 2 show the energy spectra for the six considered NaI(Tl) crystals during the first (blue) and last (green) 25 day segments of the dataset taken between October 21, 2016 and July 18, 2018. A significant reduction of peaks from short-lived cosmogenic isotopes in Crystals 4, 6, and 7 for both single- and multiple-hit events is evident, while the differences for Crystals 1 and 2 are small, as expected. To associate the specific peaks with its cosmogenic nuclide, we simulated each isotope in Table 1 as a radioactive contaminant randomly distributed inside the NaI(Tl) crystal bulk. Figure 3 shows the differences between the two data sets for Crystal-4. The subtracted spectrum is well fitted by the simulated cosmogenic components, thereby validating our selection of the main cosmogenic contributors to the low-energy single-hit distribution.

Four long-lived nuclides, $^{109}$Cd, $^{22}$Na, $^{3}$H, and $^{129}$I have low energy deposits and are, therefore, potentially troublesome. The beta-decay spectrum of tritium has an endpoint energy of 18 keV and the electron capture decay of $^{22}$Na produces ~0.8 keV X-rays. The beta decay of $^{129}$I to $^{129}$Xe* is followed by $^{129}$Xe* transitioning to the sta-
Figure 2: Background spectra for six NaI(Tl) crystals during the first (blue points) and the last (green points) 25 days of the dataset taken from October 21, 2016 to July 18, 2018. The upper plots show single-hit events and the lower ones show multiple-hit events.

Figure 3: The difference between the first and last 25 day spectra of the 1.7 year data for Crystal-4. The red histogram shows the fitted spectrum.

Once the regions where the main contributions from each cosmogenically activated isotope are identified, we look into the decay rates for each of them, integrating the rates over the specific energy ranges. We fit the decay rate over time for each component. Depending on the energy region selected for each cosmogenic, the decay rate can be fit by a constant and one or more exponential functions, as following:

$$A + B \cdot e^{-\ln(2) \frac{(x-x_0)}{t_{1/2}}}$$

where $x_0$ is the initial time, $A$ is the expected...
Table 2: Initial activity $A_0$ (mBq/kg) of $^{125}$I in each crystal as measured by the decay rate method.

| Crystal-1 | Crystal-2 | Crystal-3 | Crystal-4 | Crystal-6 | Crystal-7 |
|-----------|-----------|-----------|-----------|-----------|-----------|
| -         | -         | 9.0±0.9   | 3.4±0.1   | 5.1±0.2   | 5.4±0.2   |

flat background rate, $B$ is the rate in the units of counts/day/kg/keV in $x_0$, and $C$ is the half-life, a constant of the fit. Fig. 4 shows an example of decay rate modeling with the units given in dru (counts/day/kg/keV).

The amplitude of the exponential ($B$) can be used to calculate the activity rate (in Bq/kg) of the cosmogenic isotope at the indicated initial time:

$$Rate = \frac{\Delta E \times B}{86400 \times f_{AE}}$$

where $f_{AE}$ is the fraction of the events from that cosmogenic depositing energy in the specified integration region, which can be calculated from the simulated spectra.

3.1.1. Iodine $^{125}$I

Since iodine is one of the main components in the crystal, a significant amount of $^{125}$I is activated. However, the half-life of this isotope is short with $T_{1/2} = 59.4$ days. We define the integration region for this isotope as 60 and 70 keV of the single-hit spectrum, as shown in Fig. 4. Although there is a contribution from $^{121m}$Te in this region, it is very small due to the low activity of this isotope and more importantly, due to the very small fraction of total $^{121m}$Te events that actually deposit energy in this region. Therefore, the contribution of $^{121m}$Te in the 60 to 70 keV region is insignificant.

Fig. 5 shows the difference between the measured activities of $^{125}$I for the six crystals analyzed. The different amounts of $^{125}$I are related to the cooling time of the crystals before the start of data taking. We also compare the activities calculated in the background fit to those measured through this method.

3.1.2. Tellurium $^{121m}$Te and $^{127m}$Te

- The line chosen to investigate $^{121m}$Te is the one between 20 to 40 keV in the multiple-hit spectrum. This line is dominant in that region, unlike the $^{121m}$Te lines in the single-hit spectra. The method used is the same as described above.

- As we can see from the simulated spectra, $^{127m}$Te has only one peak in the lower energy region, around 88 keV. However, there are other components that can have significant contributions in that energy region as well, such as $^{109}$Cd and $^{121m}$Te. The contribution coming from $^{121m}$Te can be calculated based on the measurement from the multiple-hit spectrum, as described above. The contribution from $^{109}$Cd, however, is calculated based on another study, which will be described in section 3.2.2. Both of these are added to the fitting function, allowing the measurement of $^{127m}$Te activity.

Fig. 6(a), (b) show the differences between the measured activities of $^{121m}$Te and $^{127m}$Te for the six crystals analyzed. We also compare the activities calculated in the background fit to those measured through this method.

As listed in Tables 2 and 3, the initial activities when crystals
were deployed at Y2L were derived for $^{125}$I, $^{121m}$Te, and $^{127m}$Te in each crystal.

### 3.2. Long-lived isotopes

#### 3.2.1. Sodium $^{22}$Na

The decays of $^{22}$Na to $^{22}$Ne* proceed via $\beta^+$ emission (90.3%) or electron capture (9.6%) with 3.75 yr mean lifetime, followed by $^{22}$Ne* transitioning to the stable $^{22}$Ne isotope via the emission of a 1274.6 keV $\gamma$-ray with a 5.3 ps mean lifetime. The electron capture of $^{22}$Na produces $\sim$0.9 keV X-rays. As a result, $\sim$10% of the $^{22}$Na decay will simultaneously produce a 1274.6 keV $\gamma$-ray and 0.9 keV X-rays. In the case of $\beta^+$ decay, the final-state positron immediately annihilates to two 511 keV $\gamma$-rays.

![Figure 7](image-url)  
(a) Double coincidence events that deposit 511 keV of energy in Crystal-3. In the fitting, it represents the peak from $^{214}$Bi in green and the peak at 511 keV from $^{22}$Na in red.  
(b) Triple coincidence events that deposit 511 keV of energy in Crystal-3. The red line represents the peak at 511 keV from $^{22}$Na.

### Table 3: Initial activity $A_0$ (mBq/kg) of $^{121m}$Te and $^{127m}$Te in each crystal.

| Crystal-1 | Crystal-2 | Crystal-3 | Crystal-4 | Crystal-6 | Crystal-7 |
|-----------|-----------|-----------|-----------|-----------|-----------|
| $^{121m}$Te | -         | -         | 0.90±0.16 | 0.89±0.06 | 0.44±0.07 | 0.41±0.07 |
| $^{127m}$Te | -         | -         | 0.87±0.16 | 0.48±0.03 | 0.38±0.04 | 0.35±0.04 |
Table 4: The initial $^{22}$Na activity level $A_0$ (mBq/kg) as measured by triple coincidences in each crystal.

| Crystal | Activity (mBq/kg) |
|---------|------------------|
| Crystal-1 | 2.0±0.4 |
| Crystal-2 | 1.52±0.37 |
| Crystal-3 | 0.84±0.18 |
| Crystal-4 | 1.17±0.19 |
| Crystal-6 | 0.65±0.14 |
| Crystal-7 | 0.87±0.23 |

If one of the two 511 $\gamma$-rays escapes the crystal, the remaining energy deposited in the crystal will be substantially greater than 650 keV. Figure 7(a) shows the Crystal-3 energy spectrum in coincidence with a signal in the (650–1000) keV energy interval in another crystal, which are called double coincidence events. The $^{22}$Na $\beta^+$ decay events show up as the peak at 511 keV (red color).

Since the eight NaI(Tl) crystal assemblies are immersed in the scintillating liquid (LS), as described in section 2, we can also identify $^{22}$Na decay events in which the $\sim$1270 keV gamma-ray converts in the LS in coincidence with two 511 keV signals in two crystals. These are referred to as triple coincidence events. Figure 7(b) shows the peak at 511 keV (red color) in Crystal-3 contributed by triple coincidence events.

We determine the activity rate (in mBq/kg) from the double/triple coincidence event rates with the relation,

$$\text{Activity (mBq/kg)} = \frac{N \cdot \epsilon}{m \cdot t},$$

where $N$ is the number of events, $\epsilon$ is the detection efficiency obtained from a Monte Carlo simulation, $m$ is the mass of the NaI(Tl) crystal, and $t$ is the time in seconds.

Figure 8 shows the measured activities of $^{22}$Na for the six crystals analyzed by these methods, compared with the activities determined from the global background fit. The initial activities, when the crystals were first deployed at Y2L, are listed in Table 4.

3.2.2. Cadmium $^{109}$Cd

The cosmogenic isotope $^{109}$Cd decays via electron capture to an isomeric state of $^{109}$Ag, with a prompt energy deposit of 25.5 keV, the binding energy of the Ag K-shell electron. This is followed by the emission of an 88 keV $\gamma$-ray from the isomer transition of $^{109}$Ag that has a mean lifetime of 57.4 s. From the time interval distribution be-

Figure 9: Time difference between a 25.5 keV and an 88 keV signal in Crystal-3, fitted with the sum of two exponential decay functions. The $^{109}$Cd contribution with lifetime 56.29 s is shown in red.

Figure 10: Average activities of $^{109}$Cd during the first 60 days of data in each crystal.
between 25.5 keV and 88 keV signals in the same crystal, we extract the level of $^{109}$Cd from a fit with two exponential decay functions. As discussed in section 3.1.2 there are significant contributions from decay functions. As discussed in section 3.1.2 there are 88 keV and $^{121}$I around $^{109}$Cd and $^{113}$Sn and $^{121}$Te around 25 keV, which dominate the blue curve in Figure 9. The fitted mean lifetime, 59.29±13.51 s, from the exponential curve in red, as can be seen in Figure 9, is consistent with the mean lifetime of 57.4 s from the isomer transition of $^{109}$Ag.

We determined the $^{109}$Cd activity rates in mBq/kg from these measurements: Fig. 10 shows the measured activity levels for the six crystals analyzed through this method, compared with the activities determined from the global background fit. The crystal activity levels when they were first deployed at Y2L are listed in Table 5.

### 4. Comparison with ACTIVIA, MENDL-2, and other experimental results

In section 3 we describe the determination of the crystals’ cosmogenic isotope activities at the time they were first deployed underground at Y2L. However, since we do not know the details of their previous exposure conditions, such as times, locations, and altitudes, these cannot be directly related to production rates or saturation activity levels. In order to extract sea level production rates we employ a simplified mathematical model for production and decay of radionuclides.

The production rate $R$ for activation of a isotope can be expressed as

$$ R = n \cdot \sigma \cdot \Phi $$

where $n$ is the number of atomic nuclei per cm$^3$ in the activation material, $\sigma$ is the neutron capture cross section for a nucleus, and $\Phi$ is the cosmic-ray neutron flux density, i.e., the number of neutrons crossing an area of 1 cm$^2$ per second at the place of irradiation. Since cosmic-ray neutron flux $\Phi$ depends on altitude, location, and time the production rate $R$ can be calculated by scaling the reference production rate $R_0$ at sea level,

$$ R = f \cdot R_0 $$

The produced nuclide then decays according to the standard decay law, and the net rate of change for the number of existing radioactive nuclei $N$ is by the differential equation

$$ \frac{dN}{dt} = -\lambda \cdot N + R, $$

Here $\lambda$ is the decay constant: $\lambda = \frac{\ln 2}{T_{1/2}}$ ($T_{1/2}$ is the decay half-life). The solution of Eq. 6 is

$$ N = \frac{R}{\lambda} \cdot (1 - e^{-\lambda t}), $$

and the activity $A(t)$ is related to the number of existing nuclei $N$ by

$$ A(t) = \lambda \cdot N $$

When the time $t$ is sufficiently large, then a saturation activity $A_s$ is reached,

$$ A_s = f \cdot R_0 $$

The ANAIS experiment [26] realized that the cosmic-ray neutron flux $\Phi$ can be scaled from its sea level reference flux, as reported in Ref. [25], to the NaI(Tl) crystal production point in Grand Junction, Colorado (altitude=1400 m), by a factor of $f = 3.6$. Using the measured $A_0$ activity levels reported here and the exposure times listed in Table 1, we compute a production rate $R_0$ at sea level from the relation

$$ A_0 = R_0 \cdot (1 - e^{-\lambda t}) + f \cdot R_0 \cdot e^{-\lambda t}. $$

---

**Table 5: Initial activity $A_0$ (mBq/kg) of $^{109}$Cd in each crystal.**

| Crystal   | $^{109}$Cd ($\times 10^{-2}$) |
|-----------|-------------------------------|
| Crystal-1 | 4.6±3.0                       |
| Crystal-2 | 0.9±1.8                       |
| Crystal-3 | 7.5±0.9                       |
| Crystal-4 | 9.2±0.7                       |
| Crystal-6 | 1.5±0.7                       |
| Crystal-7 | 1.5±0.5                       |

**Table 6: Production rate $R_0$ (/kg/day) at sea level.**

|             | $^{22}$Na | $^{109}$Cd | $^{125}$I | $^{121}$mTe | $^{127}$mTe | $^{113}$Sn |
|-------------|-----------|------------|-----------|-------------|-------------|------------|
| Crystal-1   | 57.0±4.4  | 1.6±0.8    |           |             |             |            |
| Crystal-2   | 43.9±3.8  | 0.3±0.3    |           |             |             |            |
| Crystal-3   | 24.3±2.8  | 2.6±1.0    |           |             |             |            |
| Crystal-4   | 33.8±3.4  | 3.2±1.1    |           |             |             |            |
| Crystal-5   | 18.8±2.5  | 0.5±0.5    |           |             |             |            |
| Crystal-7   | 25.2±2.9  | 0.5±0.5    |           |             |             |            |
| ACTIVIA     | 66        | 4.8        | 226       | 93          | 9           |
| MENDL-2     | 4.8       | 208        | 102       |             |             |            |
| ANAIS measurement [26, 27] | 45.1±1.9 | 2.0±0.6    | 220±10    | 23.5±0.8    | 10.2±0.4    | 6.8±1.6    |
| DM-Ice17 measurement [25]     | 230       | < 9        | 25        |             |             | 16         |
Table 7: Estimated exposure time and initial activity ($A_0$) of $^3$H in the NaI(Tl) crystals.

| Crystal    | Exposure time [year] | $A_0$ [mBq/kg] |
|------------|----------------------|----------------|
| Crystal-1  | 2.1                  | 0.38±0.04      |
| Crystal-2  | 1.0                  | 0.20±0.04      |
| Crystal-3  | 1.3                  | 0.25±0.04      |
| Crystal-4  | 1.4                  | 0.26±0.04      |
| Crystal-6  | 0.6                  | 0.11±0.04      |
| Crystal-7  | 0.5                  | 0.09±0.04      |

Table 6 shows the production rate of cosmogenic isotopes in each NaI(Tl) crystal used for the COSINE-100 experiment compared with measurements from ANAIS [26, 27] and DM-Ice17 [25], and calculations using ACTIVIA and MENDL-2.

5. Tritium $^3$H, Iodine $^{129}I$, and Discussion

It is generally difficult to measure activity levels of long-lived cosmogenic isotopes, directly from the data due to their long half-lives. This is especially the case for $^3$H, which has no distinguishing $\gamma$-X-ray peak that can be exploited. Therefore, we simulated background spectra from $^3$H in the six NaI(Tl) crystals and used the extracted spectral shapes in the data fitting, while floating their unknown fractions [21]. In this way, we determine the initial activity $A_0$ of $^3$H from the global background fitting model [21], with results shown in Table 7. From these we computed exposures times $t$ from the relation,

$$A_0 = f \cdot R_0 \cdot (1 - e^{-\lambda t})$$

(12)

where we assumed that the production rate of $^3$H at sea level is $R_0 = (83±27)$ /kg/day, which was reported by ANAIS [28]. The resultant exposure times, listed in Table 7, are in good agreement with the time period during which they were being produced at Alpha Spectra and undergoing delivery to Y2L, as shown in Table 1(b).

The presence of cosmogenic $^{129}I$ was introduced by DAMA/LIBRA with the estimated concentration of $^{129}I/^{208}Tl = (1.7±0.1)\times10^{-13}$ [29]. It is used as a floating parameter in the global background fitting modeling for the COSINE-100 NaI(Tl) crystals, with resulting values of 1.01, 1.08, 0.75, 0.72, 0.91, and 0.94 mBq/kg for Crystal-1, 2, 3, 4, 6, and 7, respectively. These values agree well with the ANAIS result: 0.96±0.06 mBq/kg [30].

6. Conclusion

We have studied background contributions from cosmogenic isotopes activated by cosmic rays in the COSINE-100 detectors. To understand their time-dependent energy spectra we simulated responses to decays of the most abundantly produced cosmogenic isotopes in NaI(Tl) crystals and identified the energy regions where they make strong contributions to the crystals’ background spectra. Based on these simulation studies we measured decay rates of the cosmogenic isotopes using the time-dependent decrease of peaks from characteristic decays of these isotopes. We also exploited the correlations of characteristic $\gamma$/X-ray peaks in terms of time differences between sequential decays of $^{109}$Cd and double- and triple-coincidences for $^{22}$Na-decay-induced multi-gamma final states. From these measurements, we extrapolated the various isotopes’ activity levels to the times that they were first deployed underground at Y2L.

With these data we estimated production rates (at sea level) for the cosmogenic isotopes that are relevant for COSINE-100 and compared them with other experimental data and ACTIVIA/MENDL-2 calculations. As listed in Table 6, the results from different approaches are in reasonable agreement with each other. We extracted exposure times using initial $^3$H activities determined from the COSINE-100 global background fitting model and found results that are in good agreement with the times reported in Table 1(b). We also quantified the unknown $^{129}I$ activity level by including it as a free-floating parameter in the global background fit model and found consistency with an ANAIS result.

This study has given us a quantitative understanding of the cosmogenic isotopes in the NaI(Tl) crystals used for the COSINE-100 experiment. It provided important constraints on time-dependent backgrounds in our search for a time-dependent modulation that would be a characteristic signal for dark matter interactions [31, 32].

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