



Calibration HPGe detector using IAEA-U source for CBRNe

Debora Siqueira Nascimento^{1,2,a} , Andrea Chierici^{2,b}, Riccardo Ciolini^{2,c}, Stefano Chiappini^{1,d}, Francesco d'Errico^{2,e}, Massimo Chiappini^{1,f}

¹ National Institute of Geophysics and Volcanology, Via di Vigna Murata, 00143 Rome, Rome, Italy

² Department of Civil and Industrial Engineering, University of Pisa, L.go Lucio Lazzarino, 56122 Pisa, PI, Italy

Received: 31 March 2024 / Accepted: 1 July 2024

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Abstract Detecting and preventing the illicit movement of radioactive materials within a country is crucial, requiring the identification of radiologic anomalies against the normal radiation background. High-purity germanium (HPGe) detectors, known for their precision and sensitivity, have become popular for analyzing radioactive materials in CBRNe scenarios. This study focused on calibrating an HPGe detector for CBRNe applications, using gamma-ray spectra from an IAEA-U reference source. Energy calibration involved identifying peaks in the spectra and creating a calibration curve for energy and channel number data. Efficiency calibration, determined using the known activity of the source, revealed a linear relationship between energy and detector response. Over four years, systematic efficiency calibrations showed a deviation of only 3% well below the recommended limit of 5%. These findings underscore the reliability of the system as a reference spectrometry method for accurate detection of radioactive materials.

1 Introduction

Gamma-ray spectrometry is a widely utilized technique for assessing the presence of radionuclides in environmental samples. In the context of CBRNe (Chemical, Biological, Radiological, Nuclear, and explosive) scenarios, the accurate identification of radioactive materials is crucial for ensuring the safety and security of affected areas. Gamma spectroscopy plays a pivotal role in this mission, enabling the precise detection and quantification of radionuclides [1, 2]. Comprehensive energy and efficiency calibration are fundamental components of gamma spectroscopy, ensuring the accuracy needed for distinguishing between natural and anthropogenic sources [2].

Moreover, the capability of gamma spectrometry to provide real-time data and spatial mapping of radioactive contamination holds immense significance in environmental monitoring. Gamma spectroscopy is employed in various environmental monitoring approaches [3]. Spectrometers can be utilized in aircraft or motor vehicles for aerial and ground measurements, respectively [3]. Portable spectrometers are a practical choice for on-the-go applications, while borehole spectrometers cater to down-hole measurements. For analyzing rock and soil samples, laboratory spectrometers are highly reliable. Worldwide studies covering extensive areas have utilized ground and air gamma-ray measurements, and their findings are typically presented in gamma-ray dose rates or concentrations of radioelements [4–6]. To achieve accurate results, proper calibration of instruments is crucial. The International Atomic Energy Agency (IAEA) plays a pivotal role in establishing standards for the calibration of gamma-ray instruments and in preparing geological reference materials for calibration purposes.

In consideration of the importance of precise environmental mapping, we are currently preparing our high-purity germanium (HPGe) detector to serve as a reference instrument for field measurements. The initial step in this preparation involves comprehensive energy and efficiency calibrations. Energy calibration is vital to ensure the accurate identification of specific radionuclides, which is essential for precise environmental mapping. By accurately determining the energies of gamma rays emitted by radioactive sources, our detector will contribute to the creation of detailed and reliable environmental maps, allowing us to identify the distribution of radioactive materials with confidence. Efficiency calibration is equally critical, guaranteeing the correct quantification of radionuclide

Andrea Chierici, Riccardo Ciolini, Stefano Chiappini, Francesco d'Errico and Massimo Chiappini have contributed equally to this work.

^a e-mail: debora.siqueiranascimento@ingv.it (corresponding author)

^b e-mail: andrea.chierici@ing.unipi.it

^c e-mail: riccardo.ciolini@unipi.it

^d e-mail: stefano.chiappini@ingv.it

^e e-mail: francesco.derrico@unipi.it

^f e-mail: massimo.chiappini@ingv.it

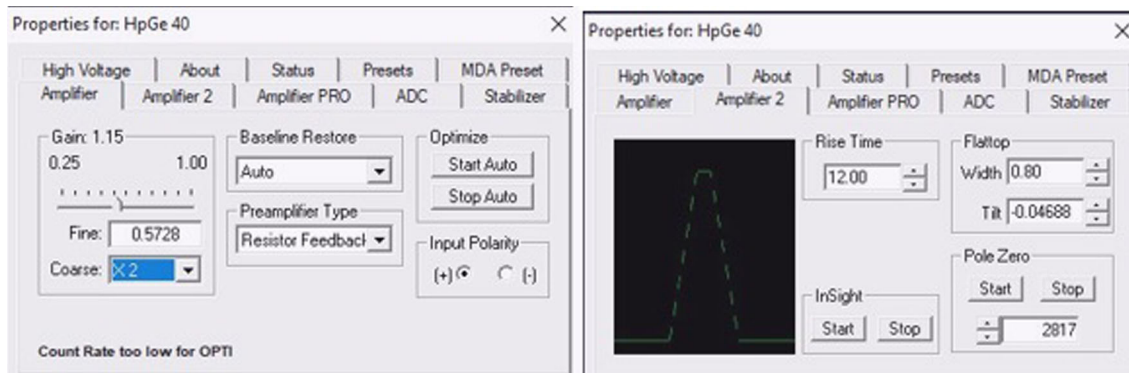


Fig. 1 Electronic setup used for the measurements

concentrations in the environment. This calibration process establishes a consistent relationship between the detector's response and the energy levels of incoming gamma radiation, enabling us to accurately measure the quantity of radioactive material present in the environment.

The efficiency curve is influenced by both the characteristics of the detection system and the shape of the sample. Theoretical determination can be achieved through Monte Carlo simulation or semi-empirically using analytical methods. In practical terms, experimental determination involves using standard samples that encompass a range of radionuclides with known activities, covering the energy range of interest.

Certified reference materials (CRMs) have been demonstrated to be a suitable source for the determination of the detection efficiency of HPGe [2, 7, 8]. Therefore, the aim of this work is to calibrate our detector HPGe using a CRMs (IAEA-U) source in energy and efficiency. In the future, enable us to better understand the spatial distribution of radioactive contaminants in the environment, supporting efforts to safeguard environmental integrity and public health.

2 Methodology

2.1 HPGe gamma-ray spectrometer

Gamma-ray spectrometry measurements were carried out using a coaxial p-type high-purity germanium (HPGe) detector, specifically the Model GEM40P4-76. The cryostat utilized in this research is the CFG-PV4 model. The crystal itself possesses a 64-mm diameter and extends to a length of 66.8 mm, demonstrating a measured relative efficiency of 49% at an energy level of 1.33 MeV. The system is meticulously shielded with a four-layer design. The initial layer comprises 1.8 mm of copper, followed by a 0.81-mm layer of tin. Subsequently, there is a shielding layer of 150 mm of lead, and lastly, an additional 0.8-mm layer of lead is added. This arrangement extends over a total length of 258 mm and serves to effectively minimize interference from laboratory background radiation. The amplifier, multi-channel analyzer and acquisition system is based on an Ortec DSPEC Jr 2.0 [9], configured as reported in Fig. 1.

2.1.1 Certified reference material

Certified reference material (CRM) is meticulously certified and supplied by the International Atomic Energy Agency (IAEA) through its Analytical Quality Control Services, serving as a trusted calibration source. These CRMs provide known concentrations of radioactivity measured in becquerel per kilogram (Bq/kg). In this study, we utilized the IAEA-U source [8] as our chosen CRM material, and its key characteristics are presented in Table 1 for reference.

The procedure for preparing IAEA source was as follows: First, the sample was weighed (100 g), and then the cap was sealed with plenty of silicone to minimize any gas exchange. After that, the samples were left for at least 21 days to achieve secular equilibrium of radon and its progeny.

2.1.2 Measurements and analyses of gamma-ray spectra

The investigation, spanning from 2020 to 2023, employed the Genie 2k software[®] for comprehensive data analysis, evaluating potential changes over a four-year timeframe. Initial efforts focused on establishing an extensive library of potential radionuclides present in the sample. Subsequently, peak identification was achieved based on energy levels and yields obtained from the Isotope Browser IAEA database application, with outcomes meticulously presented in Table 2, showcasing a high degree of consistency and coherence. The "interactive peak fit" tool played a pivotal role in peak analysis, facilitating the isolation of target peaks and providing vital information, including peak area, energy resolution, and associated uncertainties.

Table 1 IAEA-U activity

Analyte	Activity (Bq/kg)	Uncertainty
^{232}Th	4	–
^{235}U	228	± 2
^{238}U	4940	± 30
^{40}K	0.63	–
K	20	–
Th	1	–
U	400	± 2

Table 2 IAEA-U activity

Radionuclide	Energy (keV)	I_γ (%)	Activity (Bq/kg)
^{210}Pb	46.5	4.25	4940 ± 30
^{234}Th	63.3	3.7	4940 ± 30
^{235}U	143.8	10.96	228 ± 30
^{235}U	163.3	5.08	228 ± 30
$^{235}\text{U} + ^{226}\text{Ra}$	185.7 + 186.2	57 + 3.64	$(228 \pm 2) + (4940 \pm 30)$
^{214}Pb	242	7.26	4940 ± 30
^{214}Bi	295.2	18.47	4940 ± 30
^{214}Bi	609.31	45.44	4940 ± 30
^{214}Bi	768.4	4.89	4940 ± 30
^{214}Bi	934.1	3.09	4940 ± 30
^{214}Bi	1120.3	14.90	4940 ± 30
^{214}Bi	1238.1	5.83	4940 ± 30
^{214}Bi	1377.7	3.985	4940 ± 30
^{214}Bi	1764.5	15.29	4940 ± 30

Peak area determination involved a systematic process. Initially, the “calibrate” function loaded the energy calibration, selecting a pre-saved calibration file and marking the “energy/shape” square while unmarking the “efficiency” square. The calibrated energy file was then integrated into the program. Background subtraction followed, accomplished through “Options” – > “strip,” involving the identification of the background spectrum and adjustments for any discrepancies in live time between source and background measurements. Peak location was then determined using the “unidentified 2nd diff” function, and peak area calculations were performed using the “sum/nonlinear LSQ fit” function. Further isolation and analysis of the peak were executed through the “Analyze” – “Interactive peak fit” function, specifying energy, location, and generating detailed reports encompassing interactions, chi-square of the fitting, energy, channel, peak area, % error of the area, and full width at half maximum (FWHM).

2.1.3 Absolute efficiency calculations

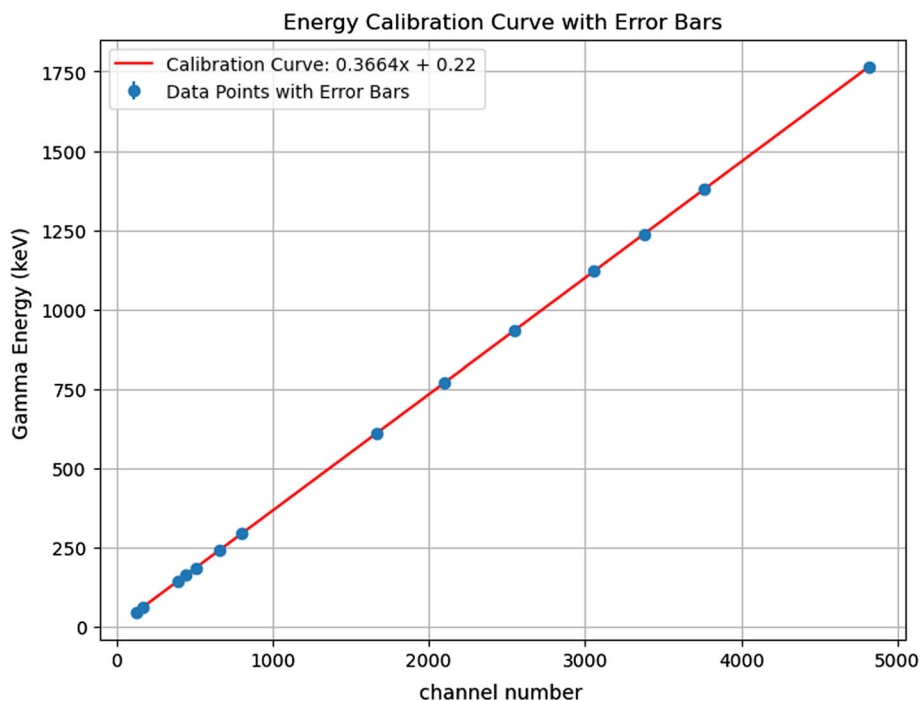
The photopeak efficiency was calculated following the formula:

$$\text{Eff} = \frac{A_{\text{peak}}}{t \times A_{\text{CRM}} \times I_\gamma \times m_{\text{CRM}}} \quad (1)$$

In the equation, A_{peak} represents the net area beneath the photopeak, t denotes the duration of the data acquisition, A_{CRM} denotes the certified activity concentration (expressed in Bq/kg) of the Certified Reference Material (CRM), I_γ denotes the probability of gamma-ray energy emission, and m_{CRM} represents the mass of the CRM (in kg). The values for activity concentration and mass of the radiation source were obtained from the manufacturer’s documentation, while the probabilities of gamma-ray energy emission were extracted from the Isotope Browser application provided by the International Atomic Energy Agency (IAEA).

A specific correction was applied to the 186 keV peak, where the total activity considered was derived from the combined contributions of the activity concentrations of ^{238}U (specifically ^{226}Ra) and ^{235}U . This combined consideration was necessary since ^{235}U exhibits a gamma peak at 185.7 keV, while ^{238}U (specifically ^{226}Ra) has a corresponding peak at 186.2 keV. Therefore, both activity contributions were factored into the analysis to accurately account for the total radiation activity at 186 keV.

Fig. 2 Energy calibration curve of IAEA-U source through 4 years with $R^2 = 1$



3 Results and discussion

3.1 Energy calibration

The initial calibration of our system involved absolute gamma energy in keV to identify unknown gamma-ray emitters. The association of energy with channels was established based on measurements taken from 2020 to 2023, and the averaged results are illustrated in Fig. 2. While good detectors typically exhibit a straight line, inherent nonlinearities in the detector or recording system may result in a less-than-perfectly linear system [10]. The energy calibration process entailed measuring gamma radiation within the range of 46.5 keV (^{210}Pb) to 1764.5 keV (^{214}Bi). This calibration yielded a well-fitted first-order equation, as shown in Fig. 2, accompanied by the equation and its corresponding error bars, although too low to discern, showcased in the same figure.

3.2 Efficiency calibration

With energy calibration done and known activity source, absolute efficiency of each photopeak can be calculated, and it is determined for the range from 46.5 to 1764.5 keV. The efficiency curve is depicted in Fig. 3, and all efficiency measurements indicate that the most suitable fit for the observed behavior is a second-degree polynomial, which is bifurcated into two segments. For low energies, ranging from 46.5 to 143 keV, the fit is expressed as Eq. 2. In contrast, for high energies, the fit is represented as Eq. 3. This crossover fitting was already cited in literature using another model of HPGe detector [8], and it was justified due to distinct attenuation and absorption processes involved. According to [8], the efficiency increases rapidly at low energies, and it has its maximum around 100 keV and then decreases afterward. The parameter values are detailed in Table 3. The fitting procedure was conducted using the Genie 2k software, inputting energy points, efficiency, and associated error values. The software then generates the plot based on these parameters.

$$\text{Eff}_{(46-143\text{ keV})} = P_1 E + P_2 E^2 \tag{2}$$

$$\text{Eff}_{(143-1776\text{ keV})} = P_3 E + P_4 E^2 \tag{3}$$

Figures 4 and 5 depict the efficiency results over the four-year measurement period. In Fig. 4, covering low energies from 46.5 to 143.8 keV, the efficiency exhibits an increasing trend with energy, while the absolute value remains constant through the years. Figure 5 presents a similar representation, focusing on high energies, demonstrating that akin to Fig. 4, the absolute efficiency value remains consistent across energy levels but diminishes with increasing energy. The observed behavior of efficiency with respect to energy aligns with expectations, displaying a rise with increasing energy and reaching its peak at 143.8 keV, subsequently declining to a constant level at high energies (Fig. 3).

Table 3 Parameters of the efficiency curve through 4 years of measurements

Energy (keV)	Eff. 2023	$U_{\text{eff.2023}}(\%)$	Eff. 2022	$U_{\text{eff.2022}}(\%)$	Eff. 2021	$U_{\text{eff.2021}}(\%)$	Eff. 2020	$U_{\text{eff.2020}}(\%)$
46.539	5.33E-05	0.48	5.53E-05	0.23	5.36E-05	0.55	5.33E-05	0.19
63.3	0.000247	0.43	0.000254	0.42	0.000241	0.44	0.000247	0.41
143.767	0.000737	0.40	0.000741	0.32	0.000745	0.63	0.000732	0.27
163.356	0.000515	0.88	0.000532	0.63	0.000535	1.44	0.000514	0.66
934.06	0.000137	0.33	0.000137	0.21	0.000136	0.30	0.000136	0.21
1120.29	0.000126	0.80	0.000125	0.80	0.000124	0.81	0.000124	0.80
1238.11	0.000117	0.15	0.000117	0.08	0.000116	0.16	0.000116	0.07

Fig. 3 Efficiency curve of HPGe using IAEA-U source measured at 24-01-23

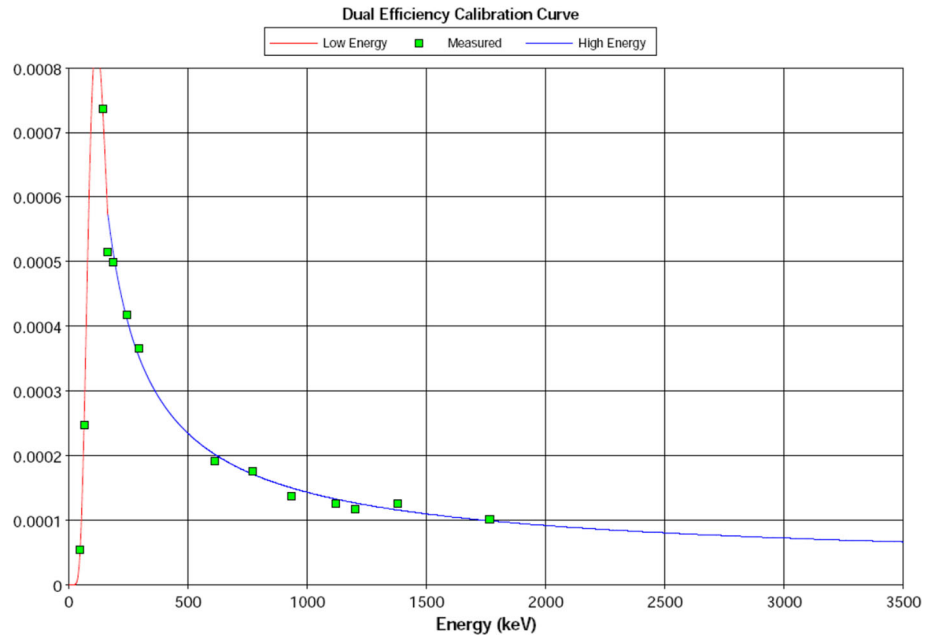


Fig. 4 Efficiency of low energies from 2020 to 2023 from IAEA-U gamma spectrum

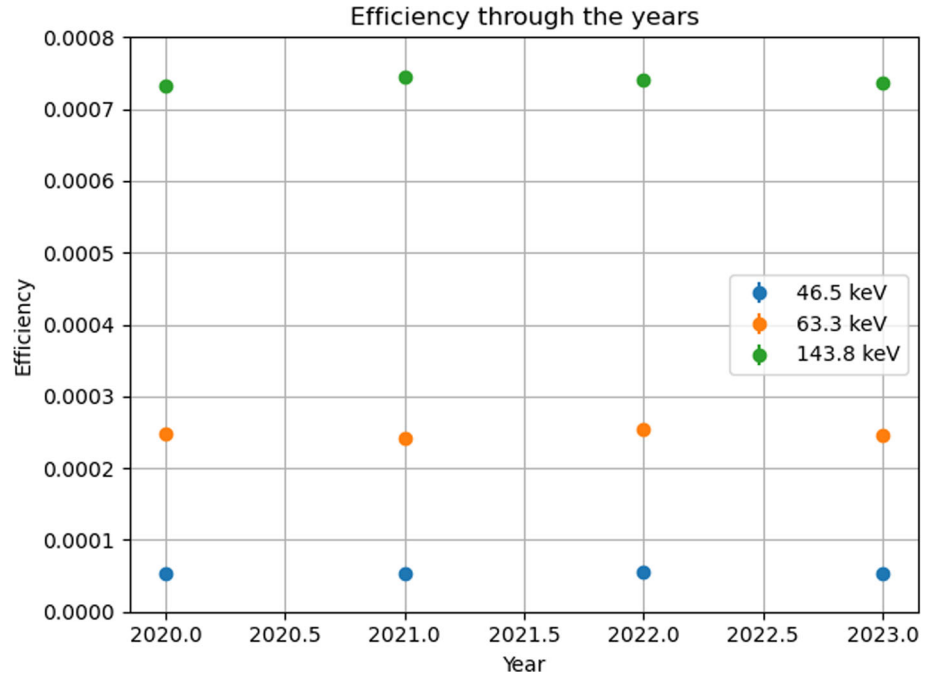
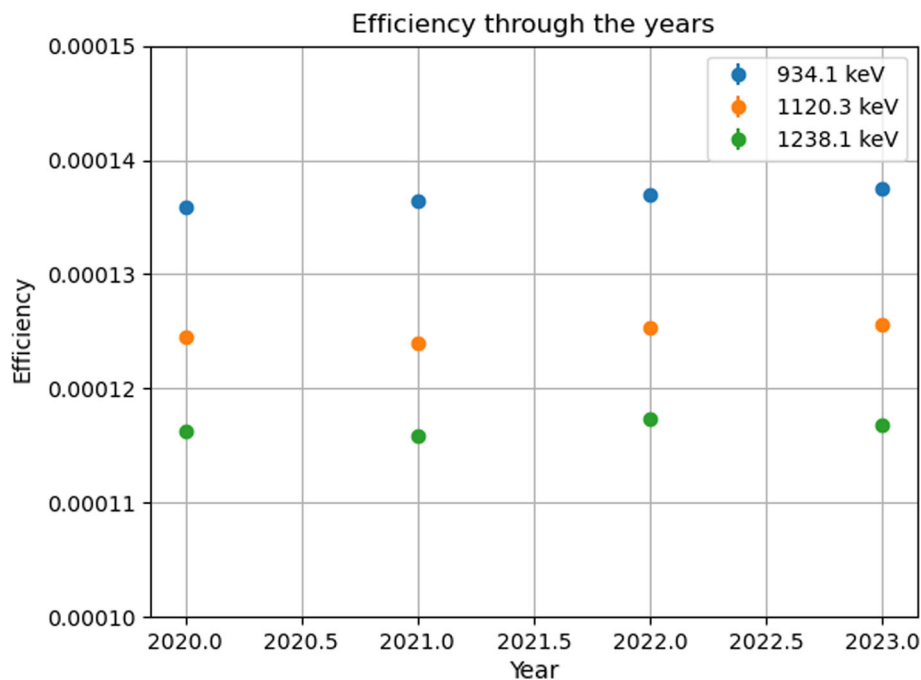


Fig. 5 Efficiency of high energies from 2020 to 2023 from IAEA-U gamma spectrum



4 Conclusion

The energy and efficiency calibration of the HPGe detector aimed to establish it as a reliable reference laboratory detector for future environmental monitoring applications. The energy calibration, spanning from 46.5 to 1764.5 keV, exhibited the anticipated linear response between energy and channel, demonstrating consistent behavior. Notably, the detector maintained a stable response over four years of measurements, with an error below 1% for all utilized energies.

Efficiency measurements indicated a fitting of two degrees for both energy ranges, aligning with the expected trends. Over the four-year period, the efficiency exhibited an error below 2%, confirming the robustness of the methodology outlined in this study. Ongoing measurements using an alternative source (IAEA-Th) are underway to validate the calibrations conducted with IAEA-U [8].

Author contributions All authors contributed to the study conception and design. Material collection was conducted by Stefano Chiappini. Analysis of data were performed by Debora Siqueira, who wrote the first draft of the manuscript, Stefano Chiappini and Riccardo Ciolini. All authors commented on previous versions of the manuscript and read and approved the final manuscript.

Funding Open access funding provided by Istituto Nazionale di Geofisica e Vulcanologia within the CRUI-CARE Agreement.

Data Availability Statement Data sets generated during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

Ethical approval Not applicable.

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