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Application of the bateman equation to analyze disequilibrium in the ^{232}Th and ^{238}U chains in low-background detector materials

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ABSTRACT

In rare-event search experiments such as AMoRE and COSINE, estimating background radioactivity levels and identifying background sources are crucial for background reduction. Typically, isotopes in the ^{238}U and ^{232}Th decay chains with relatively short half-lives are grouped together, and secular equilibrium is assumed during background measurements and estimations. During our material screening process, we observed that the ^{228}Ac–^{228}Th, and ^{238}U-^{226}Ra secular equilibrium was disrupted in several candidate materials. These measurements were conducted using a single HPGe detector at Y2L before 2023, and at Yemilab from 2024 onward.

In one notable case, an aluminum sample showed a significant increase in ^{226}Ra activity—from below the detection limit (22 mBq/kg) in 2021 to 893 ± 48 mBq/kg in 2025—despite the fact that there was no treatment or exposure that could have lead to radium contamination of the sample. Interestingly, the ^{234}Th activity remained nearly constant at around 60 Bq/kg, indicating that the ^{238}U parent activity did not change. Moreover, the ^{228}Th/^{228}Ac activity ratio decreased significantly over time. In 2021, the activity values were ^{228}Ac = 155 ± 19 mBq/kg and ^{228}Th = 4030 ± 206 mBq/kg, resulting in a ratio of approximately 27. By 2025, the measured values had shifted to ^{228}Ac = 421 ± 30 mBq/kg and ^{228}Th = 1273 ± 67 mBq/kg, corresponding to a reduced ratio of around 3.

This behavior suggests that prior radium purification influenced the relative amounts of isotopes in both the ^{238}U and ^{232}Th decay chains, resulting in a significant disruption of secular equilibrium.
A clear correlation was observed between the buildup of ^{226}Ra and the imbalance in the ^{228}Th/^{228}Ac activity ratio, highlighting the interconnected nature of these decay series.
These findings underscore the need for time-dependent modeling using the Bateman equation, which can account for such disequilibrium and provide accurate predictions of future activity levels.

By applying this method, we aim to predict the time-dependent activity of long-lived isotopes and enhance the accuracy of background level estimation in upcoming rare-event search experiments, including AMoRE-II and COSINE.

Collaboration you are representing

AMoRE, COSINE

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