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Constraint on anthropogenic ^{81}Kr in the atmosphere

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Paleo-groundwater serves as a natural archive of geological and climatic changes, preserving valuable information in its chemical and isotopic signatures. To unlock the potential of this archive, the long-lived radionuclide ^{81}Kr (half-life 229 kyr, isotopic abundance 9.3×10^{-13}) has become an ideal tracer for dating groundwater over timescales of 10^4 to 10^6 years. For this application, Atom Trap Trace Analysis (ATTA) has emerged as the primary technique for measuring ^{81}Kr in environmental samples. With ATTA now achieving analytical precision of 1%, it has become necessary to assess the potential impact of anthropogenic sources on atmospheric ^{81}Kr levels and their implications for ^{81}Kr dating. While theoretical estimates suggest that contributions from nuclear activities in the atmosphere are at the 0.15% level, these calculations carry substantial uncertainties and have not yet been tightly constrained by experimental data.

Here, we report a high-precision measurement of the anthropogenic ^{81}Kr contribution using ATTA. We analyzed a pre-nuclear atmospheric krypton sample collected in the 1940s alongside a modern atmospheric sample, acquiring over one million ^{81}Kr atom counts for each. Based on the comparison between these samples, we obtain an upper limit of 0.43% (90% confidence level) for the anthropogenic contribution, thereby confirming the stability of the atmospheric ^{81}Kr baseline for high-precision dating applications.