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Temporal changes of $^{235}U/^{238}U$ and $^{236}U/^{238}U$ isotopic ratios in Tokyo Bay from the 1960s to the 2000s

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Uranium in nature consists mainly of three radionuclides, which are a radiogenic isotope ^{234}U and two primordial nuclides ^{235}U and ^{238}U. Among the three, ^{235}U is a fissile isotope that has been used in nuclear bombs and nuclear reactors. When uranium is used for these purposes, ^{236}U is produced by both the ^{235}U (n, γ) ^{236}U reaction and the ^{238}U (n, 3n) ^{236}U reaction. Because the amount of anthropogenic ^{236}U is significantly larger than that of natural ^{236}U, $^{236}U/^{238}U$ has been used as an environmental tracer to investigate the contamination of anthropogenic uranium released from nuclear weapons tests, fuel reprocessing, nuclear power plant accidents, and the decommissioning of nuclear power plants. In this study, we examined the temporal changes of $^{235}U/^{238}U$ and $^{236}U/^{238}U$ in sediment samples collected in Tokyo Bay and elucidated the anthropogenic sources of uranium in the 1960s–2000s. Uranium-236 was detected in samples deposited in the 1960s-2000s, and the $^{236}U/^{238}U$ ratio of the sediment core shows peak values in the 1970s. The $^{235}U/^{238}U$ isotopic ratios in samples deposited in the early 1960s are almost identical to that of natural uranium, implying that the ^{236}U might have originated from global fallout. A decrease in $^{235}U/^{238}U$ was observed in the late 1960s–2000s, suggesting that depleted uranium from nuclear fuel reprocessing increased the $^{236}U/^{238}U$ ratios in the sediment. The $^{236}U/^{238}U$ values in sediments from the 1980s–2000s were lower than those in the 1970s but considerably higher than those in the 1960s, suggesting that the main source of depleted uranium still remain around Tokyo Bay. Our results demonstrated that the depleted uranium released in the 1970s should be considered as an important end-member when using uranium isotopic ratios as environmental tracers in closed aquatic environments around industrial cities.